GW (oneshot) + DMFT documentation

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Contents

1 Introduction

This document provides the prescription of the combination of a GW calculation for correlated materials with DMFT applied only to a subset of correlated orbitals. At this level, the GW calculation will be performed only once at the beginning (one-shot) based on a DFT Hamiltonian H^{DFT}, to obtain a nonlocal Selfenergy Σ^{GW} for all states. In addition, the local part of Σ^{GW} of a subset of strongly correlated orbitals will be replaced by a Selfenergy Σ^{DMFT} obtained within a selfconsistent DMFT scheme, where the selfconsistency is done including the full nonlocal effects of the combined Selfenergy.

No further selfconsistency apart from the DMFT cycle will be performed, *i.e.* no update of Σ^{GW} will be done. By this, the final interacting system will be described by a Green's function with the non-interacting DFT dispersion, corrected by a non-local Selfenergy where the non-local components correspond to $\Sigma^{GW} = G_0 W_0$, while the local components of the correlated orbitals correspond to Σ^{DMFT}. This Σ^{DMFT} is usually different to the one obtained by a standard DMFT calculation since the selfconsistency is done with the inclusion of the nonlocal parts of the Selfenergy.

Extensions to a full GW+DMFT selfconsistency will be discussed elsewhere.

2 Approximation to the free energy functional $\Gamma[G, W]$: Combination of GW and DMFT

As stated by Almbladh[ref], the free energy of a solid can be written in terms of a functional $\Gamma[G, W]$ of the fully dressed Green's function G and the screened Coulomb interaction W. While an analytic expression for Γ is not known, it can be shown that it can be separated into a Hartree part Γ^H and a correction arising from all other many-body effects Ψ

$$\Gamma[G, W] = \Gamma^H[G, W] + \Psi[G, W]. \tag{2.1}$$

The many-body correction $\Psi[G, W]$ is the sum of all skeleton diagrams that are irreducible with respect to both one-electron propagator and interaction lines. It has the properties

$$\frac{\delta \Psi}{\delta G} = \Sigma^{XC} \tag{2.2}$$

$$\frac{\delta \Psi}{\delta W} = -\frac{1}{2} P, \tag{2.3}$$

where Σ^{XC} is the exchange-correlation Selfenergy corresponding to the fully dressed Green's function G, thus excluding the Hartree part Σ^H. P is the full polarization of the system that screens the bare Coulomb interaction V down to the screened interaction W.

Since methods like Density Functional Theory already treat the Hartree contribution from the Coulomb interaction, we are interested in obtaining an (approximative) expression for the many-body correction $\Psi[G, W]$.

One possibility is the GW approximation, which expands $\Psi[G, W]$ in powers of the screened interaction W and truncates the series at first order. The resulting expression is thus

$$\Psi[G, W] \approx -\frac{1}{2}\text{Tr}(GWG). \tag{2.4}$$

Using equations (2.2) and (2.3), we immediately obtain the GW Selfenergy and polarization as

$$\Sigma^{GW} = -GW \tag{2.5}$$
$$P^{GW} = GG. \tag{2.6}$$

While this approximation goes well beyond the level of a simple Hartree approximation and usually treats all states without any separation of spaces, it is only an expansion up to first order in W and thus justified only when W is small, i.e. in case of weakly correlated systems. Thus, it is tempting to combine GW with other methods like DMFT for an improved treatment of correlated systems.

In the GW+DMFT scheme, we first separate the Ψ functional into its local and nonlocal parts

$$\Psi[G, W] = \Psi_{\text{nonloc}}[G, W] + \Psi_{\text{loc}}[G, W], \tag{2.7}$$

Is this exact? where usually the nonlocal part is approximated by GW, while the local part is usually approximated by DMFT, but right now we do not want to decide on a specific method and only focus on how to separate the local and nonlocal contributions.

We only want to impose one specific condition: First, we work in an orbital separated scheme, where we separate the full Hilbert space $L + H$ into a correlated subspace L and the remaining subspace H. Then, our defintion of Ψ_{loc} is the following: Ψ_{loc} is generated only from the <u>local</u> components of G and W in the <u>correlated subspace</u>, i.e.

$$\Psi[G, W] = \Psi_{\text{nonloc}}[G, W] + \underbrace{\Psi[G^{\text{loc},L}, W^{\text{loc},L}]}_{\Psi_{\text{loc}}}. \tag{2.8}$$

By this, all internal processes contributing to Ψ_{loc} are restricted to the smaller correlated subspace L and its local G and W. This construction already points to the usage of DMFT for Ψ_{loc}, but it is instructive not to fix on a specific method yet. All other contributions to the full Ψ are now <u>defined</u> to be originating from $\Psi_{\text{nonloc}}[G, W]$. We now have to explain what we actually mean by the two objects Ψ_{nonloc} and Ψ_{loc}.

First, let us start with the nonlocal part Ψ_{nonloc}. Rewriting Eq. (2.8) in the following way naturally leads us to its definition via

$$\Psi[G, W] = \Psi_{\text{nonloc}}[G, W] + \Psi[G^{\text{loc},L}, W^{\text{loc},L}] \tag{2.9}$$

$$= \underbrace{\Psi[G, W] - \Psi[G^{\text{loc},L}, W^{\text{loc},L}]}_{:=\Psi_{\text{nonloc}}} + \Psi[G^{\text{loc},L}, W^{\text{loc},L}]. \tag{2.10}$$

By this, we immediately see that applying any approximation A to Ψ_{nonloc} and Ψ_{loc} will give us the approximate functional $\Psi^A = \Psi^A_{\text{nonloc}} + \Psi^A_{\text{loc}}$.

Side remark: Using the same approximation on both terms will not create any doublecounting or loss of terms, regardless of whether we use an orbital separated scheme or not.

It starts to get really interesting when we use two different approximations A and B to treat the two terms

$$\Psi[G, W] \approx \Psi^A_{\text{nonloc}}[G, W] + \Psi^B_{\text{loc}}[G^{\text{loc},L}, W^{\text{loc},L}]. \tag{2.11}$$

The main point here is IS THERE REALLY NO OVERLAP/DOUBLECOUNTING IN THIS SCHEME?

In the context of GW+DMFT we will now approximate the nonlocal part by GW and the local part by DMFT. The GW approximation is usually performed as a single-shot, and thus based on the DFT Green's function G^0 and RPA screened interaction W^0, while the local functional from DMFT is obtained from the impurity Green's function and interaction, i.e.

$$\Psi[G, W] \approx \Psi^{GW}_{\text{nonloc}}[G, W] + \Psi^{DMFT}_{\text{loc}}[G^{\text{loc},L}, W^{\text{loc},L}] \tag{2.12}$$

$$= -\frac{1}{2}\Big(G^0 W^0 G^0 - G^{0,\text{loc},L} W^{0,\text{loc},L} G^{0,\text{loc},L}\Big) + \Psi^{DMFT}[G^{\text{imp},L}, W^{\text{imp},L}]. \tag{2.13}$$

Please note that in the orbital separated scheme

$$\Sigma^{GW,\text{loc},L} \neq \sum_k \frac{\delta}{\delta G_k^L}\Big(-\frac{1}{2}G^{0,\text{loc},L} W^{0,\text{loc},L} G^{0,\text{loc},L}\Big), \tag{2.14}$$

but this is not relevant here. Using equations (2.2) and (2.3) we obtain for the GW+DMFT Selfenergy and polarization

$$\Sigma_{ab} = \begin{cases} -[G^0 W^0]_{ab} + [G^{0,\text{loc},L} W^{0,\text{loc},L}]_{ab} + \Sigma^{\text{imp},XC,L}_{ab} & \text{for } a, b \in L \\ -[G^0 W^0]_{ab} & \text{for } a \text{ or/and } b \in H \end{cases} \tag{2.15}$$

$$P_{abcd} = \begin{cases} -[G^0 G^0]_{abcd} + [G^{0,\text{loc},L} G^{0,\text{loc},L}]_{abcd} + P^{\text{imp},L}_{abcd} & \text{for } a, b, c, d \in L \\ -[G^0 G^0]_{abcd} & \text{for } \{a, b, c, d\} \cap H \neq \emptyset \end{cases} \tag{2.16}$$

Limiting cases:

W small: In this case the Selfenergy will be well described already by GW, so the impurity solution will basically give the same result $G^{0,\text{loc},L}W^{0,\text{loc},L} = \Sigma^{\text{imp},L}$. The two terms cancel and we fully regain the GW result. NO doublecounting like in FLL LDA+U or LDA+DMFT!

W large: Then the GW Selfenergy contribution within the subspace L is basically given by the impurity solution. There is no mismatch of exchange terms originating from outside the space L, since only the local impurity exchange is removed from GW and replaced by the local impurity DMFT exchange. But this term can be different due to rearrangements of the local impurity charge. This contribution is not yet considered in the GW screening since no GW selfconsistency has been applied. Can we now do another GW calculation only replacing the new contribution from the impurity states? Then we perform a GW selfconsistency including the local vertex corrections from DMFT, in similar spirit as Boehnke et al., but additionally with improved transitions between L and H which they do not update.

3 Product basis

In the GW formalism we encounter objects such as the inverse dielectric function

$$\epsilon^{-1}, \tag{3.1}$$

which is a two-particle operator. The standard way to specify the action of a two-particle operator is to start from a complete orthonormal single-particle basis $\{|i\rangle\}$, where

$$\langle \mathbf{r}|i\rangle = \psi_i(\mathbf{r}), \tag{3.2}$$

with a complex valued function $\psi_i : R^3 \to \mathbb{C}$. Then one introduces a two-particle basis $\{|ij\rangle\}$, which is composed of the single-particle states via

$$\langle \mathbf{rr}'|ij\rangle = \Big(\langle \mathbf{r}| \otimes \langle \mathbf{r}'| \Big)\Big(|i\rangle \otimes |j\rangle \Big) \tag{3.3}$$

$$= \psi_i(\mathbf{r})\psi_j(\mathbf{r}'). \tag{3.4}$$

In this basis, any two-particle operator A can be represented as a rank-4 tensor by its action on the two-particle basis states

$$A_{ijkl} = \langle ij|A|kl\rangle \tag{3.5}$$

$$= \int \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \, \psi_i^*(\mathbf{r})\psi_j^*(\mathbf{r}')A(\mathbf{r}, \mathbf{r}')\psi_l(\mathbf{r}')\psi_k(\mathbf{r}), \tag{3.6}$$

where we have assumed that

$$\langle \mathbf{rr}'|A|\mathbf{r}''\mathbf{r}'''\rangle = A(\mathbf{r}, \mathbf{r}')\delta(\mathbf{r} - \mathbf{r}'')\delta(\mathbf{r}' - \mathbf{r}'''), \tag{3.7}$$

which applies to the Coulomb interaction operator and all other operators we will consider here.

Our goal is to obtain a matrix (rank-2) representation of the two-particle operator A, so that we can define a proper inverse A^{-1} or a multiplication AB between these operators. This is usually done in two ways:

3.1 Index combination

In the two-particle basis the structure of the Tensor-elements

$$A_{ijkl} = \langle ij|A|kl \rangle , \tag{3.8}$$

suggests that we could also interpret each basis state as

$$|a\rangle := |ij\rangle = |i\rangle \times |j\rangle , \tag{3.9}$$

with a single basis state $|a\rangle$, where the index a now runs over N^2 values if we have N single particle states $|i\rangle$. In this notation, we can indeed write the tensor elements as matrix elements

$$A_{ab} = \langle a|A|b\rangle \tag{3.10}$$
$$= \langle ij|A|kl \rangle \tag{3.11}$$
$$= A_{(ij)(kl)}. \tag{3.12}$$

3.1.1 Properties and consistency

The combination of the two left, the "outgoing" indices ij and the right, the "incoming" kl is in principle arbitrary. We could also combine the indices ik of the first partcile and jl of the second particle. Though, the $(ij)(kl)$ combination should be preferrable, since it does not mix vectors $|i\rangle$ with their dual counterpart $\langle i|$.

Furthermore, in cases where we want to apply this scheme, the tensor operations can indeed be rewritten as a matrix multiplication in the combined "ingoing-outgoing" index notation. For example the screened interaction W is given by

$$W_{ijkl} = [v + vPW]_{ijkl} \tag{3.13}$$
$$= v_{ijkl} + \sum_{mnop} v_{ijmn}P_{mnop}W_{opkl}, \tag{3.14}$$

with the bare interaction v and the polarization P. Using the combined index notation we get the representation

$$W_{ab} = W_{(ij)(kl)} \tag{3.15}$$
$$= v_{(ij)(kl)} + \sum_{(mn)(op)} v_{(ij)(mn)}P_{(mn)(op)}W_{(op)(kl)} \tag{3.16}$$
$$= v_{ab} + \sum_{cd} v_{ac}P_{cd}W_{da} \tag{3.17}$$
$$= [v + vPW]_{ab}, \tag{3.18}$$

where vPW is to be understood as the matrix product of v, P and W in the combined index notation.

3.1.2 Tensor inverse

Now let us try to obtain a closed expression of W satisfying this expression, which is not possible in the 4-index tensor notation. For this, we need to define first an object $\mathbb{1}$ that serves as the identity element, which then will allow us to poperly define an inverse of a matrix in the combined index notation. The identity element should have the following property

$$\mathbb{1}A = A = A\mathbb{1}, \tag{3.19}$$

where A is a two-particle tensor, which means

$$A_{(ij)(kl)} = A_{ab} \tag{3.20}$$
$$= [\mathbb{1}A]_{ab} \tag{3.21}$$
$$= \sum_c \mathbb{1}_{ac} A_{ca} \tag{3.22}$$
$$= \sum_{mn} \mathbb{1}_{(ij)(mn)} A_{(mn)(kl)}. \tag{3.23}$$

From this we conclude that

$$\mathbb{1}_{(ij)(mn)} = \delta_{im}\delta_{jn} \tag{3.24}$$
$$\Rightarrow \mathbb{1}_{ac} = \delta_{ac}, \tag{3.25}$$

which leads to the natural definition of the identity element in the combined index notation. It can be directly seen that also $A\mathbb{1} = A$ is fulfilled.

From this we can define the inverse A^{-1} as the standard matrix inverse in the combined index notation that fulfills

$$A^{-1}A = AA^{-1} = \mathbb{1}, \tag{3.26}$$

since

$$[A^{-1}A]_{(ij)(kl)} = [A^{-1}A]_{ab} \tag{3.27}$$
$$= \delta_{ab} \tag{3.28}$$
$$= \delta_{ik}\delta_{kl} \tag{3.29}$$
$$= \mathbb{1}_{(ij)(kl)} \tag{3.30}$$

With this, we can finally solve the equation above for the screened interaction

$$W = v + vPW \tag{3.31}$$
$$\Rightarrow (\mathbb{1} - vP)W = v \tag{3.32}$$
$$\Rightarrow W = (\mathbb{1} - vP)^{-1}v, \tag{3.33}$$

By contruction, the tensor elements $W_{ijkl} = [(\mathbb{1} - vP)^{-1}v]_{ijkl}$ will now satisfy the equation above for the screened interaction.

3.1.3 Problems

Possible problems are:

- A two-particle operator diagonal in position representation cannot be inverted in the combined index-notation! This can be for example a purely local Coulomb interaction!

  Consider

  $$\langle \mathbf{r}\mathbf{r}'|A|\mathbf{r}''\mathbf{r}'''\rangle = A(\mathbf{r})\delta(\mathbf{r} - \mathbf{r}')\delta(\mathbf{r} - \mathbf{r}'')\delta(\mathbf{r}' - \mathbf{r}'''), \tag{3.34}$$

  and we assume that $A(\mathbf{r}) = a > 0$ constant, *i.e.* it does not matter where the particles interact. As long as their positions are identical they pick up a factor a.

  This leads to the following tensor elements

  $$A_{ijkl} = \langle ij|A|kl\rangle \tag{3.35}$$

  $$= \int\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}'\, \psi_i^*(\mathbf{r})\psi_j^*(\mathbf{r}')a\delta(\mathbf{r} - \mathbf{r}')\psi_l(\mathbf{r}')\psi_k(\mathbf{r}) \tag{3.36}$$

  $$= a\int \mathrm{d}\mathbf{r}\, \psi_i^*(\mathbf{r})\psi_j^*(\mathbf{r})\psi_l(\mathbf{r})\psi_k(\mathbf{r}). \tag{3.37}$$

  For the case of real wave functions we see that we always get a non-zero contribution when we pair two indices with one another, leading to an integral of the form

  $$a\int \mathrm{d}\mathbf{r}\, \psi_m^2(\mathbf{r})\psi_n^2(\mathbf{r}) \neq 0 \tag{3.38}$$

  For the example of $N = 2$ single-particle states $\psi_i(\mathbf{r})$, we can choose two out of four indices to pair, then the last one has to be paired with $N = 2$ choices for the index, leading to 8 combinations which are at least non-zero. In the combined index notation we then can arrive at the following matrix (Note: I have confirmed this numerically for a few basis sets)

  $$A = a\begin{pmatrix} c_1 & 0 & 0 & c_3 \\ 0 & c_2 & c_2 & 0 \\ 0 & c_2 & c_2 & 0 \\ c_3 & 0 & 0 & c_4 \end{pmatrix}, \tag{3.39}$$

  where we can immediately see that this matrix cannot be inverted since two columns are linearly dependent (even identical).

- A "constant" operator $A(\mathbf{r}, \mathbf{r}') = c \neq 0$ can be inverted! (See discussion in next section)

3.2 Aryasetiawan-style

3.2.1 Defining the new basis set

We start by rewriting the equation for the tensor elements in the two-particle basis in the following way

$$\langle ij|A|kl\rangle = \int\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}'\, \psi_i^*(\mathbf{r})\psi_j^*(\mathbf{r}')A(\mathbf{r},\mathbf{r}')\psi_l(\mathbf{r}')\psi_k(\mathbf{r}) \tag{3.40}$$

$$= \int\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}'\, \psi_i^*(\mathbf{r})\psi_k(\mathbf{r})A(\mathbf{r},\mathbf{r}')\psi_j^*(\mathbf{r}')\psi_l(\mathbf{r}') \tag{3.41}$$

$$= \int\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}'\, \underbrace{\left(\psi_k^*(\mathbf{r})\psi_i(\mathbf{r})\right)^*}_{B_a^*(\mathbf{r})} A(\mathbf{r},\mathbf{r}') \underbrace{\psi_j^*(\mathbf{r}')\psi_l(\mathbf{r}')}_{B_b(\mathbf{r})} \tag{3.42}$$

$$= \langle B_a|A|B_b\rangle. \tag{3.43}$$

From these observation we see that we should define new basis states $\{B_a\}$ by the product of the single-particle states by

$$\langle\mathbf{r}|B_a\rangle := \psi_i^*(\mathbf{r})\psi_j(\mathbf{r}), \tag{3.44}$$

where the index $a := a(i,j)$ lables the combination of the indices i,j. As a result, for a finite number of single-particle states $N = \dim\{|i\rangle\}$, we have N^2 new basis states $\{B_a\}$, and could define the relation between the indices as

$$a(i,j) := iN + j. \tag{3.45}$$

Though, the new set $\{B_a\}$ is *not* a basis, since it is overcomplete/linearly dependent and in addition not orthonormal. For example if there exist at least two real single-particle basis functions ψ_i, ψ_j, one has

$$\psi_i(\mathbf{r})\psi_j(\mathbf{r}) = B_a(\mathbf{r}) = B_b(\mathbf{r}) = \psi_j(\mathbf{r})\psi_i(\mathbf{r}), \tag{3.46}$$

where $a \neq b$. Also, for a states with indices $a(i,i)$ and $b(i,i)$, which leads to

$$\langle B_a|B_b\rangle = \int \mathrm{d}\mathbf{r}\, \psi_i^*(\mathbf{r})\psi_i(\mathbf{r})\psi_j^*(\mathbf{r})\psi_j(\mathbf{r}) \tag{3.47}$$

$$= \int \mathrm{d}\mathbf{r}\, |\psi_i(\mathbf{r})|^2|\psi_j(\mathbf{r})|^2 \tag{3.48}$$

which is usually larger than zero for $a \neq b$, i.e. $i = j$ and not equal to 1 in case $a = b$, i.e. $i \neq j$.

The overlap matrix for the new set $\{B_a\}$ is therefore different from the unit matrix. If we define

$$v := \left(|B_1\rangle \quad |B_2\rangle \quad |B_3\rangle \quad \cdots \quad |B_{N^2}\rangle\right), \tag{3.49}$$

then the overlap matrix can be written as

$$O = v^\dagger \cdot v, \tag{3.50}$$

since

$$O_{ab} = (v^\dagger \cdot v)_{ab} \tag{3.51}$$

$$= \langle B_a | B_b \rangle . \tag{3.52}$$

Due to the overcompleteness, some columns of O will not be linear independent, i.e. some of the Eigenvalues λ_i of O will be equal to zero.

3.2.2 Reduction of the basis set and reorthonormalization

After diagonalizing O to obtain the Eigenvectors $o_1, o_2, ..., o_{N^2}$, we throw away the ones with Eigenvalue zero and set up the matrices

$$U = \begin{pmatrix} o_1 & o_2 & o_3 & \cdots o_{N_r} \end{pmatrix} \in \mathbb{C}^{N^2 \times N_r}, \tag{3.53}$$

where $N_r \leq N^2$ is the number of Eigevectors with nonzero Eigenvalue, and

$$D^{-1/2} := \mathrm{diag}(\lambda_1^{-1/2},\ \lambda_2^{-1/2},\ \lambda_3^{-1/2},\ \cdots,\ \lambda_{N_r}^{-1/2}) \in \mathbb{R}^{N_r \times N_r}, \tag{3.54}$$

where λ_i are the nonzero Eigenvalues of O.

The elements of the following vector

$$\tilde{v} := vUD^{-1/2}, \tag{3.55}$$

will then yield a new set of N_r basis functions which are complete and orthonormal, as can be seen from the new overlap matrix

$$\tilde{O} = \tilde{v}^\dagger \cdot \tilde{v} \tag{3.56}$$

$$= D^{-1/2} U^\dagger (v^\dagger \cdot v) U D^{-1/2} \tag{3.57}$$

$$= D^{-1/2} \underbrace{U^\dagger O U}_{=D} D^{-1/2} \tag{3.58}$$

$$= \mathbb{1} \in \mathbb{R}^{N_r \times N_r}. \tag{3.59}$$

Remark: To reduce the size of the basis for computational efficiency, one can also exclude Eigenvectors with a finite, but small Eigenvalue, i.e. below some treshhold $\delta > \lambda_i$. Then the new basis will only be approximately complete and orthonormal, but the error can be controlled by choosing δ sufficiently small.

3.2.3 Product basis matrix elements

After having obtained the new complete orthonormal basis $\{\tilde{B}_a\}$, the matrix elements of any two-particle operator are then given as

$$A_{ab} = \langle \tilde{B}_a | A | \tilde{B}_b \rangle \tag{3.60}$$

$$= \int \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}'\, \tilde{B}_a^*(\mathbf{r}) A(\mathbf{r}, \mathbf{r}') \tilde{B}_b(\mathbf{r}'). \tag{3.61}$$

The representation of A in the product basis is then given as

$$A = \sum_{a,b} |\tilde{B}_a\rangle A_{ab} \langle \tilde{B}_b| . \tag{3.62}$$

or in the position representation DOES THIS MAKE SENSE?

$$A(\mathbf{r}, \mathbf{r}') = \sum_{a,b} \langle \mathbf{r} | \tilde{B}_a \rangle A_{ab} \langle \tilde{B}_b | \mathbf{r}' \rangle \tag{3.63}$$

$$= \sum_{a,b} \tilde{B}_a(\mathbf{r}) A_{ab} \tilde{B}_b^*(\mathbf{r}'). \tag{3.64}$$

CHECK HERE IF BOTH REPRESENTATIONS LEAD TO THE SAME RESULT!!! The product basis should recover the right A(r-r'), shouldn't it? (I think so...) Can we actually prove this?

3.2.4 Switching between the product and the two-particle basis

If we want to obtain the original tensor representation of a two-particle operator in the product basis, we have to evaluate DOES THIS MAKE SENSE?

$$A_{ijkl} = \langle ij | A | kl \rangle \tag{3.65}$$

$$= \int \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{r}' \, \psi_i^*(\mathbf{r}) \psi_j^*(\mathbf{r}') A(\mathbf{r}, \mathbf{r}') \psi_l(\mathbf{r}') \psi_k(\mathbf{r}) \tag{3.66}$$

$$= \sum_{a,b} \int \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{r}' \, \psi_i^*(\mathbf{r}) \psi_j^*(\mathbf{r}') \tilde{B}_a(\mathbf{r}) A_{ab} \tilde{B}_b^*(\mathbf{r}') \psi_l(\mathbf{r}') \psi_k(\mathbf{r}). \tag{3.67}$$

The other way round, if we have a two-particle operator given in the two-particle basis, the product basis representation can be obtained by

$$A_{ab} = \langle \tilde{B}_a | A | \tilde{B}_b \rangle \tag{3.68}$$

$$= \int \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{r}' \, \tilde{B}_a^*(\mathbf{r}) A(\mathbf{r}, \mathbf{r}') \tilde{B}_b(\mathbf{r}') \tag{3.69}$$

$$= \sum_{ijkl} \int \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{r}' \, \tilde{B}_a^*(\mathbf{r}) \psi_i(\mathbf{r}) \psi_j(\mathbf{r}') A_{ijkl} \psi_l^*(\mathbf{r}') \psi_k^*(\mathbf{r}) \tilde{B}_b(\mathbf{r}'). \tag{3.70}$$

3.2.5 Tensor inverse

Since in the product basis any two-particle operator can now be represented as a matrix/rank-2 tensor, we can finally define its inverse $\tilde{A}^{-1}$ as the standard Matrix inverse, which then in the product basis fulfils the property

$$(\tilde{A}^{-1}A)_{ab} = \langle \tilde{B}_a | \tilde{A}^{-1} A | \tilde{B}_b \rangle \tag{3.71}$$

$$= \delta_{ab}. \tag{3.72}$$

In the position representation

$$\langle \mathbf{r} | \tilde{A}^{-1} A | \mathbf{r}' \rangle = \sum_{a,b} \langle \mathbf{r} | \tilde{B}_a \rangle \, (\tilde{A}^{-1} A)_{ab} \, \langle \tilde{B}_b | \mathbf{r}' \rangle \tag{3.73}$$

$$= \sum_{a,b} \langle \mathbf{r} | \tilde{B}_a \rangle \, \delta_{ab} \, \langle \tilde{B}_b | \mathbf{r}' \rangle \tag{3.74}$$

$$= \sum_{a} \langle \mathbf{r} | \tilde{B}_a \rangle \, \langle \tilde{B}_a | \mathbf{r}' \rangle \tag{3.75}$$

$$= \langle \mathbf{r} | \mathbf{r}' \rangle \tag{3.76}$$

$$= \delta(\mathbf{r} - \mathbf{r}'), \tag{3.77}$$

if the product basis is complete.

3.2.6 Problems

Possible problems are:

- A "constant" tensor of the form $A(\mathbf{r}, \mathbf{r}') = c \neq 0$ cannot be inverted! I'm not sure, this is maybe correct behaviour?

  Let us assume we obtain a final basis state $|\tilde{B}_o\rangle$ in a one-dimensional system which is a real odd function in position representation, *i.e.*

  $$\tilde{B}_o(r) = -\tilde{B}_o(-r). \tag{3.78}$$

  For a constant tensor this leads to the following

  $$A_{ab} = c \int \int \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{r}' \, \tilde{B}_a^*(\mathbf{r}) \tilde{B}_b(\mathbf{r}') \tag{3.79}$$

  $$= c \left(\int \mathrm{d}\mathbf{r} \, \tilde{B}_a^*(\mathbf{r}) \right) \left(\int \mathrm{d}\mathbf{r}' \, \tilde{B}_b(\mathbf{r}'), \right), \tag{3.80}$$

  i.e. the two integrations over the basis states decouple, and everytime a or b is equal to the real odd function $\tilde{B}_o$, we obtain zero. Therefore, the o-th column and row is equal to zero. Which means, we have at least one Eigenvalue equal to zero and, therefore, the tensor in the product basis cannot be inverted.

4 The GW part

On the basis of H^{DFT} a $G_0 W_0$ calculation has to be performed on the full system. By this, the Selfenergy in the Kohn-Sham basis is obtained for all states

$$\Sigma_{\nu\nu'}(k, \omega) = [G_0 W_0]_{\nu\nu'}(k, \omega). \tag{4.1}$$

By this, the GW estimate for the full interacting Green's function is given by

$$G_{\nu\nu'}^{GW}(k, \omega) = \left[\mathbb{1}(\omega + \mu + i\delta) - H^{DFT}(k) + v^{XC}(k) - \Sigma^{GW}(k, \omega) \right]_{\nu\nu'}^{-1}. \tag{4.2}$$

4.1 Output for DMFT

At this point the basis transformation to the local Wannier basis will be performed on the GW side. For the next step of the DMFT calculation one needs on a mesh in k-space in the full Brillouin zone:

- $\epsilon_m(k)$: The eigenvalues of $H^{DFT}(k)$ in the Wannier basis for all relevant orbitals

- μ: The chemical potential that yields the correct physical number of electrons N_e. It is not needed if all $\epsilon_m(k)$ are given with respect to the Fermi level. for H^{DFT}

- $v_{mm'}^{XC}(k)$: The value of the exchange-correlation potential in the Wannier basis for all relevant orbitals

- $\Sigma_{mm'}^{GW}(k, i\omega)$: The Selfenergy within GW in the Wannier basis for all relevant orbitals on imaginary frequencies ω.

- β: The inverse temperature used for defining $\omega_n = (2n + 1)\pi/\beta$.

All output from the GW calculation will be in atomic units and have to be converted to eV!!

5 The DMFT part

Within DMFT we then calculate a local correction Σ^{DMFT} for a subset of correlated Wannier orbitals.

The input of the calculation will be the output of the GW calculation. First, one will usually apply a Wannier-interpolation of the GW data to obtain a fine k-mesh since the GW output will be given on a very coarse grid.

Since the Hartree term is already incorporated on the DMFT level, it has to be excluded from the Selfenergies in GW (already taken care of) and in DMFT (has to be done in the selfconsistency).

5.1 The self-consistency cycle

We then proceed as follows:

1. Calculate the local diagonal part of the GW Selfenergy ONLY for the correlated subset of orbitals f that will be later replaced by the DMFT result

$$\Sigma_f^{GW,loc}(i\omega_n) = \frac{1}{N_k} \sum_k \Sigma_{ff}^{GW}(k, i\omega_n). \tag{5.1}$$

2. Make a first guess for the local impurity Selfenergy of the correlated orbitals Σ_{ff}^{imp}, for example one can use the GW result

$$\Sigma_{ff}^{imp}(i\omega_n) = \Sigma_f^{GW,loc}(i\omega_n). \tag{5.2}$$

3. Set up the interacting Green's function, where the local component of the GW Selfenergy for the correlated orbitals f is replaced by Σ^{imp} and the Hartree part $\Sigma_f^{H,imp}$ is substracted from Σ^{imp}. In the first step when using $\Sigma_{ff}^{imp} = \Sigma_f^{GW,loc}$ this term is zero.

$$G_{mm'}(k, i\omega_n) = \Big[\mathbb{1}(i\omega_n + \mu) - H^{DFT}(k) + v^{XC}(k) \tag{5.3}$$

$$- \Sigma^{GW}(k, i\omega_n) + \Sigma_f^{GW,loc}(i\omega_n) - \Sigma^{imp}(i\omega_n) + \Sigma_f^{H,imp}\Big]_{mm'}^{-1}. \tag{5.4}$$

Adjust the chemical potential μ in a way such that the desired filling

$$N_e = \lim_{\tau \to 0^-} \frac{1}{\beta N_k} \sum_{i\omega_n} \sum_{k,m} G_{mm}(k, i\omega_n) e^{-i\omega_n \tau}. \tag{5.5}$$

is obtained.

4. Calculate the local Green's function (for all orbitals) then and Weiss field $\mathscr{G}$ ONLY on the subset of correlated orbitals f. Neglecting offdiagonal components in the hybridization we are also only interested in the diagonal components of $\mathscr{G}$

$$G_{mm'}^{loc}(i\omega_n) = \frac{1}{N_k} \sum_k G_{mm'}(k, i\omega_n), \tag{5.6}$$

$$\Rightarrow \mathscr{G}_{ff}^{-1}(i\omega_n) = [G^{loc}]_{ff}^{-1}(i\omega_n) + \Sigma_{ff}^{imp}(i\omega_n). \tag{5.7}$$

Here, the full Σ_{ff}^{imp} has to be used, since the Hartree part is calculated in the impurity model.

The Weiss field matrix $\mathscr{G}$ is not explicitly needed, so we do not invert the last equation to obtain $\mathscr{G}$.

5. Calculate the Hybridization function

$$\Delta_{ff}(i\omega_n) = i\omega_n + \tilde{\mu}_f - \mathscr{G}_{ff}^{-1}(i\omega_n), \tag{5.8}$$

where the local chemical potential $\tilde{\mu}_f$ is given by

$$\tilde{\mu}_f = \lim_{\omega_n \to \infty} \mathrm{Re}\left[\mathscr{G}_{ff}^{-1}(i\omega_n)\right] \tag{5.9}$$

and transform $\Delta(i\omega_n)$ to the imaginary time axis τ by a Fourier transform

$$\Delta_{ff}(\tau) = \frac{1}{\beta} \sum_{i\omega_n} \Delta_{ff}(i\omega_n) e^{-i\omega_n \tau} \tag{5.10}$$

and solve the impurity model for the correlated f orbitals.

6. Obtain the new local Selfenergy $\Sigma_{ff}^{imp}(i\omega_n)$ from the impurity model and calculate the updated Hartree correction from the impurity occupations as given by Eq. (6.14) and (6.30). Then go back to step 3. Repeat until convergence is reached.

5.2 Output

After convergence, $e.g.$ the local spectral function $A_m(\omega)$ can be obtained by analytic continuation of $G_{mm}^{loc}(i\omega_n)$.

6 Hartree- and Exchange term in DMFT

The derivation follows the ideas of Haule PRL 115, 196403 (2015).

6.1 Hartree term

The Hartree energy has the general form

$$E^H[\rho] = \frac{1}{2} \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \, \rho(\mathbf{r}) V_C(\mathbf{r} - \mathbf{r}')\rho(\mathbf{r}') \tag{6.1}$$

$$= \frac{1}{2} \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \, \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}, \tag{6.2}$$

where $\rho(\mathbf{r})$ is the sum of all spin-components

$$\rho(\mathbf{r}) = \rho_\uparrow(\mathbf{r}) + \rho_\downarrow(\mathbf{r}). \tag{6.3}$$

In order to evaluate these term for DMFT we introduce a local orbital basis $|\chi_m^\sigma\rangle$, and replace the bare Coulomb interaction $V_C(\mathbf{r}-\mathbf{r}')$ by an effective screened Coulomb interaction $V_{DMFT}(\mathbf{r} - \mathbf{r}')$. This leads to

$$E^H[\rho] = \frac{1}{2} \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \, \rho(\mathbf{r}) V_C(\mathbf{r} - \mathbf{r}')\rho(\mathbf{r}') \tag{6.4}$$

$$= \frac{1}{2} \sum_{\substack{klmn \\ \sigma\sigma'}} \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \, \langle\mathbf{r}|\chi_k^\sigma\rangle \, \langle\chi_k^\sigma|\rho|\chi_l^\sigma\rangle \, \langle\chi_l^\sigma|\mathbf{r}\rangle \, V_{DMFT}(\mathbf{r} - \mathbf{r}')$$

$$\times \langle\mathbf{r}'|\chi_m^{\sigma'}\rangle \, \langle\chi_m^{\sigma'}|\rho|\chi_n^{\sigma'}\rangle \, \langle\chi_n^{\sigma'}|\mathbf{r}'\rangle \tag{6.5}$$

$$= \frac{1}{2} \sum_{\substack{klmn \\ \sigma\sigma'}} \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}'(\chi_l^\sigma)^*(\mathbf{r})(\chi_n^{\sigma'})^*(\mathbf{r}')V_{DMFT}(\mathbf{r} - \mathbf{r}')\chi_m^{\sigma'}(\mathbf{r}')\chi_k^\sigma(\mathbf{r})$$

$$\times \langle\chi_k^\sigma|\rho|\chi_l^\sigma\rangle \, \langle\chi_m^{\sigma'}|\rho|\chi_n^{\sigma'}\rangle \,. \tag{6.6}$$

In the last equation we can now identify the matrix elements of the local screened Coulomb interaction

$$\langle ln|U|km\rangle = \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}'(\chi_l^\sigma)^*(\mathbf{r})(\chi_n^{\sigma'})^*(\mathbf{r}')V_{DMFT}(\mathbf{r} - \mathbf{r}')\chi_m^{\sigma'}(\mathbf{r}')\chi_k^\sigma(\mathbf{r}), \tag{6.7}$$

and the DMFT density matrix

$$\langle\chi_k^\sigma|\rho|\chi_l^\sigma\rangle = n_{kl}^\sigma. \tag{6.8}$$

With this, the Hartree energy takes on the form

$$E^{DMFT} = \frac{1}{2} \sum_{\substack{klmn \\ \sigma\sigma'}} \langle ln|U|km \rangle \, n_{kl}^{\sigma} n_{mn}^{\sigma'}. \tag{6.9}$$

In the impurity model we restrict ourselves to diagonal density matrices, which leads to

$$E_{DMFT}^{H} = \frac{1}{2} \sum_{\substack{km \\ \sigma\sigma'}} \langle km|U|km \rangle \, n_{k}^{\sigma} n_{m}^{\sigma'}. \tag{6.10}$$

This leads to the following Hartree part of the Selfenergy in DMFT

$$\Sigma_{l\sigma}^{H,DMFT} = \frac{\partial}{\partial n_{l}^{\sigma}} E_{DMFT}^{H} \tag{6.11}$$

$$= \frac{1}{2} \sum_{m,\sigma'} \langle lm|U|lm \rangle \, n_{m}^{\sigma'} + \frac{1}{2} \sum_{k,\sigma'} \langle kl|U|kl \rangle \, n_{k}^{\sigma'} \tag{6.12}$$

$$= \sum_{m,\sigma'} \langle lm|U|lm \rangle \, n_{m}^{\sigma'} \tag{6.13}$$

$$= U_0(n_l^{\uparrow} + n_l^{\downarrow}) + \sum_{m \neq l}(U_0 - 2J_{lm})(n_m^{\uparrow} + n_m^{\downarrow}). \tag{6.14}$$

6.2 Exchange term

The exact exchange energy has the general form

$$E^{X}[\rho] = -\frac{1}{2} \sum_{\sigma} \int d\mathbf{r} d\mathbf{r}' \, \rho_{\sigma}(\mathbf{r}, \mathbf{r}') V_C(\mathbf{r} - \mathbf{r}') \rho_{\sigma}(\mathbf{r}', \mathbf{r}) \tag{6.15}$$

$$= -\frac{1}{2} \sum_{\sigma} \int d\mathbf{r} d\mathbf{r}' \, \frac{\rho_{\sigma}(\mathbf{r}, \mathbf{r}') \rho_{\sigma}(\mathbf{r}', \mathbf{r})}{|\mathbf{r} - \mathbf{r}'|}. \tag{6.16}$$

In order to evaluate these term for DMFT we introduce a local orbital basis $|\chi_m^{\sigma}\rangle$, and replace the bare Coulomb interaction $V_C(\mathbf{r} - \mathbf{r}')$ by an effective screened Coulomb interaction $V_{DMFT}(\mathbf{r} - \mathbf{r}')$. This leads to

$$E^{X}[\rho] = -\frac{1}{2} \sum_{\sigma} \int d\mathbf{r} d\mathbf{r}' \, \rho_{\sigma}(\mathbf{r}, \mathbf{r}') V_{DMFT}(\mathbf{r} - \mathbf{r}') \rho_{\sigma}(\mathbf{r}', \mathbf{r}) \tag{6.17}$$

$$= -\frac{1}{2} \sum_{\substack{klmn \\ \sigma}} \int d\mathbf{r} d\mathbf{r}' \, \langle \mathbf{r}|\chi_k^{\sigma} \rangle \langle \chi_k^{\sigma}|\rho_{\sigma}|\chi_l^{\sigma} \rangle \langle \chi_l^{\sigma}|\mathbf{r}' \rangle V_{DMFT}(\mathbf{r} - \mathbf{r}')$$

$$\times \langle \mathbf{r}'|\chi_m^{\sigma} \rangle \langle \chi_m^{\sigma}|\rho_{\sigma}|\chi_n^{\sigma} \rangle \langle \chi_n^{\sigma}|\mathbf{r} \rangle \tag{6.18}$$

$$= -\frac{1}{2} \sum_{\substack{klmn \\ \sigma}} \int d\mathbf{r} d\mathbf{r}' \, (\chi_n^{\sigma})^{*}(\mathbf{r})(\chi_l^{\sigma})^{*}(\mathbf{r}') V_{DMFT}(\mathbf{r} - \mathbf{r}') \chi_m^{\sigma}(\mathbf{r}') \chi_k^{\sigma}(\mathbf{r})$$

$$\times \langle \chi_m^{\sigma}|\rho_{\sigma}|\chi_n^{\sigma} \rangle \langle \chi_k^{\sigma}|\rho_{\sigma}|\chi_l^{\sigma} \rangle. \tag{6.19}$$

In the last equation we can now identify the matrix elements of the local screened Coulomb interaction

$$\langle nl|U|km\rangle = \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}'\,(\chi_n^\sigma)^*(\mathbf{r})(\chi_l^\sigma)^*(\mathbf{r}')V_{DMFT}(\mathbf{r}-\mathbf{r}')\chi_m^\sigma(\mathbf{r}')\chi_k^\sigma(\mathbf{r}), \qquad (6.20)$$

and the DMFT density matrix

$$\langle \chi_m^\sigma|\rho_\sigma|\chi_n^\sigma\rangle = n_{mn}^\sigma. \qquad (6.21)$$

With this, the exchange energy takes on the form

$$E_{DMFT}^X = -\frac{1}{2}\sum_{\substack{klmn\\\sigma}} \langle nl|U|km\rangle\, n_{mn}^\sigma n_{kl}^\sigma. \qquad (6.22)$$

In the impurity model we restrict ourselves to diagonal density matrices, which leads to

$$E_{DMFT}^X = -\frac{1}{2}\sum_{mk,\sigma} \langle mk|U|km\rangle\, n_m^\sigma n_k^\sigma. \qquad (6.23)$$

This leads to the following exchange part of the Selfenergy in DMFT

$$\Sigma_{l\sigma}^{X,DMFT} = \frac{\partial}{\partial n_l^\sigma}E_{DMFT}^X \qquad (6.24)$$

$$= -\frac{1}{2}\sum_k \langle lk|U|kl\rangle\, n_k^\sigma - \frac{1}{2}\sum_m \langle ml|U|lm\rangle\, n_m^\sigma \qquad (6.25)$$

$$= -\sum_k \langle lk|U|kl\rangle\, n_k^\sigma \qquad (6.26)$$

$$= -U_0\, n_l^\sigma - \sum_{k\neq l} J_{lk}\, n_k^\sigma. \qquad (6.27)$$

6.3 Hartree + exchange Selfenergy

For consistency checks, we add up the Hartree and the exchange contribution to the Selfenergy and obtain

$$\Sigma_{l\sigma}^{H,DMFT} + \Sigma_{l\sigma}^{X,DMFT} = U_0 n_l^{\bar\sigma} + \sum_{m\neq l}(U_0 - 2J_{lm})n_m^{\bar\sigma}$$
$$+ \sum_{m\neq l}(U_0 - 3J_{lm})n_m^\sigma \qquad (6.28)$$
$$= \lim_{\omega_n\to\infty} \Sigma^{DMFT}(i\omega_n), \qquad (6.29)$$

which is identical to the high-frequency limit of the true DMFT Selfenergy. This term is also equal to the sum of all first order diagrams to the DMFT Selfenergy, i.e. the Hartree- and the Fock diagram.

6.4 Dynamical interactions

In the case of dynamical interactions $U(\omega)$ in the Hartree and exchange part the screened Coulomb matrix elements recover their bare values is this correct?, i.e. U_0 has to be replaced by the bare V (assuming no frequency dependence of the Hund's coupling)

$$\Sigma_{l\sigma}^{H,DMFT} = V(n_l^\uparrow + n_l^\downarrow) + \sum_{m \neq l}(V - 2J_{lm})(n_m^\uparrow + n_m^\downarrow) \tag{6.30}$$

$$\Sigma_{l\sigma}^{X,DMFT} = -V\,n_l^\sigma - \sum_{k \neq l} J_{lk}\,n_k^\sigma. \tag{6.31}$$

CAUTION! Does U_0 or F_0 recover the bare interaction? If $F_0 = V$, then U_0 has to be replaced in the 5-orbital model by $V + \frac{8}{7}J_{avg}$.

7 Implementation details

7.1 Impurity solver input

The CT-HYB impurity solver by Yusuke needs the following input files

dmft.input Includes information about U,J, number of frequencies, etc. At the moment possible: Only 3-fold degenerate orbitals. No freq. dependent U.

hyb_tau.dat The hybridization function as a matrix for imaginary time. It needs to be diagonal!
Only real part, one column. Seperate matrix elements via two line breaks and `# hyb    2    1` etc. We need `Nmesh+1` points where the endpoints $\tau = 0, \beta$ are included! By convention has negative sign. The local orbital levels are assumed to be $\tilde{\mu} = 0$ and any shift is absorbed in the chemical potential! This has to be checked for consistency!!!

omega_mesh.dat Specifies the bosonic frequency grid for some correlation functions. Just reuse the standard template file. Not important for us.

fort.10* Includes information about the Monte-Carlo configuration used for starting the sampling. Is initialized once with Yusuke's code and then overwritten by the solver. No change required here.