The DMFT object of the CP-PAW code

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Chapter 1

Purpose and theoretical background

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

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

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

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

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 $\text{stat}_x Y$, we denote the stationary condition, which requires that the partial derivatives of Y with respect to the variable x is obeyed. It explicitly 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. 33)

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

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**

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

where $Q_\beta^{\hat{W}}$ is the density-matrix functional without the entropy contribution of a non-interacting electron gas with the same density matrix. The entropy contribution is taken care of with the **Mermin functional**[3] to describe DFT calculations with electrons at finite temperature.

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

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

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

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 a density-functional contribution $Q^{DFT,\hat{W}}[\hat{\rho}]$ acting as double counting term and a screened Hartree-Fock contribution $Q^{HF,\hat{W}}[\hat{\rho}]$ described later, we obtain

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HELMHOLTZ POTENTIAL

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

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

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

$$\left. - \mu \Big[\sum_n f_n - N \Big] - \sum_{m,n} \Lambda_{m,n} \Big(\langle \psi_n | \psi_m \rangle - \delta_{m,n} \Big) \right\} \tag{1.10}$$

where

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

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

$$\rho_{a,b} = \sum_n \langle \pi_a | \psi_n \rangle f_n \langle \psi | \pi_b \rangle \tag{1.11}$$

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)

   $$Q^{HF,\hat{W}}_\beta[\boldsymbol{\rho}] = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \Big[\rho_{d,a}\rho_{c,b} - \rho_{c,a}\rho_{d,b} \Big] \tag{1.12}$$

   (See also appendix D on p.46.)

   The first term, E_H, is the **Hartree energy** and the second term, E_X, is the **exchange**

energy

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

$$E_X = -\frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \rho_{c,a} \rho_{d,b} \tag{1.13}$$

For the Hartree Fock contribution 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** $Q_\beta^{DFT,\hat{W}_1}$:

$$Q^{DFT,\hat{W}}[\hat{\rho}] = \frac{1}{2} \int d^3r \int d^3r' \frac{e^2 n(\vec{r})n(\vec{r}')}{4\pi\epsilon_0 |\vec{r}-\vec{r}'|} + \underbrace{E_{xc}[n] + T_s[n] - \text{Tr}[\frac{\hat{\vec{p}}^2}{2m_e}\hat{\rho}]}_{U_{xc}}, \quad (1.14)$$

where $n(r) = \langle \vec{r} | \hat{\rho} | \vec{r} \rangle$. U_{xc} is the interaction part at full interaction strength.

In practice the we use E_{xc} instead of U_{xc}

The term $Q_\beta^{DFT,\hat{W}_1}$ describes the DFT contribution when the interaction is limited to $\hat{W}_1$. The basic ideas behind the double-counting correction have been described in a previous paper[4].

Both, the Hartree-Fock term and the DFT double-counting term contain the same Hartree energy, which cancels exactly.

3. a **dynamic-correlation correction**

$$Q_\beta^{dyn,\hat{W}_2} \overset{\text{def}}{=} Q_\beta^{\hat{W}_2} - Q_\beta^{HF,\hat{W}_2} \tag{1.15}$$

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. 1.10: 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 $|\chi_a\rangle$.

$$\left[\frac{\hat{\vec{p}}^2}{2m_e} + v_{ext} + v_{Hartree} + v_{xc} + \sum_{a,b}|\pi_a\rangle\left(\frac{\delta Q_{HF,\beta}^{\hat{W}_1}}{\delta\rho_{a,b}} - \frac{\delta Q_{DFT,\beta}^{\hat{W}_1}}{\delta\rho_{a,b}} + \frac{\delta Q_{dyn,\beta}^{\hat{W}_2}}{\delta\rho_{a,b}}\right)\langle\pi_b|\right]|\psi_n\rangle$$
$$= \sum_m |\psi_m\rangle\Lambda_{m,n}\frac{1}{f_n} \tag{1.16}$$

$$\langle\psi_n|\left[\frac{\hat{\vec{p}}^2}{2m_e} + v_{ext} + v_{Hartree} + v_{xc} + \sum_{a,b}|\pi_a\rangle\left(\frac{\delta Q_{HF,\beta}^{\hat{W}_1}}{\delta\rho_{a,b}} - \frac{\delta Q_{DFT,\beta}^{\hat{W}_1}}{\delta\rho_{a,b}} + \frac{\delta Q_{dyn,\beta}^{\hat{W}_2}}{\delta\rho_{a,b}}\right)\langle\pi_b|\right]|\psi_n\rangle$$
$$- \mu + \frac{1}{\beta}\ln\left[\frac{f_n}{1-f_n}\right] = 0 \tag{1.17}$$

These equations specify the optimum density matrix as

EQUILIBRIUM DENSITY MATRIX

$$\hat{\rho} = \left[\hat{1} + e^{\beta(\hat{h}_{eff}-\mu)}\right]^{-1} \tag{1.18}$$

with the effective Hamiltonian

$$\hat{h}_{eff} = \frac{\hat{\vec{p}}^2}{2m_e} + v_{ext} + v_{Hartree} + v_{xc} + \sum_{a,b}|\pi_a\rangle\left(\frac{\delta Q_{HF,\beta}^{\hat{W}_1}}{\delta\rho_{a,b}} - \frac{\delta Q_{DFT,\beta}^{\hat{W}_1}}{\delta\rho_{a,b}} + \frac{\delta Q_{dyn,\beta}^{\hat{W}_2}}{\delta\rho_{a,b}}\right)\langle\pi_b|$$
$$\tag{1.19}$$

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

Eq. 1.18 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 $Q_{\text{dyn},\beta}^{\hat{W}}$ is simply the difference of the complete term minus the Hartree-Fock contribution.

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

$$= \underset{\boldsymbol{h}'_{dyn}}{\text{stat}}\ \underset{\boldsymbol{G},\boldsymbol{\Sigma}_{dyn}}{\text{stat}}\ \Big\{\Phi_{\beta}^{LW}[\boldsymbol{G},\hat{W}_2] - \Phi_{\beta}^{HF,LW}[\boldsymbol{G},\hat{W}_2]$$

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

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

$$(1.20)$$

We introduce a new variable

$$\boldsymbol{\Gamma} \stackrel{\text{def}}{=} \bar{\boldsymbol{h}} - \boldsymbol{h}'_{dyn} \tag{1.21}$$

so that

DYNAMIC CORRELATION CORRECTION

$$Q_{dyn,\beta}^{\hat{W}_2}[\boldsymbol{\rho}] = \underset{\boldsymbol{\Gamma}}{\text{stat}}\ \underset{\boldsymbol{G},\boldsymbol{\Sigma}_{dyn}}{\text{stat}}\ \Big\{\Phi_{\beta}^{LW}[\boldsymbol{G},\hat{W}_2] - \Phi_{\beta}^{HF,LW}[\boldsymbol{G},\hat{W}_2]$$

$$-\frac{1}{\beta}\sum_{\nu}\text{Tr}\Big\{\ln\Big[\mathbf{1} - \Big((i\hbar\omega_{\nu}+\mu)\mathbf{1}-\bar{\boldsymbol{h}}\Big)^{-1}\Big(\boldsymbol{\Sigma}_{dyn}(i\omega_{\nu})-\boldsymbol{\Gamma}\Big)\Big]$$

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

$$(1.22)$$

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 $\boldsymbol{h}'_{dyn}$ and $\boldsymbol{\Sigma}_{dyn}$ 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 $Q_{\text{dyn},\beta}^{\hat{W}_2}[\boldsymbol{\rho}]$ in all detail to avoid using inappropriate assumptions.

In order to simplify the discussion we introduce the non-interacting density matrix, which produces a given density matrix as

$$\bar{\boldsymbol{G}}(i\omega_\nu) = \left[(i\hbar\omega_\nu + \mu)\boldsymbol{1} - \bar{\boldsymbol{h}}\right]^{-1} \tag{1.23}$$

with $\bar{\boldsymbol{h}}$ defined in Eq. 1.5 as a functional of the density matrix.

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

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

$$
\begin{aligned}
dQ_\beta^{dyn,\hat{W}_2} = -\frac{1}{\beta}\sum_\nu \mathrm{Tr}\Bigg\{ &\left(-\frac{\beta\delta\Phi_\beta^{LW}}{\delta\boldsymbol{G}(i\omega_\nu)} + \frac{\beta\delta\Phi_{\beta,HF}^{LW}}{\delta\boldsymbol{G}(i\omega_\nu)}\right)d\boldsymbol{G} \\
&+ \left[\boldsymbol{1} - \bar{\boldsymbol{G}}\left(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma}\right)\right]^{-1}\left(-d\bar{\boldsymbol{G}}(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma}) - \bar{\boldsymbol{G}}(d\boldsymbol{\Sigma}_{dyn} - d\boldsymbol{\Gamma})\right) \\
&+ (d\boldsymbol{\Sigma}_{dyn} - \underbrace{d\boldsymbol{\Gamma}}_{(A)})\boldsymbol{G} + (\boldsymbol{\Sigma}_{dyn} - \underbrace{\boldsymbol{\Gamma}}_{(B)})d\boldsymbol{G} + (\underbrace{d\boldsymbol{G}}_{(B)} - d\bar{\boldsymbol{G}})\boldsymbol{\Gamma} + (\underbrace{\boldsymbol{G}}_{(A)} - \bar{\boldsymbol{G}})d\boldsymbol{\Gamma}\Bigg\} \\
= -\frac{1}{\beta}\sum_\nu \mathrm{Tr}\Bigg\{ &\left[\boldsymbol{\Sigma}_{dyn} - \frac{\beta\delta\Phi_\beta^{LW}}{\delta\boldsymbol{G}(i\omega_\nu)} + \frac{\beta\delta\Phi_{\beta,HF}^{LW}}{\delta\boldsymbol{G}(i\omega_\nu)}\right]d\boldsymbol{G} \\
&+ \left[-\left[\boldsymbol{1} - \bar{\boldsymbol{G}}\left(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma}\right)\right]^{-1}\bar{\boldsymbol{G}} + \boldsymbol{G}\right]d\boldsymbol{\Sigma}_{dyn} \\
&+ \left[\left[\boldsymbol{1} - \bar{\boldsymbol{G}}\left(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma}\right)\right]^{-1}\bar{\boldsymbol{G}} - \bar{\boldsymbol{G}}\right]d\boldsymbol{\Gamma} \\
&- \left[\bar{\boldsymbol{G}}(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma})\left[\boldsymbol{1} - \bar{\boldsymbol{G}}\left(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma}\right)\right]^{-1}\bar{\boldsymbol{G}}\right]d\bar{\boldsymbol{h}} \\
&- d\bar{\boldsymbol{G}}\boldsymbol{\Gamma}\Bigg\} \\
= -\frac{1}{\beta}\sum_\nu \mathrm{Tr}\Bigg\{ &\left[\boldsymbol{\Sigma}_{dyn} - \frac{\beta\delta\Phi_\beta^{LW}}{\delta\boldsymbol{G}(i\omega_\nu)} + \frac{\beta\delta\Phi_{\beta,HF}^{LW}}{\delta\boldsymbol{G}(i\omega_\nu)}\right]d\boldsymbol{G} \\
&+ \left[\boldsymbol{G} - \left[\boldsymbol{1} - \bar{\boldsymbol{G}}\left(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma}\right)\right]^{-1}\bar{\boldsymbol{G}}\right]d\boldsymbol{\Sigma}_{dyn} \\
&+ \left[\left[\boldsymbol{1} - \bar{\boldsymbol{G}}\left(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma}\right)\right]^{-1}\bar{\boldsymbol{G}} - \bar{\boldsymbol{G}}\right]\left(d\boldsymbol{\Gamma} - d\bar{\boldsymbol{h}}\right) \\
&+ \underbrace{\left[\left[\boldsymbol{1} - \bar{\boldsymbol{G}}\left(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma}\right)\right]^{-1}\bar{\boldsymbol{G}} - \bar{\boldsymbol{G}} - \bar{\boldsymbol{G}}(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma})\left[\boldsymbol{1} - \bar{\boldsymbol{G}}\left(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma}\right)\right]^{-1}\bar{\boldsymbol{G}}\right]}_{=0}d\bar{\boldsymbol{h}} \\
&- d\bar{\boldsymbol{G}}\boldsymbol{\Gamma}\Bigg\} \tag{1.25}
\end{aligned}
$$

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[1]

$$\boldsymbol{A}\boldsymbol{A}^{-1} = \boldsymbol{1} \quad \Rightarrow \quad \left(d\boldsymbol{A}\right)\boldsymbol{A}^{-1} + \boldsymbol{A}\left(d\boldsymbol{A}^{-1}\right) = 0 \quad \Rightarrow \quad d\boldsymbol{A}^{-1}\right) = -\boldsymbol{A}^{-1}\left(d\boldsymbol{A}\right)\boldsymbol{A}^{-1} \tag{1.24}$$

Thus we obtain

$$
\begin{aligned}
d\mathcal{Q}_{\beta}^{dyn,\hat{W}_2} = &-\frac{1}{\beta}\sum_{\nu}\mathrm{Tr}\Bigg\{\left[\boldsymbol{\Sigma}_{dyn}(i\omega_{\nu}) - \frac{\beta\delta\Phi_{\beta}^{LW}}{\delta\boldsymbol{G}(i\omega_{\nu})} + \frac{\beta\delta\Phi_{\beta,HF}^{LW}}{\delta\boldsymbol{G}(i\omega_{\nu})}\right]d\boldsymbol{G}(i\omega_{\nu})\Bigg\} \\
&-\frac{1}{\beta}\sum_{\nu}\mathrm{Tr}\Bigg\{\left[\boldsymbol{G}(i\omega_{\nu}) - \left[\boldsymbol{1} - \bar{\boldsymbol{G}}(i\omega_{\nu})\left(\boldsymbol{\Sigma}_{dyn}(i\omega_{\nu}) - \boldsymbol{\Gamma}\right)\right]^{-1}\bar{\boldsymbol{G}}(i\omega_{\nu})\right]d\boldsymbol{\Sigma}_{dyn}(i\omega_{\nu})\Bigg\} \\
&-\frac{1}{\beta}\sum_{\nu}\mathrm{Tr}\Bigg\{\left[\boldsymbol{1} - \bar{\boldsymbol{G}}\left(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma}\right)\right]^{-1}\bar{\boldsymbol{G}} - \bar{\boldsymbol{G}}\Bigg\}\left(d\boldsymbol{\Gamma} - d\bar{\boldsymbol{h}}\right) \\
&+\mathrm{Tr}\left[\boldsymbol{\Gamma}d\boldsymbol{\rho}\right]
\end{aligned}
\tag{1.26}
$$

Thus the stationary conditions are

- the definition of the dynamic self energy

$$
\boldsymbol{\Sigma}_{dyn}(i\omega_{\nu}) = \frac{\beta\delta\Phi_{\beta}^{LW}}{\delta\boldsymbol{G}(i\omega_{\nu})} - \frac{\beta\delta\Phi_{\beta,HF}^{LW}}{\delta\boldsymbol{G}(i\omega_{\nu})}
\tag{1.27}
$$

- Dyson's equation defining the Green's function

$$
\boldsymbol{G}(i\omega_{\nu}) = \left[\boldsymbol{1} - \bar{\boldsymbol{G}}(i\omega_{\nu})\left(\boldsymbol{\Sigma}_{dyn}(i\omega_{\nu}) - \boldsymbol{\Gamma}\right)\right]^{-1}\bar{\boldsymbol{G}}(i\omega_{\nu})
\tag{1.28}
$$

- and the density-matrix constraint

$$
\frac{1}{\beta}\sum_{\nu}\mathrm{Tr}\Bigg\{\left[\boldsymbol{1} - \bar{\boldsymbol{G}}\left(\boldsymbol{\Sigma}_{dyn} - \boldsymbol{\Gamma}\right)\right]^{-1}\bar{\boldsymbol{G}} - \bar{\boldsymbol{G}}\Bigg\}
\tag{1.29}
$$

If the stationary conditions are obeyed, the derivative with respect to the density matrix is simply the matrix $\boldsymbol{\Gamma}$.

DENSITY-MATRIX DERIVATIVE OF THE DYNAMIC CORRELATION CORRECTION

$$
\frac{\delta\boldsymbol{Q}_{\beta}^{dyn,\hat{W}_2}[\boldsymbol{\rho}]}{\delta\boldsymbol{\rho}} = \boldsymbol{\Gamma}
\tag{1.30}
$$

The matrix $\boldsymbol{\Gamma}$ 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   Local orbitals

1.4.1   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

$$|\psi_n\rangle = \sum_a |\chi_a\rangle\langle\pi_a|\psi_n\rangle + |\delta\psi_n\rangle \tag{1.31}$$

where the projector functions obey the bi-orthogonality condition

$$\langle\pi_a|\chi_b\rangle = \delta_{a,b} \ , \tag{1.32}$$

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

$$\langle\pi_a|\delta\psi_n\rangle = 0 \tag{1.33}$$

1.4.2   Energy correction

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

$$Q_\beta^{\hat{W}}[\hat{\rho}] = Q_\beta^{\hat{W}}\left[\sum_n |\psi_n\rangle f_n \langle\psi_n|\right] = Q_\beta^{\hat{W}}\left[\sum_{a,b} |\chi_a\rangle \underbrace{\left(\sum_n \langle\pi_a|\psi_n\rangle f_n \langle\psi_n|\pi_b\rangle\right)}_{\rho_{a,b}} \langle\chi_b|\right] \tag{1.34}$$

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

$$Q_\beta^{\hat{W}}[\boldsymbol{\rho}] \stackrel{\text{def}}{=} Q_\beta^{\hat{W}}\left[\sum_{a,b} |\chi_a\rangle\rho_{a,b}\langle\chi_b|\right] \tag{1.35}$$

The derivatives are

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

$$\frac{\delta Q_\beta^{\hat{W}}}{\delta f_n} = \sum_{a,b} \langle\psi_n|\pi_b\rangle\langle\chi_b|\frac{\delta Q_\beta^{\hat{W}}}{\delta\hat{\rho}}|\chi_a\rangle\langle\pi_a|\psi_n\rangle \tag{1.36}$$

In the following we introduce a new symbol, namely

$$\frac{\delta Q_\beta^{\hat{W}}}{\delta\rho_{b,a}} \stackrel{\text{def}}{=} \langle\chi_b|\frac{\delta Q_\beta^{\hat{W}}}{\delta\hat{\rho}}|\chi_a\rangle \tag{1.37}$$

1.4.3   U-tensor

The interaction $\hat{W}$

$$\hat{W} = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \hat{c}_a^\dagger \hat{c}_b^\dagger \hat{c}_c \hat{c}_d \tag{1.38}$$

is expressed by the **U-tensor**

$$U_{a,b,c,d} = \sum_{\sigma,\sigma'} \int d^3r \int d^3r \, \frac{e^2 \chi_a^*(\vec{r},\sigma) \chi_b^*(\vec{r'},\sigma') \chi_c(\vec{r},\sigma) \chi_d(\vec{r'},\sigma')}{4\pi\epsilon_0 |\vec{r} - \vec{r'}|} \tag{1.39}$$

.

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.

1.4.4   Non-orthonormal orbitals

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

$$\left[\hat{c}_a^\dagger, \hat{c}_b\right]_+ = \langle \pi_b | \pi_a \rangle \tag{1.40}$$

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 $\boldsymbol{AA}^{-1}$ 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

$$|\psi_n\rangle = \sum_a |\chi_a\rangle \langle \pi_a | \psi_n \rangle \tag{1.41}$$

Then the Green's function has the form

$$\hat{G} = \sum_{m,n} |\psi_m\rangle \langle \psi_m | \hat{G} | \psi_n \rangle \langle \psi_n | = \sum_{a,b} |\chi_a\rangle \underbrace{\langle \pi_a | \hat{G} | \pi_b \rangle}_{G_{a,b}} \langle \chi_b | \tag{1.42}$$

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.

1.4.5   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

$$|\kappa_n\rangle \overset{\text{def}}{=} \sum_a |\chi_a\rangle \langle \pi_a | \psi_n \rangle \tag{1.43}$$

as complete orthonormal basisset.

Thus,

$$\delta_{m,n} = \langle \kappa_m | \kappa_n \rangle = \sum_{a,b} \langle \psi_m | \pi_a \rangle \langle \chi_a | \chi_b \rangle \langle \pi_b | \psi_n \rangle \tag{1.44}$$

and

$$\hat{1} = \sum_{a,b,n} |\chi_a \rangle \langle \pi_a | \psi_n \rangle \langle \psi_n | \pi_b \rangle \langle \chi_b | \tag{1.45}$$

I introduce

$$S_{a,b}^{-1} \stackrel{\text{def}}{=} \sum_n \langle \pi_a | \psi_n \rangle \langle \psi_n | \pi_b \rangle \tag{1.46}$$

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 $|\kappa_n \rangle$.

1.5   Variational parameters for the Green's function

Representation of the Green's function

The Green's function is expressed in a Bloch representation.

$$\mathbf{G}_{\vec{t},\vec{t}'}(i\omega_\nu) = \frac{1}{N_k} \sum_{\vec{k}} \mathbf{G}_{\vec{k}}(i\omega_\nu) e^{i\vec{k}(\vec{t}-\vec{t}')} \tag{1.47}$$

where N_k is the number of $\vec{k}$ points on the chosen numerical grid in the first unit cell. $\mathbf{G}_{\vec{t},\vec{t}'}$ is a matrix with dimension equal to the number of local orbitals in a given unit cell, and it connects these orbitals in two unit cells specified by the translation vectors $\vec{t}$ and $\vec{t}$. Check the sign in the exponential!

The Green's function can be expressed by the values at the positive Matsubara frequencies up to a maximum value ω_x and the first three Laurent expansion terms.

$$\mathbf{G}_{\vec{k}}(i\omega_\nu) = \begin{cases} \mathbf{G}_{\vec{k}}(i\omega_\nu) & \text{for } 0 < \omega_\nu \leq \omega_x \\ \mathbf{G}_{\vec{k}}^{\dagger}(-i\omega_\nu) & \text{for } -\omega_x \leq \omega_\nu < 0 \\ \sum_{j=1,2} \frac{1}{(i\hbar\omega_\nu)^j} \mathcal{G}_{\vec{k}}^{(j)} & \text{for } |\omega_\nu| > \omega_x \end{cases} \tag{1.48}$$

where $\mathcal{G}_{\vec{k}}^{(0)} = \mathbf{S}_{\vec{k}}^{-1}$.

Variational parameters

However, we treat this representation as the derived form. By employing the stationary condition for the self energy, we obtain Dyson's equation, which expresses the Green's function by the self energy. Using the same stationary condition with respect to the Laurent expansion terms of the self energy, one obtaines a consisent definition for the Laurent expansion terms of the Green's function.

The parameters entering in the representation Eq. 1.48 of the Greens function given above are obtained from the fundamental variables as.

$$
\begin{aligned}
\boldsymbol{G}_{\vec{k}}(i\omega_\nu) \quad &= \quad \left[(i\hbar\omega_\nu + \mu)\boldsymbol{S}_{\vec{k}} - \bar{\boldsymbol{h}}_{\vec{k}} - \left[\boldsymbol{\Sigma}_{dyn}(i\omega_\nu) - \boldsymbol{\Gamma}_k\right]\right]^{-1} \qquad \text{for } 0 < \omega_\nu \leq \omega_x \\
\mathcal{G}_{\vec{k}}^{(1)} \;\overset{\text{Eq. }B.26}{=}\; &\boldsymbol{S}_{\vec{k}}^{-1} \\
\mathcal{G}_{\vec{k}}^{(2)} \;\overset{\text{Eq. }B.26}{=}\; &-\boldsymbol{S}_{\vec{k}}^{-1}\left[\mu\boldsymbol{S}_{\vec{k}} - \boldsymbol{h}_{\vec{k}} + \boldsymbol{\Gamma}_{\vec{k}} - \mathcal{S}_{dyn}^{(0)}\right]\boldsymbol{S}_{\vec{k}}^{-1}
\end{aligned} \tag{1.49}
$$

The leading order in the Laurent expansion of the Green's function is not a dynamical variable, because it does not depend on any of the variational parameters in the definition of the dynamical correlation.

Thus the self energy is treated as variable that is iteratively optimized. This optimization is guided by the derivative with respect to the Green's function.

$$
\boldsymbol{\Sigma}_{dyn}(+) = \boldsymbol{\Sigma}_{dyn}(0) + \alpha\left[\frac{\beta\delta\Phi^{LW,dyn}}{\delta\boldsymbol{G}}\bigg|_{\boldsymbol{G}[\boldsymbol{\Sigma}(0)]} - \boldsymbol{\Sigma}_{dyn}(0)\right] \tag{1.50}
$$

The fundamental variables are

- the site-diagonal and therefore k-independent dynamic self energy $\boldsymbol{\Sigma}_{dyn,R}(i\omega_\nu) = \boldsymbol{\Sigma}_R(i\omega_\nu) - \boldsymbol{\Sigma}_{HF,R}(i\omega_\nu)$. The self energy for positive and negative Matsubara frequencies are related by a hermitean conjugation $\boldsymbol{\Sigma}_{dyn,R}(-i\omega_\nu) = \boldsymbol{\Sigma}_{dyn,R}^{\dagger}(i\omega_\nu)$

- the Hamiltonian $\bar{\boldsymbol{h}}_{\vec{k}}$, which produces the one-particle reduced density matrix as $\boldsymbol{\rho}_{\vec{k}} = \left[\boldsymbol{1} + e^{\beta(\bar{\boldsymbol{h}}_{\vec{k}} - \mu\boldsymbol{1})}\right]^{-1}$.

- the static correction $\boldsymbol{\Gamma}_{\vec{k}}$, that ensures that the original density matrix is obtained from the Green's function, if the dynamic self energy is taken into account.

From the site-diagonal self energies, we obtain as direct sum. The self energy that connects each orbital in a unit cell with each other.

$$
\boldsymbol{\Sigma}_{dyn}(i\omega_\nu) = \bigoplus_{R \in \text{unit cell}} \boldsymbol{\Sigma}_{dyn,R}(i\omega_\nu) \tag{1.51}
$$

This self energy is independent of $\vec{k}$.

Local Green's function

Then local Green's function is obtains as Brillouin-zone integral

$$
\boldsymbol{G}_R(i\omega_\nu) = \frac{1}{N_k}\sum_{\vec{k}}\boldsymbol{P}_R\boldsymbol{G}_{\vec{k}}(i\omega_\nu)\boldsymbol{P}_R^{\dagger} \tag{1.52}
$$

where $\boldsymbol{P}_R$ is a non-square diagonal matrix, which projects the matrix onto the subspace of orbitals on a given site.

1.5.1   High-Freqency expansion of Matsubara sum

A consistent choice for the Laurent expansion for the Green's function and self energy has turned out to be non-trivial. The most consistent approach apepars to write down an approximate form for the high-frequency tail of the Matsubara sum in the dynanic correlation correction, and to extract the Laurent expansion coefficients from the stationary conditions.

With the high-frequency tail, we obtain

$$
\ln\left[\mathbf{1} - \bar{\mathbf{G}}\left(\mathbf{\Sigma} - \mathbf{\Gamma}\right)\right] + \left(\mathbf{\Sigma} - \mathbf{\Gamma}\right)\mathbf{G} + \left(\mathbf{G} - \bar{\mathbf{G}}\right)\mathbf{\Gamma}
$$

$$
= -\bar{\mathbf{G}}\left(\mathbf{\Sigma} - \mathbf{\Gamma}\right) - \sum_{n=2}^{\infty}\frac{1}{n}\left[\bar{\mathbf{G}}\left(\mathbf{\Sigma} - \mathbf{\Gamma}\right)\right]^{n} + \left(\mathbf{\Sigma} - \mathbf{\Gamma}\right)\mathbf{G} + \left(\mathbf{G} - \bar{\mathbf{G}}\right)\mathbf{\Gamma}
$$

$$
= \left(\mathbf{G} - \bar{\mathbf{G}}\right)\left(\mathbf{\Sigma} - \mathbf{\Gamma}\right) - \sum_{n=2}^{\infty}\frac{1}{n}\left[\bar{\mathbf{G}}\left(\mathbf{\Sigma} - \mathbf{\Gamma}\right)\right]^{n} + \underbrace{\left(\mathbf{G} - \bar{\mathbf{G}}\right)\mathbf{\Gamma}}_{\rightarrow\text{number constraint}}
$$

$$
= \left(\mathbf{G} - \bar{\mathbf{G}}\right)\mathbf{\Sigma} - \sum_{n=2}^{\infty}\frac{1}{n}\left[\bar{\mathbf{G}}\left(\mathbf{\Sigma} - \mathbf{\Gamma}\right)\right]^{n} \tag{1.53}
$$

Side Remark:[2]

---

[2]In order to rationalize the form of the Kadanoff-Baym functional, we look for a functional X , that provides Dyson's equation as functional derivative of the self energy. We do not make any reference regarding the representation of the Green's function. We onmly require $d\text{Tr}[A^n] = nA^{n-1}dA$ and $\ln[1 - A] = -\sum_{n=1}^{\infty}\frac{1}{n}A^n$.

We start with Dyson's equation

$$
G = G_0 + G_0\Sigma G = G_0 + \sum_{n=1}G_0\left(\Sigma G_0\right)^n = \sum_{n=0}G_0\left(\Sigma G_0\right)^n \tag{1.54}
$$

We define a functional $X[G, \Sigma$ so that

$$
\left.\frac{\delta X[G, \Sigma]}{\delta\Sigma}\right|_{G_0} = G - \sum_{n=0}G_0\left(\Sigma G_0\right)^n \tag{1.55}
$$

The functional has the form

$$
X[G, \Sigma] \quad = \quad \text{Tr}\left[G\Sigma - \sum_{n=0}\frac{1}{n+1}\left(\Sigma G_0\right)^{n+1}\right] = \text{Tr}\left[G\Sigma - \sum_{n=1}\frac{1}{n}\left(\Sigma G_0\right)^{n}\right]
$$

$$
\overset{\text{Eq. } B.16}{=} \text{Tr}\left[G\Sigma + \ln\left(1 - \Sigma G_0\right)\right] \tag{1.56}
$$

The functional derivative of X with respect to the Green's function is the self energy

$$
\left.\frac{\delta X[G, \Sigma]}{\delta G}\right|_{G_0} = \Sigma \tag{1.57}
$$

Thus, if one finds a functional $\Phi[G]$ of the Green's function alone, which produces the self energy as derivative,

$$
\frac{\delta\Phi[G]}{\delta G} = \Sigma \tag{1.58}
$$

we can produce a functional

$$
\Phi[G] - X[G, \Sigma] \tag{1.59}
$$

which produces Dyson's equation as stationary condition with respect to Green's function and self energy. The connection to the total energy is, however, no clear from this argument.

Now we insert the Laurent expansions into Eq. 1.53

$$\left(\boldsymbol{G} - \bar{\boldsymbol{G}}\right)\boldsymbol{\Sigma} - \sum_{n=2}^{\infty} \frac{1}{n}\left[\bar{\boldsymbol{G}}\left(\boldsymbol{\Sigma} - \boldsymbol{\Gamma}\right)\right]^n$$

$$= \left[\sum_{j=2}^{\infty} \frac{1}{(i\hbar\omega_\nu)^j}\left(\mathcal{G}^{(j)} - \bar{\mathcal{G}}^{(j)}\right)\right]\left[\sum_{j'=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j'}}\mathcal{S}^{(j')}\right]$$

$$- \sum_{n=2}^{\infty} \frac{1}{n}\left[\left(\sum_{j=1}^{\infty} \frac{1}{(i\hbar\omega_\nu)^j}\bar{\mathcal{G}}^{(j)}\right)\left(\mathcal{S}^{(0)} - \boldsymbol{\Gamma} + \sum_{j'=1}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j'}}\mathcal{S}^{(j')}\right)\right]^n \quad (1.60)$$

So-far, no approximations have been made: Now we introduce an approximation by truncating the Laurent expansions.

- We choose to include terms in the Matsubara sum up to the second order of the inverse Matsubara frequency.

- we include only the leading order of the Laurent expansion, namely $\mathcal{G}^{(2)}$ and $\mathcal{S}^{(0)}$ for the self energy.

Alternatively one could include terms in the Matsubara sum up to fourth order in the inverse temperature and to include the also $\mathcal{G}^{(3)}$ and $\mathcal{S}^{(1)}$. Note that the third order term in inverse temperature of the Matsubara sum does not contribute. However we will postpone this for the time being.

Another alternative would be to truncate the power-series expansion of thelogarithm beyond a certain value and to inlcude all Laurent expansion terms of the Matsubara sum which are produced by the Green's-function and self-energy expansion up to a certain order.

Then, the functional X

$$X = -\frac{1}{\beta}\sum_{\nu,|\omega_\nu|>\omega_x} \text{Tr}\left\{\frac{1}{(i\hbar\omega_\nu)^2}\left\{\left(\mathcal{G}^{(2)} - \bar{\mathcal{G}}^{(2)}\right)\mathcal{S}^{(0)} - \frac{1}{2}\left[\bar{\mathcal{G}}^{(1)}\left(\mathcal{S}^{(0)} - \boldsymbol{\Gamma}\right)\right]^2\right\}\right\}$$

$$\overset{\text{Eq. } B.16}{=} -\frac{1}{\beta}\sum_{\nu,|\omega_\nu|>\omega_x} \text{Tr}\left\{\frac{1}{(i\hbar\omega_\nu)^2}\left\{\left(\mathcal{G}^{(2)} - \bar{\mathcal{G}}^{(2)}\right)\mathcal{S}^{(0)} - \frac{1}{2}\left[\boldsymbol{S}^{-1}\left(\mathcal{S}^{(0)} - \boldsymbol{\Gamma}\right)\right]^2\right\}\right\} \quad (1.61)$$

The functional derivatives with respect to the self energy is

$$\frac{dX}{d\mathcal{S}^{(0)}} = \left\{-\frac{1}{\beta}\sum_{\nu,|\omega_\nu|>\omega_x} \frac{1}{(i\hbar\omega_\nu)^2}\right\}\left[\mathcal{G}^{(2)} - \bar{\mathcal{G}}^{(2)} - \boldsymbol{S}^{-1}\left(\mathcal{S}^{(0)} - \boldsymbol{\Gamma}\right)\boldsymbol{S}^{-1}\right] \quad (1.62)$$

With the Laurent expansion of the non-interacting Green's function, Eq. B.16, we obtain

$$\mathcal{G}^{(2)} = \boldsymbol{S}^{-1}\left(\bar{\boldsymbol{h}} - \mu\boldsymbol{S} - \left(\mathcal{S}^{(0)} - \boldsymbol{\Gamma}\right)\right)\boldsymbol{S}^{-1} \quad (1.63)$$

The derivative with respect to the Green's function yields

$$\frac{dX}{d\mathcal{G}^{(2)}} = \left\{-\frac{1}{\beta}\sum_{\nu,|\omega_\nu|>\omega_x} \frac{1}{(i\hbar\omega_\nu)^2}\right\}\mathcal{S}^{(0)} \quad (1.64)$$

The stationary condition with respect to the Laurent term of the Green's function yields

$$\frac{1}{\beta} \sum_{\nu;|\omega_\nu|>\omega_x} \left[\frac{\beta \delta \Phi^{LW}}{\delta \boldsymbol{G}(i\omega_\nu)} - \frac{\beta \delta \Phi^{LW,HF}}{\delta \boldsymbol{G}(i\omega_\nu)} \right] \underbrace{\frac{\delta \boldsymbol{G}(i\omega_\nu)}{\delta \mathcal{G}^{(2)}}}_{\frac{1}{(i\hbar\omega_\nu)^2}} + \left\{ -\frac{1}{\beta} \sum_{\nu,|\omega_\nu|>\omega_x} \frac{1}{(i\hbar\omega_\nu)^2} \right\} \mathcal{S}^{(0)} = 0$$

$$\mathcal{S}^{(0)} = \left\{ +\frac{1}{\beta} \sum_{\nu,|\omega_\nu|>\omega_x} \frac{1}{(i\hbar\omega_\nu)^2} \right\}^{-1} \left\{ \frac{1}{\beta} \sum_{\nu;|\omega_\nu|>\omega_x} \frac{1}{(i\hbar\omega_\nu)^2} \left[\frac{\beta \delta \Phi^{LW}}{\delta \boldsymbol{G}(i\omega_\nu)} - \frac{\beta \delta \Phi^{LW,HF}}{\delta \boldsymbol{G}(i\omega_\nu)} \right] \right\} \quad (1.65)$$

1.6   Dynamic-correlation correction in non-orthonormal basis and in Bloch representation

---

DYNAMIC CORRELATION CORRECTION

$$Q_{dyn,\beta}^{\hat{W}_2}[\rho] \overset{\text{Eq. 1.22}}{=} \underset{\mathbf{\Gamma}_{\vec{k}}}{\text{stat}} \underset{\mathbf{G}_{\vec{k}},\mathbf{\Sigma}_{dyn,R}}{\text{stat}} \left\{ \sum_R \left(\Phi_\beta^{LW}[\mathbf{G}_R, \hat{W}_{2,R}] - \Phi_\beta^{HF,LW}[\mathbf{G}_R, \hat{W}_{2,R}] \right) \right.$$

$$- \frac{1}{N_k} \sum_k \frac{1}{\beta} \sum_{\nu, |\omega_\nu| < \omega_x} \text{Tr}\left\{ \ln\left[\mathbf{1} - \left((i\hbar\omega_\nu + \mu)\mathbf{S}_{\vec{k}} - \bar{\mathbf{h}}_{\vec{k}} \right)^{-1} \left(\mathbf{\Sigma}_{dyn}(i\omega_\nu) - \mathbf{\Gamma}_{\vec{k}} \right) \right] \right.$$

$$+ \left(\mathbf{\Sigma}_{dyn}(i\omega_\nu) - \mathbf{\Gamma}_{\vec{k}} \right) \mathbf{G}_{\vec{k}}(i\omega_\nu)$$

$$\left. + \left[\mathbf{G}_{\vec{k}}(i\omega_\nu) - \left((i\hbar\omega_\nu + \mu)\mathbf{S}_{\vec{k}} - \bar{\mathbf{h}}_{\vec{k}} \right)^{-1} \right] \mathbf{\Gamma}_{\vec{k}} \right\} \right\}$$

$$- \left[\frac{1}{\beta} \sum_{\nu, |\omega_\nu| > \omega_x} \frac{1}{(i\hbar\omega_\nu)^2} \right] \frac{1}{N_k} \sum_k \text{Tr}\left\{ \left[\mathcal{G}_{\vec{k}}^{(2)} - \mathbf{S}_{\vec{k}}^{-1}\left(\bar{\mathbf{h}}_{\vec{k}} - \mu\mathbf{S}_{\vec{k}} \right)\mathbf{S}_{\vec{k}}^{-1} \right] \mathcal{S}_{\vec{k}}^{(0)} \right.$$

$$\left.\left. - \frac{1}{2}\left[\mathbf{S}_{\vec{k}}^{-1}\left(\mathcal{S}_{\vec{k}}^{(0)} - \mathbf{\Gamma}_{\vec{k}} \right) \right]^2 \right\} \right\} \tag{1.66}$$

$\mathbf{G}_R$ is obtained from $\mathbf{G}_{\vec{k}}$ via Eq. 1.52 and $\mathbf{\Sigma}_{dyn}$ is obtained from the local contributions $\mathbf{\Sigma}_{dyn,R}$ via Eq. 1.51. The overlap matrix $\mathbf{S}_{\vec{k}}$ is obtained from Eq. 1.46.
N_k is the number of k-points in considered for the grid in the Brillouin zone integration.

$$\bar{h}_{a,b,\vec{k}} = \mu\mathbf{S}_{a,b,\vec{k}} + \sum_n \langle \chi_a|\psi'_n\rangle \frac{1}{\beta} \ln\left[\frac{1 - f'_n}{f'_n} \right] \langle \psi'_n|\chi_b\rangle \tag{1.67}$$

where

$$\left(\sum_{a,b} |\chi_a\rangle \rho_{\vec{k},a,b} \langle \chi_b| \right)\left(\sum_c |\chi_c\rangle \langle \pi_c|\psi'_{\vec{k},n}\rangle \right) = \left(\sum_a |\chi_a\rangle \langle \pi_a|\psi'_{\vec{k},n}\rangle \right) f'_{\vec{k},n}$$

$$\Rightarrow \qquad \mathbf{S}_{\vec{k}} \boldsymbol{\rho}_{\vec{k}} \mathbf{S}_{\vec{k}} \vec{\psi}'_{\vec{k},n} = \mathbf{S}_{\vec{k}} \vec{\psi}'_{\vec{k},n} f'_n \tag{1.68}$$

and $\langle \chi_a|\chi_b\rangle = \mathbf{S}_k$.
In the high-frequency part, i.e. for $|\omega_\nu| > \omega_x$, the Green's function and the self energy are given by

$$\mathbf{G}_{\vec{k}}(i\omega_\nu) = \frac{1}{i\hbar\omega}\mathbf{S}_{\vec{k}}^{-1} + \frac{1}{(i\hbar\omega)^2}\mathcal{G}^{(2)}$$

$$\mathbf{\Sigma}_{dyn,R}(i\omega_\nu) = \mathcal{S}_{dyn,R}^{(0)} \tag{1.69}$$

The result is correct for $N_k \to \infty$ and for $\omega_x \to \infty$.

1.7   Description of Subroutines

Data structures

$\langle \pi_\alpha | \psi_{n,\vec{k}} \rangle$	`KSET(IKPT)%PIPSI(:NDIM,:NCHI,:NB,NSPIN)`
$\boldsymbol{\rho}_{\vec{k}}$	`KSET(IKPT)%RHO(:NCHI,:NCHI,:NDIMD)`
$\boldsymbol{S}_{\vec{k}}$	`KSET(IKPT)%SMAT(:NCHI,:NCHI,:NDIMD)`
$\boldsymbol{S}_{\vec{k}}^{-1}$	`KSET(IKPT)%SINV(:NCHI,:NCHI,:NDIMD)`
$\boldsymbol{\Sigma}_{dyn,R}(i\omega_\nu)$	`ATOMSET(IAT)%SLOC(:NLOC,:NLOC,:NDIMD,:NOMEGA)`
$\mathcal{S}_{dyn,R}^{(0)}$	`ATOMSET(IAT)%SLOClaur(:NLOC,:NLOC,:NDIMD,1)`
$\mathcal{S}_{dyn,R}^{(1)}$	`ATOMSET(IAT)%SLOClaur(:NLOC,:NLOC,:NDIMD,2)`
$\bar{h}_{\vec{k}}$	`KSET(IKPT)%HRHO(:NCHI,:NCHI,:NDIMD)`
$\boldsymbol{\Gamma}_{\vec{k}}$	`KSET(IKPT)%GAMMA(:NCHI,:NCHI,:NDIMD)`

1.7.1   Data exchange of the Object wit the outer world

1.7.2   Flowchart
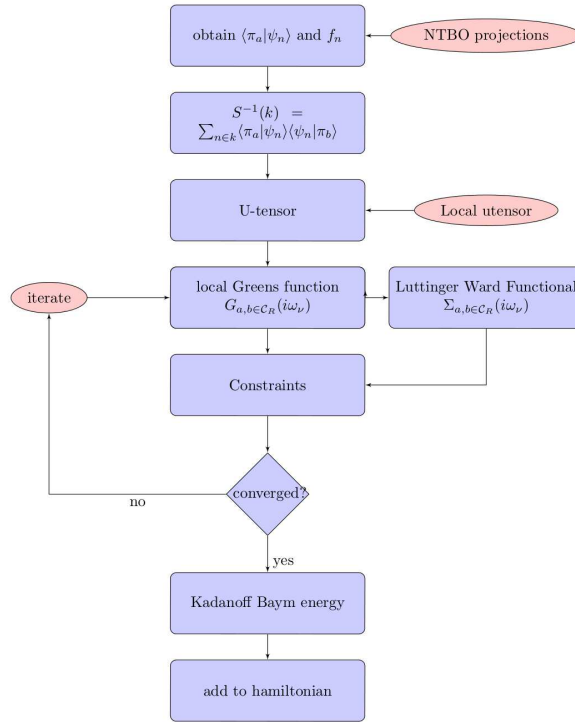
1.7.3   DMFT$GREEN

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.

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

```
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`.

$$\langle\pi_a|\psi_n\rangle \tag{1.70}$$

$$\rho_{a,b}(\vec{k}) = \sum_n \langle\pi_a|\psi_n(\vec{k})\rangle f_n(\vec{k})\langle\psi_n(\vec{k})|\pi_b\rangle \tag{1.71}$$

   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.

$$U_{a,b,c,d} = \alpha \int d^4x \int d^4x' \frac{e^2\chi_a^*(\vec{x})\chi_b^*(\vec{x'})\chi_c(\vec{x})\chi_d(\vec{x'})}{4\pi\epsilon_0|\vec{r}-\vec{r'}|} \tag{1.72}$$

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

$$\boldsymbol{S}^{-1}(\vec{k}) = \sum_n \langle \pi_a | \psi_n(\vec{k}) \rangle \langle \psi_n(\vec{k}) | \pi_b \rangle \tag{1.73}$$

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_HRHO`: (see also section 1.7.7 on p. 27.) Calculate the non-interacting Hamiltonian $\bar{h}_{\vec{k}}$ that produces the specified density matrix.

$$\bar{h}_{\vec{k},a,b} = \mu S_{\vec{k},a,b} + \sum_n \langle \chi_a | \psi'_{\vec{k},n} \rangle \frac{1}{\beta} \ln \left[\frac{1 - f'_{\vec{k},n}}{f'_{\vec{k},n}} \right] \langle \psi'_{\vec{k},n} | \chi_b \rangle \tag{1.74}$$

$\bar{h}_{\vec{k}}$ is kept as `KSET(IKPT)%HRHO`.

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

$$\hat{\rho}_R |\phi_{R,i}\rangle = |\phi_{R,i}\rangle f'_{R,i} \tag{1.75}$$

These states will be called local natural orbitals.

The orbital coefficients of the local natural orbitals are calculated as [3]

$$\sum_b \langle \pi_a | \hat{\rho}_R | \pi_b \rangle y_{b,j} = \sum_b S^{-1}_{R,a,b} y_{b,j} f'_{R,j} \qquad \text{with } \vec{y}_i^\dagger \boldsymbol{S}_R^{-1} \vec{y}_j = \delta_{i,j} \tag{1.80}$$

$$\langle \pi_a | \phi_{R,j} \rangle = \sum_b S^{-1}_{R,a,b} y_{b,j} \tag{1.81}$$

---

[3]

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

$$\sum_{b,c} \langle \pi_a | \hat{\rho}_R | \pi_b \rangle \langle \chi_b | \chi_c \rangle x_{c,j} = x_{a,j} f'_{R,j} \tag{1.76}$$

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

$$\sum_b \langle \pi_a | \hat{\rho}_R | \pi_b \rangle y_{b,j} = \sum_b S^{-1}_{a,b} y_{b,j} f'_{R,j} \tag{1.77}$$

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

$$|\phi_{R,j}\rangle = \sum_{a,b} |\chi_a\rangle S^{-1}_{a,b} y_{b,j} \tag{1.78}$$

The local natural orbitals are orthonormal in the sense

$$\langle \phi_{R,i} | \phi_{R,j} \rangle = \sum_{a,b,c,d} y^*_{a,i} S^{-1}_{a,c} \underbrace{\langle \chi_c | \chi_d \rangle}_{(\boldsymbol{S}^{-1})^{-1}} S^{-1}_{d,b} y_{b,j} = \sum_{a,b} y^*_{a,i} S^{-1}_{a,b} y_{b,j} = \delta_{i,j} \tag{1.79}$$

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

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

$$\boldsymbol{S}_R^{-1} = \boldsymbol{P}_R \left[\sum_{\vec{k}} w_{\vec{k}} \boldsymbol{S}^{-1}(\vec{k}) \right] \boldsymbol{P}_R \tag{1.82}$$

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?

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.

$$\boldsymbol{G}_R(i\omega_\nu) = \sum_k w_{\vec{k}} \left[(i\omega_\nu + \mu) \boldsymbol{S}_{\vec{k}} - \bar{\boldsymbol{h}}_{\vec{k}} + \boldsymbol{\Gamma}_{\vec{k}} - \boldsymbol{\Sigma}_{dyn}^{\hat{W}_2}(i\omega_\nu) \right]_R^{-1} \tag{1.83}$$

The Laurent expansion is obtained as

$$\mathcal{G}_R^{(1)}(i\omega_\nu) = \sum_k w_{\vec{k}} \boldsymbol{S}_{\vec{k}}^{-1}$$

$$\mathcal{G}_R^{(2)}(i\omega_\nu) = \sum_k w_{\vec{k}} \boldsymbol{S}_{\vec{k}}^{-1} \left(\bar{\boldsymbol{h}} - \mu \boldsymbol{S}_{\vec{k}} + \boldsymbol{\Gamma}_{\vec{k}} - \mathcal{S}_{dyn}^{(0)} \right) \boldsymbol{S}_{\vec{k}}^{-1} \tag{1.84}$$

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

8. `DMFT_SOLVER`: Calculates the dynamic contribution $\Phi_\beta^{LW,\hat{W}_2} - \Phi_\beta^{HF,\hat{W}_2}$ 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.

$$\Phi_{dyn}^{LW}[\mathbf{G}(i\omega_\nu), \hat{W}_2] = \Phi^{LW}[\mathbf{G}(i\omega_\nu), \hat{W}_2] - \frac{1}{2}\sum_{a,b,c,d} U_{a,b,d,c}^{\hat{W}_2}\Big(\rho_{d,a}\rho_{c,b} - \rho_{c,a}\rho_{d,b}\Big)\Big)$$

$$\Sigma_{dyn,a,b}^{\hat{W}_2}(i\omega_\nu) = \frac{\beta\partial\Phi^{LW}}{\partial G_{b,a}(i\omega_\nu)} - \sum_{c,d}\Big(U_{a,c,b,d}^{\hat{W}_2} - U_{a,c,d,b}^{\hat{W}_2}\Big)\rho_{d,c}$$

$$\mathcal{S}_{dyn,a,b}^{(0)} = \frac{1}{\beta}\sum_{|\nu|>\nu_x}\frac{1}{(i\hbar\omega_\nu)^2}\left[\frac{\beta\partial\Phi^{LW}}{\partial G_{b,a}(i\omega_\nu)} - \sum_{c,d}\Big(U_{a,c,b,d}^{\hat{W}_2} - U_{a,c,d,b}^{\hat{W}_2}\Big)\rho_{d,c}\right]$$

$$\frac{\delta\Phi_\beta^{dyn,LW}}{\delta U_{a,b,c,d}} = \frac{\delta\Phi_\beta^{LW}}{\delta U_{a,b,d,c}} - \frac{1}{2}\Big(\rho_{d,a}\rho_{c,b} - \rho_{c,a}\rho_{d,b}\Big)\Big) \tag{1.85}$$

pay attention to the reversed order of the indices c, d!
$\frac{\delta\Phi_\beta^{dyn,LW}}{\delta U_{a,b,c,d}}$ is the difference of the two-particle density for the interacting and the non-interacting electron gas at the same density matrix. In contrast to the corresponding term in the density functional theory, this term does not contain a kinetic energy contribution as does $n(\vec{r})\int_0^1 d\lambda\, h_\lambda(\vec{r}, \vec{r'})$, but rather $n(\vec{r})\Big[h_1(\vec{r}, \vec{r'}) - h_0(\vec{r}, \vec{r'})\Big]$

(d) After `DMFT_SOLVERIO` is finished, 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_MIX` mixes 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 E on p. 48.

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

$$\mathcal{S}_{dyn,a,b}^{(0)} \leftarrow \mathcal{S}_{dyn,a,b}^{(0)} + \frac{(\hbar\omega_x)^2}{C}\left(\mathcal{S}_{dyn,a,b}^{(0)} - \frac{\delta\Phi_{dyn}^{LW}}{\delta\mathcal{G}_{b,a}^{(2)}}\right) \tag{1.86}$$

It seems that the sign for the mixing is incorrect!! I am changing it in the code to see what happens. $\boldsymbol{\Sigma}^{dyn}$ 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.

10. `DMFT_CONSTRAINTS` enforced the density matrix constraint: The new Green's function $\mathbf{G}^{new}$ has the form

$$\mathbf{G}^{new}(i\omega_\nu) = \Big[(i\hbar\omega_\nu + \mu)\mathbf{1} - \bar{\mathbf{h}} + \boldsymbol{\Gamma} - \boldsymbol{\Sigma}^{dyn}(i\omega_\nu)\Big]^{-1} \tag{1.87}$$

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

$$\boldsymbol{\rho} = \frac{1}{\beta} \sum_\nu e^{i\beta\hbar\omega_\nu 0^+} \boldsymbol{G}^{new}(i\omega_\nu) \tag{1.88}$$

is fulfilled.

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

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

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

$$\delta\Gamma_{c,d} = \underbrace{\left[\frac{1}{\beta} \sum_\nu G^{new}_{a,c}(i\omega_\nu) G^{new}_{d,b}(i\omega_\nu) \right]^{-1}_{cd,ab}}_{\rightsquigarrow \quad \alpha\delta_{c,a}\delta_{d,b}} \left(\underbrace{\left[\frac{1}{\beta} \sum_\nu e^{i\beta\hbar\omega_\nu 0^+} G^{new}_{a,b}(i\omega_\nu) \right]}_{\rho^{new}_{a,b}} - \rho_{a,b} \right) \tag{1.89}$$

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

Thus, we iterate the coupled equations

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

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

$$\mathbf{\Gamma} = \mathbf{\Gamma} + \alpha\delta\boldsymbol{\rho} \tag{1.90}$$

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.

11. `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.

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

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

$$
+\Big(\mathbf{\Sigma}^{\hat{W}_2}_{dyn}(i\omega_\nu)-\mathbf{\Gamma}\Big)\mathbf{G}(i\omega_\nu)+\Big[\mathbf{G}(i\omega_\nu)-\big(i\hbar\omega_\nu+\mu)\mathbf{1}-\bar{h}\big)^{-1}\Big[\mathbf{\Gamma}\Big\} \quad (1.91)
$$

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

13. `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

$$
\omega_\nu = (2\nu-1)\pi k_B T \qquad \text{for } \nu = 1,\dots,N_\omega \tag{1.92}
$$

- 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 U-tensor for all local orbitals from subroutine `dmft_ulocal` reduces it to the entries for the correlated orbitals.

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

$$|\chi_a\rangle = |\phi_a\rangle - \sum_{b;R_b=R_a} |\dot{\phi}_b\rangle \bar{S}_{a,b} \tag{1.93}$$

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

$$u_{\ell,a,b,c,d} = \frac{2\ell+1}{4\pi} \int dr\, r^2 R_c(r) R_d(r) \left[\int d^3r'\, V_\ell(r,r') R_a(r') R_b(r') \right] \tag{1.94}$$

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

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

$$U_{a,b,c,d} = \sum_{ell} \frac{4\pi}{2\ell+1} \sum_{m=-\ell}^{\ell} u_{\ell,b,d,c,a} C_{L,L_b,L_d} C_{L,L_c,L_a} \tag{1.95}$$

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[4] in Eq. 1.72, i.e. as

$$U_{a,b,c,d} = \int d^4x \int d^4x' \frac{e^2 \chi_a^*(\vec{x}) \chi_b^*(\vec{x'}) \chi_c(\vec{x}) \chi_d(\vec{x'})}{4\pi\epsilon_0 |\vec{r}-\vec{r'}|} \tag{1.96}$$

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

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

$$E_X = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,c,d}\, \rho_{c,b}\rho_{d,a} \tag{1.97}$$

---

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

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.

$$\hat{1}_{\mathcal{C}_R} = \sum_{\alpha,\beta} \sum_n |\chi_\alpha\rangle \langle \pi_\alpha | \psi_n \rangle \langle \psi_n | \pi_\beta \rangle \langle \chi_\beta| \tag{1.98}$$

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

$$\boldsymbol{S}^{-1}_{\alpha,\beta} = \sum_{\alpha,\beta} \sum_n \langle \pi_\alpha | \psi_n \rangle \langle \psi_n | \pi_\beta \rangle \tag{1.99}$$

Note,however, that this matrix lives only on a single site! We can obtain it from the k-dependent $\boldsymbol{S}^{-1}$ 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

$$\hat{\rho}_{\mathcal{C}_R} = \sum_{\alpha,\beta} \sum_n |\chi_\alpha\rangle \langle \pi_\alpha | \psi_n \rangle f_n \langle \psi_n | \pi_\beta \rangle \langle \chi_\beta| \tag{1.100}$$

The eigenvalue equation

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

$$\sum_{\alpha,\beta} |\chi_\alpha\rangle \langle \pi_\alpha | \hat{\rho} | \pi_\beta \rangle \langle \chi_\beta | \phi_j \rangle = \sum_{\alpha,\beta} |\chi_\alpha\rangle \langle \pi_\alpha | \pi_\beta \rangle \langle \chi_\beta | \phi_j \rangle f_j = \sum_{\alpha,\beta} |\chi_\alpha\rangle \boldsymbol{S}^{-1}_{\alpha,\beta} \langle \chi_\beta | \phi_j \rangle \bar{f}_j \tag{1.101}$$

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

$$\rho V = \boldsymbol{S}^{-1} V \bar{f} \qquad \text{with} \qquad V^\dagger \boldsymbol{S}^{-1} V = 1 \tag{1.102}$$

The natural orbitals are

$$|\phi_j\rangle = \sum_{\alpha,\beta \in \mathcal{C}_R} \underbrace{|\chi_\alpha\rangle \boldsymbol{S}^{-1}_{\alpha,\beta} \langle \chi_\beta|}_{\boldsymbol{1}_{\mathcal{C}_R}} \phi_j\rangle = \sum_{\alpha \in \mathcal{C}_R} |\chi_\alpha\rangle \underbrace{\left(\boldsymbol{S}^{-1} V \right)_{\alpha,j}}_{V^{\dagger,-1}_{\alpha,j}} \tag{1.103}$$

The matrix $\boldsymbol{V}$ is placed into the structure `atomset%natorb%chiphi`, together with the occupations $\bar{f}$ which are in `atomset%natorb%f`. In addition we store $\boldsymbol{S}^{-1}V = V^{\dagger,-1}$ 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.

$$\bar{h}_{a,b}(\vec{k}) = \mu S_{a,b}(\vec{k}) + \sum_n \langle \chi_a | \psi'_n \rangle \frac{1}{\beta} \ln \left[\frac{1 - f'_n}{f'_n} \right] \langle \psi'_n | \chi_b \rangle \tag{1.104}$$

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

$$|\psi'_n\rangle = \sum_a |\chi_a\rangle\langle\pi_a|\psi'_n\rangle \tag{1.105}$$

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

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

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

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

$$\rho\left(\mathbf{S}\vec{x}_n\right) = \mathbf{S}^{-1}\left(\mathbf{S}\vec{c}_n\right)f'_n \tag{1.106}$$

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

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

where $\mathbf{f}'(\vec{k})$ 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 $\bar{\mathbf{h}}$ in the representation of the natural orbitals $|\psi'_n\rangle$.

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

$$\epsilon = \mu + k_B \begin{cases} \ln\left(\frac{1-f}{f}\right) & \text{for } f_0 < f < 1 - f_0 \\ a + b(f - f_0) & \text{for } f < f_0 \\ -a + b(f - (1 - f_0)) & \text{for } f > 1 - f_0 \end{cases} \tag{1.107}$$

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 $\hat{\bar{j}} = \sum_{a,b}|\pi_a\rangle h_{a,b}\langle\pi_b|$.

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

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

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

$$\bar{h}(\vec{k}) = \mathbf{V}\epsilon\mathbf{V}^{\dagger} \tag{1.108}$$

1.7.8 DMFT_CONSTRAINTS

Green's function

The Green's function is calculated as follows

$$\boldsymbol{G}(\vec{k}, i\omega_\nu) = \left[(i\omega_\nu + \mu)\boldsymbol{S}(\vec{k}) - \bar{\boldsymbol{h}}(\vec{k}) + \boldsymbol{\Gamma}(\vec{k}) - \sum_R \boldsymbol{\Sigma}_R^{dyn}(i\omega_\nu)\right]^{-1} \tag{1.109}$$

As described in section B.2.2 on p. 41, the Laurent-expansion coefficients are

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

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

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

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

$$\tag{1.110}$$

Green's function difference

In order to calculate the constraint, it may be adviseable to directly evaluate the difference of te Green's function from the non-interacting Greens function $\bar{\boldsymbol{G}}$.

$$\Delta\mathcal{G}^{(1)}(\vec{k}) = 0$$

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

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

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

$$\left.-\left(-\mu\boldsymbol{S}(\vec{k}) + \bar{\boldsymbol{h}}(\vec{k})\right)\boldsymbol{S}^{-1}(\vec{k})\left(-\mu\boldsymbol{S}(\vec{k}) + \bar{\boldsymbol{h}}(\vec{k})\right)\right]\boldsymbol{S}^{-1}(\vec{k}) \tag{1.111}$$

Update Γ

UPDATE Γ

The following two equations are solved iteratively[a]

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

$$-\underbrace{\left[(i\omega_\nu + \mu)\boldsymbol{S}(\vec{k}) - \bar{\boldsymbol{h}}(\vec{k})\right]^{-1}}_{\bar{\boldsymbol{G}}(\vec{k})}\Bigg)$$

$$\boldsymbol{\Gamma}(\vec{k}) = \boldsymbol{\Gamma}(\vec{k}) - \frac{4}{\beta}\boldsymbol{S}(\vec{k})\delta\rho(\vec{k})\boldsymbol{S}(\vec{k}) \qquad (1.112)$$

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

---

[a]Instead of subtracting the density matrix from the Matsubara sum for the Green'sfunction, we subtract the Greens function $\bar{\boldsymbol{G}}$ inside the Matsubara sum to avoid numerical problems due to the finite Matsubara sum.

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.

$$\delta\boldsymbol{\rho}(\boldsymbol{\Gamma}) = \delta\boldsymbol{\rho}(\boldsymbol{\Gamma}_0) - \frac{1}{\beta}\sum_\nu \boldsymbol{G}(\boldsymbol{\Gamma} - \boldsymbol{\Gamma}_0)\boldsymbol{G} + ... \qquad (1.113)$$

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

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

$$\Rightarrow \quad \boldsymbol{\Gamma} = \boldsymbol{\Gamma}_0 + \frac{4}{\beta}\boldsymbol{S}\left(\underbrace{\delta\boldsymbol{\rho}(\boldsymbol{\Gamma})}_{=0} - \delta\boldsymbol{\rho}(\boldsymbol{\Gamma}_0)\right)\boldsymbol{S} \qquad (1.114)$$

Should there be convergence problems, it may be improved by using the static Green's function $\boldsymbol{G}_\rho$ instead.

1.7.9   DMFT_SOLVER

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

1.7.10   DMFT_ETOT

When the loop is converged, we evaluate

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

$$(1.115)$$

Evaluation of the logarithm using a power series expansion

We use the Taylor expansion of the logarithm

$$\ln[1 - x] = -\sum_{n=1}^\infty \frac{1}{n} x^n \qquad \text{for } |x| < 1 \tag{1.116}$$

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

$$(1.117)$$

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

$$\mathrm{Tr}\ln(\boldsymbol{A}\boldsymbol{A}^\dagger) \overset{?}{=} \mathrm{Tr}\ln(\boldsymbol{A}) + Tr\ln(\boldsymbol{A}^\dagger) \tag{1.118}$$

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

Consider the singular value decompostion of $\boldsymbol{A} = \boldsymbol{U}\boldsymbol{\Sigma}\boldsymbol{V}^\dagger$, 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.

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

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

$$\mathrm{Tr}\ln(\boldsymbol{A}) + Tr\ln(\boldsymbol{A}^\dagger) = \mathrm{Tr}\ln(\boldsymbol{U}\boldsymbol{\Sigma}\boldsymbol{V}^\dagger) + \mathrm{Tr}\ln(\boldsymbol{V}\boldsymbol{\Sigma}^\dagger\boldsymbol{U}^\dagger) \tag{1.119}$$

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.

Instead of the singular value decomposition, we can also simply use the eigenvalue equation

$$\boldsymbol{A}\boldsymbol{V} = \boldsymbol{V}\boldsymbol{\Sigma} \tag{1.120}$$

so that

$$\boldsymbol{A} = \boldsymbol{V}\boldsymbol{\Sigma}\boldsymbol{V}^{-1}$$
$$\boldsymbol{A}^\dagger = \boldsymbol{V}^{-1,\dagger}\boldsymbol{\Sigma}^*\boldsymbol{V}^\dagger \tag{1.121}$$

Unfortunately, the same problem arises, because, in general, $\boldsymbol{V}\boldsymbol{V}^\dagger \neq \boldsymbol{1}$.

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

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

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]

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

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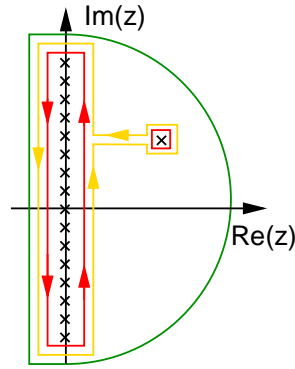
The integration is closed in the half plane with $\pm\text{Re}(z) > 0$, because we there the corresponding weighting function $h^\pm$ decays exponentially for $\text{Re}(z) \to \pm\infty$.

---

[1]Cauchy's Integral formula:

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

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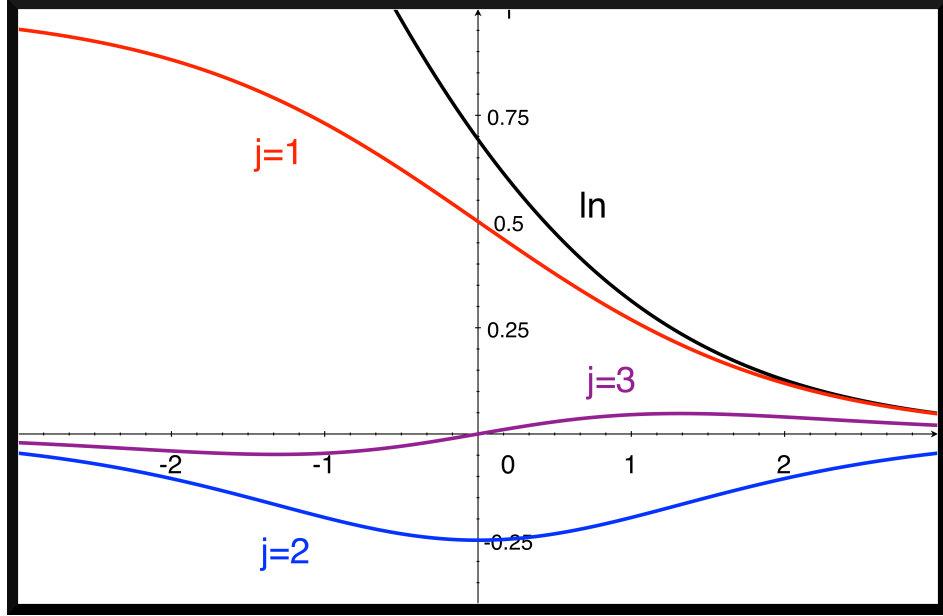
Fig. A.1: The result of Matsubara sums $-k_B T \sum_\nu \ln[-i\hbar\omega_\nu + \epsilon]$, $k_B T \sum_\nu \frac{1}{(i\hbar\omega_\nu - \epsilon)} e^{i\omega_\nu 0^+}$, $k_B T \sum_\nu \frac{1}{(i\hbar\omega_\nu - \epsilon)^2}$ as function of energy ϵ.
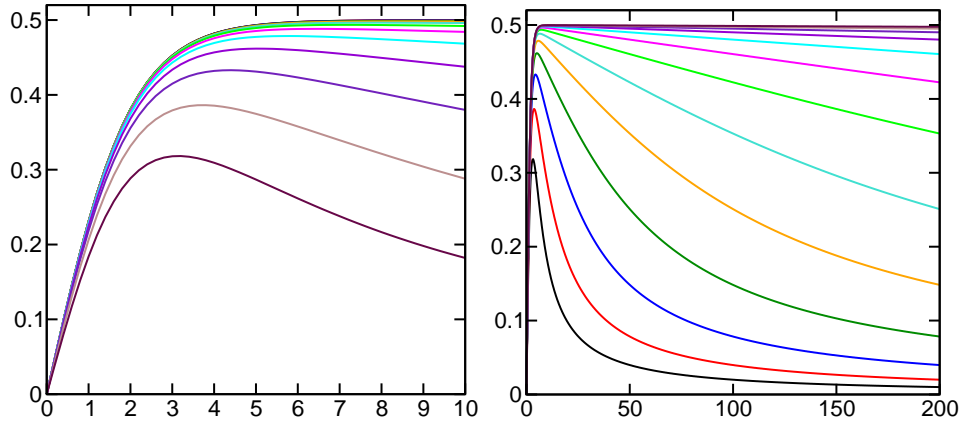

Fig. A.2: Fermi function calculated from a finite Matsubara sum. The function is shifted by $-\frac{1}{2}$ because no relgularization has been done. The sums have been performed with 2^n grid points with $n = 0, \ldots, 12$ for $k_B T = 1$.

Appendix B

The Green function

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

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

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

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

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

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

$$\boldsymbol{G}(-i\omega_\nu) = \boldsymbol{G}^\dagger(i\omega) \qquad (B.3)$$

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

Proof:

$$
\begin{aligned}
G_{\alpha,\beta}(i\omega_n) &= -\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\, e^{i\omega_n\tau}\left\langle \mathcal{T}\hat{c}_\alpha(\tau)\hat{c}_\beta^\dagger(0)\right\rangle && \text{(B.4)}\\
\Rightarrow G_{\alpha,\beta}(-i\omega_n) &= -\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\, e^{-i\omega_n\tau}\left\langle \mathcal{T}\hat{c}_\alpha(\tau)\hat{c}_\beta^\dagger(0)\right\rangle \\
&= \left\{-\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\, e^{i\omega_n\tau}\left\langle \left(e^{\beta(\hat{H}-\mu\hat{N})}\hat{c}_\alpha e^{-\beta(\hat{H}-\mu\hat{N})}\hat{c}_\beta^\dagger\right)^\dagger\right\rangle\right\}^* \\
&= \left\{-\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\, e^{i\omega_n\tau}\left\langle \hat{c}_\beta e^{-\beta(\hat{H}-\mu\hat{N})}\hat{c}_\alpha^\dagger e^{\beta(\hat{H}-\mu\hat{N})}\right\rangle\right\}^* \\
&\stackrel{cycl.perm}{=} \left\{-\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\, e^{i\omega_n\tau}\left\langle e^{\beta(\hat{H}-\mu\hat{N})}\hat{c}_\beta e^{-\beta(\hat{H}-\mu\hat{N})}\hat{c}_\alpha^\dagger\right\rangle\right\}^* \\
&= \left\{-\frac{1}{\hbar}\int_0^{\hbar\beta} d\tau\, e^{i\omega_n\tau}\left\langle \mathcal{T}\hat{c}_\beta(\tau)\hat{c}_\alpha^\dagger\right\rangle\right\}^* \\
&= G_{\beta,\alpha}^*(i\omega_\nu) && \text{(B.5)}
\end{aligned}
$$

B.1.2   Density of states

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

$$
N(\mu) = \int_{-\infty}^{\infty} d\epsilon\, f_\mu(\epsilon) D(\epsilon)\,, \tag{B.6}
$$

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

$$
\frac{dN}{d\mu} = \int_{-\infty}^{\infty} d\epsilon\, \frac{\partial f_\mu(\epsilon)}{\partial\mu} D(\epsilon) = \underbrace{\int_{-\infty}^{\infty} d\epsilon\, \frac{\beta}{\cosh^2\left(\frac{1}{2}\beta(\epsilon-\mu)\right)} D(\epsilon)}_{\tilde{D}(\mu)} \tag{B.7}
$$

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

$$
N_{a,b}(\mu) = \frac{1}{\beta}\sum_\nu G_{a,b}(i\omega_\nu,\mu)e^{i\omega_\nu 0^+} = \frac{1}{\beta}\sum_\nu \left[\left(G(i\omega_\nu,\mu_0)\right)^{-1}+(\mu-\mu_0)\right]^{-1}_{a,b} e^{i\omega_\nu 0^+} \tag{B.8}
$$

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

$$
\begin{aligned}
\tilde{D}_{a,b}(\mu) &\stackrel{\text{def}}{=} \frac{\partial N_{a,b}}{\partial\mu} \\
&= -\frac{1}{\beta}\sum_\nu\sum_c \left[\left(G(i\omega_\nu,\mu_0)\right)^{-1}+(\mu-\mu_0)\mathbf{1}\right]^{-1}_{a,c}\left[\left(G(i\omega_\nu,\mu_0)\right)^{-1}+(\mu-\mu_0)\mathbf{1}\right]^{-1}_{c,b}
\end{aligned}
$$
$$
\tag{B.9}
$$

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

$$
\hat{G}(i\omega_\nu) = \sum_{a,b} |\chi_a\rangle \Bigg\{ \sum_n \langle \pi_a|\psi_n\rangle \Bigg[(i\hbar\omega_\nu + \mu)\delta_{n,n'}
$$
$$
- \sum_{c,d} \langle \psi_n|\pi_c\rangle \Big(h_{c,d} + \Sigma_{c,d}(i\omega_\nu) \Big) \langle \pi_d|\psi_{n'}\rangle \Bigg]^{-1}_{n,n'} \langle \psi_{n'}|\pi_b\rangle \Bigg\} \langle \chi_b| \tag{B.10}
$$

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

$$
\boldsymbol{G}_{a,b}(i\omega_\nu) = \Big[(i\hbar\omega_\nu + \mu)\boldsymbol{S} - \boldsymbol{h} - \boldsymbol{\Sigma}(i\omega_\nu) \Big]^{-1}_{a,b} \tag{B.11}
$$

where

$$
S^{-1}_{a,b} = \sum_k \langle \pi_a|\psi_n\rangle\langle \psi_n|\pi_b\rangle \tag{B.12}
$$

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

$$
\boldsymbol{G}_{a,b}(i\omega_\nu) = \sum_n \langle \pi_a|\psi_n\rangle \Bigg[(i\hbar\omega_\nu + \mu)\delta_{n,n'} - \sum_{c,d} \langle \psi_n|\pi_c\rangle \Big(\boldsymbol{h}_{c,d} + \boldsymbol{\Sigma}_{c,d}(i\omega_\nu) \Big) \langle \pi_d|\psi_{n'}\rangle \Bigg]^{-1}_{n,n'} \langle \psi_{n'}|\pi_b\rangle
$$
$$
\tag{B.13}
$$

can be expressed in the form of Eq. B.11.

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

both can be absorbed in the non-interacting Hamiltonian.

$$
\begin{aligned}
G_{a,b}(i\omega_\nu) &= \sum_{n,n'} \langle \pi_a | \psi_n \rangle \left(i\hbar\omega_\nu \delta_{n,n'} - \langle \psi_n | \pi \rangle \boldsymbol{h} \langle \pi | \psi_{n'} \rangle \right)^{-1}_{n,n'} \langle \psi_{n'} | \pi_b \rangle \\
&= \sum_{n,n'} \langle \pi_a | \psi_n \rangle \left[\sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \left(\langle \psi | \pi \rangle \boldsymbol{h} \langle \pi | \psi \rangle \right)^{j} \right]_{n,n'} \langle \psi_{n'} | \pi_b \rangle \\
&= \sum_{n,n'} \langle \pi_a | \psi_n \rangle \left[\sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \left(\sum_{n_2,\ldots,n_j} \langle \psi_n | \pi \rangle \boldsymbol{h} \langle \pi | \psi_{n_2} \rangle \langle \psi_{n_2} | \pi \rangle \ldots \boldsymbol{h} \langle \pi | \psi_{n'} \rangle \right) \right] \langle \psi_{n'} | \pi_b \rangle \\
&= \sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \left(\sum_{n_1,\ldots,n_{j+1}} \underbrace{\langle \pi | \psi_{n_1} \rangle \langle \psi_{n_1} | \pi \rangle}_{\boldsymbol{S}^{-1}} \boldsymbol{h} \underbrace{\langle \pi | \psi_{n_2} \rangle \langle \psi_{n_2} | \pi \rangle}_{\boldsymbol{S}^{-1}} \ldots \boldsymbol{h} \underbrace{\langle \pi | \psi_{n_{j+1}} \rangle \langle \psi_{n_{j+1}} | \pi \rangle}_{\boldsymbol{S}^{-1}} \right)_{a,b} \\
&= \left(\sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \left(\boldsymbol{S}^{-1} \boldsymbol{h} \right)^{j} \boldsymbol{S}^{-1} \right)_{a,b} \\
&= \left[i\hbar\omega_\nu \boldsymbol{S} \left(1 - \frac{1}{i\hbar\omega_\nu} \boldsymbol{S}^{-1} \boldsymbol{h} \right) \right]^{-1}_{a,b} \\
&= \left[i\hbar\omega_\nu \boldsymbol{S} - \boldsymbol{h} \right]^{-1}_{a,b}
\end{aligned}
\tag{B.14}
$$

q.e.d.

B.2.1 Laurentexpansion of the non-interacting Green's function

We determine the Laurent expansion terms of $\bar{\boldsymbol{G}}$

$$
\bar{\boldsymbol{G}} = \left[i\hbar\omega_\nu \boldsymbol{S} - \left(\bar{\boldsymbol{h}} - \mu \boldsymbol{S} \right) \right]^{-1} = \sum_{j=1}^{\infty} \frac{1}{(i\hbar\omega_\nu)^j} \bar{\mathcal{G}}^{(j)}
\tag{B.15}
$$

and therefore

$$
\begin{aligned}
\boldsymbol{1} &= \sum_{j=1} \frac{1}{(i\hbar\omega_\nu)^j} \bar{\mathcal{G}}^{(j)} \left[i\hbar\omega_\nu \boldsymbol{S} - \left(\bar{\boldsymbol{h}} - \mu \boldsymbol{S} \right) \right] \\
&= \sum_{j=0} \frac{1}{(i\hbar\omega_\nu)^j} \bar{\mathcal{G}}^{(j+1)} \boldsymbol{S} - \sum_{j=1} \frac{1}{(i\hbar\omega_\nu)^j} \bar{\mathcal{G}}^{(j)} \left(\bar{\boldsymbol{h}} - \mu \boldsymbol{S} \right) \\
\bar{\mathcal{G}}^{(1)} &= \boldsymbol{S}^{-1} \\
\bar{\mathcal{G}}^{(j+1)} &= \bar{\mathcal{G}}^{(j)} \left(\bar{\boldsymbol{h}} - \mu \boldsymbol{S} \right) \boldsymbol{S}^{-1} \qquad \text{for } j > 1 \\
\bar{\mathcal{G}}^{j} &= \boldsymbol{S}^{-1} \left[\left(\bar{\boldsymbol{h}} - \mu \boldsymbol{S} \right) \boldsymbol{S}^{-1} \right]^{j-1}
\end{aligned}
\tag{B.16}
$$

The first term in the Laurent expansion of $\boldsymbol{G} - \bar{\boldsymbol{G}}$ vanishes

$$
\mathcal{G}^{(1)} - \bar{\mathcal{G}}^{(1)} = 0
\tag{B.17}
$$

B.2.2 Laurent expansion of the Green's function (new)

In order to regularize the Matsubara sums, respectively, to limit the extent of the Matsubara sum evaluated numerically, we need to represent the Green's function for high frequencies in terms of Matsubara sums.

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

$$\mathbf{G} = \left[(i\hbar\omega_\nu + \mu)\mathbf{S} - \mathbf{h} - \mathbf{\Sigma}(i\omega_\nu) \right]^{-1} \tag{B.18}$$

where the self energy has the Laurent expansion

$$\mathbf{\Sigma}(i\omega_\nu) = \sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^j} \mathcal{S}^{(j)} \tag{B.19}$$

Thus the numerical representation of the Green's function is

$$\mathbf{G}(i\omega_\nu) = \begin{cases} \left[(i\hbar\omega_\nu + \mu)\mathbf{S} - \mathbf{h} - \mathbf{\Sigma}(i\omega_\nu) \right]^{-1} & \text{for } |\nu| \le \nu_x \\[2ex] \left[(i\hbar\omega_\nu + \mu)\mathbf{S} - \mathbf{h} - \sum_{j'=0}^{j'_x} \mathcal{S}^{(j')} \frac{1}{(i\hbar\omega_\nu)^{j'}} \right]^{-1} & \text{for } |\nu| > \nu_x \end{cases} \tag{B.20}$$

The high-frequency representation of the Green's function shall itself be represented by a Laurent expansion

$$\mathbf{G}(i\omega_\nu) = \sum_{j=1}^{\infty} \mathcal{G}^{(j)} \frac{1}{(i\hbar\omega_\nu)^j} \tag{B.21}$$

The identity

$$\sum_{j=1}^{\infty} \mathcal{G}^{(j)} \frac{1}{(i\hbar\omega_\nu)^j} = \left[(i\hbar\omega_\nu + \mu)\mathbf{S} - \mathbf{h} - \sum_{j'=0}^{j'_x} \mathcal{S}^{(j')} \frac{1}{(i\hbar\omega_\nu)^{j'}} \right]^{-1} \tag{B.22}$$

yields

$$\sum_{j=1}^{\infty} \mathcal{G}^{(j)} \frac{1}{(i\hbar\omega_\nu)^j} \left[(i\hbar\omega_\nu + \mu)\mathbf{S} - \mathbf{h} - \sum_{j'=0}^{j'_x} \mathcal{S}^{(j')} \frac{1}{(i\hbar\omega_\nu)^{j'}} \right] = \mathbf{1} \tag{B.23}$$

which we can be arranged in orders of inverse Matsubara frequencies

$$\sum_{j=0}^{\infty} \mathcal{G}^{(j+1)} \mathbf{S} \frac{1}{(i\hbar\omega_\nu)^j} + \sum_{j=1}^{\infty} \mathcal{G}^{(j)} \left[\mu\mathbf{S} - \mathbf{h} - \mathcal{S}^{(0)} \right] \frac{1}{(i\hbar\omega_\nu)^j}$$

$$- \sum_{j=2}^{\infty} \sum_{n=1}^{j'_x} \mathcal{G}^{(j-n)} \mathcal{S}^{(n)} \frac{1}{(i\hbar\omega_\nu)^j} = \mathbf{1} \tag{B.24}$$

Thus, the expansion coefficients are

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

$$\mathcal{G}^{(j+1)} = -\mathcal{G}^{(j)}\left[\mu\boldsymbol{S} - \boldsymbol{h} - \mathcal{S}^{(0)}\right]\boldsymbol{S}^{-1} + \sum_{n=1}^{min(j'_x, j-1)} \mathcal{G}^{(j-n)}\mathcal{S}^{(n)}\boldsymbol{S}^{-1} \qquad \text{for } j > 0 \quad \text{(B.25)}$$

Let us make the higher-order terms more explicit

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

$$\mathcal{G}^{(2)} = -\mathcal{G}^{(1)}\left[\mu\boldsymbol{S} - \boldsymbol{h} - \mathcal{S}^{(0)}\right]\boldsymbol{S}^{-1} = -\boldsymbol{S}^{-1}\left[\mu\boldsymbol{S} - \boldsymbol{h} - \mathcal{S}^{(0)}\right]\boldsymbol{S}^{-1}$$

$$\mathcal{G}^{(3)} = -\mathcal{G}^{(2)}\left[\mu\boldsymbol{S} - \boldsymbol{h} - \mathcal{S}^{(0)}\right]\boldsymbol{S}^{-1} + \mathcal{G}^{(1)}\mathcal{S}^{(1)}\boldsymbol{S}^{-1}$$

$$= \boldsymbol{S}^{-1}\left[\mu\boldsymbol{S} - \boldsymbol{h} - \mathcal{S}^{(0)}\right]\boldsymbol{S}^{-1}\left[\mu\boldsymbol{S} - \boldsymbol{h} - \mathcal{S}^{(0)}\right]\boldsymbol{S}^{-1} + \boldsymbol{S}^{-1}\mathcal{S}^{(1)}\boldsymbol{S}^{-1} \qquad \text{(B.26)}$$

In practice, we expand the Green's function up to $\mathcal{G}^{(3)}$ and the self energy up to $\mathcal{S}^{(1)}$.

Thus the Green's function will be represented in the form

$$\boldsymbol{G}(i\omega_\nu) = \begin{cases} \left[(i\hbar\omega_\nu + \mu)\boldsymbol{S} - \boldsymbol{h} - \boldsymbol{\Sigma}(i\omega_\nu)\right]^{-1} & \text{for } |\nu| \leq \nu_x \\ \sum_{j=1}^{3}\mathcal{G}^{(j)}\frac{1}{(i\hbar\omega_\nu)^j} & \text{for } |\nu| > \nu_x \end{cases} \qquad \text{(B.27)}$$

where the Laurent-expansion coefficients of the Green's function are defined in eqeq:Lauenttermsgreenappendix. The variational degrees of freedom for the Greens functrion is the Green's function for $0 < \omega_\nu \leq \omega_x$ and the first two Laurent-expansion terms $\mathcal{S}^{(0)}$ and $\mathcal{S}^{(1)}$ of the self energy. The Green's functiuon for negative Matsubara frequencies are obtained by Eq. B.3, $\boldsymbol{G}(-\text{ß}\omega_\nu) = \boldsymbol{G}^\dagger(\text{ß}\omega_\nu)$.

Laurent-expansion coefficients are Hermitean

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

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

$$\Rightarrow \qquad \mathcal{G}^{(j)} = \left(\mathcal{G}^{(j)}\right)^\dagger \qquad \text{(B.28)}$$

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

Difference of Green's functions

The difference of two Green's function with different self energies has the form

$$\Delta\mathcal{G}^{(1)} = 0 \qquad \text{(B.29)}$$

$$\Delta\mathcal{G}^{(2)} = \boldsymbol{S}^{-1}\mathcal{S}^{(0)}\boldsymbol{S}^{-1} \qquad \text{(B.30)}$$

$$\Delta\mathcal{G}^{(3)} = -\boldsymbol{S}^{-1}\left[\mu\boldsymbol{S} - \boldsymbol{h}\right]\boldsymbol{S}^{-1}\mathcal{S}^{(0)}\boldsymbol{S}^{-1} - \boldsymbol{S}^{-1}\mathcal{S}^{(0)}\boldsymbol{S}^{-1}\left[\mu\boldsymbol{S} - \boldsymbol{h}\right]\boldsymbol{S}^{-1}$$

$$+ \boldsymbol{S}^{-1}\mathcal{S}^{(0)}\boldsymbol{S}^{-1}\mathcal{S}^{(0)}\boldsymbol{S}^{-1} + \boldsymbol{S}^{-1}\mathcal{S}^{(1)}\boldsymbol{S}^{-1} \qquad \text{(B.31)}$$

Laurent expansion for the density matrix

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

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

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

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

Derivative of the Luttinger Ward functional

For $j' \in \{0,1\}$

$$\frac{\partial\Phi_\beta^{LW}}{\partial\mathcal{S}^{(j')}} = \sum_{j=1}^3 \frac{1}{\beta}\sum_{\nu;|\nu|>\nu_x}\frac{\beta\partial\Phi_\beta^{LW}}{\partial\boldsymbol{G}(i\omega_\nu)}\frac{\partial\boldsymbol{G}(i\omega_\nu)}{\partial\mathcal{G}^{(j)}}\frac{\partial\mathcal{G}^{(j)}}{\partial\mathcal{S}^{(j')}}$$

$$= \sum_{j=1}^3\left[\frac{1}{\beta}\sum_{\nu;|\nu|>\nu_x}\frac{\beta\partial\Phi_\beta^{LW}}{\partial\boldsymbol{G}(i\omega_\nu)}\frac{1}{(i\hbar\omega_\nu)^j}\right]\frac{\partial\mathcal{G}^{(j)}}{\partial\mathcal{S}^{(j')}}$$

$$\frac{\partial\Phi_\beta^{LW}}{\partial\mathcal{S}^{(0)}} = \boldsymbol{S}^{-1}\underbrace{\left[\frac{1}{\beta}\sum_{\nu;|\nu|>\nu_x}\frac{\beta\partial\Phi_\beta^{LW}}{\partial\boldsymbol{G}(i\omega_\nu)}\frac{1}{(i\hbar\omega_\nu)^2}\right]}_{\delta\Phi_{\beta,dyn}^{LW}/\delta\mathcal{G}^{(2)}}\boldsymbol{S}^{-1}$$

$$- \boldsymbol{S}^{-1}\underbrace{\left[\frac{1}{\beta}\sum_{\nu;|\nu|>\nu_x}\frac{\beta\partial\Phi_\beta^{LW}}{\partial\boldsymbol{G}(i\omega_\nu)}\frac{1}{(i\hbar\omega_\nu)^3}\right]}_{\delta\Phi_{\beta,dyn}^{LW}/\delta\mathcal{G}^{(3)}}\boldsymbol{S}^{-1}\left[\mu\boldsymbol{S} - \boldsymbol{h} - \mathcal{S}^{(0)}\right]\boldsymbol{S}^{-1}$$

$$- \boldsymbol{S}^{-1}\left[\mu\boldsymbol{S} - \boldsymbol{h} - \mathcal{S}^{(0)}\right]\boldsymbol{S}^{-1}\underbrace{\left[\frac{1}{\beta}\sum_{\nu;|\nu|>\nu_x}\frac{\beta\partial\Phi_\beta^{LW}}{\partial\boldsymbol{G}(i\omega_\nu)}\frac{1}{(i\hbar\omega_\nu)^3}\right]}_{\delta\Phi_{\beta,dyn}^{LW}/\delta\mathcal{G}^{(3)}}\boldsymbol{S}^{-1}$$

$$\frac{\partial\mathcal{G}^{(j)}}{\partial\mathcal{S}^{(1)}} = \boldsymbol{S}^{-1}\underbrace{\left[\frac{1}{\beta}\sum_{\nu;|\nu|>\nu_x}\frac{\beta\partial\Phi_\beta^{LW}}{\partial\boldsymbol{G}(i\omega_\nu)}\frac{1}{(i\hbar\omega_\nu)^3}\right]}_{\delta\Phi_{\beta,dyn}^{LW}/\delta\mathcal{G}^{(3)}}\boldsymbol{S}^{-1} \tag{B.32}$$

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.

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

The stationary conditions are

$$0 = -\frac{\beta\delta\Phi^{LW,1}}{\delta\boldsymbol{G}(i\omega_\nu)} - \frac{\beta\delta\Phi^{LW,2}}{\delta\boldsymbol{G}(i\omega_\nu)} + \Big(\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \bar{\boldsymbol{h}} \Big) - \Big(\boldsymbol{h}' - \bar{\boldsymbol{h}} \Big)$$
$$\Rightarrow \quad \boldsymbol{\Sigma}(i\omega_\nu) = \frac{\beta\delta\Phi^{LW,1}}{\delta\boldsymbol{G}(i\omega_\nu)} + \frac{\beta\delta\Phi^{LW,2}}{\delta\boldsymbol{G}(i\omega_\nu)}$$
$$0 = \Big[\mathbf{1} - \bar{\boldsymbol{G}}(i\omega_\nu)\Big(\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \bar{\boldsymbol{h}} \Big) \Big]^{-1} \Big(-\bar{\boldsymbol{G}}(i\omega_\nu) \Big) + \boldsymbol{G}(i\omega_\nu) \quad\quad \text{(C.2)}$$
$$\Rightarrow \quad \boldsymbol{G}(i\omega_\nu) = \Big[\mathbf{1} - \bar{\boldsymbol{G}}(i\omega_\nu)\Big(\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \bar{\boldsymbol{h}} \Big) \Big]^{-1} \bar{\boldsymbol{G}}(i\omega_\nu)$$
$$0 = -\frac{1}{\beta} \sum_\nu \underbrace{\Big\{ \Big[\mathbf{1} - \bar{\boldsymbol{G}}(i\omega_\nu)\Big(\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \bar{\boldsymbol{h}} \Big) \Big]^{-1} \Big(-\bar{\boldsymbol{G}}(i\omega_\nu) \Big) + \boldsymbol{G}(i\omega_\nu) - \Big(\boldsymbol{G}(i\omega_\nu) - \bar{\boldsymbol{G}}(i\omega_\nu) \Big) \Big\}}_{=0 \quad \text{Eq. C.2}}$$
$$\Rightarrow \quad \frac{1}{\beta} \sum_\nu \boldsymbol{G}(i\omega_\nu) e^{i\hbar\omega_\nu\beta0^+} = \frac{1}{\beta} \sum_\nu \bar{\boldsymbol{G}}(i\omega_\nu) e^{i\hbar\omega_\nu\beta0^+} = \boldsymbol{\rho} \quad\quad \text{(C.3)}$$

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

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

(C.4)

The stationary conditions are

$$\boldsymbol{\Sigma}_2(i\omega_\nu) = \frac{\beta\delta\Phi^{LW,2}}{\delta\boldsymbol{G}(i\omega_\nu)}$$

$$\boldsymbol{G}(i\omega_\nu) = \Big[\boldsymbol{1} - \bar{\boldsymbol{G}}(i\omega_\nu)\Big(\boldsymbol{h}_2' + \boldsymbol{\Sigma}_2(i\omega_\nu) - \bar{\boldsymbol{h}}\Big)\Big]^{-1}\bar{\boldsymbol{G}}(i\omega_\nu)$$

(C.5)

We see that

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

(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

$$\boldsymbol{h}' + \boldsymbol{\Sigma} = \bar{\boldsymbol{h}}$$
$$\boldsymbol{G}(i\omega_\nu) = \bar{\boldsymbol{G}}(i\omega_\nu)$$

(C.7)

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

Appendix D

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

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

- 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 Structure of the self-consistency

The self-consisten scheme has the following structure

$$\mathbf{\Sigma}_{dyn}(+) = \mathbf{\Sigma}_{dyn}(0) + \alpha \left[\frac{\beta \delta \Phi^{LW,dyn}}{\delta \mathbf{G}} \bigg|_{\mathbf{G}[\mathbf{\Sigma}_{dyn}(0)]} - \mathbf{\Sigma}_{dyn}(0) \right] \tag{E.1}$$

and we search for the fixed point of this dynamics. $\mathbf{G}[\mathbf{\Sigma}_{dyn}]$ is given by Dyson's equation, that is by the stationary condition of the dynamic-correlation correction with respect to the self energy.

This problem is equivalent to the search of a zero of $\frac{\beta \delta \Phi^{LW,dyn}_\beta}{\delta \mathbf{G}} - \mathbf{\Sigma}_{dyn}$. More precisely, we search $\delta \mathbf{\Sigma}_{dyn}$ so that

$$\frac{\beta \delta \Phi^{LW,dyn}_\beta}{\delta \mathbf{G}} \bigg|_{\mathbf{G}[\mathbf{\Sigma}_{dyn} + \delta \mathbf{\Sigma}_{dyn}]} - \mathbf{\Sigma}_{dyn} - \delta \mathbf{\Sigma}_{dyn} = 0 \tag{E.2}$$

This yields to first order in $\delta \Sigma_{dyn}$

$$\left[\frac{\beta \delta \Phi^{LW,dyn}_\beta}{\delta \mathbf{G}} \bigg|_{\mathbf{G}[\mathbf{\Sigma}_{dyn}]} - \mathbf{\Sigma}_{dyn} \right] + \frac{\beta \delta^2 \Phi^{LW,dyn}_\beta}{\delta \mathbf{G} \delta \mathbf{G}'} \bigg|_{\mathbf{G}[\mathbf{\Sigma}_{dyn}]} \frac{\delta \mathbf{G}'}{\delta \mathbf{\Sigma}_{dyn}} \delta \mathbf{\Sigma}_{dyn} - \delta \mathbf{\Sigma}_{dyn} = 0 \tag{E.3}$$

I write out the indices

$$\left[\frac{\beta \delta \Phi^{LW,dyn}_\beta}{\delta G_{b,a}} \bigg|_{\mathbf{G}[\mathbf{\Sigma}_{dyn}]} - \Sigma_{dyn,a,b} \right] + \sum_{e,f} \left\{ \left[\sum_{c,d} \frac{\beta \delta^2 \Phi^{LW,dyn}_\beta}{\delta G_{b,a} \delta G_{d,c}} \bigg|_{\mathbf{G}[\mathbf{\Sigma}_{dyn}]} \frac{\delta G_{d,c}}{\delta \Sigma_{dyn,e,f}} \right] - \delta_{a,e} \delta_{b,f} \right\} \delta \Sigma_{dyn,e,f} = 0$$

$$\tag{E.4}$$

We can now evaluate the derivative of the Green's function with respect to the self energy.

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

$$\Rightarrow \quad \delta \mathbf{G} = \mathbf{G} \left(\delta \mathbf{\Sigma}_{dyn} - \delta \mathbf{\Gamma} \right) \mathbf{G}$$

$$\Rightarrow \quad \delta G_{d,c} = G_{d,e} \left(\delta \Sigma_{dyn,e,f} - \delta \Gamma_{e,f} \right) \mathbf{G}_{f,c} \tag{E.5}$$

and obtain

$$\left[\frac{\beta\delta\Phi_\beta^{LW,dyn}}{\delta G_{b,a}}\bigg|_{\mathbf{G}[\boldsymbol{\Sigma}_{dyn}]} - \Sigma_{dyn,a,b}\right] + \sum_{e,f}\left\{\left[\sum_{c,d}G_{c,e}\frac{\beta\delta^2\Phi_\beta^{LW,dyn}}{\delta G_{b,a}\delta G_{c,d}}\bigg|_{\mathbf{G}[\boldsymbol{\Sigma}_{dyn}]}G_{f,d}\right] - \delta_{a,e}\delta_{b,f}\right\}\delta\Sigma_{dyn,e,f} = 0$$

(E.6)

In order to devise an efficient preconditioning scheme, we need a model Green's function and a model dynamical Luttinger-Ward functional. A model Green's function is developed below in section E.3 on p. 50.

$$G_{a,b}(i\omega_\nu)G_{c,d}(i\omega_\nu) \approx \mathbf{S}_{a,b}^{-1}\mathbf{S}_{c,d}^{-1}\frac{1}{A + B(\hbar\omega_\nu)^2}$$

(E.7)

Without a good approximation for the Luttinger Ward functional, we make the following simple approximation

$$\sum_{c,d}G_{c,e}\frac{\beta\delta^2\Phi_\beta^{LW,dyn}}{\delta G_{b,a}\delta G_{c,d}}\bigg|_{\mathbf{G}[\boldsymbol{\Sigma}_{dyn}]}G_{f,d} \approx \delta_{a,e}\delta_{b,f}\frac{1}{A + B(\hbar\omega_\nu)^2}$$

(E.8)

With this approximation, I obtain the prediction for $\delta\boldsymbol{\Sigma}_{dyn}$ as

$$\left[\frac{\beta\delta\Phi_\beta^{LW,dyn}}{\delta G_{b,a}}\bigg|_{\mathbf{G}[\boldsymbol{\Sigma}_{dyn}]} - \Sigma_{dyn,a,b}\right] + \sum_{e,f}\left\{\delta_{a,e}\delta_{b,f}\frac{1}{A + B(\hbar\omega_\nu)^2} - \delta_{a,e}\delta_{b,f}\right\}\delta\Sigma_{dyn,e,f} = 0$$

$$\left[\frac{\beta\delta\Phi_\beta^{LW,dyn}}{\delta G_{b,a}}\bigg|_{\mathbf{G}[\boldsymbol{\Sigma}_{dyn}]} - \Sigma_{dyn,a,b}\right] + \left\{\frac{1}{A + B(\hbar\omega_\nu)^2} - 1\right\}\delta\Sigma_{dyn,a,b} = 0 \quad \text{(E.9)}$$

so that

$$\delta\Sigma_{dyn,a,b} \approx \frac{1}{1 - \frac{1}{A+B(\hbar\omega_\nu)^2}}\left[\frac{\beta\delta\Phi_\beta^{LW,dyn}}{\delta G_{b,a}}\bigg|_{\mathbf{G}[\boldsymbol{\Sigma}_{dyn}]} - \Sigma_{dyn,a,b}\right]$$

(E.10)

Thus, in the high-frequency limit, it seems that a mixing factor equal to one seems to be ideal, while the low frequency limit should have an independent mixing. To avoid a singularity, A must be chosen larher than one. A value $A = 1$ would lead to infinite mixing for $\omega_\nu = 0$. Therefore, I introduce a new variable α with ($A = \alpha/(\alpha - 1)$ for the low-frequency mixing factor α is a mixing factor for the low-frequency part, and B controls how fast the mixing approaches the high-frequency value.

$$\boldsymbol{\Sigma}_{dyn}(+) = \boldsymbol{\Sigma}_{dyn}(0) + \frac{1}{1 - \frac{1}{\frac{\alpha}{\alpha-1}+B(\hbar\omega_\nu)^2}}\left[\frac{\beta\delta\Phi_\beta^{LW,dyn}}{\delta G_{b,a}}\bigg|_{\mathbf{G}[\boldsymbol{\Sigma}_{dyn}(0)]} - \Sigma_{dyn,a,b}(0)\right]$$

(E.11)

The Laurent expansions are mixed with a mixing factor equal to unity consistent with the high-frequency mixing.

E.2   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.3   Model Green's function

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

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

$$G_{a,b}(i\omega_\nu) = \sum_n \frac{\langle \pi_a | \psi_n \rangle \langle \psi_n | \pi_b \rangle}{i\hbar\omega_\nu + \mu - \epsilon_n} = \int d\epsilon \, \frac{D_{a,b}(\epsilon)}{i\hbar\omega_\nu + \mu - \epsilon} \tag{E.12}$$

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

$$D_{a,b}(\epsilon) = S_{a,b}^{-1} \frac{1}{W} \cdot \underbrace{\theta\left(\epsilon - \left(E - \frac{W}{2}\right)\right)}_{=1 \quad \text{above lower bound}} \cdot \underbrace{\theta\left(\left(E + \frac{W}{2}\right) - \epsilon\right)}_{=1 \quad \text{below upper bound}} \tag{E.13}$$

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

$$\begin{aligned}
G_{a,b}(i\omega_\nu) &= \int_{E-W/2}^{E+W/2} d\epsilon \, \frac{\frac{1}{W} S_{a,b}^{-1}}{i\hbar\omega_\nu - \epsilon} \\
&= \frac{S_{a,b}^{-1}}{W} \int_{E-W/2}^{E+W/2} d\epsilon \, \frac{-\epsilon - i\hbar\omega_\nu}{(\hbar\omega)^2 + \epsilon^2} \\
&= -\frac{S_{a,b}^{-1}}{W} \int_{E-W/2}^{E+W/2} d\epsilon \, \frac{\epsilon + i\hbar\omega_\nu}{(\hbar\omega)^2 + \epsilon^2} \\
&= -\frac{S_{a,b}^{-1}}{W} \int_{E-W/2}^{E+W/2} dx \, \frac{x + i\hbar\omega_\nu}{(\hbar\omega)^2 + x^2} \\
&= -\frac{S_{a,b}^{-1}}{W} \left\{ \int_{E-W/2}^{E+W/2} dx \, \frac{x}{(\hbar\omega)^2 + x^2} + i\hbar\omega_\nu \int_{E-W/2}^{E+W/2} dx \, \frac{1}{(\hbar\omega)^2 + x^2} \right\} \\
&= -\frac{S_{a,b}^{-1}}{W} \left\{ \frac{1}{2} \ln\left[(\hbar\omega)^2 + (E + W/2)^2\right] - \frac{1}{2} \ln\left[(\hbar\omega)^2 + (E - W/2)^2\right] \right. \\
&\qquad \left. + i\hbar\omega_\nu \left[\frac{1}{\hbar\omega} \text{atan}\left(\frac{E + W/2}{\hbar\omega}\right) - \frac{1}{\hbar\omega} \text{atan}\left(\frac{E - W/2}{\hbar\omega}\right) \right] \right\} \\
&= \frac{S_{a,b}^{-1}}{W} \left\{ +\frac{1}{2} \ln\left[\frac{(\hbar\omega)^2 + (E - W/2)^2}{(\hbar\omega)^2 + (E + W/2)^2} \right] - i \left[\text{atan}\left(\frac{E + W/2}{\hbar\omega}\right) - \text{atan}\left(\frac{E - W/2}{\hbar\omega}\right) \right] \right\}
\end{aligned} \tag{E.14}$$

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

---

[1] Integral formula's:

$$\int dx \, \frac{1}{a^2 + x^2} = \frac{1}{a} \text{atan}\left(\frac{x}{a}\right)$$

$$\int dx \, \frac{x}{a^2 + x^2} = \frac{1}{2} \ln\left(a^2 + x^2\right) \tag{E.15}$$

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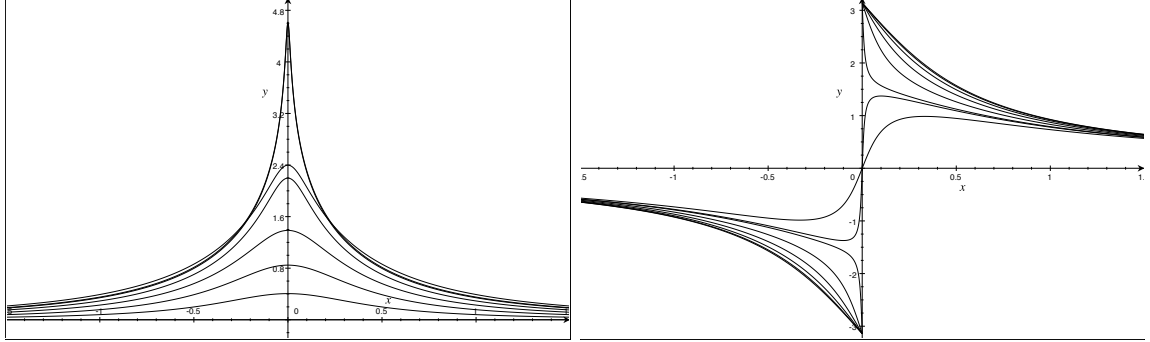

Fig. E.1: Negative real part (left) and negative imaginary part (right) of the Greens function with a constant density of states of width $W = 1$, centered at $E = \{0, 0.1, 0.2, 0.3, 0.49, 0.51, 0.6\}$.

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

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 expansion of the logarithm [3].

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

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

$$\left. -i\left[\frac{\pi}{2} - \frac{\hbar\omega}{E+W/2}\underbrace{-\frac{\pi}{2} + \frac{\hbar\omega}{E-W/2}}_{\text{times -1 for insulators}}\right]\right\}$$

$$\approx \frac{S_{a,b}^{-1}}{W}\left\{+\ln\left[\frac{(E-W/2)}{(E+W/2)}\right] + \frac{EW}{\left(E^2-\left(\frac{W}{2}\right)^2\right)^2}\hbar\omega^2 - i\begin{cases}\frac{W}{E^2-\left(\frac{W}{2}\right)^2}\hbar\omega & \text{for insulators}\\ \pm\pi - \frac{4|E|}{E^2-\left(\frac{W}{2}\right)^2}\hbar\omega & \text{for metals}\end{cases}\right\}$$

(E.18)

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

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

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

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

(E.19)

Remark: We can approximate

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

(E.20)

---

[2]

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

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

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

(E.16)

[3]

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

(E.17)

E.4   Preconditioning

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

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

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

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

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

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

$$m_\Sigma(\omega_\nu) = A \left(\frac{\mathrm{atan}(\frac{W/2}{\hbar\omega})}{(W/2)} \right)^2 \tag{E.21}$$

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.

$$M_\Sigma = m_\Sigma(0) \tag{E.22}$$

$$C_\Sigma = \lim_{\omega \to \infty} m_\Sigma(\omega) \cdot (\hbar\omega)^2 \tag{E.23}$$

With these parameters we obtain

$$m_\Sigma(\omega_\nu) \approx M_\Sigma \left[\frac{2}{\pi} \mathrm{atan}\left(\frac{\pi}{2} \sqrt{\frac{C_\Sigma}{M_\Sigma}} \frac{1}{\hbar\omega} \right) \right]^2 \tag{E.24}$$

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

$$m_\Sigma(\omega_\nu) \approx m_\Sigma \left[\left(\frac{2}{\pi W/2} \right)^2 + (\hbar\omega)^2 \right]^{-1} = C_\Sigma \left[\frac{C_\Sigma}{M_\Sigma} + (\hbar\omega)^2 \right]^{-1} \tag{E.25}$$

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.

Appendix F

Usage

F.0.1   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
```

F.0.2   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
```

Appendix G

Bloch representation

Appendix H

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[1]: Caution, I generalized Dieter's Equations without checking. Signs, factors, indices, etc. may be wrong!

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

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

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

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

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

$$\Sigma_{\alpha,\beta}^{out}(i\omega_\nu) := -\Big(G^{out}(i\omega_\nu) \Big)_{\alpha,\beta}^{-1} + i\hbar\omega_\nu \delta_{\alpha,\beta} - \Delta_{\alpha,\beta}^{in}(i\omega_\nu) \qquad (H.1)$$

---

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

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

$$\Delta_{\alpha,\beta}^{out}(i\omega_\nu) := -\left(G(i\omega_\nu)\right)_{\alpha,\beta}^{-1} + i\hbar\omega_\nu\delta_{\alpha,\beta} - \Sigma_{\alpha,\beta}^{out}(i\omega_\nu) \tag{H.2}$$

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.

$$\boldsymbol{\Delta}^{out} = F[\boldsymbol{\Delta}^{in}, \boldsymbol{G}, \boldsymbol{U}] \tag{H.3}$$

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. H.2 and Eq. H.1 yield

$$G_{\alpha,\beta}^{out}(i\omega_\nu) = G_{\alpha,\beta}(i\omega_\nu) \tag{H.4}$$

and

$$G_{\alpha,\beta}(i\omega_\nu) = \left[i\hbar\omega_\nu\boldsymbol{1} - \boldsymbol{\Sigma}^{out}(i\omega_\nu) - \boldsymbol{\Delta}^{in}(i\omega_\nu)\right]_{\alpha,\beta}^{-1}. \tag{H.5}$$

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