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# 1 GW (oneshot) + DMFT documentation

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  $\Sigma^{GW}$  for all states. In addition, the local part of  $\Sigma^{GW}$  of a subset of strongly correlated orbitals will be replaced by a Selfenergy  $\Sigma^{DMFT}$  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  $\Sigma^{GW}$  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  $\Sigma^{DMFT}$ . This  $\Sigma^{DMFT}$  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.

## 1.1 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

$$\Sigma_{\nu\nu'}(k, \omega) = [G_0 W_0]_{\nu\nu'}(k, \omega). \quad (1.1.1)$$

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

$$G_{\nu\nu'}^{GW}(k, \omega) = [\mathbb{1}(\omega + \mu + i\delta) - H^{DFT}(k) + v^{XC}(k) - \Sigma^{GW}(k, \omega)]_{\nu\nu'}^{-1}. \quad (1.1.2)$$

### 1.1.1 Output

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
- $\mu$ : 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  $\omega$ .

- $\beta$ : 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!!*

## 1.2 The DMFT part

Within DMFT we then calculate a local correction  $\Sigma^{DMFT}$  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).

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

$$\Sigma_f^{GW,loc}(i\omega_n) = \frac{1}{N_k} \sum_k \Sigma_{ff}^{GW}(k, i\omega_n). \quad (1.2.1)$$

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

$$\Sigma_{ff}^{imp}(i\omega_n) = \Sigma_f^{GW,loc}(i\omega_n). \quad (1.2.2)$$

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

$$G_{mm'}(k, i\omega_n) = [\mathbb{1}(i\omega_n + \mu) - H^{DFT}(k) + v^{XC}(k) \quad (1.2.3)$$

$$- \Sigma^{GW}(k, i\omega_n) + \Sigma_f^{GW,loc}(i\omega_n) - \Sigma^{imp}(i\omega_n) + \Sigma_f^{H,imp}]_{mm'}^{-1}. \quad (1.2.4)$$

Adjust the chemical potential  $\mu$  in a way such that the desired filling

$$N_e = \lim_{\tau \rightarrow 0^-} \frac{1}{\beta N_k} \sum_{i\omega_n} \sum_{k,m} G_{mm}(k, i\omega_n) e^{-i\omega_n \tau}. \quad (1.2.5)$$

is obtained.

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

$$G_{mm'}^{loc}(i\omega_n) = \frac{1}{N_k} \sum_k G_{mm'}(k, i\omega_n), \quad (1.2.6)$$

$$\Rightarrow \mathcal{G}_{ff}^{-1}(i\omega_n) = [G^{loc}]_{ff}^{-1}(i\omega_n) + \Sigma_{ff}^{imp}(i\omega_n). \quad (1.2.7)$$

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

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

5. Calculate the Hybridization function

$$\Delta_{ff}(i\omega_n) = i\omega_n + \tilde{\mu}_f - \mathcal{G}_{ff}^{-1}(i\omega_n), \quad (1.2.8)$$

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

$$\tilde{\mu}_f = \lim_{\omega_n \rightarrow \infty} \text{Re} [\mathcal{G}_{ff}^{-1}(i\omega_n)] \quad (1.2.9)$$

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

$$\Delta_{ff}(\tau) = \frac{1}{\beta} \sum_{i\omega_n} \Delta_{ff}(i\omega_n) e^{-i\omega_n \tau} \quad (1.2.10)$$

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. (2.1.14) and (2.4.1). Then go back to step 3. Repeat until convergence is reached.

### 1.2.1 Output

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

## 2 Hartree- and Exchange term in DMFT

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

## 2.1 Hartree term

The Hartree energy has the general form

$$E^H[\rho] = \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' \rho(\mathbf{r}) V_C(\mathbf{r} - \mathbf{r}') \rho(\mathbf{r}') \quad (2.1.1)$$

$$= \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' \frac{\rho(\mathbf{r}) \rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}, \quad (2.1.2)$$

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

$$\rho(\mathbf{r}) = \rho_\uparrow(\mathbf{r}) + \rho_\downarrow(\mathbf{r}). \quad (2.1.3)$$

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

$$E^H[\rho] = \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' \rho(\mathbf{r}) V_C(\mathbf{r} - \mathbf{r}') \rho(\mathbf{r}') \quad (2.1.4)$$

$$= \frac{1}{2} \sum_{\substack{klmn \\ \sigma\sigma'}} \int d\mathbf{r} d\mathbf{r}' \langle \mathbf{r} | \chi_k^\sigma \rangle \langle \chi_k^\sigma | \rho | \chi_l^\sigma \rangle \langle \chi_l^\sigma | \mathbf{r}' \rangle V_{DMFT}(\mathbf{r} - \mathbf{r}') \\ \times \langle \mathbf{r}' | \chi_m^{\sigma'} \rangle \langle \chi_m^{\sigma'} | \rho | \chi_n^{\sigma'} \rangle \langle \chi_n^{\sigma'} | \mathbf{r}' \rangle \quad (2.1.5)$$

$$= \frac{1}{2} \sum_{\substack{klmn \\ \sigma\sigma'}} \int d\mathbf{r} d\mathbf{r}' (\chi_l^\sigma)^*(\mathbf{r}) (\chi_n^{\sigma'})^*(\mathbf{r}') V_{DMFT}(\mathbf{r} - \mathbf{r}') \chi_m^{\sigma'}(\mathbf{r}') \chi_k^\sigma(\mathbf{r}) \\ \times \langle \chi_k^\sigma | \rho | \chi_l^\sigma \rangle \langle \chi_m^{\sigma'} | \rho | \chi_n^{\sigma'} \rangle. \quad (2.1.6)$$

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

$$\langle ln|U|km \rangle = \int d\mathbf{r} d\mathbf{r}' (\chi_l^\sigma)^*(\mathbf{r}) (\chi_n^{\sigma'})^*(\mathbf{r}') V_{DMFT}(\mathbf{r} - \mathbf{r}') \chi_m^{\sigma'}(\mathbf{r}') \chi_k^\sigma(\mathbf{r}), \quad (2.1.7)$$

and the DMFT density matrix

$$\langle \chi_k^\sigma | \rho | \chi_l^\sigma \rangle = n_{kl}^\sigma. \quad (2.1.8)$$

With this, the Hartree energy takes on the form

$$E^{DMFT} = \frac{1}{2} \sum_{\substack{klmn \\ \sigma\sigma'}} \langle ln|U|km \rangle n_{kl}^\sigma n_{mn}^{\sigma'}. \quad (2.1.9)$$

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

$$E_{DMFT}^H = \frac{1}{2} \sum_{\substack{km \\ \sigma\sigma'}} \langle km|U|km \rangle n_k^\sigma n_m^{\sigma'}. \quad (2.1.10)$$

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

$$\Sigma_{l\sigma}^{H,DMFT} = \frac{\partial}{\partial n_l^\sigma} E_{DMFT}^H \quad (2.1.11)$$

$$= \frac{1}{2} \sum_{m,\sigma'} \langle lm|U|lm\rangle n_m^{\sigma'} + \frac{1}{2} \sum_{k,\sigma'} \langle kl|U|kl\rangle n_k^{\sigma'} \quad (2.1.12)$$

$$= \sum_{m,\sigma'} \langle lm|U|lm\rangle n_m^{\sigma'} \quad (2.1.13)$$

$$= U_0(n_l^\uparrow + n_l^\downarrow) + \sum_{m \neq l} (U_0 - 2J_{lm})(n_m^\uparrow + n_m^\downarrow). \quad (2.1.14)$$

## 2.2 Exchange term

The exact exchange energy has the general form

$$E^X[\rho] = -\frac{1}{2} \sum_{\sigma} \int d\mathbf{r} d\mathbf{r}' \rho_{\sigma}(\mathbf{r}, \mathbf{r}') V_C(\mathbf{r} - \mathbf{r}') \rho_{\sigma}(\mathbf{r}', \mathbf{r}) \quad (2.2.1)$$

$$= -\frac{1}{2} \sum_{\sigma} \int d\mathbf{r} d\mathbf{r}' \frac{\rho_{\sigma}(\mathbf{r}, \mathbf{r}') \rho_{\sigma}(\mathbf{r}', \mathbf{r})}{|\mathbf{r} - \mathbf{r}'|}. \quad (2.2.2)$$

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

$$E^X[\rho] = -\frac{1}{2} \sum_{\sigma} \int d\mathbf{r} d\mathbf{r}' \rho_{\sigma}(\mathbf{r}, \mathbf{r}') V_{DMFT}(\mathbf{r} - \mathbf{r}') \rho_{\sigma}(\mathbf{r}', \mathbf{r}) \quad (2.2.3)$$

$$= -\frac{1}{2} \sum_{\substack{klmn \\ \sigma}} \int d\mathbf{r} d\mathbf{r}' \langle \mathbf{r} | \chi_k^\sigma \rangle \langle \chi_k^\sigma | \rho_{\sigma} | \chi_l^\sigma \rangle \langle \chi_l^\sigma | \mathbf{r}' \rangle V_{DMFT}(\mathbf{r} - \mathbf{r}') \\ \times \langle \mathbf{r}' | \chi_m^\sigma \rangle \langle \chi_m^\sigma | \rho_{\sigma} | \chi_n^\sigma \rangle \langle \chi_n^\sigma | \mathbf{r} \rangle \quad (2.2.4)$$

$$= -\frac{1}{2} \sum_{\substack{klmn \\ \sigma}} \int d\mathbf{r} d\mathbf{r}' (\chi_n^\sigma)^*(\mathbf{r}) (\chi_l^\sigma)^*(\mathbf{r}') V_{DMFT}(\mathbf{r} - \mathbf{r}') \chi_m^\sigma(\mathbf{r}') \chi_k^\sigma(\mathbf{r}) \\ \times \langle \chi_m^\sigma | \rho_{\sigma} | \chi_n^\sigma \rangle \langle \chi_k^\sigma | \rho_{\sigma} | \chi_l^\sigma \rangle. \quad (2.2.5)$$

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

$$\langle nl|U|km\rangle = \int d\mathbf{r} d\mathbf{r}' (\chi_n^\sigma)^*(\mathbf{r}) (\chi_l^\sigma)^*(\mathbf{r}') V_{DMFT}(\mathbf{r} - \mathbf{r}') \chi_m^\sigma(\mathbf{r}') \chi_k^\sigma(\mathbf{r}), \quad (2.2.6)$$

and the DMFT density matrix

$$\langle \chi_m^\sigma | \rho_{\sigma} | \chi_n^\sigma \rangle = n_{mn}^\sigma. \quad (2.2.7)$$

With this, the exchange energy takes on the form

$$E_{DMFT}^X = -\frac{1}{2} \sum_{\substack{klmn \\ \sigma}} \langle nl|U|km\rangle n_{mn}^\sigma n_{kl}^\sigma. \quad (2.2.8)$$

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

$$E_{DMFT}^X = -\frac{1}{2} \sum_{mk,\sigma} \langle mk|U|km\rangle n_m^\sigma n_k^\sigma. \quad (2.2.9)$$

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

$$\Sigma_{l\sigma}^{X,DMFT} = \frac{\partial}{\partial n_l^\sigma} E_{DMFT}^X \quad (2.2.10)$$

$$= -\frac{1}{2} \sum_k \langle lk|U|kl\rangle n_k^\sigma - \frac{1}{2} \sum_m \langle ml|U|lm\rangle n_m^\sigma \quad (2.2.11)$$

$$= -\sum_k \langle lk|U|kl\rangle n_k^\sigma \quad (2.2.12)$$

$$= -U_0 n_l^\sigma - \sum_{k \neq l} J_{lk} n_k^\sigma. \quad (2.2.13)$$

## 2.3 Hartree + exchange Selfenergy

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

$$\begin{aligned} \Sigma_{l\sigma}^{H,DMFT} + \Sigma_{l\sigma}^{X,DMFT} &= U_0 n_l^\sigma + \sum_{m \neq l} (U_0 - 2J_{lm}) n_m^\sigma \\ &\quad + \sum_{m \neq l} (U_0 - 3J_{lm}) n_m^\sigma \end{aligned} \quad (2.3.1)$$

$$= \lim_{\omega_n \rightarrow \infty} \Sigma^{DMFT}(i\omega_n), \quad (2.3.2)$$

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.

## 2.4 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)

$$\Sigma_{l\sigma}^{H,DMFT} = V(n_l^\uparrow + n_l^\downarrow) + \sum_{m \neq l} (V - 2J_{lm})(n_m^\uparrow + n_m^\downarrow) \quad (2.4.1)$$

$$\Sigma_{l\sigma}^{X,DMFT} = -V n_l^\sigma - \sum_{k \neq l} J_{lk} n_k^\sigma. \quad (2.4.2)$$

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