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 nonlocal 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 can treat the Hartree contribution from the Coulomb interaction quite well, 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 truncated 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

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

Is this exact? and approximate the nonlocal part by GW, while the local part is approximated by DMFT

$$\Psi[G, W] \approx \Psi_{\mathrm{nonloc}}^{GW}[G, W] + \Psi_{\mathrm{loc}}^{DMFT}[G, W]. \tag{2.8}$$

We now have to define what we actually mean by the two objects $\Psi_{\mathrm{nonloc}}^{GW}[G, W]$ and $\Psi_{\mathrm{loc}}^{DMFT}[G, W]$.

3 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

$$\Sigma_{\nu\nu'}(k, \omega) = [G_0 W_0]_{\nu\nu'}(k, \omega). \tag{3.1}$$

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

$$G_{\nu\nu'}^{GW}(k, \omega) = \left[\mathbb{1}(\omega + \mu + i\delta) - H^{DFT}(k) + v^{XC}(k) - \Sigma^{GW}(k, \omega)\right]_{\nu\nu'}^{-1}. \tag{3.2}$$

3.1 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!!

4 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).

4.1 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

$$\Sigma_f^{GW,loc}(i\omega_n) = \frac{1}{N_k} \sum_k \Sigma_{ff}^{GW}(k, i\omega_n). \tag{4.1}$$

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

$$\Sigma_{ff}^{imp}(i\omega_n) = \Sigma_f^{GW,loc}(i\omega_n). \tag{4.2}$$

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.

$$G_{mm'}(k, i\omega_n) = \big[\mathbb{1}(i\omega_n + \mu) - H^{DFT}(k) + v^{XC}(k) \tag{4.3}$$

$$- \Sigma^{GW}(k, i\omega_n) + \Sigma_f^{GW,loc}(i\omega_n) - \Sigma^{imp}(i\omega_n) + \Sigma_f^{H,imp} \big]_{mm'}^{-1}. \tag{4.4}$$

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

$$N_e = \lim_{\tau \to 0^-} \frac{1}{\beta N_k} \sum_{i\omega_n} \sum_{k,m} G_{mm}(k, i\omega_n) e^{-i\omega_n \tau}. \tag{4.5}$$

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 $\mathscr{G}$

$$G_{mm'}^{loc}(i\omega_n) = \frac{1}{N_k} \sum_k G_{mm'}(k, i\omega_n), \tag{4.6}$$

$$\Rightarrow \mathscr{G}_{ff}^{-1}(i\omega_n) = [G^{loc}]_{ff}^{-1}(i\omega_n) + \Sigma_{ff}^{imp}(i\omega_n). \tag{4.7}$$

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

$$\Delta_{ff}(i\omega_n) = i\omega_n + \tilde{\mu}_f - \mathscr{G}_{ff}^{-1}(i\omega_n), \tag{4.8}$$

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

$$\tilde{\mu}_f = \lim_{\omega_n \to \infty} \mathrm{Re}\left[\mathscr{G}_{ff}^{-1}(i\omega_n)\right] \tag{4.9}$$

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

$$\Delta_{ff}(\tau) = \frac{1}{\beta} \sum_{i\omega_n} \Delta_{ff}(i\omega_n)\mathrm{e}^{-i\omega_n\tau} \tag{4.10}$$

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. (5.14) and (5.30). Then go back to step 3. Repeat until convergence is reached.

4.2 Output

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

5 Hartree- and Exchange term in DMFT

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

5.1 Hartree term

The Hartree energy has the general form

$$E^H[\rho] = \frac{1}{2} \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \, \rho(\mathbf{r})V_C(\mathbf{r} - \mathbf{r}')\rho(\mathbf{r}') \tag{5.1}$$

$$= \frac{1}{2} \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \, \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}, \tag{5.2}$$

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

$$\rho(\mathbf{r}) = \rho_\uparrow(\mathbf{r}) + \rho_\downarrow(\mathbf{r}). \tag{5.3}$$

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

$$E^H[\rho] = \frac{1}{2} \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \, \rho(\mathbf{r}) V_C(\mathbf{r}-\mathbf{r}') \rho(\mathbf{r}') \tag{5.4}$$

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

$$\times \langle\mathbf{r}'|\chi_m^{\sigma'}\rangle \langle\chi_m^{\sigma'}|\rho|\chi_n^{\sigma'}\rangle \langle\chi_n^{\sigma'}|\mathbf{r}'\rangle \tag{5.5}$$

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

$$\times \langle\chi_k^\sigma|\rho|\chi_l^\sigma\rangle \langle\chi_m^{\sigma'}|\rho|\chi_n^{\sigma'}\rangle . \tag{5.6}$$

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

$$\langle ln|U|km\rangle = \int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' (\chi_l^\sigma)^*(\mathbf{r})(\chi_n^{\sigma'})^*(\mathbf{r}') V_{DMFT}(\mathbf{r}-\mathbf{r}') \chi_m^{\sigma'}(\mathbf{r}') \chi_k^\sigma(\mathbf{r}), \tag{5.7}$$

and the DMFT density matrix

$$\langle\chi_k^\sigma|\rho|\chi_l^\sigma\rangle = n_{kl}^\sigma. \tag{5.8}$$

With this, the Hartree energy takes on the form

$$E^{DMFT} = \frac{1}{2} \sum_{\substack{klmn \\ \sigma\sigma'}} \langle ln|U|km\rangle \, n_{kl}^\sigma n_{mn}^{\sigma'}. \tag{5.9}$$

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

$$E_{DMFT}^H = \frac{1}{2} \sum_{\substack{km \\ \sigma\sigma'}} \langle km|U|km\rangle \, n_k^\sigma n_m^{\sigma'}. \tag{5.10}$$

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

$$\Sigma_{l\sigma}^{H,DMFT} = \frac{\partial}{\partial n_l^\sigma} E_{DMFT}^H \tag{5.11}$$

$$= \frac{1}{2} \sum_{m,\sigma'} \langle lm|U|lm\rangle \, n_m^{\sigma'} + \frac{1}{2} \sum_{k,\sigma'} \langle kl|U|kl\rangle \, n_k^{\sigma'} \tag{5.12}$$

$$= \sum_{m,\sigma'} \langle lm|U|lm\rangle \, n_m^{\sigma'} \tag{5.13}$$

$$= U_0(n_l^\uparrow + n_l^\downarrow) + \sum_{m\neq l}(U_0 - 2J_{lm})(n_m^\uparrow + n_m^\downarrow). \tag{5.14}$$

5.2 Exchange term

The exact exchange energy has the general form

$$E^X[\rho] = -\frac{1}{2} \sum_\sigma \int d\mathbf{r} d\mathbf{r}' \, \rho_\sigma(\mathbf{r}, \mathbf{r}') V_C(\mathbf{r} - \mathbf{r}') \rho_\sigma(\mathbf{r}', \mathbf{r}) \tag{5.15}$$

$$= -\frac{1}{2} \sum_\sigma \int d\mathbf{r} d\mathbf{r}' \, \frac{\rho_\sigma(\mathbf{r}, \mathbf{r}') \rho_\sigma(\mathbf{r}', \mathbf{r})}{|\mathbf{r} - \mathbf{r}'|}. \tag{5.16}$$

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

$$E^X[\rho] = -\frac{1}{2} \sum_\sigma \int d\mathbf{r} d\mathbf{r}' \, \rho_\sigma(\mathbf{r}, \mathbf{r}') V_{DMFT}(\mathbf{r} - \mathbf{r}') \rho_\sigma(\mathbf{r}', \mathbf{r}) \tag{5.17}$$

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

$$\times \langle \mathbf{r}'|\chi_m^\sigma\rangle \langle \chi_m^\sigma|\rho_\sigma|\chi_n^\sigma\rangle \langle \chi_n^\sigma|\mathbf{r}\rangle \tag{5.18}$$

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

$$\times \langle \chi_m^\sigma|\rho_\sigma|\chi_n^\sigma\rangle \langle \chi_k^\sigma|\rho_\sigma|\chi_l^\sigma\rangle. \tag{5.19}$$

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

$$\langle nl|U|km\rangle = \int d\mathbf{r} d\mathbf{r}' \, (\chi_n^\sigma)^*(\mathbf{r})(\chi_l^\sigma)^*(\mathbf{r}') V_{DMFT}(\mathbf{r} - \mathbf{r}') \chi_m^\sigma(\mathbf{r}') \chi_k^\sigma(\mathbf{r}), \tag{5.20}$$

and the DMFT density matrix

$$\langle \chi_m^\sigma|\rho_\sigma|\chi_n^\sigma\rangle = n_{mn}^\sigma. \tag{5.21}$$

With this, the exchange energy takes on the form

$$E_{DMFT}^X = -\frac{1}{2} \sum_{\substack{klmn \\ \sigma}} \langle nl|U|km\rangle \, n_{mn}^\sigma n_{kl}^\sigma. \tag{5.22}$$

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

$$E_{DMFT}^X = -\frac{1}{2} \sum_{mk,\sigma} \langle mk|U|km\rangle \, n_m^\sigma n_k^\sigma. \tag{5.23}$$

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

$$\Sigma_{l\sigma}^{X,DMFT} = \frac{\partial}{\partial n_l^\sigma} E_{DMFT}^X \tag{5.24}$$

$$= -\frac{1}{2}\sum_k \langle lk|U|kl\rangle\, n_k^\sigma - \frac{1}{2}\sum_m \langle ml|U|lm\rangle\, n_m^\sigma \tag{5.25}$$

$$= -\sum_k \langle lk|U|kl\rangle\, n_k^\sigma \tag{5.26}$$

$$= -U_0\, n_l^\sigma - \sum_{k\neq l} J_{lk}\, n_k^\sigma. \tag{5.27}$$

5.3 Hartree + exchange Selfenergy

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

$$\Sigma_{l\sigma}^{H,DMFT} + \Sigma_{l\sigma}^{X,DMFT} = U_0 n_l^{\bar\sigma} + \sum_{m\neq l}(U_0 - 2J_{lm})n_m^{\bar\sigma}$$

$$+ \sum_{m\neq l}(U_0 - 3J_{lm})n_m^\sigma \tag{5.28}$$

$$= \lim_{\omega_n\to\infty} \Sigma^{DMFT}(i\omega_n), \tag{5.29}$$

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.

5.4 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)

$$\Sigma_{l\sigma}^{H,DMFT} = V(n_l^\uparrow + n_l^\downarrow) + \sum_{m\neq l}(V - 2J_{lm})(n_m^\uparrow + n_m^\downarrow) \tag{5.30}$$

$$\Sigma_{l\sigma}^{X,DMFT} = -V\, n_l^\sigma - \sum_{k\neq l} J_{lk}\, n_k^\sigma. \tag{5.31}$$

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 $V + \frac{8}{7}J_{avg}$.

6 Implementation details

6.1 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.