The DMFT object of the CP-PAW code

Peter E. Blöchl

Contents

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. The derivations here follow closely the paper of Blöchl, Pruschke and Potthoff[1], in the following referred to **BPP**.

1.1   Grand potential and density-matrix functional as starting point

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

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

where the **density-matrix functional** $\tilde{F}_{\beta}^{\hat{W}}$ is expressed[1] by the **Luttinger-Ward functional** [2] $\Phi^{LW}[\boldsymbol{G}, \hat{W}]$ as

$$
\tilde{F}_{\beta}^{\hat{W}}[\boldsymbol{\rho}] = \frac{1}{\beta} \operatorname{Tr} \Big[\boldsymbol{\rho} \ln(\boldsymbol{\rho}) + (\mathbf{1} - \boldsymbol{\rho}) \ln(\mathbf{1} - \boldsymbol{\rho}) \Big]
$$
$$
+ \operatorname*{stat}_{\boldsymbol{h}'} \operatorname*{stat}_{\boldsymbol{G}, \boldsymbol{\Sigma}} \left\{ \Phi_{\beta}^{LW}[\boldsymbol{G}, \hat{W}] - \frac{1}{\beta} \sum_{\nu} \operatorname{Tr} \left\{ \ln \Big[\mathbf{1} - \big(i\hbar\omega_{\nu} + \mu \big) \mathbf{1} - \bar{\boldsymbol{h}} \big)^{-1} \big(\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_{\nu}) - \bar{\boldsymbol{h}} \big) \Big] \right. \right.
$$
$$
\left. \left. + (\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_{\nu}) - \bar{\boldsymbol{h}}) \boldsymbol{G}(i\omega_{\nu}) - \Big[\boldsymbol{G}(i\omega_{\nu}) - \big(i\hbar\omega_{\nu} + \mu \big) \mathbf{1} - \bar{\boldsymbol{h}} \big)^{-1} \Big] \big(\boldsymbol{h}' - \bar{\boldsymbol{h}} \big) \right\} \right\} \tag{1.2}
$$

Here, $|\psi_n\rangle$ are one-particle wave functions. They play the role of **natural orbitals**, the eigenstates of the **one-particle density matrix $\boldsymbol{\rho}$**. f_n are the occupations, the eigenvalues of the one-particle density matrix. The orthonormality of the natural orbitals is enforced with the method of Lagrange multipliers. The Lagrange multipliers are Λ. 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 $\hat{W}$.

With $\operatorname{stat}_x Y$, we denote the stationary condition, which requires that the partial derivatives of Y with respect to the variable x is obeyed. It explicitely does not require the stationary point to be an extremum.

$\boldsymbol{G}(i\omega)$ is the Green's function and $\boldsymbol{\Sigma}(i\omega_\nu)$ is the self energy. The Matsubara sum runs over the **Matsubara frequencies** (see appendix A on p. 29)

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

$\boldsymbol{h}'$ are the Lagrange multipliers for the density-matrix constraint.

$$\boldsymbol{\rho} = \frac{1}{\beta}\sum_\nu e^{i\beta\hbar\omega_\nu 0^+}\boldsymbol{G}(i\omega_\nu) \tag{1.4}$$

The Hamiltonian $\bar{\boldsymbol{h}}$ is a non-local Hamiltonian directly related to the one-particle density matrix via (see Eq. BPP24)

$$\bar{\boldsymbol{h}} = \mu\boldsymbol{1} + k_B T \ln\left[\frac{\boldsymbol{1} - \boldsymbol{\rho}}{\boldsymbol{\rho}}\right] \tag{1.5}$$

In practice, we perform calculations at fixed particle number. Therefore, we calculate the **Helmholtz potential**

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

where $Q_\beta^{\hat{W}}$ 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.

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

1.2   Separate out static correlations: Towards DFT+

In appendix C, it is shown that the correlation contribution $Q_\beta^{\hat{W}}$ can be divided into one part having a frequency-dependent self energy and several contributions having a frequency-independent self energy. The Luttinger-Ward functional with frequency-independent self energy can be expressed by the density matrix alone, that is the Green's function enters always in the form of the integral

$$\frac{1}{\beta}\sum_\nu \boldsymbol{G}(i\omega_\nu)e^{i\hbar\omega\beta 0^+} \tag{1.8}$$

This notion allows us to split the interaction contribution $Q^{\hat{W}}$ into different parts as

$$Q^{\hat{W}}[\boldsymbol{\rho}] \approx Q^{DFT,\hat{W}}[\boldsymbol{\rho}] - Q^{DFT,\hat{W}_1}[\boldsymbol{\rho}] + Q^{HF,\hat{W}_1}[\boldsymbol{\rho}] - Q^{HF,\hat{W}_2}[\boldsymbol{\rho}] + Q^{\hat{W}_2}[\boldsymbol{\rho}] \qquad (1.9)$$

of which only the last term has a frequency dependent self energy.

In order to keep the computational effort at a reasonable level, we treat the full interaction on the most simple level of the description, namely DFT, while the most complex many-particle treatment is limited to the most approximate interaction, namely $\hat{W}_2$.

With[1]

$$Q^{DFT,\hat{W}}[\hat{\rho}] = \frac{1}{2} \int d^3r \int d^3r' \frac{e^2 n(\vec{r}) n(\vec{r'})}{4\pi\epsilon_0 |\vec{r} - \vec{r'}|} + E_{xc}[n] \;, \qquad (1.10)$$

and a screened Hartree-Fock contribution $Q^{HF,\hat{W}}[\hat{\rho}]$ described later, we obtain

---

HELMHOLTZ POTENTIAL

$$A_{\beta,N}[\hat{h} + \hat{W}] = \min_{|\psi_n\rangle, f_n \in [0,1]} \operatorname*{stat}_{\mu,\Lambda} \left\{ \sum_n f_n \langle \psi_n | \frac{\hat{\vec{p}}^2}{2m_e} | \psi_n \rangle + \int d^3r \, n(\vec{r}) v_{ext}(\vec{r'}) \right.$$

$$+ \frac{1}{2} \int d^3r \int d^3r' \frac{e^2 n(\vec{r}) n(\vec{r'})}{4\pi\epsilon_0 |\vec{r} - \vec{r'}|} + E_{xc}[n] + \frac{1}{\beta} \sum_n \Big[f_n \ln(f_n) + (1 - f_n) \ln(1 - f_n) \Big]$$

$$+ \underbrace{Q^{HF,\hat{W}_1}_{\beta}[\boldsymbol{\rho}] - Q^{DFT,\hat{W}_1}_{\beta}[\boldsymbol{\rho}, n]}_{\text{local hybrid functional}} \quad + \quad \underbrace{Q^{\hat{W}_2}_{\beta}[\boldsymbol{\rho}] - Q^{HF,\hat{W}_2}_{\beta}[\boldsymbol{\rho}]}_{Q^{dyn,\hat{W}_2}_{\beta}[\boldsymbol{\rho}]}$$

$$\left. - \mu \Big[\sum_n f_n - N \Big] - \sum_{m,n} \Lambda_{m,n} \Big(\langle \psi_n | \psi_m \rangle - \delta_{m,n} \Big) \right\} \qquad (1.11)$$

where

$$n(\vec{r}, \sigma, \sigma') = \sum_n \langle \vec{r}, \sigma | \psi_n \rangle f_n \langle \psi | \vec{r}, \sigma' \rangle$$

$$n(\vec{r}) = \sum_\sigma n(\vec{r}, \sigma, \sigma)$$

$$\rho_{a,b} = \sum_n \langle \pi_a | \psi_n \rangle f_n \langle \psi | \pi_b \rangle \qquad (1.12)$$

We distinguish two different corrections:

1. a screened Hartree-Fock correction with an interaction $\hat{W}_1$.

   The Hartree-Fock term $Q^{HF,\hat{W}}_{\beta}$ is equal to $Q^{\hat{W}}_{\beta}$ when only the first-order term of the Luttinger-Ward functional in the interaction is considered. It is obtained as (BPP-Eq.43)

$$Q^{HF,\hat{W}}_{\beta}[\boldsymbol{\rho}] = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \Big[\rho_{d,a} \rho_{c,b} - \rho_{c,a} \rho_{d,b} \Big] \qquad (1.13)$$

---

[1]A proper description should replace E_{xc} with U_{xc}, the interaction part at full interaction strength. E_{xc} contains the difference of the kinetic energy of the fully interacting and the non-interacting electron gas at the same density. Because the density matrix functional works with the full interaction, this contribution must be removed.

(See also appendix D on p.40.)

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 evaluated in the `paw_lmto`-object.

2. a DFT double counting term $Q_\beta^{DFT,\hat{W}_1}$: It mimicks the DFT contribution when the interactiopn is limited to $\hat{W}_1$. The basic ideas behind the double counting correction has been described in a previous paper[4].

3. a dynamic-correlation correction

$$Q_\beta^{dyn,\hat{W}_2} \overset{\text{def}}{=} Q_\beta^{\hat{W}_2} - Q_\beta^{HF,\hat{W}_2} \qquad (1.14)$$

contains an interaction term calculated on the highest level of theory such as a many-body calculation. The corresponding Hartree-Fock term is immediately substracted as double counting correction.

The dynamic-correlation correction uses an interaction $\hat{W}_2$. In the spirit of the local approximation, the interaction of this term is limited to on-site terms only. The evaluation of the dynamic correlations is the only contribution that requires expensive many-particle calculations.

This term is further elaborated in section 1.3

Stationary conditions for wave functions and occupations

Let us now explore the stationary conditions in Eq. 1.11: We use the representation of the wave functions $|\psi_n\rangle = \sum_a |\chi_a\rangle\langle\pi_a|\psi_n\rangle$ in terms of local orbitals $|\chi_a\rangle$.

$$\left[\frac{\hat{\tilde{p}}^2}{2m_e} + v_{ext} + v_{Hartree} + v_{xc} + \sum_{a,b}|\pi_a\rangle\left(\frac{\delta Q_{HF,\beta}^{\hat{W}_1}}{\delta\rho_{a,b}} - \frac{\delta Q_{DFT,\beta}^{\hat{W}_1}}{\delta\rho_{a,b}} + \frac{\delta Q_{dyn,\beta}^{\hat{W}_2}}{\delta\rho_{a,b}}\right)\langle\pi_b|\right]|\psi_n\rangle$$

$$= \sum_m |\psi_m\rangle\Lambda_{m,n}\frac{1}{f_n} \qquad (1.15)$$

$$\langle\psi_n|\left[\frac{\hat{\tilde{p}}^2}{2m_e} + v_{ext} + v_{Hartree} + v_{xc} + \sum_{a,b}|\pi_a\rangle\left(\frac{\delta Q_{HF,\beta}^{\hat{W}_1}}{\delta\rho_{a,b}} - \frac{\delta Q_{DFT,\beta}^{\hat{W}_1}}{\delta\rho_{a,b}} + \frac{\delta Q_{dyn,\beta}^{\hat{W}_2}}{\delta\rho_{a,b}}\right)\langle\pi_b|\right]|\psi_n\rangle$$

$$- \mu + \frac{1}{\beta}\ln\left[\frac{f_n}{1-f_n}\right] = 0 \qquad (1.16)$$

These equations specify the optimum density matrix as

EQUILIBRIUM DENSITY MATRIX

$$\hat{\rho} = \left[\hat{1} + e^{\beta(\hat{h}_{eff} - \mu)} \right]^{-1} \tag{1.17}$$

with the effective Hamiltonian

$$\hat{h}_{eff} = \frac{\hat{\bar{p}}^2}{2m_e} + v_{ext} + v_{Hartree} + v_{xc} + \sum_{a,b} |\pi_a\rangle \left(\frac{\delta Q_{HF,\beta}^{\hat{W}_1}}{\delta \rho_{a,b}} - \frac{\delta Q_{DFT,\beta}^{\hat{W}_1}}{\delta \rho_{a,b}} + \frac{\delta Q_{dyn,\beta}^{\hat{W}_2}}{\delta \rho_{a,b}} \right) \langle \pi_b | \tag{1.18}$$

Note, that, in equilibrium, $\hat{h}_{eff} = \hat{h}_\rho$.

Eq. 1.17 can be veryfied from the stationary conditions by going into a basis of eigenstates of $\hat{h}_{eff}$.

1.3 Dynamic correlation correction

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

DYNAMIC CORRELATION CORRECTION

$$Q_{dyn,\beta}^{\hat{W}_2}[\rho] = \tilde{F}_\beta^{\hat{W}_2}[\rho] - \frac{1}{\beta} \mathrm{Tr}\Big[\rho \ln(\rho) + (1 - \rho)\ln(1 - \rho) \Big] - \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c}^{\hat{W}_2} \Big[\rho_{d,a}\rho_{c,b} - \rho_{c,a}\rho_{d,b} \Big]$$

$$= \mathop{\mathrm{stat}}_{h'_{dyn}} \mathop{\mathrm{stat}}_{G,\Sigma_{dyn}} \Bigg\{ \Phi_\beta^{LW}[G, \hat{W}_2] - \Phi_\beta^{HF,LW}[G, \hat{W}_2]$$

$$- \frac{1}{\beta} \sum_\nu \mathrm{Tr}\Big\{ \ln\Big[1 - \big((i\hbar\omega_\nu + \mu)1 - \bar{h} \big)^{-1} \big(h'_{dyn} + \Sigma_{dyn}(i\omega_\nu) - \bar{h} \big) \Big]$$

$$+ (h'_{dyn} + \Sigma_{dyn}(i\omega_\nu) - \bar{h})G(i\omega_\nu) - \Big[G(i\omega_\nu) - \big(i\hbar\omega_\nu + \mu)1 - \bar{h} \big)^{-1} \Big]\big(h'_{dyn} - \bar{h} \big) \Big\} \Bigg\} \tag{1.19}$$

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.

According to appendix C, the Hartree-Fock contribution can also be moved out of the variational part, where it can be calculated directly from the density matrix. In that case, however, the values of h'_{dyn} and Σ_{dyn} have different values than if they are kept inside the variational part. It is beneficial to keep it inside the variational principle, because the values of the self energy and the Lagrange multipliers will be smaller.

Derivative of the dynamical correlation

Let us form the derivative of $Q_{\mathrm{dyn},\beta}^{\hat{W}_2}[\boldsymbol{\rho}]$ in all detail to avoid using inappropriate assumptions.

During the derivation we use the rule for derivatives of an inverse of a matrix[2] $d(\bar{\boldsymbol{G}}^{-1}) = +\bar{\boldsymbol{G}}\,d\bar{\boldsymbol{h}}\,\bar{\boldsymbol{G}}$.

We form the first variation with respect to the density matrix, keeping $\boldsymbol{h}'_{dyn}$, $\boldsymbol{\Sigma}_{dyn}$ and $\boldsymbol{G}$ fixed.

$$
dQ_\beta^{dyn,\hat{W}_2} = -\frac{1}{\beta}\sum_\nu \mathrm{Tr}\Bigg\{ \Big[\mathbf{1} - \bar{\boldsymbol{G}}\Big(\boldsymbol{h}'_{dyn} + \boldsymbol{\Sigma}_{dyn} - \bar{\boldsymbol{h}}\Big)\Big]^{-1} \underbrace{\Big[-\bar{\boldsymbol{G}}\,d\bar{\boldsymbol{h}}\,\bar{\boldsymbol{G}}\Big(\boldsymbol{h}'_{dyn} + \boldsymbol{\Sigma}_{dyn} - \bar{\boldsymbol{h}}\Big) + \bar{\boldsymbol{G}}\,d\bar{\boldsymbol{h}}\Big]}_{\bar{\boldsymbol{G}}\,d\bar{\boldsymbol{h}}\Big[\mathbf{1}-\bar{\boldsymbol{G}}\Big(\boldsymbol{h}'_{dyn}+\boldsymbol{\Sigma}_{dyn}-\bar{\boldsymbol{h}}\Big)\Big]}
$$

$$
- \underbrace{d\bar{\boldsymbol{h}}\boldsymbol{G}}_{(A)} - \Big[\bar{\boldsymbol{G}}\,d\bar{\boldsymbol{h}}\,\bar{\boldsymbol{G}}\Big(\boldsymbol{h}'_{dyn} - \bar{\boldsymbol{h}}\Big) - \Big(\underbrace{\boldsymbol{G}}_{(A)} - \bar{\boldsymbol{G}}\Big)d\bar{\boldsymbol{h}}\Big]\Bigg\}
$$

$$
= -\frac{1}{\beta}\sum_\nu \mathrm{Tr}\Bigg\{ \underbrace{\bar{\boldsymbol{G}}\,d\bar{\boldsymbol{h}}}_{(B)} - \Big[-\bar{\boldsymbol{G}}\,d\bar{\boldsymbol{h}}\,\bar{\boldsymbol{G}}\Big(\boldsymbol{h}'_{dyn} - \bar{\boldsymbol{h}}\Big) + \underbrace{\bar{\boldsymbol{G}}\,d\bar{\boldsymbol{h}}}_{(B)}\Big]\Bigg\}
$$

$$
= -\mathrm{Tr}\Bigg\{ \Big(\boldsymbol{h}'_{dyn} - \bar{\boldsymbol{h}}\Big) \underbrace{\frac{1}{\beta}\sum_\nu d\bar{\boldsymbol{G}}}_{d\rho} \Bigg\} \tag{1.21}
$$

Thus, we obtain the derivative with respect to the density matrix as

$$
\frac{\delta Q_\beta^{dyn,\hat{W}_2}}{\delta\boldsymbol{\rho}} = -\Big(\boldsymbol{h}'_{dyn} - \bar{\boldsymbol{h}}\Big) \stackrel{\mathrm{Eq.}\ 1.5}{=} \mu\mathbf{1} + k_B T\ln\Big[\frac{\mathbf{1}-\boldsymbol{\rho}}{\boldsymbol{\rho}}\Big] - \boldsymbol{h}'_{dyn} \tag{1.22}
$$

which is the desired result.

The stationary condition with respect to $\boldsymbol{G}$ is

$$
\frac{\beta\delta\Phi^{LW}}{\delta\boldsymbol{G}} - \frac{\beta\delta\Phi^{HF,LW}}{\delta\boldsymbol{G}} - \boldsymbol{\Sigma}_{dyn} = 0 \qquad \Rightarrow \qquad \boldsymbol{\Sigma}_{dyn} = \frac{\beta\delta\Phi^{LW}}{\delta\boldsymbol{G}} - \frac{\beta\delta\Phi^{HF,LW}}{\delta\boldsymbol{G}} \tag{1.23}
$$

and the one with respect to $\boldsymbol{\Sigma}_{dyn}$ is

$$
\Big[\mathbf{1} - \bar{\boldsymbol{G}}\Big(\boldsymbol{h}'_{dyn} + \boldsymbol{\Sigma}_{dyn} - \bar{\boldsymbol{h}}\Big)\Big]^{-1}\Big(-\bar{\boldsymbol{G}}\Big) + \boldsymbol{G} = 0
$$

$$
\Rightarrow \qquad \boldsymbol{G} = \Big[\bar{\boldsymbol{G}}^{-1} - \Big(\boldsymbol{h}'_{dyn} + \boldsymbol{\Sigma}_{dyn} - \bar{\boldsymbol{h}}\Big)\Big]^{-1}
$$

$$
= \Big[(i\hbar\omega_\nu + \mu)\mathbf{1} - \bar{\boldsymbol{h}} - \Big(\boldsymbol{h}'_{dyn} - \bar{\boldsymbol{h}} + \boldsymbol{\Sigma}_{dyn}\Big)\Big]^{-1} \tag{1.24}
$$

---

[2]

$$
\boldsymbol{A}\boldsymbol{A}^{-1} = \mathbf{1} \quad \Rightarrow \quad \Big(d\boldsymbol{A}\Big)\boldsymbol{A}^{-1} + \boldsymbol{A}\Big(d\boldsymbol{A}^{-1}\Big) = 0 \quad \Rightarrow \quad d\boldsymbol{A}^{-1}\Big) = -\boldsymbol{A}^{-1}\Big(d\boldsymbol{A}\Big)\boldsymbol{A}^{-1} \tag{1.20}
$$

With the new symbol

$$\mathbf{\Gamma} \overset{\text{def}}{=} \bar{\boldsymbol{h}} - \boldsymbol{h}'_{dyn} \tag{1.25}$$

the derivative of the correlation contribution is

$$\frac{\delta Q_{\beta}^{dyn,\hat{W}_2}}{\delta \boldsymbol{\rho}} = \mathbf{\Gamma} \tag{1.26}$$

and the Greens function has the form

$$\boldsymbol{G}(i\omega_\nu) = \left[(i\hbar\omega_\nu + \mu)\mathbf{1} - \bar{\boldsymbol{h}} + \mathbf{\Gamma} - \boldsymbol{\Sigma}_{dyn}(i\omega_\nu) \right]^{-1} \tag{1.27}$$

The full self energy is

$$\Sigma^{\hat{W}_2} = \Sigma^{HF,\hat{W}_2} + \Sigma^{dyn,\hat{W}_2} \tag{1.28}$$

Comparing with the Green's function

$$\boldsymbol{G} = \left[(i\hbar\omega_\nu + \mu)\mathbf{1} - \bar{\boldsymbol{h}} - \left(\boldsymbol{h}' - \bar{\boldsymbol{h}} + \boldsymbol{\Sigma} \right) \right]^{-1} \tag{1.29}$$

we can make the identification

$$\boldsymbol{h}' + \boldsymbol{\Sigma} = \boldsymbol{h}'_{dyn} + \underbrace{\boldsymbol{\Sigma} - \boldsymbol{\Sigma}^{HF}}_{\boldsymbol{\Sigma}_{dyn}}$$

$$\boldsymbol{h}' = \boldsymbol{h}'_{dyn} - \boldsymbol{\Sigma}^{HF} \tag{1.30}$$

The matrix $\mathbf{\Gamma}$ replaces $\boldsymbol{h}'$ as Lagrange parameter for the density matrix constraint. It vanishes in the Hartree-Fock approximation as seen from BPP-Eq.45-46[1]. Instead of the Hartree-Fock self energy $\boldsymbol{\Sigma}^{HF}$, we could have taken any other static self energy.

One may ask, where the self energy contribution from static correlations is, because both, $\boldsymbol{\Sigma}$ and $\boldsymbol{\Sigma}^{HF}$ mentioned here refer to the second interaction using $\hat{W}_2$. The Terms related to $\hat{W}_1$ are contained in the density matrix and $\bar{\boldsymbol{h}}$.

1.4   Solver for dynamic correlations

1.4.1   Loop for the dynamic correlation contribution

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

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

$$\bar{\boldsymbol{h}} = \mu\mathbf{1} + k_B T \ln \left[\frac{\mathbf{1} - \boldsymbol{\rho}}{\boldsymbol{\rho}} \right] \tag{1.31}$$

This is done in `DMFT_HRHO`.

2. Construct the Greens function $\bar{G}$, which is the non-interacting Green's function with the correct density matrix.

$$\bar{G}(i\omega_\nu) = \left[(i\hbar\omega_\nu + \mu)\mathbf{1} - \bar{h} \right]^{-1} \tag{1.32}$$

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. The solver only contributes the dynamic correlations.

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.

$$\Phi^{LW}[G, \hat{W}_2] - \Phi^{LW,HF}[G, \hat{W}_2] \tag{1.33}$$

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

$$\Sigma_{a,b}^{dyn}(i\omega_\nu) = \Sigma_{a,b}(i\omega_\nu) - \Sigma_{a,b}^{HF}(i\omega_\nu) = \frac{\beta\delta\Phi^{LW}[G,\hat{W}_2]}{\delta G_{b,a}(i\omega_\nu)} - \frac{\beta\delta\Phi^{LW,HF}[G,\hat{W}_2]}{\delta G_{b,a}(i\omega_\nu)} \tag{1.34}$$

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.

$$N_{a,b,c,d}^{dyn} = \frac{\beta\delta\Phi^{LW}[G,\hat{W}_2]}{\delta U_{a,b,c,d}} - \frac{\beta\delta\Phi^{LW,HF}[G,\hat{W}_2]}{\delta U_{a,b,c,d}} \tag{1.35}$$

---

5. convert the self energy and the derivative of the U-tensor back to the non-orthonormal set of local orbitals.

6. Mix the self energy: The self energy cannot be simply inserted into the Green's function for the next iteration, because the changes for small Matsubara frequencies would be too violent. Therefore it needs some preconditioning.

   The underlying ideas related to the mixing are described in appendix

$$\Sigma_{a,b}^{dyn}(i\omega_\nu) = \Sigma_{a,b}^{dyn}(i\omega_\nu) + \frac{1}{C}\left(\frac{C}{M} + (\hbar\omega)^2\right)\left(\Sigma_{a,b}^{dyn}(i\omega_\nu) - \frac{\beta\delta\Phi_{dyn}^{LW}}{\partial G_{b,a}(i\omega_\nu)}\right) \tag{1.36}$$

$\mathbf{\Sigma}^{dyn}$ is the self energy that has been accumulated, and that is zero in the first iteration. $\frac{\beta\delta\Phi_{dyn}^{LW}}{\partial G_{b,a}(i\omega_\nu)}$ is the dynamic self-energy contribution produced by the solver. Note that this

self energy does not contain the Hartree-Fock term. C and M are two mixing parameters, that control the behavior at small (M) and large (C) Matsubara frequencies.

7. The new Green's function $\boldsymbol{G}^{new}$ has the form

$$\boldsymbol{G}^{new}(i\omega_\nu) = \left[(i\hbar\omega_\nu + \mu)\mathbf{1} - \bar{\boldsymbol{h}} + \boldsymbol{\Gamma} - \boldsymbol{\Sigma}^{dyn}(i\omega_\nu) \right]^{-1} \tag{1.37}$$

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

$$\boldsymbol{\rho} = \frac{1}{\beta} \sum_\nu e^{i\beta\hbar\omega_\nu 0^+} \boldsymbol{G}^{new}(i\omega_\nu) \tag{1.38}$$

is fulfilled.

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

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

$$0 = \left[\frac{1}{\beta} \sum_\nu e^{i\beta\hbar\omega_\nu 0^+} G^{new}_{a,b}(i\omega_\nu) \right] - \rho_{a,b} - \sum_{c,d} \left[\frac{1}{\beta} \sum_\nu G^{new}_{a,c}(i\omega_\nu)G^{new}_{d,b}(i\omega_\nu) \right] \delta\Gamma_{c,d}$$

$$\delta\Gamma_{c,d} = \underbrace{\left[\frac{1}{\beta} \sum_\nu G^{new}_{a,c}(i\omega_\nu)G^{new}_{d,b}(i\omega_\nu) \right]^{-1}_{cd,ab}}_{\rightsquigarrow \quad \alpha\delta_{c,a}\delta_{d,b}} \left(\underbrace{\left[\frac{1}{\beta} \sum_\nu e^{i\beta\hbar\omega_\nu 0^+} G^{new}_{a,b}(i\omega_\nu) \right]}_{\rho^{new}_{a,b}} - \rho_{a,b} \right) \tag{1.39}$$

In this equation we can use the model Green's function to avoid the calculation and inversion of $\boldsymbol{G}^{new} \otimes \boldsymbol{G}^{new}$

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

Thus, we iterate the coupled equations

$$\boldsymbol{G}^{new}(i\omega_\nu) = \left[(i\hbar\omega_\nu + \mu)\mathbf{1} - \bar{\boldsymbol{h}} + \boldsymbol{\Gamma} - \boldsymbol{\Sigma}^{dyn}(i\omega_\nu) \right]^{-1}$$

$$\delta\boldsymbol{\rho} = \frac{1}{\beta} \sum_\nu e^{i\beta\hbar\omega_\nu 0^+} \boldsymbol{G}^{new}(i\omega_\nu) - \boldsymbol{\rho}$$

$$\boldsymbol{\Gamma} = \boldsymbol{\Gamma} + \alpha\delta\boldsymbol{\rho} \tag{1.40}$$

until $\delta\rho$ vanishes.

In practice we do not use α, but precondition with a Green's function with zero one-particle level. see section 1.7.8.

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

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

1.4.2   Energy contribution

When the loop is converged, we evaluate

$$
Q_{dyn,\beta}^{\hat{W}_2}[\boldsymbol{\rho}] = \left\{ \Phi_\beta^{LW}[\boldsymbol{G}, \hat{W}_2] - \Phi_\beta^{HF,LW}[\boldsymbol{G}, \hat{W}_2] \right.
$$
$$
- \frac{1}{\beta} \sum_\nu \mathrm{Tr} \left\{ \ln \left[\mathbf{1} - \bar{\boldsymbol{G}}(i\omega_\nu) \left(\boldsymbol{\Sigma}_{dyn}(i\omega_\nu) - \boldsymbol{\Gamma} \right) \right] \right.
$$
$$
\left. + \left(\boldsymbol{\Sigma}_{dyn}(i\omega_\nu) - \boldsymbol{\Gamma} \right) \boldsymbol{G}(i\omega_\nu) + \left[\boldsymbol{G}(i\omega_\nu) - \bar{\boldsymbol{G}}(i\omega_\nu) \right] \boldsymbol{\Gamma} \right\}
$$

$$(1.41)$$

1.4.3   Evaluation of the logarithm using a power series expansion

We use the Taylor expansion of the logarithm

$$
\ln[1 - x] = -\sum_{n=1}^{\infty} \frac{1}{n} x^n \qquad \text{for } |x| < 1 \tag{1.42}
$$

$$
Q_{dyn,\beta}^{\hat{W}_2}[\boldsymbol{\rho}] = \Phi_\beta^{LW}[\boldsymbol{G}, \hat{W}_2] - \Phi_\beta^{HF,LW}[\boldsymbol{G}, \hat{W}_2]
$$
$$
- \frac{1}{\beta} \sum_\nu \mathrm{Tr} \left\{ -\sum_{n=1}^{\infty} \frac{1}{n} \left[\bar{\boldsymbol{G}}(i\omega_\nu) \left(\boldsymbol{\Sigma}_{dyn}(i\omega_\nu) - \boldsymbol{\Gamma} \right) \right]^n \right.
$$
$$
\left. + \left(\boldsymbol{\Sigma}_{dyn}(i\omega_\nu) - \boldsymbol{\Gamma} \right) \boldsymbol{G}(i\omega_\nu) + \left[\boldsymbol{G}(i\omega_\nu) - \bar{\boldsymbol{G}}(i\omega_\nu) \right] \boldsymbol{\Gamma} \right\}
$$
$$
= \Phi_\beta^{LW}[\boldsymbol{G}, \hat{W}_2] - \Phi_\beta^{HF,LW}[\boldsymbol{G}, \hat{W}_2]
$$
$$
- \frac{1}{\beta} \sum_\nu \mathrm{Tr} \left\{ \boldsymbol{\Sigma}_{dyn}(i\omega_\nu) \left[\boldsymbol{G}(i\omega_\nu) - \bar{\boldsymbol{G}}(i\omega_\nu) \right] - \sum_{n=2}^{\infty} \frac{1}{n} \left[\bar{\boldsymbol{G}}(i\omega_\nu) \left(\boldsymbol{\Sigma}_{dyn}(i\omega_\nu) - \boldsymbol{\Gamma} \right) \right]^n \right\}
$$

$$(1.43)$$

1.4.4   Evaluation of the logarithm using 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

$$
\mathrm{Tr}\ln(\boldsymbol{A}\boldsymbol{A}^\dagger) \overset{?}{=} \mathrm{Tr}\ln(\boldsymbol{A}) + Tr\ln(\boldsymbol{A}^\dagger) \tag{1.44}
$$

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

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

diagonal.

$$\text{Tr}\ln(\boldsymbol{A}\boldsymbol{A}^\dagger) = \text{Tr}\ln(\boldsymbol{U}\boldsymbol{\Sigma}\boldsymbol{V}^\dagger\boldsymbol{V}\boldsymbol{\Sigma}^\dagger\boldsymbol{U}^\dagger) = \text{Tr}\ln(\boldsymbol{U}\boldsymbol{\Sigma}\boldsymbol{\Sigma}^\dagger\boldsymbol{U}^\dagger) = \text{Tr}\left[\boldsymbol{U}\ln(\boldsymbol{\Sigma}\boldsymbol{\Sigma}^\dagger)\boldsymbol{U}^\dagger\right]$$

$$= \text{Tr}\left[\ln(\boldsymbol{\Sigma}\boldsymbol{\Sigma}^\dagger)\right] = \sum_i \ln(s_i) + \sum_i \ln(s_i) + = \text{Tr}\ln[\boldsymbol{\Sigma}] + \text{Tr}\ln[\boldsymbol{\Sigma}^\dagger]$$

$$\text{Tr}\ln(\boldsymbol{A}) + \mathcal{T}r\ln(\boldsymbol{A}^\dagger) = \text{Tr}\ln(\boldsymbol{U}\boldsymbol{\Sigma}\boldsymbol{V}^\dagger) + \text{Tr}\ln(\boldsymbol{V}\boldsymbol{\Sigma}^\dagger\boldsymbol{U}^\dagger) \tag{1.45}$$

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

1.4.5 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 $\boldsymbol{G}^{out}$ as follows[3]: Caution, I generalized Dieter's Equations without checking. Signs, factors, indices, etc. may be wrong!

$$G^{out}_{\alpha,\beta}(i\omega_\nu) := -\frac{1}{Z}\int \prod_\gamma Dc^*_\gamma Dc_\gamma \; c_\alpha(i\omega_\nu)c^*_\beta(i\omega_\nu)e^{-S_{loc}}$$

$$Z = \int \prod_\gamma Dc^*_\gamma Dc_\gamma \; e^{-S_{loc}}$$

$$S_{loc} = -\int_0^\beta d\tau_1 \int_0^\beta d\tau_2 \sum_{\alpha,\beta} c^*_\alpha(\tau_1)\left[i\hbar\omega_\nu\delta_{\alpha,\beta} - \Delta^{in}_{\alpha,\beta}(i\omega_\nu)\right]c_\beta(\tau_2)$$

$$- \int_0^\beta d\tau \sum_{\alpha,\beta,\gamma,\delta} U_{\alpha,\beta,\delta,\gamma} c^*_\alpha c^*_\beta c_\gamma c_\delta$$

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

3. Convert the Green's function into a self energy $\boldsymbol{\Sigma}^{out}$ using Dyson's equation

$$\Sigma^{out}_{\alpha,\beta}(i\omega_\nu) := -\left(G^{out}(i\omega_\nu)\right)^{-1}_{\alpha,\beta} + i\hbar\omega_\nu\delta_{\alpha,\beta} - \Delta^{in}_{\alpha,\beta}(i\omega_\nu) \tag{1.46}$$

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

$$\Delta^{out}_{\alpha,\beta}(i\omega_\nu) := -\left(G(i\omega_\nu)\right)^{-1}_{\alpha,\beta} + i\hbar\omega_\nu\delta_{\alpha,\beta} - \Sigma^{out}_{\alpha,\beta}(i\omega_\nu) \tag{1.47}$$

---

[3](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)

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.

$$\mathbf{\Delta}^{out} = F[\mathbf{\Delta}^{in}, \mathbf{G}, \mathbf{U}] \tag{1.48}$$

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. 1.47 and Eq. 1.46 yield

$$G^{out}_{\alpha,\beta}(i\omega_\nu) = G_{\alpha,\beta}(i\omega_\nu) \tag{1.49}$$

and

$$G_{\alpha,\beta}(i\omega_\nu) = \left[i\hbar\omega_\nu \mathbf{1} - \mathbf{\Sigma}^{out}(i\omega_\nu) - \mathbf{\Delta}^{in}(i\omega_\nu) \right]^{-1}_{\alpha,\beta}. \tag{1.50}$$

1.4.6   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 basisset[4]. 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 $\boldsymbol{\rho}$.

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. 1.4 as

$$\Phi^{LW,HF} = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \left(\rho_{b,a}\rho_{c,b} - \rho_{c,a}\rho_{d,b} \right) \tag{1.51}$$

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

$$\begin{aligned} \Delta\Sigma_{a,b}(i\omega_\nu) &= \beta \frac{\partial\Phi^{LW}}{\partial G_{b,a}(i\omega_\nu)} - \beta \frac{\partial\Phi^{LW,HF}}{\partial G_{b,a}(i\omega_\nu)} \\ &= \beta \frac{\partial\Phi^{LW}[\mathbf{G}, \hat{W}]}{\partial G_{b,a}(i\omega_\nu)} - \sum_{c,d} \left(U_{a,c,b,d} - U_{a,c,d,b} \right) \rho_{c,d} \end{aligned} \tag{1.52}$$

---

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

1.5   Local orbitals

1.5.1   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

$$|\psi_n\rangle = \sum_a |\chi_a\rangle\langle\pi_a|\psi_n\rangle + |\delta\psi_n\rangle \tag{1.53}$$

where the projector functions obey the bi-orthogonality condition

$$\langle\pi_a|\chi_b\rangle = \delta_{a,b} \ , \tag{1.54}$$

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

$$\langle\pi_a|\delta\psi_n\rangle = 0 \tag{1.55}$$

1.5.2   Energy correction

Here we consider changes of the correlation corrections when we move from a Bra-Ket notation to a matrix notation.

$$Q_\beta^{\hat{W}}[\hat{\rho}] = Q_\beta^{\hat{W}}\left[\sum_n |\psi_n\rangle f_n \langle\psi_n|\right] = Q_\beta^{\hat{W}}\left[\sum_{a,b} |\chi_a\rangle \underbrace{\left(\sum_n \langle\pi_a|\psi_n\rangle f_n \langle\psi_n|\pi_b\rangle\right)}_{\rho_{a,b}} \langle\chi_b|\right] \tag{1.56}$$

Below we consider also the energy contribution as functional of the density matrix in matrix form

$$Q_\beta^{\hat{W}}[\boldsymbol{\rho}] \overset{\text{def}}{=} Q_\beta^{\hat{W}}\left[\sum_{a,b} |\chi_a\rangle \rho_{a,b} \langle\chi_b|\right] \tag{1.57}$$

The derivatives are

$$\frac{\delta Q_\beta^{\hat{W}}}{\delta\langle\psi_n|} \frac{1}{f_n} = \sum_{a,b} |\pi_b\rangle\langle\chi_b| \frac{\delta Q_\beta^{\hat{W}}}{\delta\hat{\rho}} |\chi_a\rangle\langle\pi_a|\psi_n\rangle$$

$$\frac{\delta Q_\beta^{\hat{W}}}{\delta f_n} = \sum_{a,b} \langle\psi_n|\pi_b\rangle\langle\chi_b| \frac{\delta Q_\beta^{\hat{W}}}{\delta\hat{\rho}} |\chi_a\rangle\langle\pi_a|\psi_n\rangle \tag{1.58}$$

In the following we introduce a new symbol, namely

$$\frac{\delta Q_\beta^{\hat{W}}}{\delta\rho_{b,a}} \overset{\text{def}}{=} \langle\chi_b| \frac{\delta Q_\beta^{\hat{W}}}{\delta\hat{\rho}} |\chi_a\rangle \tag{1.59}$$

1.5.3 U-tensor

The interaction $\hat{W}$

$$\hat{W} = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \hat{c}_a^\dagger \hat{c}_b^\dagger \hat{c}_c \hat{c}_d \tag{1.60}$$

is expressed by the **U-tensor**

$$U_{a,b,c,d} = \sum_{\sigma,\sigma'} \int d^3r \int d^3r \, \frac{e^2 \chi_a^*(\vec{r},\sigma) \chi_b^*(\vec{r}',\sigma') \chi_c(\vec{r},\sigma) \chi_d(\vec{r}',\sigma')}{4\pi\epsilon_0 |\vec{r} - \vec{r}'|} \tag{1.61}$$

.

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 leavingf certain elements out completely.

1.5.4 Non-orthonormal orbitals

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

$$\left[\hat{c}_a^\dagger, \hat{c}_b \right]_+ = \langle \pi_b | \pi_a \rangle \tag{1.62}$$

Feynman diagrams

A Feynmandiagram in terms of orthonormal orbitals can be transformed easily into any other one-particle basis, by introducing a unit matrix in the form $\boldsymbol{A}\boldsymbol{A}^{-1}$ in between every Green's function and every interaction.

Let the original basis be $\{|\psi_n\rangle\}$ and a new basis $\{|\chi_a\rangle\}$ so that

$$|\psi_n\rangle = \sum_a |\chi_a\rangle \langle \pi_a | \psi_n \rangle \tag{1.63}$$

Then the Green's function has the form

$$\hat{G} = \sum_{m,n} |\psi_m\rangle \langle \psi_m | \hat{G} | \psi_n \rangle \langle \psi_n| = \sum_{a,b} |\chi_a\rangle \underbrace{\langle \pi_a | \hat{G} | \pi_b \rangle}_{G_{a,b}} \langle \chi_b| \tag{1.64}$$

Thus if the U-tensor is evaluated using the orbitals $|\chi\rangle$, it can directly be used with the Green's function $\langle \pi_a | \hat{G} | \pi_b \rangle$. The final result will be invariant if all sums are traced out.

1.5.5 Incompleteness

One difficulty stems from the fact that the natural orbitals, the local orbitals and the projector functions span different spaces.

We choose a solution by introducing a new type of algebra, that is designed to give certain sum rules exactly. In this algebra we consider the states

$$|\kappa_n\rangle \stackrel{\text{def}}{=} \sum_a |\chi_a\rangle \langle \pi_a | \psi_n \rangle \tag{1.65}$$

as complete orthonormal basisset.

Thus,

$$\delta_{m,n} = \langle \kappa_m | \kappa_n \rangle = \sum_{a,b} \langle \psi_m | \pi_a \rangle \langle \chi_a | \chi_b \rangle \langle \pi_b | \psi_n \rangle \tag{1.66}$$

and

$$\hat{1} = \sum_{a,b,n} | \chi_a \rangle \langle \pi_a | \psi_n \rangle \langle \psi_n | \pi_b \rangle \langle \chi_b | \tag{1.67}$$

I introduce

$$S_{a,b}^{-1} \stackrel{\text{def}}{=} \sum_{n} \langle \pi_a | \psi_n \rangle \langle \psi_n | \pi_b \rangle \tag{1.68}$$

and redefine the overlap $\langle \chi_a | \chi_b \rangle$ as the inverse of $\mathbf{S}$. We could consider this as a redefinition of the scalar product between the states $| \kappa_n \rangle$.

1.6   Usage

1.6.1   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
!end
!EOB
```

1.6.2   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
```

1.7   Description of Subroutines

1.7.1   Flowchart
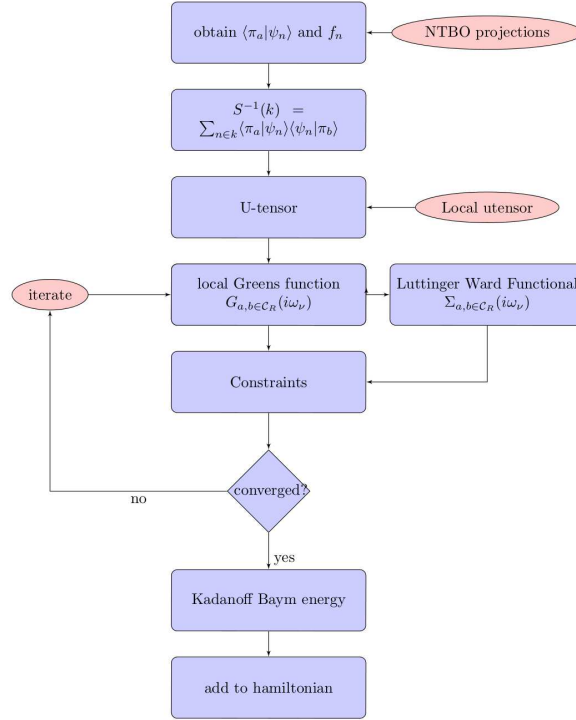
1.7.2   Data exchange of the Object wit the outer world

1.7.3   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()
do iter=1,3
  call dmft_gloc()
  call dmft_solver(etot)
  call dmft_constraints()
enddo ! end of loop over iterations to enforce constraint
call dmft_staticsolver(svar)
etot=etot+svar
```

```
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`: The orbital coefficients $\langle \pi_a | \psi_n \rangle$ have been calculated by the LMTO object and they are kept in the module of `paw_waves`.

$$\langle \pi_a | \psi_n \rangle \tag{1.69}$$

$$\rho_{a,b}(\vec{k}) = \sum_n \langle \pi_a | \psi_n(\vec{k}) \rangle f_n(\vec{k}) \langle \psi_n(\vec{k}) | \pi_b \rangle \tag{1.70}$$

The orbital coefficients are kept as `kset(ik)%pipsi(ndim,nchi,nb,nspin)`. The k-dependent density matrix is kept as `kset(ik)%rho(nchi,nchi,ndimd)`.

The name DMFT_COLLECTHAMILTONIAN is misleading, because we only collect the orbital coeffcients and the density matrix.

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

This still uses the orbital coefficients and occupations from the PAW_WAVES module, instead of using the orbital coefficients or dthe k-dependent density matrix from KSET.

The name DMFT_COLLECTFULLDENMAT is misleading.

3. `DMFT_UTENSOR`: (see also section 1.7.5) The local U-tensor is collected from the onsite elements `POTPAR(ISP)%TAILED%U` calculated in the `paw_LMTO` object. It is directly converted from the "tailed representation" into the basis of local orbitals.

$$U_{a,b,c,d} = \alpha \int d^4x \int d^4x' \, \frac{e^2 \chi_a^*(\vec{x}) \chi_b^*(\vec{x'}) \chi_c(\vec{x}) \chi_d(\vec{x'})}{4\pi\epsilon_0 |\vec{r} - \vec{r'}|} \tag{1.71}$$

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`.

The scaled U-tensor is kept as `ATOMSET(iat)%U`

4. `DMFT_SMAT`: The inverse overlap matrix $\boldsymbol{S}^{-1}$ is calculated as

$$\boldsymbol{S}^{-1}(\vec{k}) = \sum_n \langle \pi_a | \psi_n(\vec{k}) \rangle \langle \psi_n(\vec{k}) | \pi_b \rangle \tag{1.72}$$

Note that the inverse overlap matrix is spin and orbital dependent. $\boldsymbol{S}(\vec{k})$ is pot on the KSET structure as `KSET%SINV` and $\boldsymbol{S}^{-1}(\vec{k})$ as `KSET%SMAT`

5. `DMFT_NATORB`: (see also section 1.7.6) Construct the eigenstates $|\phi_j\rangle$ of the single site density matrix

$$\hat{\rho}_R |\phi_{R,i}\rangle = |\phi_{R,i}\rangle f'_{R,i} \tag{1.73}$$

These states will be called local natural orbitals.

The orbital coefficients of the local natural orbitals are calculated as [5]

$$\sum_b \langle \pi_a | \hat{\rho}_R | \pi_b \rangle y_{b,j} = \sum_b S_{R,a,b}^{-1} y_{b,j} f_{R,j}' \qquad \text{with } \vec{y}_i^\dagger \mathbf{S}_R^{-1} \vec{y}_j = \delta_{i,j} \tag{1.78}$$

$$\langle \pi_a | \phi_{R,j} \rangle = \sum_b S_{R,a,b}^{-1} y_{b,j} \tag{1.79}$$

The local density matrix $\boldsymbol{\rho}_R$ andf the local inverse overlap matrix $\mathbf{S}_R^{-1}$ are obtained as Brillouin zone intergral projected onto the lcoal site

$$\boldsymbol{\rho}_R = \boldsymbol{P}_R \left[\sum_{\vec{k}} w_{\vec{k}} \boldsymbol{\rho}(\vec{k}) \right] \boldsymbol{P}_R$$

$$\mathbf{S}_R^{-1} = \boldsymbol{P}_R \left[\sum_{\vec{k}} w_{\vec{k}} \mathbf{S}^{-1}(\vec{k}) \right] \boldsymbol{P}_R \tag{1.80}$$

The transformation matrices are stored in the fully non-collinear data model in `atomset%natorb%piphi` and the vectors $\vec{y}_j$ are stored as `atomset%natorb%chiphi`.

The vector y is used for the transformation to the local natural orbitals in the solver interface. Check if this is correct. should it be the vector x?

6. `DMFT_HRHO`: (see also section 1.7.7 on p. 26).) Calculate the non-interacting Hamiltonian $\bar{h}$ that produces the specified density matrix.

$$\bar{h}_{a,b}(\vec{k}) = \mu S_{a,b}(\vec{k}) + \sum_n \langle \chi_a | \psi_n' \rangle \frac{1}{\beta} \ln \left[\frac{1 - f_n'}{f_n'} \right] \langle \psi_n' | \chi_b \rangle \tag{1.81}$$

---

[5]

$$\underbrace{\sum_{a,b} |\chi_a\rangle \langle \pi_a | \hat{\rho}_R | \pi_b \rangle \langle \chi_b|}_{\hat{\rho}_R} \underbrace{\sum_c |\chi_c\rangle x_{c,j}}_{|\phi_{R,j}\rangle} = \underbrace{\sum_a |\chi_a\rangle x_{a,j}}_{|\phi_{R,j}\rangle} f_{R,j}'$$

$$\sum_{b,c} \langle \pi_a | \hat{\rho}_R | \pi_b \rangle \langle \chi_b | \chi_c \rangle x_{c,j} = x_{a,j} f_{R,j}' \tag{1.74}$$

Now, introduce $y_{a,j} = \sum_b \langle \chi_a | \chi_b \rangle x_{b,j}$

$$\sum_b \langle \pi_a | \hat{\rho}_R | \pi_b \rangle y_{b,j} = \sum_b S_{a,b}^{-1} y_{b,j} f_{R,j}' \tag{1.75}$$

which is a generalized eigenvalue problem for the vectors $\vec{y}_j$, which are converted into orbital coefficients by multiplication with $\mathbf{S}^{-1}$.

$$|\phi_{R,j}\rangle = \sum_{a,b} |\chi_a\rangle S_{a,b}^{-1} y_{b,j} \tag{1.76}$$

The local natural orbitals are orthonormal in the sense

$$\langle \phi_{R,i} | \phi_{R,j} \rangle = \sum_{a,b,c,d} y_{a,i}^* S_{a,c}^{-1} \underbrace{\langle \chi_c | \chi_d \rangle}_{(\mathbf{S}^{-1})^{-1}} S_{d,b}^{-1} y_{b,j} = \sum_{a,b} y_{a,i}^* S_{a,b}^{-1} y_{b,j} = \delta_{i,j} \tag{1.77}$$

where the natural orbitals $|\psi'_n\rangle$ and the occupations f'_n differ from the original natural orbitals.

h' is kept as `KSET%H0`.

7. `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.

$$\boldsymbol{G}_R(i\omega_\nu) = \sum_k w(\vec{k}) \left[(i\omega_\nu + \mu)\boldsymbol{S}(\vec{k}) - \bar{\boldsymbol{h}} + \boldsymbol{\Gamma} - \boldsymbol{\Sigma}^{\hat{W}_2}_{dyn}(i\omega_\nu) \right]^{-1}_R \qquad (1.82)$$

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

8. `DMFT_SOLVER`: Calculates the dynamic contribution $\Phi^{LW,\hat{W}_2}_\beta - \Phi^{HF,\hat{W}_2}_\beta$ to the Luttinger-Ward functional and the corresponding contribution $\boldsymbol{\Sigma}_{dyn}, \hat{W}_2$ to the self energy for the specified Green's function.

   (a) In a first loop over atoms the atoms are individually adressed and the routine `dmft_dynamicsolver` is called.

   (b) Inside `dmft_dynamicsolver` the U-tensor and the Green's function are converted into local natural orbitals and in a up-down spinor representation with all up-spin components first and the down-spin components second.

   (c) then the actual interface routine `DMFT_SOLVERIO` is called, which returns the value of the dynamic contribution to the Luttinger-Ward functional (HF subtracted), the coresponding contribution to the self energy and the derivative of the U-tensor.

$$\Phi^{dyn,LW}[\boldsymbol{G}(i\omega_\nu), \hat{W}_2] = \Phi^{LW}[\boldsymbol{G}(i\omega_\nu), \hat{W}_2] - \frac{1}{2} \sum_{a,b,c,d} U^{\hat{W}_2}_{a,b,d,c} \left(\rho_{d,a}\rho_{c,b} - \rho_{c,a}\rho_{d,b} \right))$$

$$\Sigma^{\hat{W}_2}_{dyn,a,b}(i\omega_\nu) = \frac{\beta \partial \Phi^{LW}}{\partial G_{b,a}(i\omega_\nu)} - \sum_{c,d} \left(U^{\hat{W}_2}_{a,c,b,d} - U^{\hat{W}_2}_{a,c,d,b} \right) \rho_{d,c}$$

$$\frac{\delta \Phi^{dyn,LW}_\beta}{\delta U_{a,b,c,d}} = \frac{\delta \Phi^{LW}_\beta}{\delta U_{a,b,d,c}} - \frac{1}{2} \left(\rho_{d,a}\rho_{c,b} - \rho_{c,a}\rho_{d,b} \right)) \qquad (1.83)$$

   pay attention to the reversed order of the indices c, d!

   (d) After `DMFT_SOLVERIO` the self energy and the derivative with respect to the U-tensor are transformed back.

The external solver may make additional approximations to the U-tensor or to the Kadanoff-Baym functional. However, these approximationa must be consistent across the dynamical correlation correction.

9. `DMFT_STATICSOLVER`: Calculates the static contribution to the correlation energy, i.e. the screened Hartree Fock term and the DFT double counting correction and its derivatives. These terms do not affect the density matrix constraint.

   This term must be removed if the corresponding terms are treated in the PAW_LMTO object. The double counting for the correlation must however still be included.

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 the correlation 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.

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

$$
-\frac{1}{\beta}\sum_\nu \mathrm{Tr}\Bigg\{ \ln\Big[\mathbf{1}-\Big(i\hbar\omega_\nu+\mu)\mathbf{1}-\bar{\mathbf{h}}\Big)^{-1}\Big(\mathbf{\Sigma}^{\hat{W}_2}_{dyn}(i\omega_\nu)-\mathbf{\Gamma}\Big)\Big]
$$
$$
+\Big(\mathbf{\Sigma}^{\hat{W}_2}_{dyn}(i\omega_\nu)-\mathbf{\Gamma}\Big)\mathbf{G}(i\omega_\nu)+\Big[\mathbf{G}(i\omega_\nu)-\Big(i\hbar\omega_\nu+\mu)\mathbf{1}-\bar{\mathbf{h}}\Big)^{-1}\Big[\mathbf{\Gamma}\Big]\Bigg\} \qquad (1.84)
$$

At this point, I am still using the Dahlen trick, which is probably not quite correct.

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.

1.7.4   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 `DYNOCC` Object
- KBT from `dynocc$getr8a('temp')` from `paw_occupations.f90`
- `atomset(iat)%lhfweight` 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

$$\omega_\nu = (2\nu - 1)\pi k_B T \qquad \text{for } \nu = 1, \ldots, N_\omega \tag{1.85}$$

- 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

1.7.5   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

$$|\chi_a\rangle = |\phi_a\rangle - \sum_{b;R_b=R_a} |\dot{\bar{\phi}}_b\rangle \bar{S}_{a,b} \tag{1.86}$$

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

$$u_{\ell,a,b,c,d} = \frac{2\ell+1}{4\pi} \int dr\, r^2 R_c(r) R_d(r) \left[\int d^3r'\, V_\ell(r,r') R_a(r') R_b(r') \right] \tag{1.87}$$

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

$$U_{a,b,c,d} = \sum_{\ell\ell} \frac{4\pi}{2\ell+1} \sum_{m=-\ell}^{\ell} u_{\ell,b,d,c,a} C_{L,L_b,L_d} C_{L,L_c,L_a} \tag{1.88}$$

The U-tensor is screened by a factor `LHFWEIGHT`. It is obtained from the `lmto_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 as[6] in Eq. 1.71, i.e. as

$$U_{a,b,c,d} = \int d^4x \int d^4x' \frac{e^2 \chi_a^*(\vec{x}) \chi_b^*(\vec{x'}) \chi_c(\vec{x}) \chi_d(\vec{x'})}{4\pi\epsilon_0 |\vec{r} - \vec{r'}|} \tag{1.89}$$

---

[6]$\vec{x} = (\vec{r}, \sigma)$

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

$$E_H = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,c,d} \, \rho_{c,a}\rho_{d,b}$$

$$E_X = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,c,d} \, \rho_{c,b}\rho_{d,a} \qquad (1.90)$$

1.7.6 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.

$$\hat{1}_{\mathcal{C}_R} = \sum_{\alpha,\beta} \sum_n |\chi_\alpha\rangle\langle\pi_\alpha|\psi_n\rangle\langle\psi_n|\pi_\beta\rangle\langle\chi_\beta| \qquad (1.91)$$

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

$$S^{-1}_{\alpha,\beta} = \sum_{\alpha,\beta} \sum_n \langle\pi_\alpha|\psi_n\rangle\langle\psi_n|\pi_\beta\rangle \qquad (1.92)$$

Note, however, that this matrix lives only on a single site! We can obtain it from the k-dependent 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

$$\hat{\rho}_{\mathcal{C}_R} = \sum_{\alpha,\beta} \sum_n |\chi_\alpha\rangle\langle\pi_\alpha|\psi_n\rangle f_n\langle\psi_n|\pi_\beta\rangle\langle\chi_\beta| \qquad (1.93)$$

The eigenvalue equation

$$\hat{\rho}|\phi_n\rangle = |\phi_n\rangle f_n$$

$$\sum_{\alpha,\beta} |\chi_\alpha\rangle\langle\pi_\alpha|\hat{\rho}|\pi_\beta\rangle\langle\chi_\beta|\phi_j\rangle = \sum_{\alpha,\beta} |\chi_\alpha\rangle\langle\pi_\alpha|\pi_\beta\rangle\langle\chi_\beta|\phi_j\rangle f_j = \sum_{\alpha,\beta} |\chi_\alpha\rangle S^{-1}_{\alpha,\beta}\langle\chi_\beta|\phi_j\rangle\bar{f}_j \quad (1.94)$$

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

$$\rho V = S^{-1}V\bar{f} \qquad \text{with} \qquad V^\dagger S^{-1}V = \mathbf{1} \qquad (1.95)$$

The natural orbitals are

$$|\phi_j\rangle = \sum_{\alpha,\beta\in\mathcal{C}_R} \underbrace{|\chi_\alpha\rangle S^{-1}_{\alpha,\beta}\langle\chi_\beta|}_{\mathbf{1}_{\mathcal{C}_R}} \phi_j\rangle = \sum_{\alpha\in\mathcal{C}_R} |\chi_\alpha\rangle \underbrace{\left(S^{-1}V\right)_{\alpha,j}}_{V^{\dagger,-1}_{\alpha,j}} \qquad (1.96)$$

The matrix 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 $S^{-1}V = V^{\dagger,-1}$ in `atomset%natorb%piphi` All data in `atomset%natorb` are stored in the fully non-collinear model.

1.7.7 DMFT_HRHO

Calculate the non-interacting Hamiltonian $\bar{h}$ that produces the specified density matrix.

$$\bar{h}_{a,b}(\vec{k}) = \mu S_{a,b}(\vec{k}) + \sum_n \langle \chi_a | \psi'_n \rangle \frac{1}{\beta} \ln \left[\frac{1 - f'_n}{f'_n} \right] \langle \psi'_n | \chi_b \rangle \tag{1.97}$$

where the natural orbitals $|\psi'_n\rangle$ and the occupations f'_n differ from the original natural orbitals.

1. First, we transform onto natural orbitals

$$|\psi'_n\rangle = \sum_a |\chi_a\rangle\langle\pi_a|\psi'_n\rangle \tag{1.98}$$

   These natural orbitals may be slighly different from the original natural orbitals, because they are restricted to the space of local orbitals.

$$\hat{\rho}|\psi'_n\rangle = |\psi'_n\rangle f'_n$$

$$\langle\chi_a| \underbrace{\sum_{b,c} |\chi_b\rangle \overbrace{\left(\sum_m \langle\pi_b|\psi_m\rangle f_m \langle\psi_m|\pi_c\rangle\right)}^{\rho_{b,c}} \langle\chi_c|}_{\hat{\rho}} \underbrace{\sum_d |\chi_d\rangle\langle\pi_d|\psi'_n\rangle}_{|\psi'_n\rangle} = \langle\chi_a| \underbrace{\sum_d |\chi_d\rangle\langle\pi_d|\psi'_n\rangle}_{|\psi'_n\rangle} f'_n$$

$$S_{a,b}\rho_{b,c}S_{c,d}\langle\pi_d|\psi'_n\rangle = S_{a,d}\langle\pi_d|\psi'_n\rangle f'_n$$

$$\rho\left(S\vec{x}_n\right) = S^{-1}\left(S\vec{c}_n\right)f'_n \tag{1.99}$$

   Thus, for each k-point solve the generalized eigenvalue problem

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

   where $f'(\vec{k})$ is a diagonal matrix that contains k-dependent occupations. (They differ from the occupations f_n of the natural orbitals $|\psi_n\rangle$.)

2. Next we evaluate the hamiltonian $\bar{h}$ in the representation of the natural orbitals $|\psi'_n\rangle$.

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

$$\epsilon = \mu + k_B \begin{cases} \ln\left(\frac{1-f}{f}\right) & \text{for } f_0 < f < 1 - f_0 \\ a + b(f - f_0) & \text{for } f < f_0 \\ -a + b(f - (1 - f_0)) & \text{for } f > 1 - f_0 \end{cases} \tag{1.100}$$

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

3. next we transform this Hamiltonian back into the basis of local orbitals, where the Hamiltonian has the form $\bar{h}_{a,b} = \langle\chi_a|\hat{\bar{h}}|\chi_b\rangle$, respectively $\hat{\bar{j}} = \sum_{a,b} |\pi_a\rangle h_{a,b}\langle\pi_b|$.

$$\hat{\bar{h}} = \sum_n |\psi'_n\rangle\epsilon_n\langle\psi'_n|$$

$$\langle\chi_a|\hat{\bar{h}}|\chi_b\rangle = \sum_n \langle\chi_a|\psi'_n\rangle\epsilon_n\langle\psi'_n|\chi_b\rangle$$

$$= \sum_{n,c,d} \langle\chi_a|\chi_c\rangle\langle\pi_c|\psi'_n\rangle\epsilon_n\langle\psi'_n|\pi_d\rangle\langle\chi_d|\chi_b\rangle = V_{a,n}\epsilon_n V^\dagger_{n,b}$$

$$\bar{h}(\vec{k}) = V\epsilon V^\dagger \tag{1.101}$$

1.7.8 DMFT_CONSTRAINTS

Green's function

The Green's function is calculated as follows

$$\boldsymbol{G}(\vec{k}, i\omega_\nu) = \left[(i\omega_\nu + \mu)\boldsymbol{S}(\vec{k}) - \bar{\boldsymbol{h}}(\vec{k}) + \boldsymbol{\Gamma}(\vec{k}) - \sum_R \boldsymbol{\Sigma}_R^{dyn}(i\omega_\nu)\right]^{-1} \qquad (1.102)$$

As described in section B.2.1 on p. 36, the Laurent-expansion coefficients are

$$\boldsymbol{G}^{(1)}(\vec{k}) = \boldsymbol{S}^{-1}(\vec{k})$$

$$\boldsymbol{G}^{(2)}(\vec{k}) = \boldsymbol{S}^{-1}(\vec{k})\left(-\mu\boldsymbol{S}(\vec{k}) + \bar{\boldsymbol{h}}(\vec{k}) - \boldsymbol{\Gamma}(\vec{k}) + \mathcal{S}_{dyn}^{(1)}(\vec{k})\right)\boldsymbol{S}^{-1}(\vec{k})$$

$$\boldsymbol{G}^{(3)}(\vec{k}) = \boldsymbol{S}^{-1}(\vec{k})\left[\mathcal{S}_{dyn}^{(2)}(\vec{k}) + \left(-\mu\boldsymbol{S}(\vec{k}) + \bar{\boldsymbol{h}}(\vec{k}) - \boldsymbol{\Gamma}(\vec{k}) + \mathcal{S}_{dyn}^{(1)}(\vec{k})\right)\right.$$

$$\left.\times\boldsymbol{S}^{-1}(\vec{k})\left(-\mu\boldsymbol{S}(\vec{k}) + \bar{\boldsymbol{h}}(\vec{k}) - \boldsymbol{\Gamma}(\vec{k}) + \mathcal{S}_{dyn}^{(1)}(\vec{k})\right)\right]\boldsymbol{S}^{-1}(\vec{k})$$

$$(1.103)$$

Update Γ

UPDATE Γ

The following two equations are solved iteratively

$$\delta\boldsymbol{\rho}(\vec{k}) = \left(\frac{1}{\beta}\sum_\nu \underbrace{\left[(i\omega_\nu + \mu)\boldsymbol{S}(\vec{k}) - \bar{\boldsymbol{h}}(\vec{k}) + \boldsymbol{\Gamma}(\vec{k}) - \sum_R \boldsymbol{\Sigma}_R^{dyn}(i\omega_\nu)\right]^{-1}}_{\boldsymbol{G}(\vec{k})}\right) - \boldsymbol{\rho}(\vec{k})$$

$$\boldsymbol{\Gamma}(\vec{k}) = \boldsymbol{\Gamma}(\vec{k}) - \frac{4}{\beta}\boldsymbol{S}(\vec{k})\delta\rho(\vec{k})\boldsymbol{S}(\vec{k}) \qquad (1.104)$$

until $\delta\boldsymbol{\rho}$ vanishes.

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.

$$\delta\boldsymbol{\rho}(\boldsymbol{\Gamma}) = \delta\boldsymbol{\rho}(\boldsymbol{\Gamma}_0) - \frac{1}{\beta}\sum_\nu \boldsymbol{G}(\boldsymbol{\Gamma} - \boldsymbol{\Gamma}_0)\boldsymbol{G} + \dots \qquad (1.105)$$

Now we approximate $\boldsymbol{G}$ by $(i\hbar\omega_\nu)^{-1}\boldsymbol{S}^{-1}$

$$\delta\boldsymbol{\rho}(\boldsymbol{\Gamma}) = \delta\boldsymbol{\rho}(\boldsymbol{\Gamma}_0) - \frac{1}{\beta}\underbrace{\left[\sum_\nu \frac{1}{(i\hbar\omega_\nu)^2}\right]}_{-\beta/4}\boldsymbol{S}^{-1}(\boldsymbol{\Gamma} - \boldsymbol{\Gamma})\boldsymbol{S}^{-1} + \dots$$

$$\Rightarrow \qquad \boldsymbol{\Gamma} = \boldsymbol{\Gamma}_0 + \frac{4}{\beta}\boldsymbol{S}\left(\underbrace{\delta\boldsymbol{\rho}(\boldsymbol{\Gamma})}_{=0} - \delta\boldsymbol{\rho}(\boldsymbol{\Gamma}_0)\right)\boldsymbol{S} \qquad (1.106)$$

Should there be convergence problems, it may be improved by using the static Green's function G_ρ instead.

1.7.9   DMFT_SOLVER

The solver first calculates the Hartree-Fock contribution to the Luttinger-Ward functional as defined in Eq. 1.51 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

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

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

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

The poles of the Matsubara weighting functions obey

$$1 + e^{\pm\beta\hbar z} = 0 \tag{A.2}$$

With the help of

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

Eq. A.2 can be rewrfitten in the form

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

The second equation is obeyed for

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

The first equation yields

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

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

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

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 x_0 is the position of the pole. For this purpose we expand the inverse
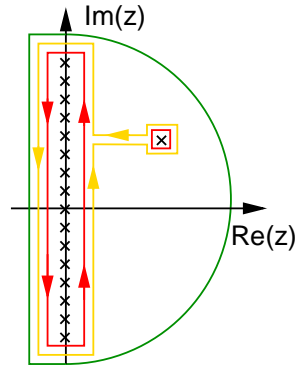
$$\frac{1}{\mp\hbar\beta}(1 + e^{\pm\hbar\beta z}) = \frac{1}{\mp\hbar\beta}(\underbrace{1 + e^{\pm\hbar\beta z_0}}_{=0} \pm \hbar\beta \underbrace{e^{\pm\hbar\beta z_0}}_{-1}(z - z_0) + O(|z - z_0|^2)) \tag{A.7}$$

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

With the help of the Matsubara weighting function we can evaluate the Matsubara sum as[1]

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

The minus sign occurs because the c ounter 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 $\pm Re(z) > 0$, because we there the corresponding weighting function $h^\pm$ decays exponentially for $Re(z) \to \pm\infty$.

---

[1]Cauchy's Integral formula:

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

where γ is a counter-clockwise countour around a pole z_0 of a function f, that can be expanded into a Laurent series about z_0. The Residuum 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 are[2]

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

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

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

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

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 e level, that lies far from the chemical potential, the finite Matsubara sum falls off to $\frac{1}{2}$.

---

[2]They differ from those of the source by a factor $1/\hbar$.
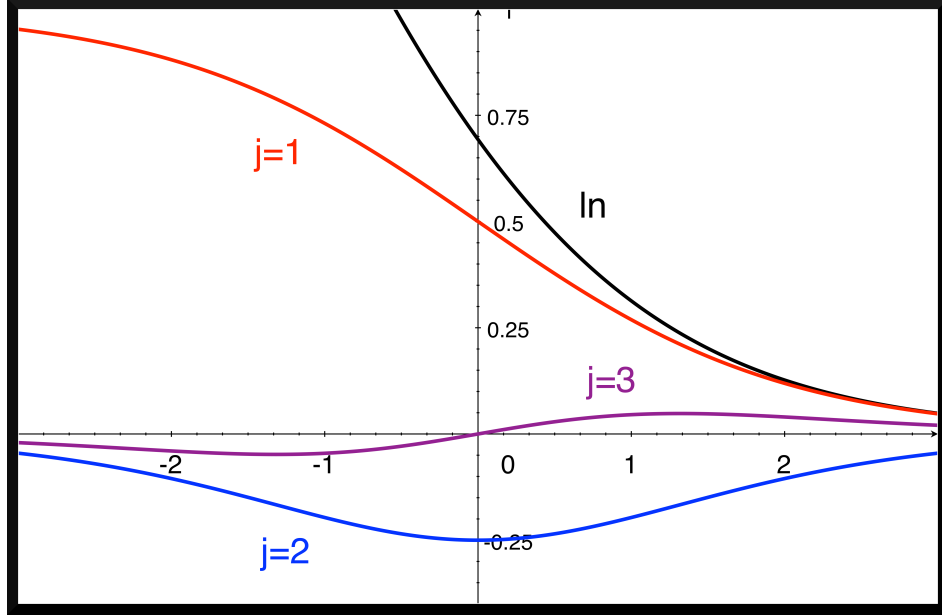
Fig. A.1: The result of Matsubara sums $-k_B T \sum_\nu \ln[-i\hbar\omega_\nu + \epsilon]$, $k_B T \sum_\nu \frac{1}{(i\hbar\omega_\nu - \epsilon)} e^{i\omega_\nu 0^+}$, $k_B T \sum_\nu \frac{1}{(i\hbar\omega_\nu - \epsilon)^2}$ as function of energy ϵ.
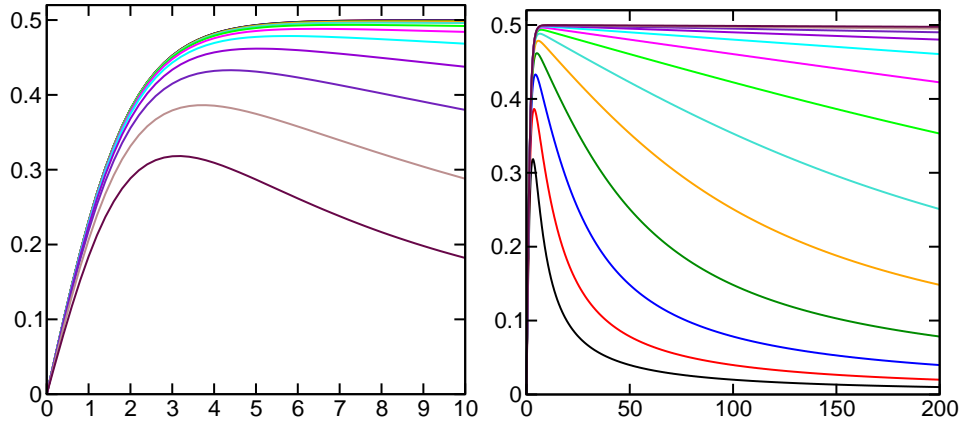

Fig. A.2: Fermi function calculated from a finite Matsubara sum. The function is shifted by $-\frac{1}{2}$ because no relgularization has been done. The sums have been performed with 2^n grid points with $n = 0, \ldots, 12$ for $k_B T = 1$.

Appendix B

The Green function

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

$$\hat{G}^{lat}(i\omega_\nu) = \left[(i\hbar\omega_\nu + \mu)\hat{1} - \hat{h} - \Delta\hat{\Sigma}(i\omega_\nu) \right]^{-1}$$

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

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

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

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

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

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

Proof:

$$
\begin{aligned}
G_{\alpha,\beta}(i\omega_n) \;&=\; -\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\; e^{i\omega_n\tau}\left\langle \mathcal{T}\hat{c}_\alpha(\tau)\hat{c}_\beta^\dagger(0)\right\rangle &\text{(B.3)}\\[4pt]
\Rightarrow G_{\alpha,\beta}(-i\omega_n) \;&=\; -\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\; e^{-i\omega_n\tau}\left\langle \mathcal{T}\hat{c}_\alpha(\tau)\hat{c}_\beta^\dagger(0)\right\rangle\\[4pt]
&=\; \left\{-\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\; e^{i\omega_n\tau}\left\langle \left(e^{\beta(\hat{H}-\mu\hat{N})}\hat{c}_\alpha e^{-\beta(\hat{H}-\mu\hat{N})}\hat{c}_\beta^\dagger\right)^\dagger\right\rangle\right\}^*\\[4pt]
&=\; \left\{-\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\; e^{i\omega_n\tau}\left\langle \hat{c}_\beta e^{-\beta(\hat{H}-\mu\hat{N})}\hat{c}_\alpha^\dagger e^{\beta(\hat{H}-\mu\hat{N})}\right\rangle\right\}^*\\[4pt]
&\overset{cycl.perm}{=\!=}\; \left\{-\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\; e^{i\omega_n\tau}\left\langle e^{\beta(\hat{H}-\mu\hat{N})}\hat{c}_\beta e^{-\beta(\hat{H}-\mu\hat{N})}\hat{c}_\alpha^\dagger\right\rangle\right\}^*\\[4pt]
&=\; \left\{-\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\; e^{i\omega_n\tau}\left\langle \mathcal{T}\hat{c}_\beta(\tau)\hat{c}_\alpha^\dagger\right\rangle\right\}^*\\[4pt]
&=\; G_{\beta,\alpha}^*(i\omega_\nu) &\text{(B.4)}
\end{aligned}
$$

B.1.2   Density of states

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

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

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

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

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

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

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

$$
\begin{aligned}
\tilde{D}_{a,b}(\mu) &\overset{\text{def}}{=} \frac{\partial N_{a,b}}{\partial\mu}\\[4pt]
&= -\frac{1}{\beta}\sum_\nu\sum_c \left[\left(\boldsymbol{G}(i\omega_\nu,\mu_0)\right)^{-1} + (\mu-\mu_0)\mathbf{1}\right]_{a,c}^{-1}\left[\left(\boldsymbol{G}(i\omega_\nu,\mu_0)\right)^{-1} + (\mu-\mu_0)\mathbf{1}\right]_{c,b}^{-1}
\end{aligned}
$$
$$\tag{B.8}$$

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

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

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

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

where

$$
S^{-1}_{a,b} = \sum_k \langle\pi_a|\psi_n\rangle f_n \langle\psi_n|\pi_b\rangle \tag{B.11}
$$

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

$$
\boldsymbol{G}_{a,b}(i\omega_\nu) = \sum_n \langle\pi_a|\psi_n\rangle \Bigg[(i\hbar\omega_\nu + \mu)\delta_{n,n'} - \sum_{c,d} \langle\psi_n|\pi_c\rangle \Big(\boldsymbol{h}_{c,d} + \boldsymbol{\Sigma}_{c,d}(i\omega_\nu) \Big) \langle\pi_d|\psi_{n'}\rangle \Bigg]^{-1}_{n,n'} \langle\psi_{n'}|\pi_b\rangle
$$
$$
\tag{B.12}
$$

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

I 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.

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

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

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

where the self energy has the Laurent expansion

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

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

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

with

$$\mathcal{G}^{(1)} = \boldsymbol{S}^{-1}$$
$$\mathcal{G}^{(2)} = \boldsymbol{S}^{-1}\left(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)}\right)\boldsymbol{S}^{-1}$$
$$\mathcal{G}^{(3)} = \boldsymbol{S}^{-1}\left(\mathcal{S}^{(1)} + \left(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)}\right)\boldsymbol{S}^{-1}\left(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)}\right)\right)\boldsymbol{S}^{-1} \tag{B.18}$$

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

$$\sum_{j=1}^{\infty} \frac{1}{(-i\hbar\omega_\nu)^j} \mathcal{G}^{(j)} = \boldsymbol{G}(-i\omega_\nu) = \boldsymbol{G}^\dagger(i\omega_\nu) = \sum_{j=1}^{\infty} \frac{1}{(-i\hbar\omega_\nu)^j} \left(\mathcal{G}^{(j)}\right)^\dagger$$

$$\Rightarrow \qquad \mathcal{G}^{(j)} = \left(\mathcal{G}^{(j)}\right)^\dagger \tag{B.19}$$

From this requirement we can derive that $\boldsymbol{S}$, $\boldsymbol{h}$, $\mathcal{S}^{(0)}$ 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

$$\frac{1}{\beta}\sum_\nu \boldsymbol{G}e^{i\omega_\nu 0^+} = \sum_{j=1}^{\infty} \mathcal{G}^{(j)} \frac{1}{(j-1)!} \partial_\epsilon^{j-1}\Big|_{\epsilon=0} (1 + e^{\beta\epsilon})^{-1}$$

$$= \frac{1}{2}\mathcal{G}^{(1)} - \frac{1}{4}\beta\mathcal{G}^{(2)} - 0\cdot\beta^2\mathcal{G}^{(3)} + \underbrace{\left(\frac{1}{3!}\frac{1}{8}\beta^3\mathcal{G}^{(4)} - \frac{1}{5!}\frac{1}{4}\beta^5\mathcal{G}^{(6)} + \frac{1}{7!}1.0?\beta^7\mathcal{G}^{(8)}\right)}_{\text{from grapher}} + O(\beta^9)$$

$$= \frac{1}{2}\mathcal{G}^{(1)} - \frac{1}{4}\beta\mathcal{G}^{(2)} + \underbrace{\left(\frac{1}{48}\beta^3\mathcal{G}^{(4)} - \frac{1}{480}\beta^5\mathcal{G}^{(6)} + \frac{1.0?}{4320}\beta^7\mathcal{G}^{(8)}\right)}_{\text{from grapher}} + O(\beta^9)$$

Appendix C

Dividing the correlation contribution into a static and a dynamic part

We will divide the correlation contribution $Q_\beta^{\hat{W}}$ in Eq. 1.7 on p. 4 into two separate terms. It seems that division is only possible if only one of the terms has a frequency-dependent self energy.

For this purpose, we express the Luttinger-Ward functional as a sum of two terms $\Phi_\beta^{LW} = \Phi_\beta^{LW,2} + \Phi_\beta^{LW,2}$, of which only the second has the frequency dependent self energy.

$$Q_\beta^{\hat{W}}[\boldsymbol{\rho}] \overset{\text{Eq. }1.7}{=} \underset{h'}{\text{stat}}\, \underset{\boldsymbol{G},\boldsymbol{\Sigma}}{\text{stat}} \Bigg\{ \Phi_\beta^{LW,1}[\boldsymbol{G}, \hat{W}_1] + \Phi_\beta^{LW,2}[\boldsymbol{G}, \hat{W}_2]$$
$$- \frac{1}{\beta}\sum_\nu \text{Tr}\Big\{ \ln\Big[\mathbf{1} - \Big((i\hbar\omega_\nu + \mu)\mathbf{1} - \bar{\boldsymbol{h}}\Big)^{-1}\Big(\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \bar{\boldsymbol{h}}\Big)\Big]$$
$$+ (\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \bar{\boldsymbol{h}})\boldsymbol{G}(i\omega_\nu) - \Big[\boldsymbol{G}(i\omega_\nu) - \Big((i\hbar\omega_\nu + \mu)\mathbf{1} - \bar{\boldsymbol{h}}\Big)^{-1}\Big]\Big(\boldsymbol{h}' - \bar{\boldsymbol{h}}\Big)\Big\}\Bigg\}$$
(C.1)

The stationary conditions are

$$0 = -\frac{\beta\delta\Phi^{LW,1}}{\delta\boldsymbol{G}(i\omega_\nu)} - \frac{\beta\delta\Phi^{LW,2}}{\delta\boldsymbol{G}(i\omega_\nu)} + \Big(\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \bar{\boldsymbol{h}}\Big) - \Big(\boldsymbol{h}' - \bar{\boldsymbol{h}}\Big)$$

$$\Rightarrow \quad \boldsymbol{\Sigma}(i\omega_\nu) = \frac{\beta\delta\Phi^{LW,1}}{\delta\boldsymbol{G}(i\omega_\nu)} + \frac{\beta\delta\Phi^{LW,2}}{\delta\boldsymbol{G}(i\omega_\nu)}$$

$$0 = \Big[\mathbf{1} - \bar{\boldsymbol{G}}(i\omega_\nu)\Big(\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \bar{\boldsymbol{h}}\Big)\Big]^{-1}\Big(-\bar{\boldsymbol{G}}(i\omega_\nu)\Big) + \boldsymbol{G}(i\omega_\nu) \qquad (\text{C.2})$$

$$\Rightarrow \quad \boldsymbol{G}(i\omega_\nu) = \Big[\mathbf{1} - \bar{\boldsymbol{G}}(i\omega_\nu)\Big(\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \bar{\boldsymbol{h}}\Big)\Big]^{-1}\bar{\boldsymbol{G}}(i\omega_\nu)$$

$$0 = -\frac{1}{\beta}\sum_\nu \underbrace{\Big\{\Big[\mathbf{1} - \bar{\boldsymbol{G}}(i\omega_\nu)\Big(\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \bar{\boldsymbol{h}}\Big)\Big]^{-1}\Big(-\bar{\boldsymbol{G}}(i\omega_\nu)\Big) + \boldsymbol{G}(i\omega_\nu) - \Big(\boldsymbol{G}(i\omega_\nu) - \bar{\boldsymbol{G}}(i\omega_\nu)\Big)\Big\}}_{=0 \quad \text{Eq. C.2}}$$

$$\Rightarrow \quad \frac{1}{\beta}\sum_\nu \boldsymbol{G}(i\omega_\nu)e^{i\hbar\omega_\nu\beta 0^+} = \frac{1}{\beta}\sum_\nu \bar{\boldsymbol{G}}(i\omega_\nu)e^{i\hbar\omega_\nu\beta 0^+} = \boldsymbol{\rho} \qquad (\text{C.3})$$

Whenever the Luttinger-Ward functional can be expressed directly by the density matrix, the self energy is independent of the Matsubara frequencies. In this case the Luttinger-Ward functional depends on the Green'sfunction only via the integral $\boldsymbol{\rho} = \frac{1}{\beta}\sum_\nu \boldsymbol{G}(i\omega_\nu)e^{i\hbar\omega_\nu\beta0^+}$. This is the case for the Hartree-Fock approximation and it is the case for the Density functional theory.

In the following, we assume that $\Phi_\beta^{LW,1}$ has this form, i.e. that it depends on the Green's function only via the density matrix.

Divided form

We rewrite the expression as

$$
Q_\beta^{\hat{W}}[\boldsymbol{\rho}] = \Phi_\beta^{LW,1}[\bar{\boldsymbol{G}},\hat{W}_1] + \underset{h_2'}{\mathrm{stat}}\,\underset{\boldsymbol{G},\boldsymbol{\Sigma}_2}{\mathrm{stat}}\Bigg\{\Phi_\beta^{LW,2}[\boldsymbol{G},\hat{W}_2]
$$
$$
-\frac{1}{\beta}\sum_\nu \mathrm{Tr}\Big\{\ln\Big[\boldsymbol{1} - \big(i\hbar\omega_\nu + \mu\big)\boldsymbol{1} - \bar{\boldsymbol{h}}\big)^{-1}\big(\boldsymbol{h}_2' + \boldsymbol{\Sigma}_2(i\omega_\nu) - \bar{\boldsymbol{h}}\big)\Big]
$$
$$
+(\boldsymbol{h}_2' + \boldsymbol{\Sigma}_2(i\omega_\nu) - \bar{\boldsymbol{h}})\boldsymbol{G}(i\omega_\nu) - \Big[\boldsymbol{G}(i\omega_\nu) - \big(i\hbar\omega_\nu + \mu\big)\boldsymbol{1} - \bar{\boldsymbol{h}}\big)^{-1}\Big]\big(\boldsymbol{h}_2' - \bar{\boldsymbol{h}}\big)\Big\}\Bigg\}
$$
$$
\text{(C.4)}
$$

The stationary conditions are

$$
\boldsymbol{\Sigma}_2(i\omega_\nu) = \frac{\beta\delta\Phi^{LW,2}}{\delta\boldsymbol{G}(i\omega_\nu)}
$$
$$
\boldsymbol{G}(i\omega_\nu) = \Big[\boldsymbol{1} - \bar{\boldsymbol{G}}(i\omega_\nu)\big(\boldsymbol{h}_2' + \boldsymbol{\Sigma}_2(i\omega_\nu) - \bar{\boldsymbol{h}}\big)\Big]^{-1}\bar{\boldsymbol{G}}(i\omega_\nu)
\qquad \text{(C.5)}
$$

We see that

$$
\boldsymbol{h}' = \boldsymbol{h}_2' + \frac{\beta\delta\Phi^{LW,1}}{\delta\boldsymbol{G}(i\omega_\nu)}
\qquad \text{(C.6)}
$$

leads to the same Green's function as in the former case. That is, both forms, i.e. Eq. C.1 and Eq. C.4, have the same stationary state. At the stationary state both forms have furthermore the same value.

No Matsubara sum with a stationary self energy

Here, I show that $Q_\beta^{\hat{W}}[\boldsymbol{\rho}] = \Phi_\beta^{LW}[\bar{\boldsymbol{G}},\hat{W}]$, if the Luttinger Ward functional only depends on the Greens function via the density matrix.

In that case, the self energy is frequency independent. All stationary conditions can be fulfilled by choosing

$$
\boldsymbol{h}' + \boldsymbol{\Sigma} = \bar{\boldsymbol{h}}
$$
$$
\boldsymbol{G}(i\omega_\nu) = \bar{\boldsymbol{G}}(i\omega_\nu)
\qquad \text{(C.7)}
$$

Thus we may decompose the correlation contribution into several terms of which only one has a frequency dependent self energy

Appendix D

Form of the Hartree Fock energy

. 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 (BPP-Eq.43)

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

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

$$Q_{X,\beta}^{\hat{W}}[\boldsymbol{\rho}] = \frac{1}{2} \sum_{m,n} f_m f_n \int d^3r \int d^3r' \frac{e^2 \left(\phi_m^*(\vec{r})\phi_n^*(\vec{r'})\phi_n(\vec{r'})\phi_m(\vec{r}) - \phi_m^*(\vec{r})\phi_n^*(\vec{r'})\phi_m(\vec{r'})\phi_n(\vec{r}) \right)}{4\pi\epsilon_0|\vec{r}-\vec{r'}|}$$

$$= \frac{1}{2} \sum_{a,b,c,d} \underbrace{\left(\sum_m \langle\pi_a|\phi_m\rangle f_m \langle\phi_m|\pi_b\rangle \right)}_{\rho_{a,b}} \underbrace{\left(\sum_n \langle\pi_c|\phi_n\rangle f_n \langle\phi_n|\pi_d\rangle \right)}_{\rho_{c,d}}$$

$$\times \int d^3r \int d^3r' \frac{e^2 \left(\chi_b^*(\vec{r})\chi_d^*(\vec{r'})\chi_c(\vec{r'})\chi_a(\vec{r}) - \chi_b^*(\vec{r})\chi_d^*(\vec{r'})\chi_a(\vec{r'})\chi_c(\vec{r}) \right)}{4\pi\epsilon_0|\vec{r}-\vec{r'}|} \tag{D.2}$$

$$= \frac{1}{2} \sum_{a,b,c,d} \rho_{a,b}\rho_{c,d} \left(U_{b,d,a,c} - U_{b,d,c,a} \right) \tag{D.3}$$

$$= \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \left(\rho_{d,a}\rho_{c,b} - \rho_{c,a}\rho_{d,b} \right) \tag{D.4}$$

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

$$\Psi(\vec{x}_1, \ldots, \vec{x}_n) = C \det|\boldsymbol{M}| = C \sum_{i_1,\ldots,i_N=1}^{N} \epsilon_{i_1,\ldots,i_N} \chi_{i_1}(\vec{x}_1) \cdots \chi_{i_N}(\vec{x}_N) \tag{D.5}$$

with $M_{i,j} = \chi_i(\vec{x}_j)$ and the normalization constant C. $\epsilon_{i_1,\ldots,i_N}$ is the fully antisymmetric tensor defined by $\epsilon_{1,2,\ldots,N} = 1$, 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.

Appendix E

Mixing of the self energy

E.1 Euler-Lagrange equations

In order to arrive at a reasonable mixing scheme, we explore a dynamics from a Lagrangian formalism. It is however important to note that this dynamics is useless, because there is no minimum prinicple, so that applying a friction will not lead to a stationary state.

$$m_\Sigma(\omega_\nu)\ddot{\Sigma}_{a,b}(i\omega_\nu) = -\beta\frac{\partial E}{\partial\Sigma^\dagger_{a,b}(i\omega_\nu)}$$

The equation for the self energy is a little complicated. Therefore we obtain it directly from the action principle.

$$\mathcal{L} = \frac{1}{\beta}\sum_\nu m_\Sigma\sum_{a,b}\left\{\dot{\Sigma}_{a,b}(i\omega_\nu)\dot{\Sigma}^\dagger_{b,a}(i\omega_\nu)\right\} - \Phi(\boldsymbol{G})$$

$$0 = \frac{1}{\beta}\sum_\nu m_\Sigma\sum_{a,b}\left\{\dot{\Sigma}^\dagger_{a,b}(i\omega_\nu)\delta\dot{\Sigma}_{b,a}(i\omega_\nu) + \dot{\Sigma}_{a,b}(i\omega_\nu)\delta\dot{\Sigma}^\dagger_{b,a}(i\omega_\nu)\right\}$$

$$-\frac{1}{\beta}\sum_\nu\sum_{a,b,c,d}\left[\frac{\beta\partial\Phi}{\partial G_{a,b}(i\omega_\nu)}\frac{\partial G_{a,b}(i\omega_\nu)}{\partial\Sigma_{c,d}(i\omega_\nu)}\delta\Sigma_{c,d}(i\omega_\nu) + \frac{\beta\partial\Phi}{\partial G^\dagger_{a,b}(i\omega_\nu)}\frac{\partial G^\dagger_{a,b}(i\omega_\nu)}{\partial\Sigma^\dagger_{c,d}(i\omega_\nu)}\delta\Sigma^\dagger_{c,d}(i\omega_\nu)\right]$$

$$= \frac{1}{\beta}\sum_\nu m_\Sigma\sum_{a,b}\left\{-\ddot{\Sigma}^\dagger_{a,b}(i\omega_\nu)\delta\Sigma_{b,a}(i\omega_\nu) - \ddot{\Sigma}_{a,b}(i\omega_\nu)\delta\Sigma^\dagger_{b,a}(i\omega_\nu)\right\}$$

$$-\frac{1}{\beta}\sum_\nu\sum_{a,b,c,d}\frac{\beta\partial\Phi}{\partial G_{a,b}(i\omega_\nu)}G_{a,c}(i\omega_\nu)\delta\Sigma_{c,d}(i\omega_\nu)G_{d,b}(i\omega_\nu)$$

$$-\frac{1}{\beta}\sum_\nu\sum_{a,b,c,d}\frac{\beta\partial\Phi}{\partial G^\dagger_{a,b}(i\omega_\nu)}G^\dagger_{a,c}(i\omega_\nu)\delta\Sigma^\dagger_{c,d}(i\omega_\nu)G^\dagger_{d,b}(i\omega_\nu) + \text{boundary terms at } t_1, t_2$$

$$= -\frac{1}{\beta}\sum_\nu\left[\sum_{a,b} m_\Sigma\ddot{\Sigma}_{a,b}(i\omega_\nu)\delta\Sigma^\dagger_{b,a}(i\omega_\nu) + \sum_{c,d}\left(\sum_{a,b}G^\dagger_{d,b}(i\omega_\nu)\frac{\beta\partial\Phi}{\partial G^\dagger_{a,b}(i\omega_\nu)}G^\dagger_{a,c}(i\omega_\nu)\right)\delta\Sigma^\dagger_{c,d}(i\omega_\nu)\right] + \text{c.c.}$$

$$= -\frac{1}{\beta}\sum_\nu\sum_{a,b}\left[m_\Sigma\ddot{\Sigma}_{a,b}(i\omega_\nu) + \sum_{c,d}G^\dagger_{a,c}(i\omega_\nu)\frac{\beta\partial\Phi}{\partial G^\dagger_{d,c}(i\omega_\nu)}G^\dagger_{d,b}(i\omega_\nu)\right]\delta\Sigma^\dagger_{b,a}(i\omega_\nu) + \text{c.c.}$$

Thus the equation of motion has the form

$$m_\Sigma \ddot{\Sigma}_{a,b}(i\omega_\nu) = -\sum_{c,d} G^\dagger_{a,c}(i\omega_\nu) \frac{\beta\partial\Phi}{\partial G^\dagger_{d,c}(i\omega_\nu)} G^\dagger_{d,b}(i\omega_\nu)$$

With $G(i\omega) = G^\dagger(-i\omega)$ and $\Sigma(i\omega) = \Sigma^\dagger(-i\omega)$, I obtain

$$m_\Sigma \ddot{\Sigma}^\dagger_{a,b}(i\omega_\nu) = -\sum_{c,d} G_{a,c}(i\omega_\nu) \frac{\beta\partial\Phi}{\partial G_{d,c}(i\omega_\nu)} G_{d,b}(i\omega_\nu)$$

E.2   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

$$G_{a,b}(i\omega_\nu) = \sum_n \frac{\langle\pi_a|\psi_n\rangle\langle\psi_n|\pi_b\rangle}{i\hbar\omega_\nu + \mu - \epsilon_n} = \int d\epsilon \frac{D_{a,b}(\epsilon)}{i\hbar\omega_\nu + \mu - \epsilon} \tag{E.1}$$

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

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

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 chemicall 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 allo expand the argument to multi-band situations. The

resulting Green's function has the form

$$
\begin{aligned}
G_{a,b}(i\omega_\nu) &= \int_{E-W/2}^{E+W/2} d\epsilon\, \frac{\frac{1}{W}\delta_{a,b}}{i\hbar\omega_\nu - \epsilon} \\
&= \frac{\delta_{a,b}}{W} \int_{E-W/2}^{E+W/2} d\epsilon\, \frac{-\epsilon - i\hbar\omega_\nu}{(\hbar\omega)^2 + \epsilon^2} \\
&= -\frac{\delta_{a,b}}{W} \int_{E-W/2}^{E+W/2} d\epsilon\, \frac{\epsilon + i\hbar\omega_\nu}{(\hbar\omega)^2 + \epsilon^2} \\
&= -\frac{\delta_{a,b}}{W} \int_{E-W/2}^{E+W/2} dx\, \frac{x + i\hbar\omega_\nu}{(\hbar\omega)^2 + x^2} \\
&= -\frac{\delta_{a,b}}{W} \left\{ \int_{E-W/2}^{E+W/2} dx\, \frac{x}{(\hbar\omega)^2 + x^2} + i\hbar\omega_\nu \int_{E-W/2}^{E+W/2} dx\, \frac{1}{(\hbar\omega)^2 + x^2} \right\} \\
&= -\frac{\delta_{a,b}}{W} \left\{ \frac{1}{2} \ln\left[(\hbar\omega)^2 + (E+W/2)^2\right] - \frac{1}{2} \ln\left[(\hbar\omega)^2 + (E-W/2)^2\right] \right. \\
&\qquad \left. + i\hbar\omega_\nu \left[\frac{1}{\hbar\omega}\mathrm{atan}\left(\frac{E+W/2}{\hbar\omega}\right) - \frac{1}{\hbar\omega}\mathrm{atan}\left(\frac{E-W/2}{\hbar\omega}\right) \right] \right\} \\
&= \frac{\delta_{a,b}}{W} \left\{ +\frac{1}{2} \ln\left[\frac{(\hbar\omega)^2 + (E-W/2)^2}{(\hbar\omega)^2 + (E+W/2)^2}\right] - i \left[\mathrm{atan}\left(\frac{E+W/2}{\hbar\omega}\right) - \mathrm{atan}\left(\frac{E-W/2}{\hbar\omega}\right) \right] \right\}
\end{aligned}
$$
(E.3)

We used the integral formula's from Bronstein[1]

The result as function of ω is shown in Fig. E.1 The real part is symmetric about the origin,
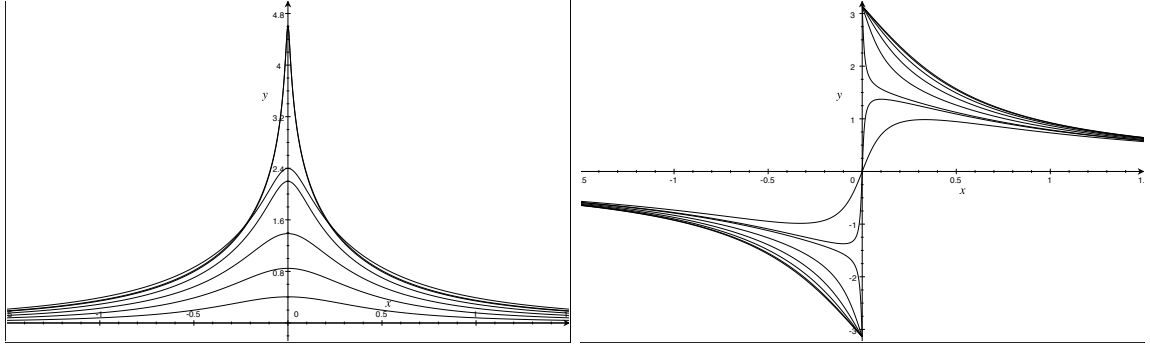

Fig. E.1: Negative real part (left) and negative imaginary part (right) of the Greens function with a constant density of states of width $W = 1$, centered at $E = \{0, 0.1, 0.2, 0.3, 0.49, 0.51, 0.6\}$.

while the imaginary part is antisymmetric. This is consistent with $\boldsymbol{G}(-i\omega_\nu) = \boldsymbol{G}^\dagger(-i\omega_\nu)$.

[1]Integral formula's:

$$
\int dx\, \frac{1}{a^2 + x^2} = \frac{1}{a}\mathrm{atan}\left(\frac{x}{a}\right)
$$
$$
\int dx\, \frac{x}{a^2 + x^2} = \frac{1}{2} \ln\left(a^2 + x^2\right)
$$
(E.4)

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.

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

- $\hbar\omega \to 0$:

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

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

  (E.7)

---

[2]

$$
\mathrm{atan}(x)=y \quad\Rightarrow\quad y=\tan(x)=\frac{\sin(x)}{\cos(x)}
$$

$$
x=\frac{1}{\frac{\pi}{2}-y}+O(y-\frac{\pi}{2})^2 \quad\Rightarrow\quad y\approx\frac{\pi}{2}-\frac{1}{x} \quad\text{for } x\to+\infty
$$

$$
x=\frac{1}{y+\frac{\pi}{2}}+O(y+\frac{\pi}{2})^2 \quad\Rightarrow\quad y\approx-\frac{\pi}{2}+\frac{1}{x} \quad\text{for } x\to-\infty \tag{E.5}
$$

[3]

$$
\ln\left[\frac{a+x}{b+x}\right]\approx\ln\left[\frac{a}{b}\right]+\left(\frac{1}{a}+\frac{1}{b}\right)x+O(x^2) \tag{E.6}
$$

- $\hbar\omega \to \infty$:

$$G_{a,b}(i\omega_\nu) = \frac{\delta_{a,b}}{W}\left\{+\frac{1}{2}\ln\left[\frac{(\hbar\omega)^2 + (E - W/2)^2}{(\hbar\omega)^2 + (E + W/2)^2}\right] - i\left[\text{atan}\left(\frac{E + W/2}{\hbar\omega}\right) - \text{atan}\left(\frac{E - W/2}{\hbar\omega}\right)\right]\right\}$$

$$\approx \frac{\delta_{a,b}}{W}\left\{-EW(\hbar\omega)^{-2} - iW(\hbar\omega)^{-1}\right\}$$

$$\approx \delta_{a,b}\left\{-E(\hbar\omega)^{-2} - i(\hbar\omega)^{-1}\right\} \tag{E.8}$$

Remark: We can approximate

$$\frac{1}{G_{a,a}^2} \approx \left(\frac{1}{W}\ln\left[\frac{E - W/2}{E + W/2}\right]\right)^{-2} + (\hbar\omega)^2 \tag{E.9}$$

E.3 Preconditioning

With this model and using $E = 0$ we obtain

$$\ddot{\Sigma}_{a,b}(i\omega_\nu) = \frac{1}{m_\Sigma(\omega_\nu)}\sum_{c,d} G_{a,c}^{corr}(i\omega_\nu)\left[\Sigma_{c,d}(i\omega) - \frac{\beta\partial\Phi^{LW}}{\partial G_{d,c}^{loc}(i\omega_\nu)}\right] G_{d,b}^{corr}(i\omega_\nu)$$

$$= \frac{1}{m_\Sigma(\omega_\nu)}\sum_{c,d}\left[-i\delta_{a,c}\frac{1}{W/2}\text{atan}(\frac{W/2}{\hbar\omega})\right]\left[\Delta\Sigma_{c,d}(i\omega) - \frac{\beta\partial\Delta\Phi^{LW}}{\partial G_{d,c}^{loc}(i\omega_\nu)}\right]$$

$$\left[-i\delta_{d,b}\frac{1}{W/2}\text{atan}(\frac{W/2}{\hbar\omega})\right]$$

$$= -\frac{1}{m_\Sigma(\omega_\nu)}\left(\frac{\text{atan}(\frac{W/2}{\hbar\omega})}{(W/2)}\right)^2\left[\Delta\Sigma_{a,b}(i\omega) - \frac{\beta\partial\Delta\Phi^{LW}}{\partial G_{b,a}^{loc}(i\omega_\nu)}\right.$$

Thus we make the ansatz for the ω-dependent mass for the self energy as

$$m_\Sigma(\omega_\nu) = A\left(\frac{\text{atan}(\frac{W/2}{\hbar\omega})}{(W/2)}\right)^2 \tag{E.10}$$

where A is a parameter. For $\omega \to \infty$ the mass approaches $A/(\hbar\omega)^2$. Near the origin, however the mass does not vanish, but it approaches a constant, namely $A(\frac{\pi}{W})^2$.

Let us define two parameters that determine the ω-dependent mass.

$$M_\Sigma = m_\Sigma(0) \tag{E.11}$$

$$C_\Sigma = \lim_{\omega\to\infty} m_\Sigma(\omega) \cdot (\hbar\omega)^2 \tag{E.12}$$

With these parameters we obtain

$$m_\Sigma(\omega_\nu) \approx M_\Sigma\left[\frac{2}{\pi}\text{atan}\left(\frac{\pi}{2}\sqrt{\frac{C_\Sigma}{M_\Sigma}}\frac{1}{\hbar\omega}\right)\right]^2 \tag{E.13}$$

An approximation for the mass (which is not used, though!) is

$$m_\Sigma(\omega_\nu) \approx m_\Sigma \left[\left(\frac{2}{\pi W/2} \right)^2 + (\hbar\omega)^2 \right]^{-1} = C_\Sigma \left[\frac{C_\Sigma}{M_\Sigma} + (\hbar\omega)^2 \right]^{-1} \tag{E.14}$$

which has the same value at the origin and the same tail for $\omega \to \infty$. At the origin it does not have the kink of the original function but remains larger.

The mass for the low frequency region is M_Σ and the mass of the high-frequency region is $C_\Sigma/(\hbar\omega)^2$. The high-frequency tail of the Greens function is system independent, which allows us to determine a canonical value for C_Σ, that only depends in the time step. If the period of the high-frequency tail shall be 10 time steps, C_Σ should have the value $C_\Sigma = \left(\frac{10\Delta t}{2\pi} \right)^2 =$, which is about 250 for $\Delta t = 10$.

If the period is fixed, we can determine also the optimum friction as $a_{opt} = \frac{\alpha_{opt}}{2} \Delta t = 2\pi \frac{\Delta t}{T}$ which is with our choices $a_{opt} = \frac{2\pi}{10}$. It is not adviseable tio choose such a factor, because it would overdamp the low-frequency region.

E.4   Bloch representation

Bibliography

[1] Peter E. Blöchl, Thomas Pruschke, and Michael Potthoff. Density-matrix functionals from green's functions. Phys. Rev. B, 88:205139, Nov 2013. doi: $10.1103/\mathrm{PhysRevB}.88.205139$. URL http://link.aps.org/doi/10.1103/PhysRevB.88.205139.

[2] J. M. Luttinger and J. C. Ward. Ground-state energy of a many-fermion system ii. Phys. Rev., 118:1417, 1960. doi: $10.1103/\mathrm{PhysRev}.118.1417$. URL http://link.aps.org/doi/10.1103/PhysRev.118.1417.

[3] N.D. Mermin. Thermal properties of the inhomogeneous electron gas. Physical Review, 137:A1441, 1965. doi: $10.1103/\mathrm{PhysRev}.137.\mathrm{A}1441$. URL http://link.aps.org/doi/10.1103/PhysRev.137.A1441.

[4] P. E. Blöchl, C. F. J. Walther, and Th. Pruschke. Method to include explicit correlations into density-functional calculations based on density-matrix functional theory. Phys. Rev. B, 84:205101, 2011. doi: $10.1103/\mathrm{PhysRevB}.84.205101$. URL http://link.aps.org/doi/10.1103/PhysRevB.84.205101.

[5] Nils Erik Dahlen, Robert van Leeuwen, and Ulf von Barth. Variational energy functionals of the green function and of the density tested on molecules. Phys. Rev. A, 73:012511, Jan 2006. doi: $10.1103/\mathrm{PhysRevA}.73.012511$. URL http://link.aps.org/doi/10.1103/PhysRevA.73.012511.