The DMFT object of the CP-PAW code

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Chapter 1

DMFT object

1.1 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.

Grand potential and density-matrix functional as starting point

The grand potential has the form

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

where the density-matrix functional is expressed with the Luttinger-Ward functional[1] $\Phi^{LW}[\boldsymbol{G},\hat{W}]$ as

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

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 chemical potential is μ and $\beta = 1/(k_B T)$. The Hamiltonian consists of a non-interacting part $\hat{h} = \frac{\hat{p}^2}{2m_0} + \hat{v}_{ext}$ and an electron-electron interaction $\hat{W}$.

The Matsubara sum runs over the Matsubara frequencies

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

The Hamiltonian $\boldsymbol{h}_\rho$ is a non-local Hamiltonian directly related to the one-particle density matrix via

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

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

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

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

as a consequence of the startionarity with respect to the Lagrange multiplier $\boldsymbol{h}'$

Helmholtz potential

In practice, we calculate the Helmholtz potential

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

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

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

1.1.1 Integration into DFT

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 in general 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

$$|\psi_n\rangle = \sum_a |\chi_a\rangle\langle\pi_a|\psi_n\rangle + |\delta\psi_n\rangle \tag{1.8}$$

where the projector functions obey the bi-orthogonality condition

$$\langle\pi_a|\chi_b\rangle = \delta_{a,b}\ , \tag{1.9}$$

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

$$\langle\pi_a|\delta\psi_n\rangle = 0 \tag{1.10}$$

Double counting

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

$$\begin{aligned} A_{\beta,N}[\hat{h}+\hat{W}] = \min_{|\psi_n\rangle, f_n\in[0,1]} \operatorname*{stat}_{\mu,\Lambda} \Bigg\{ & \sum_n f_n \langle\psi_n|\frac{\hat{\vec{p}}^2}{2m_e}|\psi_n\rangle + \int d^3r\ n(\vec{r})v_{ext}(\vec{r'}) \\ & + \frac{1}{2}\int d^3r \int d^3r' \frac{e^2 n(\vec{r})n(\vec{r'})}{4\pi\epsilon_0|\vec{r}-\vec{r'}|} + E_{xc}[n] + \frac{1}{\beta}\sum_n \Big[f_n \ln(f_n) + (1-f_n)\ln(1-f_n)\Big] \\ & - Q^{\hat{W}}_{\mathrm{DFT},\beta}[\boldsymbol{\rho}, n] + Q^{\hat{W}}_{\mathrm{X},\beta}[\boldsymbol{\rho}] + Q^{\hat{W}}_{\mathrm{dyn},\beta}[\boldsymbol{\rho}] \\ & - \mu\Big[\sum_n f_n - N\Big] - \sum_{m,n}\Lambda_{m,n}\Big(\langle\psi_n|\psi_m\rangle - \delta_{m,n}\Big)\Bigg\} \end{aligned} \tag{1.11}$$

where

$$\begin{aligned} n(\vec{r},\sigma,\sigma') &= \sum_n \langle\vec{r},\sigma|\psi_n\rangle f_n \langle\psi|\vec{r},\sigma'\rangle \\ n(\vec{r}) &= \sum_\sigma n(\vec{r},\sigma,\sigma) \\ \rho_{a,b} &= \sum_n \langle\pi_a|\psi_n\rangle f_n \langle\psi|\pi_b\rangle \end{aligned} \tag{1.12}$$

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

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

where the U-tensor is defined as

$$U_{a,b,c,d} = \frac{e^2\chi^*_a(\vec{r})\chi^*_b(\vec{r'})\chi_c(\vec{r})\chi_d(\vec{r'})}{4\pi\epsilon_0|\vec{r}-\vec{r'}|} \tag{1.14}$$

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

The purpose of the DMFT object is to add

$$Q^{\hat{W}}_{\text{X},\beta}[\boldsymbol{\rho}] + Q^{\hat{W}}_{\text{dyn},\beta}[\boldsymbol{\rho}] \tag{1.15}$$

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

Solver

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!`

$$\begin{aligned}
G^{out}_{\alpha,\beta}(i\omega_\nu) &:= -\frac{1}{Z}\int \prod_\gamma Dc^*_\gamma Dc_\gamma \; c_\alpha(i\omega_\nu)c^*_\beta(i\omega_\nu)\mathrm{e}^{-S_{loc}} \\
Z &= \int \prod_\gamma Dc^*_\gamma Dc_\gamma \; \mathrm{e}^{-S_{loc}} \\
S_{loc} &= -\int_0^\beta d\tau_1 \int_0^\beta d\tau_2 \sum_{\alpha,\beta} c^*_\alpha(\tau_1)\Big[i\hbar\omega_\nu\delta_{\alpha,\beta} - \Delta^{in}_{\alpha,\beta}(i\omega_\nu)\Big]c_\beta(\tau_2) \\
&\quad -\int_0^\beta d\tau \sum_{\alpha,\beta,\gamma,\delta} U_{\alpha,\beta,\delta,\gamma}c^*_\alpha c^*_\beta c_\gamma c_\delta
\end{aligned}$$

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

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

$$\Sigma^{out}_{\alpha,\beta}(i\omega_\nu) := -\Big(G^{out}(i\omega_\nu)\Big)^{-1}_{\alpha,\beta} + i\hbar\omega_\nu\delta_{\alpha,\beta} - \Delta^{in}_{\alpha,\beta}(i\omega_\nu) \tag{1.16}$$

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

$$\Delta^{out}_{\alpha,\beta}(i\omega_\nu) := -\Big(G(i\omega_\nu)\Big)^{-1}_{\alpha,\beta} + i\hbar\omega_\nu\delta_{\alpha,\beta} - \Sigma^{out}_{\alpha,\beta}(i\omega_\nu) \tag{1.17}$$

[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)

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.

$$\boldsymbol{\Delta}^{out} = F[\boldsymbol{\Delta}^{in}, \boldsymbol{G}, \boldsymbol{U}] \tag{1.18}$$

The mapping depends on exactly two quantities, namely the input Green's function and the U-tensor. Self-consistency yields the fixed point $\Delta^{out} = \Delta^{in}$ of this mapping. At this fixed point, Eq. 1.17 and Eq. 1.16 yield

$$G^{out}_{\alpha,\beta}(i\omega_\nu) = G_{\alpha,\beta}(i\omega_\nu) \tag{1.19}$$

and

$$G_{\alpha,\beta}(i\omega_\nu) = \left[i\hbar\omega_\nu \mathbf{1} - \boldsymbol{\Sigma}^{out}(i\omega_\nu) - \boldsymbol{\Delta}^{in}(i\omega_\nu) \right]^{-1}_{\alpha,\beta} . \tag{1.20}$$

1.2 Usage

1.2.1 Control file

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

```
!CONTROL
  !GENERIC NSTEP=500  dt=5. STart=F  !END
  !FOURIER EPWPSI=30. CDUAL=2.0 !END
  !DFT TYPE=10
     !NTBO MODUS='DMFT' !END
  !END
  !PSIDYN STOP=T FRIC=.05 SAFEORTHO=F
    !auto fric(-)=0.3 fact(-)=0.97 fric(+)=0.3 fact(+)=1.0 minfric=0.05 !end
  !END
  !MERMIN T[K]=4000. ADIABATIC=T RETARD=10. !END
!end
!EOB
```

1.2.2 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 TORB=F F F F F
           CV=F FOCKSETUP=F TAILLAMBDA=4.0 2.0 !END
  !END
  !SPECIES NAME='MN' ID='MN_HBS' NPRO=1 1 1
     !NTBO TORB=F F T   LHFWeiGHT=0.25
           CV=F FOCKSETUP=F TAILLAMBDA=4.0 2.0 !END
  !END
  !SPECIES NAME='O_' ID='O_.75_6.0'  NPRO=1 1 0
     !NTBO TORB=F F   TORB_X=T F T F F
           CV=F FOCKSETUP=F TAILLAMBDA=4.0 2.0 !END
  !END
  !LATTICE T=  1.0000        0.0000        0.0000000000
              0.0000        1.0000        0.0000000000
              0.0000        0.0000        1.0000000000 !END
  !ATOM NAME= 'CA1'   R=   0.0  0.0 0.0 !END
  !ATOM NAME= 'MN1'   R=   0.5  0.5 0.5 !END
  !ATOM NAME= 'O_1'   R=   0.0  0.5 0.5 !END
  !ATOM NAME= 'O_2'   R=   0.5  0.0 0.5 !END
  !ATOM NAME= 'O_3'   R=   0.5  0.5 0.0 !END
  !ORBPOT_X
    !POT ATOM='MN(UP)1' VALUE=+.2 TYPE='D' RC=1.5 S=1 !END
  !END
!END
!EOB
```

1.2.3 The interface routine

The solver is linked into the subroutine `DMFT_SOLVERIO`.

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

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

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

$$\Phi^{LW,HF} = \frac{1}{2} \sum_{a,b,c,d} U_{a,b,d,c} \Big(\rho_{b,a}\rho_{c,b} - \rho_{c,a}\rho_{d,b} \Big) \tag{1.21}$$

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

$$\begin{aligned}\Delta\Sigma_{a,b}(i\omega_\nu) &= \beta\frac{\partial\Phi^{LW}}{\partial G_{b,a}(i\omega_\nu)} - \beta\frac{\partial\Phi^{LW,HF}}{\partial G_{b,a}(i\omega_\nu)} \\ &= \beta\frac{\partial\Phi^{LW}[\boldsymbol{G},\hat{W}]}{\partial G_{b,a}(i\omega_\nu)} - \sum_{c,d}\Big(U_{a,c,b,d} - U_{a,c,d,b}\Big)\rho_{c,d}\end{aligned} \tag{1.22}$$

1.3 Description of Subroutines

1.3.1 Index arrays

The local orbitals arranged on a long array indexed by `IPRO`. The the subset of local orbitals considered in the DMFT module, in the following called correlated orbitals, are indexed by by `ICHI`. They are arranged by the following pseudocode:

```
ipro=0
ichi=0
do iat=1,nat
  isp=isp(iat)
  do ln=1,lnx(isp)
    l=lox(ln,isp)
    do im=1,2*l+1
      ipro=ipro+1
      if(torb(ln,isp(iat))ichi=ichi+1
    enddo
  enddo
enddo
```

`ISP`,`LNX`,`LOX` and `TORB` are obtained from the `SETUPS` project.

1.3.2 DMFT$GREEN

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

```
call dmft_ini()
call dmft_collecthamiltonian()
call dmft_collectfulldenmat()
call dmft_utensor()
call dmft_smat()
call dmft_natorb()
call dmft_hrho()
call dmft_constraints('hrho')
do iter=1,3
  call dmft_gloc_withatomset()
  call dmft_solver(etot)
```

```
  call dmft_constraints('h0')
enddo ! end of loop over iterations to enforce constraint
call dmft_detot(svar)
etot=etot+svar
call energylist$set('dmft interface',etot)
call energylist$add('local correlation',etot)
call energylist$add('total energy',etot)
call dmft_addtohpsi()
```

1. DMFT_COLLECTHAMILTONIAN:

$$\langle \pi_a | \psi_n \rangle \tag{1.23}$$

$$\rho_{a,b}(\vec{k}) = \sum_n \langle \pi_a | \psi_n(\vec{k}) \rangle f_n(\vec{k}) \langle \psi_n(\vec{k}) | \pi_b \rangle \tag{1.24}$$

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

3. DMFT_UTENSOR:

$$U_{a,b,c,d} = \alpha \int d^4x \int d^4x' \, \frac{e^2 \chi_a^*(\vec{x}) \chi_b^*(\vec{x'}) \chi_c(\vec{x}) \chi_d(\vec{x'})}{4\pi\epsilon_0 |\vec{r} - \vec{r'}|} \tag{1.25}$$

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

4. DMFT_SMAT:

$$\boldsymbol{S}^{-1}(\vec{k}) = \sum_n \langle \pi_a | \psi_n(\vec{k}) \rangle \langle \psi_n(\vec{k}) | \pi_b \rangle \tag{1.26}$$

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

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

$$\sum_{a,b} \langle \phi_i | \phi_a \rangle \Big(\boldsymbol{S}_R \Big)_{a,b} \langle \pi_b | \phi_j \rangle = \delta_{i,j} \tag{1.27}$$

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

   The two matrices are related by

$$\langle \pi | \phi \rangle = \boldsymbol{S}_R^{-1} \langle \pi | \phi \rangle \tag{1.28}$$

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

$$\boldsymbol{\rho}_R \langle \chi | \phi \rangle = \boldsymbol{S}_R^{-1} \langle \chi | \phi \rangle \vec{x} \tag{1.29}$$

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

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

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

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

$$\boldsymbol{h}_\rho(\vec{k}) - \mu = \boldsymbol{V}^{\dagger,-1}(\vec{k})\ k_B \ln\left[\frac{\mathbf{1} - \boldsymbol{V}^\dagger(\vec{k})\boldsymbol{\rho}(\vec{k})\boldsymbol{V}(\vec{k})}{\boldsymbol{V}^\dagger(\vec{k})\boldsymbol{\rho}(\vec{k})\boldsymbol{V}(\vec{k})}\right]\boldsymbol{V}^{-1}(\vec{k}) \tag{1.30}$$

$\boldsymbol{h}_\rho$ is kept as `KSET%HRHO`.

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

$$\boldsymbol{\rho}(\vec{k}) = k_B T \sum_\nu \Big[(i\omega_\nu + \mu)\boldsymbol{S}(\vec{k}) - \boldsymbol{h}' - \boldsymbol{\Sigma}(i\omega_\nu)\Big]^{-1} \tag{1.31}$$

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

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

$$\boldsymbol{G}_R(i\omega_\nu) = \sum_k w(\vec{k})\Big[(i\omega_\nu + \mu)\boldsymbol{S}(\vec{k}) - \boldsymbol{h}' - \boldsymbol{\Sigma}(i\omega_\nu)\Big]_R^{-1} \tag{1.32}$$

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

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

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

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

The external solver shall evaluate

$$\Phi^{LW}[\boldsymbol{G}(i\omega_\nu), \hat{W}] - \Phi^{LW,HF}[\boldsymbol{G}(i\omega_\nu), \hat{W}] \tag{1.33}$$

where

$$\begin{aligned}\Phi^{LW,HF}[\boldsymbol{G}, \hat{W}] &= \frac{1}{2}\frac{1}{\beta^2}\sum_{\nu,\nu'} e^{i\beta\hbar\omega_\nu 0^+} e^{i\beta\hbar\omega_{\nu'} 0^+} \sum_{a,b,c,d} U_{a,b,d,c} \\ &\quad\times\Big(G_{d,a}(i\omega_\nu)G_{c,b}(i\omega_{\nu'}) - G_{c,a}(i\omega_\nu)G_{d,b}(i\omega_{\nu'})\Big) \\ &= \frac{1}{2}\sum_{a,b,c,d} U_{a,b,d,c}\Big(\rho_{d,a}\rho_{c,b} - \rho_{c,a}\rho_{d,b}\Big)\Big)\end{aligned} \tag{1.34}$$

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

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

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

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

$$-\frac{1}{\beta}\sum_{\nu}\mathrm{Tr}\Big\{\ln\Big[\mathbf{1} - \Big(i\hbar\omega_\nu + \mu)\mathbf{1} - \boldsymbol{h}_\rho\Big)^{-1}\Big(\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \boldsymbol{h}_\rho\Big)\Big] + (\boldsymbol{h}' + \boldsymbol{\Sigma}(i\omega_\nu) - \boldsymbol{h}_\rho)\boldsymbol{G}(i\omega_\nu) - \Big[\boldsymbol{G}(i\omega_\nu) - \Big(i\hbar\omega_\nu + \mu)\mathbf{1} - \boldsymbol{h}_\rho\Big)^{-1}\Big]\Big(\boldsymbol{h}' - \boldsymbol{h}_\rho\Big)\Big\} \quad (1.35)$$

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

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

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

1.3.3 DMFT_INI

Hardwired data

- NOMEGA (N_ω)

Inherited data

- NDIM from `waves_module`
- NSPIN from `waves_module`
- NKPTL from `waves_module`
- NAT from `ATOMLIST` Object
- isp(IAT)
- WKPT and (NKPT,KMAP) from `DYNOCC` Object
- KBT from `dynocc$getr8a('temp')` from `paw_occupations.f90`
- `atomset(iat)%lhfweight` from `lmto_module`

Derived data

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

$$\omega_\nu = (2\nu - 1)\pi k_B T \qquad \text{for } \nu = 1, \ldots, N_\omega \tag{1.36}$$

- `IPROOFCHI` maps the indices ICHI to indices IPRO. ICHI and IPRO are explained in section 1.3.1.
- 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
`atomset\%DENMAT\%LNX`	number of radial partial waves for this atom
`atomset\%DENMAT\%LOX(lnx)`	main angular momentum quantum numbers ℓ
`atomset\%DENMAT\%LMNX`	number (including m quantum number) of partial waves for this atom
`atomset\%DENMAT\%LMN(lmnx)`	IPRO index relative to the first index for this atom

- KSET structure

`KSET\%WKPT`	geometric k-point weight

1.3.4 DMFT_UTENSOR

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

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

$$|\chi_a\rangle = |\phi_a\rangle - \sum_{b;R_b=R_a} |\dot{\bar{\phi}}_b\rangle \bar{S}_{b,a} \tag{1.37}$$

The U-tensor is constructed in `LMTO\_MAKETAILEDPARTIALWAVES`. `LMTO\_ULITTLE` constructs

$$u_{\ell,a,b,c,d} = \frac{2\ell+1}{4\pi} \int dr\, r^2 R_c(r) R_d(r) \Big[\int d^3r'\, V_\ell(r,r') R_a(r') R_b(r') \Big] \tag{1.38}$$

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

$$U_{a,b,c,d} = \sum_{ell} \frac{4\pi}{2\ell+1} \sum_{m=-\ell}^{\ell} u_{\ell,b,d,c,a} C_{L,L_b,L_d} C_{L,L_c,L_a} \tag{1.39}$$

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[2] in Eq. 1.25, i.e. as

$$U_{a,b,c,d} = \int d^4x \int d^4x' \, \frac{e^2 \chi_a^*(\vec{x}) \chi_b^*(\vec{x'}) \chi_c(\vec{x}) \chi_d(\vec{x'})}{4\pi\epsilon_0 |\vec{r} - \vec{r'}|} \tag{1.40}$$

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

$$\begin{aligned} E_H &= \frac{1}{2} \sum_{a,b,c,d} U_{a,b,c,d} \; \rho_{c,a} \rho_{d,b} \\ E_X &= \frac{1}{2} \sum_{a,b,c,d} U_{a,b,c,d} \; \rho_{c,b} \rho_{d,a} \end{aligned} \tag{1.41}$$

1.3.5 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.

$$\hat{1}_{\mathcal{C}_R} = \sum_{\alpha,\beta} \sum_n |\chi_\alpha\rangle\langle\pi_\alpha|\psi_n\rangle\langle\psi_n|\pi_\beta\rangle\langle\chi_\beta| \tag{1.42}$$

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

$$\boldsymbol{S}^{-1}_{\alpha,\beta} = \sum_{\alpha,\beta} \sum_n \langle\pi_\alpha|\psi_n\rangle\langle\psi_n|\pi_\beta\rangle \tag{1.43}$$

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

$$\hat{\rho}_{\mathcal{C}_R} = \sum_{\alpha,\beta} \sum_n |\chi_\alpha\rangle\langle\pi_\alpha|\psi_n\rangle f_n \langle\psi_n|\pi_\beta\rangle\langle\chi_\beta| \tag{1.44}$$

The eigenvalue equation

$$\begin{aligned} \hat{\rho}|\phi_n\rangle = |\phi_n\rangle f_n \\ \sum_{\alpha,\beta} |\chi_\alpha\rangle\langle\pi_\alpha|\hat{\rho}|\pi_\beta\rangle\langle\chi_\beta|\phi_j\rangle = \sum_{\alpha,\beta} |\chi_\alpha\rangle\langle\pi_\alpha|\pi_\beta\rangle\langle\chi_\beta|\phi_j\rangle f_j = \sum_{\alpha,\beta} |\chi_\alpha\rangle \boldsymbol{S}^{-1}_{\alpha,\beta} \langle\chi_\beta|\phi_j\rangle \bar{f}_j \end{aligned} \tag{1.45}$$

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

$$\boldsymbol{\rho V} = \boldsymbol{S}^{-1} \boldsymbol{V} \bar{f} \qquad \text{with} \qquad \boldsymbol{V}^\dagger \boldsymbol{S}^{-1} \boldsymbol{V} = \boldsymbol{1} \tag{1.46}$$

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

The natural orbitals are

$$|\phi_j\rangle = \sum_{\alpha,\beta\in\mathcal{C}_R} \underbrace{|\chi_\alpha\rangle \boldsymbol{S}^{-1}_{\alpha,\beta}\langle\chi_\beta|}_{\mathbf{1}_{\mathcal{C}_R}} \phi_j\rangle = \sum_{\alpha\in\mathcal{C}_R} |\chi_\alpha\rangle \underbrace{\left(\boldsymbol{S}^{-1}\boldsymbol{V}\right)_{\alpha,j}}_{\boldsymbol{V}^{\dagger,-1}_{\alpha,j}} \tag{1.47}$$

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

1.3.6 DMFT_HRHO

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

$$\epsilon = k_B \begin{cases} \ln\left(\frac{1-f}{f}\right) & \text{for } f_0 < f < 1-f_0 \\ a + b(f-f_0) & \text{for } f < f_0 \\ -a + b(f-(1-f_0)) & \text{for } f > 1-f_0 \end{cases} \tag{1.48}$$

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

1.3.7 DMFT_CONSTRAINTS

The two equations are solved iteratively

$$\bar{\boldsymbol{\rho}}(\vec{k}) = k_B T \sum_\nu \underbrace{\left[(i\omega_\nu + \mu)\boldsymbol{S}(\vec{k}) - \boldsymbol{h}(\vec{k}) - \sum_R \Sigma_R(i\omega_\nu)\right]^{-1}}_{\boldsymbol{G}(\vec{k})} \tag{1.49}$$

$$\delta\boldsymbol{h} = 4k_B T \boldsymbol{S}(\vec{k})\left[\bar{\rho}(\vec{k}) - \rho(\vec{k})\right]\boldsymbol{S}(\vec{k}) \tag{1.50}$$

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

The Laurent expansion terms are

$$\begin{aligned} \boldsymbol{G}^{(1)}(\vec{k}) &= \boldsymbol{S}^{-1}(\vec{k}) \\ \boldsymbol{G}^{(2)}(\vec{k}) &= \boldsymbol{S}^{-1}(\vec{k})\Big(-\mu\boldsymbol{S}(\vec{k}) + \boldsymbol{h}'(\vec{k}) + \boldsymbol{\Sigma}^{(1)}(\vec{k})\Big)\boldsymbol{S}^{-1}(\vec{k}) \\ \boldsymbol{G}^{(2)}(\vec{k}) &= \boldsymbol{S}^{-1}(\vec{k})\Big[\boldsymbol{S}^{-1}(\vec{k})\Big(-\mu\boldsymbol{S}(\vec{k}) + \boldsymbol{h}'(\vec{k}) + \boldsymbol{\Sigma}^{(1)}(\vec{k})\Big)\boldsymbol{S}^{-1}(\vec{k}) + \boldsymbol{\Sigma}^{(2)}(\vec{k})\Big]\boldsymbol{S}^{-1}(\vec{k}) \end{aligned} \tag{1.51}$$

1.3.8 DMFT_SOLVER

The solver first calculates the Hartree-Fock contribution to the Luttinger-Ward functional as defined in Eq. 1.21 and the corresponding self energy. Because the Hartree-Fock self energy is

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

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

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

Appendix A

The Green function

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

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

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

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

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

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

A.1 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 onto the local orbitals, the Green's function can be expressed as

$$\hat{G}(i\omega_\nu) = \sum_{a,b} |\chi_a\rangle \Bigg\{ \sum_n \langle\pi_a|\psi_n\rangle \Bigg[(i\hbar\omega_\nu + \mu)\delta_{n,n'}$$
$$- \sum_{c,d} \langle\psi_n|\pi_c\rangle \Big(h_{c,d} + \Sigma_{c,d}(i\omega_\nu)\Big) \langle\pi_d|\psi_{n'}\rangle \Bigg]^{-1}_{n,n'} \langle\psi_{n'}|\pi_b\rangle \Bigg\} \langle\chi_b| \tag{A.3}$$

We converted Eq. (A.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

$$\boldsymbol{G}_{a,b}(i\omega_\nu) = \Big[(i\hbar\omega_\nu + \mu)\boldsymbol{S} - \boldsymbol{h} - \boldsymbol{\Sigma}(i\omega_\nu)\Big]^{-1}_{a,b} \tag{A.4}$$

where

$$S^{-1}_{a,b} = \sum_k \langle \pi_a | \psi_n \rangle f_n \langle \psi_n | \pi_b \rangle \tag{A.5}$$

Proof: Here, I will show that the Green's function matrix expressed in local orbitals Eq. A.4

$$\boldsymbol{G}_{a,b}(i\omega_\nu) = \sum_n \langle \pi_a | \psi_n \rangle \left[(i\hbar\omega_\nu + \mu)\delta_{n,n'} - \sum_{c,d} \langle \psi_n | \pi_c \rangle \Big(\boldsymbol{h}_{c,d} + \boldsymbol{\Sigma}_{c,d}(i\omega_\nu) \Big) \langle \pi_d | \psi_{n'} \rangle \right]^{-1}_{n,n'} \langle \psi_{n'} | \pi_b \rangle \tag{A.6}$$

can be expressed in the form of Eq. A.4.

I order to simplify the proof, I will not write out the self energy, nor the chemical potential. This is possible because the proof works for each Matsubara frequency independently, so that both can be absorbed in the non-interacting Hamiltonian.

$$\begin{aligned}
G_{a,b}(i\omega_\nu) &= \sum_{n,n'} \langle \pi_a | \psi_n \rangle \Big(i\hbar\omega_\nu \delta_{n,n'} - \langle \psi_n | \pi \rangle \boldsymbol{h} \langle \pi | \psi_{n'} \rangle \Big)^{-1}_{n,n'} \langle \psi_{n'} | \pi_b \rangle \\
&= \sum_{n,n'} \langle \pi_a | \psi_n \rangle \Big(\sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \Big(\langle \psi | \pi \rangle \boldsymbol{h} \langle \pi | \psi \rangle \Big)^{j}_{n,n'} \langle \psi_{n'} | \pi_b \rangle \\
&= \sum_{n,n'} \langle \pi_a | \psi_n \rangle \Big(\sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \Big(\sum_{n_2,\dots,n_j} \langle \psi_n | \pi \rangle \boldsymbol{h} \langle \pi | \psi_{n_2} \rangle \langle \psi_{n_2} | \pi \rangle \dots \boldsymbol{h} \langle \pi | \psi_{n'} \rangle \Big) \langle \psi_{n'} | \pi_b \rangle \\
&= \sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \Big(\sum_{n_1,\dots,n_{j+1}} \underbrace{\langle \pi | \psi_{n_1} \rangle \langle \psi_{n_1} | \pi \rangle}_{\boldsymbol{S}^{-1}} \boldsymbol{h} \underbrace{\langle \pi | \psi_{n_2} \rangle \langle \psi_{n_2} | \pi \rangle}_{\boldsymbol{S}^{-1}} \dots \boldsymbol{h} \underbrace{\langle \pi | \psi_{n_{j+1}} \rangle \langle \psi_{n_{j+1}} | \pi \rangle}_{\boldsymbol{S}^{-1}} \Big)_{a,b} \\
&= \Big(\sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}} \Big(\boldsymbol{S}^{-1}\boldsymbol{h} \Big)^{j} \boldsymbol{S}^{-1} \Big)_{a,b} \\
&= \left[i\hbar\omega_\nu \boldsymbol{S} \Big(\boldsymbol{1} - \frac{1}{i\hbar\omega_\nu} \boldsymbol{S}^{-1}\boldsymbol{h} \Big) \right]^{-1}_{a,b} \\
&= \Big[i\hbar\omega_\nu \boldsymbol{S} - \boldsymbol{h} \Big]^{-1}_{a,b}
\end{aligned} \tag{A.7}$$

q.e.d.

A.1.1 Laurent expansion of the Green's function

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

$$\boldsymbol{G} = \Big[(i\hbar\omega_\nu + \mu)\boldsymbol{S} - \boldsymbol{h} - \boldsymbol{\Sigma}(i\omega_\nu) \Big]^{-1} \tag{A.8}$$

where the self energy has the Laurent expansion

$$\boldsymbol{\Sigma}(i\omega_\nu) = \sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^j} \mathcal{S}^{(j)} \tag{A.9}$$

$$
\begin{aligned}
\boldsymbol{G} &= \left[(i\hbar\omega_\nu)\boldsymbol{S}\Big(\boldsymbol{1} - \frac{1}{i\hbar\omega_\nu}\boldsymbol{S}^{-1}\Big(\boldsymbol{h} - \mu\boldsymbol{S} + \boldsymbol{\Sigma}(i\omega_\nu)\Big)\Big)\right]^{-1} \\
&= \sum_{j=0}^{\infty} \frac{1}{(i\hbar\omega_\nu)^{j+1}}\Big(\boldsymbol{S}^{-1}\Big(\boldsymbol{h} - \mu\boldsymbol{S} + \boldsymbol{\Sigma}(i\omega_\nu)\Big)\Big)^j \boldsymbol{S}^{-1} \\
&= \frac{1}{(i\hbar\omega_\nu)}\boldsymbol{S}^{-1} + \frac{1}{(i\hbar\omega_\nu)^2}\boldsymbol{S}^{-1}\Big(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)} + \frac{1}{i\hbar\omega_\nu}\mathcal{S}^{(1)}\Big)\boldsymbol{S}^{-1} \\
&\quad + \frac{1}{(i\hbar\omega_\nu)^3}\boldsymbol{S}^{-1}\Big(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)}\Big)\boldsymbol{S}^{-1}\Big(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)}\Big)\Big)\boldsymbol{S}^{-1} + O(\omega_\nu^{-4}) \\
&= \frac{1}{(i\hbar\omega_\nu)}\boldsymbol{S}^{-1} + \frac{1}{(i\hbar\omega_\nu)^2}\boldsymbol{S}^{-1}\Big(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)}\Big)\boldsymbol{S}^{-1} \\
&\quad + \frac{1}{(i\hbar\omega_\nu)^3}\boldsymbol{S}^{-1}\Big(\mathcal{S}^{(1)} + \Big(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)}\Big)\boldsymbol{S}^{-1}\Big(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)}\Big)\Big)\boldsymbol{S}^{-1} + O(\omega_\nu^{-4})
\end{aligned}
\tag{A.10}
$$

$$
\boldsymbol{G} = \sum_{j=1}^{\infty} \frac{1}{(i\hbar\omega_\nu)^j}\mathcal{G}^{(j)} \tag{A.11}
$$

with

$$
\begin{aligned}
\mathcal{G}^{(1)} &= \boldsymbol{S}^{-1} \\
\mathcal{G}^{(2)} &= \boldsymbol{S}^{-1}\Big(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)}\Big)\boldsymbol{S}^{-1} \\
\mathcal{G}^{(3)} &= \boldsymbol{S}^{-1}\Big(\mathcal{S}^{(1)} + \Big(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)}\Big)\boldsymbol{S}^{-1}\Big(\boldsymbol{h} - \mu\boldsymbol{S} + \mathcal{S}^{(0)}\Big)\Big)\boldsymbol{S}^{-1}
\end{aligned}
\tag{A.12}
$$

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

$$
\begin{aligned}
\sum_{j=1}^{\infty} \frac{1}{(-i\hbar\omega_\nu)^j}\mathcal{G}^{(j)} = \boldsymbol{G}(-i\omega_\nu) = \boldsymbol{G}^\dagger(i\omega_\nu) = \sum_{j=1}^{\infty} \frac{1}{(-i\hbar\omega_\nu)^j}\left(\mathcal{G}^{(j)}\right)^\dagger \\
\Rightarrow \qquad \mathcal{G}^{(j)} = \left(\mathcal{G}^{(j)}\right)^\dagger
\end{aligned}
\tag{A.13}
$$

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

A.1.2 Regularization

From http://en.wikipedia.org/wiki/Matsubara_frequency we obtain the following expression for the **fermionic** summations. In these expressions the Matsubara frequencies are[1]

$$
\omega_\nu = (2\nu + 1)\pi/(\hbar\beta) \tag{A.14}
$$

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

$$-k_BT\sum_\nu \ln[-i\hbar\omega_\nu+\epsilon]e^{i\omega_\nu 0^+} = -k_BT\ln[1+e^{-\beta\epsilon}] \tag{A.15}$$

$$k_BT\sum_\nu \frac{1}{(i\hbar\omega_\nu-\epsilon)}e^{i\omega_\nu 0^+} = (1+e^{\beta\epsilon})^{-1} \tag{A.16}$$

$$k_BT\sum_\nu \frac{1}{(i\hbar\omega_\nu-\epsilon)^n} = \frac{1}{(k_BT)^{n-1}(n-1)!}\,\partial_\epsilon^{n-1}\big|_{x=\beta\epsilon}(1+e^x)^{-1} \tag{A.17}$$

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

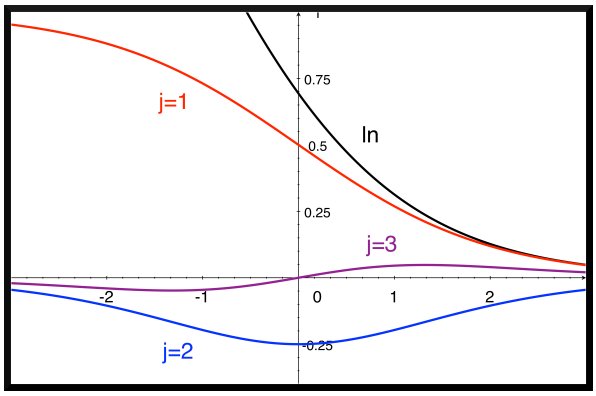

Fig. A.1: The result of Matsubara sums $-k_BT\sum_\nu \ln[-i\hbar\omega_\nu+\epsilon]$, $k_BT\sum_\nu \frac{1}{(i\hbar\omega_\nu-\epsilon)}e^{i\omega_\nu 0^+}$, $k_BT\sum_\nu \frac{1}{(i\hbar\omega_\nu-\epsilon)^2}$ as function of energy ϵ.

Laurent expansion for the density matrix

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

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

Bibliography

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