Self-consistent implementation of DMFT in SIESTA with DMFTwDFT - User Manual

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This document is a manual that summarizes the method to implement DFT+DMFT full charge self-consistency using the DMFTwDFT code to the DFT program Siesta [1]. For a full derivation please refer to Dr. Park's article in Ref. [2]. This tutorial is based on his article titled, "*DMFTwDFT Tutorial*" written in May, 2019 and Dr. Junquera's article "*Notes on the self-consistent implementation of Dynamical Mean Dynamical Mean Field Theory in siesta*" written in January, 2020.

This tutorial will follow a DFT+DMFT calculation performed for the correlated material $SrVO_3$ using 512 k-points, 28 bands and 14 Wannier bands for the V-d orbitals and O-p orbitals. The file structure of the test directory is displayed below.

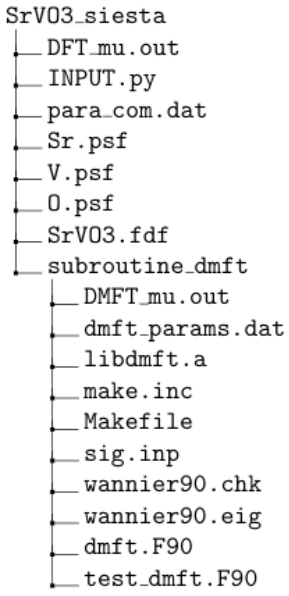

Figure 1: File tree

The root directory contains the files necessary to run an initial DMFT calculation with Siesta and DMFTwDFT. **DFT_mu.out** is an initial guess for the chemical potential which can be set to the Fermi energy of the system. **INPUT.py** contains parameters that govern the DFT+DMFT calculation. **SrVO3.fdf** is the input file for the Siesta DFT calculation. It should contain the following block to allow the Wannier90 calculation.

```
Siesta2Wannier90.WriteMmn       .true.
Siesta2Wannier90.WriteAmn       .true.
Siesta2Wannier90.WriteEig       .true.
Siesta2Wannier90.WriteUnk       .false.
Siesta2Wannier90.NumberOfBands 28
```

A siesta+DMFT calculation can be launched easily through the DMFTwDFT program as follows:

```
$DMFT.py -dft siesta -structurename SrVO3 -dmft
```

DMFTwDFT automatically generates a **SrVO3.win** file for the Wannier90 calculation based on these inputs. The subdirectory **subroutine_dmft** contains some of the outputs from a siesta+DMFT calculation with a single DFT step and multiple DMFT steps. To develop a siesta+DMFTwDFT interface for full DFT+DMFT charge self-consistency, the DFT charge density must be updated from the DMFT charge density. The formalism to achieve this is explained briefly below.

The total DFT+DMFT charge density $\rho(r)$ can be given by

$$\rho(\mathbf{r}) = \sum_{i \notin W} \rho_i^{DFT}(\mathbf{r}) + \sum_{i,j \in W} \rho_{ij}^{DMFT}(\mathbf{r}) \tag{1}$$

where $\rho(r)^{DMFT}$ is the charge density within the DMFT subspace, W is the energy window for constructing Wannier functions, and i,j are the band indices. The DMFT charge density $\rho(r)^{DMFT}$ for the correlated subspace can represented using DFT KS wavefunctions as follows:

$$\rho_{ij}^{\mathrm{DMFT}}(\vec{r}) = \frac{1}{N_{\vec{k}}} \sum_{\vec{k}} n_{\vec{k}ij} \left\langle \psi_{i\vec{k}}^{\mathrm{KS}} | \vec{r} \right\rangle \left\langle \vec{r} | \psi_{j\vec{k}}^{\mathrm{KS}} \right\rangle \tag{2}$$

where $n_{\vec{k}ij}$ is the DMFT occupancy matrix in the KS basis (calculated in **dmft.F90** as the 3-d array, **n**) and $|\psi_{\vec{k}i}^{KS}\rangle$ is the KS wavefunction at momentum $\vec{k}$ and band i.

Following Eq. (43) of Ref. [1], at every sampled $\vec{k}$-point the eigenfunctions of the hamiltonian are written as

$$\psi_{i\vec{k}}^{\mathrm{KS}}(\vec{r}) = \left\langle \vec{r} | \psi_{i\vec{k}}^{\mathrm{KS}} \right\rangle = \sum_{\mu'} e^{i\vec{k}\cdot\vec{R}_{\mu'}} \phi_{\mu'}(\vec{r}) c_{\mu' i}(\vec{k}) \tag{3}$$

where the sum in μ' extends to all basis orbitals in space (each of them centered in an atom at position $\vec{R}_{\mu'}$), i labels the different bands, and $c_{\mu' i} \equiv c_{\mu i}$ if $\mu' \equiv \mu$.

Inserting Eq. 3 into Eq. 2,

$$\begin{aligned}
\rho_{ij}^{\mathrm{DMFT}}(\vec{r}) &= \frac{1}{N_{\vec{k}}} \sum_{\vec{k}} n_{\vec{k}ij} \left(\sum_{\nu'} e^{-i\vec{k}\cdot\vec{R}_{\nu'}} \phi_{\nu'}^*(\vec{r}) c_{i\nu'}(\vec{k}) \right) \left(\sum_{\mu'} e^{i\vec{k}\cdot\vec{R}_{\mu'}} \phi_{\mu'}(\vec{r}) c_{\mu' j}(\vec{k}) \right) \\
&= \sum_{\nu'} \sum_{\mu'} \phi_{\nu'}^*(\vec{r}) \phi_{\mu'}(\vec{r}) \left(\frac{1}{N_{\vec{k}}} \sum_{\vec{k}} n_{\vec{k}ij} e^{i\vec{k}\cdot(\vec{R}_{\mu'} - \vec{R}_{\nu'})} c_{i\nu'}(\vec{k}) c_{\mu' j}(\vec{k}) \right) \\
&= \sum_{\nu'} \sum_{\mu'} \phi_{\nu'}^*(\vec{r}) \phi_{\mu'}(\vec{r}) \rho_{\nu'\mu' ij},
\end{aligned}$$

where we have defined the "density matrix" $\rho_{\nu'\mu' ij}$ as

$$\rho_{\nu'\mu' ij} = \frac{1}{N_{\vec{k}}} \sum_{\vec{k}} n_{\vec{k}ij} e^{i\vec{k}\cdot(\vec{R}_{\mu'} - \vec{R}_{\nu'})} c_{i\nu'}(\vec{k}) c_{\mu' j}(\vec{k}), \tag{4}$$

and $c_{i\nu'} \equiv c^*_{\nu' i}$.

The sum over band indices i,j in Eq. 2 can be simplified to the sum over one index because the DMFT occupancy matrix $n_{\vec{k}ij}$ is Hermitian and so can be written in terms of eigenvalues $w_{\vec{k}\lambda}$ and eigenfunctions ϕ_λ as

$$n_{\vec{k}ij} = \sum_\lambda U^{DMFT}_{\vec{k}i\lambda} \cdot w_{\vec{k}\lambda} \cdot U^{DMFT*}_{\vec{k}j\lambda} \tag{5}$$

where, $U^{DMFT}_{\vec{k}i\lambda}$ are unitary matrices whose rows are ϕ_λ's. $n_{\vec{k}ij}$ is calculated in **dmft.F90**. These matrices need to be updated at every self-consistent step.

The following section discusses the two Fortran files and the library used to perform the calculations relating to the above formulation.

1. test_dmft.F90 -

   This mimics a subroutine in Siesta which calculates the DFT charge density. Inputs from Siesta namely kgrid, k-mesh weights, number of wannier bands, number of kpoints and charge density grid dimensions are passed into the subroutine dmft() from here. For now we have included sample values which should be replaced by corresponding values obtained from Siesta. Once the DMFT charge density is passed back here it can be added to the DFT charge density as portrayed in Eq. 1.

2. dmft.F90 -

   This subroutine is meant to calculate the DMFT charge density using Eq. 2 and Eq. 5. So far we have computed the DMFT occupancy matrix, $n_{\vec{k}ij}$ in here. What we have to achieve now is to calculate the DMFT charge density $\rho(r)^{DMFT}$ using this occupancy matrix $n_{\vec{k}ij}$ and the Siesta Kohn-Sham wave functions as depicted in equations 2 - 5. Using the above we can then obtain the full DFT+DMFT charge density which is used as the starting charge density for the DFT calculation. This subroutine calls **Compute_DMFT()** from the dmft library **(libdmft.a)** to compute the DMFT weight $w_{\vec{k}\lambda}$ and Unitary matrix $U^{DMFT}_{\vec{k}i\lambda}$ at each k-point. The variables of the dmft() subroutine is as follows:

```
INPUT:
------
integer n_kpts              : The number of k-points in DFT
integer n_wann              : The number of Wannier orbitals
real kpt_dft(3, n_kpts)     : List of k-points in DFT using fractional coordinates
real wght_dft(n_kpts)       : List of k-point weights. The sum should be one.
integer Nx                  : Number of grid points in x direction for charge density grid
integer Ny                  : Number of grid points in y direction for charge density grid
integer Nz                  : Number of grid points in z direction for charge density grid

INTERMEDIATE:
--------------
These variables are calculated from Compute_DMFT() using the above inputs
and are then used to calculate the DMFT charge density:
integer band_win(2, n_kpts)               : Band range of the Wannier subspace at
                                            each k-point
real DMFT_eval(n_wann, n_kpts)            : Eigenvalues W_{k,lambda} of the DMFT
                                            occupancy matrix n_{k,i,j}
complex DMFT_evec(n_wann, n_wann, n_kpts) : DMFT eigen vectors of the Unitary matrix
                                        U_{k,i,lambda}
complex DMFT_evec_k(n_wann,n_wann)        : Unitary matrix U_{k,i,lambda} at a given
                                            k-point
complex DMFT_evec_T(n_wann,n_wann)        : Transpose Conjugate of the Unitary matrix
                                            U_{k,i,lambda} at a given k-point
```

```
complex n(n_wann,n_wann,n_kpts)          : DMFT Occupancy Matrix n_{k,i,j}

OUTPUT:
-------
real DMFT_chg(Nx, Ny, Nz)                : DMFT charge density rho_{i,j}, where i,j is
                                             in the Wannier window
```

3. libdmft.a -

   This contains the subroutine **Compute_DMFT()** which is called from dmft.F90 and calculates the DMFT Unitary matrix that is used in dmft.F90 to calculate the Occupancy matrix needed for the DMFT charge density calculation.

To compile these files, simply type **make** in the directory. To run,

```
mpirun -np nprocs ./test_dmft.x
```

FAQ

1. Where should I find $n_{\vec{k}ij}$?

A: Inside **dmft.F90**. It is called **n**.

2. Is it already implemented? If so, where and how is it stored?

   A: Yes, it is stored as a complex (n_wann, n_wann, n_kpts) dimensional array as mentioned in the previous question.

3. In the computation of the standard density matrix, I guess we need to remove the contributions from the bands in the correlated manifold. Am I right?

   A: Correct. As portrayed in Eq. 1's first quantity ρ^{DFT} is for theb ands that are not in the correlated subspace.

4. In your notes, you have Eq. (5) and Eq. (6), where$\langle\vec{r}|\psi^{\mathrm{DMFT}}_{\vec{k}\lambda}\rangle$ is defined. Should I use the KS eigenvectors or those DMFT eigenvectors?

   A: In Dr. Park's manual, equations (4) and (5) describe another method to calculate the DMFT charge density. The formulas supplied in this tutorial are sufficient to calculate this quantity.

References

[1] José M Soler, Emilio Artacho, Julian D Gale, Alberto García, Javier Junquera, Pablo Ordejón, and Daniel Sánchez-Portal. The SIESTA method forab initioorder-nmaterials simulation. *Journal of Physics: Condensed Matter*, 14(11):2745–2779, mar 2002.

[2] Hyowon Park, Andrew J. Millis, and Chris A. Marianetti. Computing total energies in complex materials using charge self-consistent dft + dmft. *Phys. Rev. B*, 90:235103, Dec 2014.