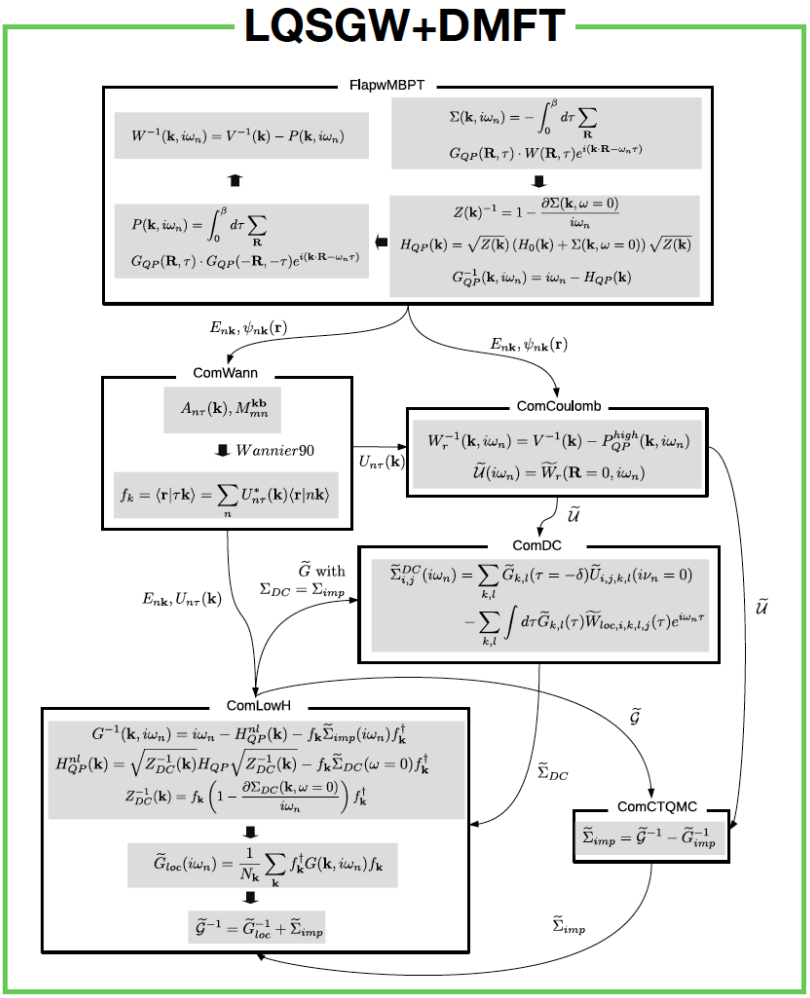
Example: MnO

LQSGW+DMFT in COMSUITE

We will calculate the electronic structure of a charge transfer insulator MnO within LQSGW+DMFT. The COMSUITE package for LQSGW+DMFT is composed of six components (software packages) and its work flow is described below.



1. Construction of a quasi-particle Hamiltonian within *ab initio* LQSGW by **FlapwMBPT** (see https://doi.org/10.1016/j.cpc.2017.06.012, https://www.bnl.gov/cmpmsd/flapwmbpt).
2. Construction of the atom-centered local basis set spanning the low energy Hilbert space by **ComWann utilizing the Wannier90 package.**
3. Calculation of the bosonic Weiss field within the constrained Random Phase approximation(cRPA) and evaluation of the Slater's integrals associated with the impurity orbitals by **ComCoulomb.**
4. Calculation of the double-counted self-energy associated with the impurity orbitals within the local GW approximation by **ComDC.**
5. Wannier interpolation of the mean-field Hamiltonian and solving the DMFT self-consistent equation by **ComLowH** and **ComCTQMC.**

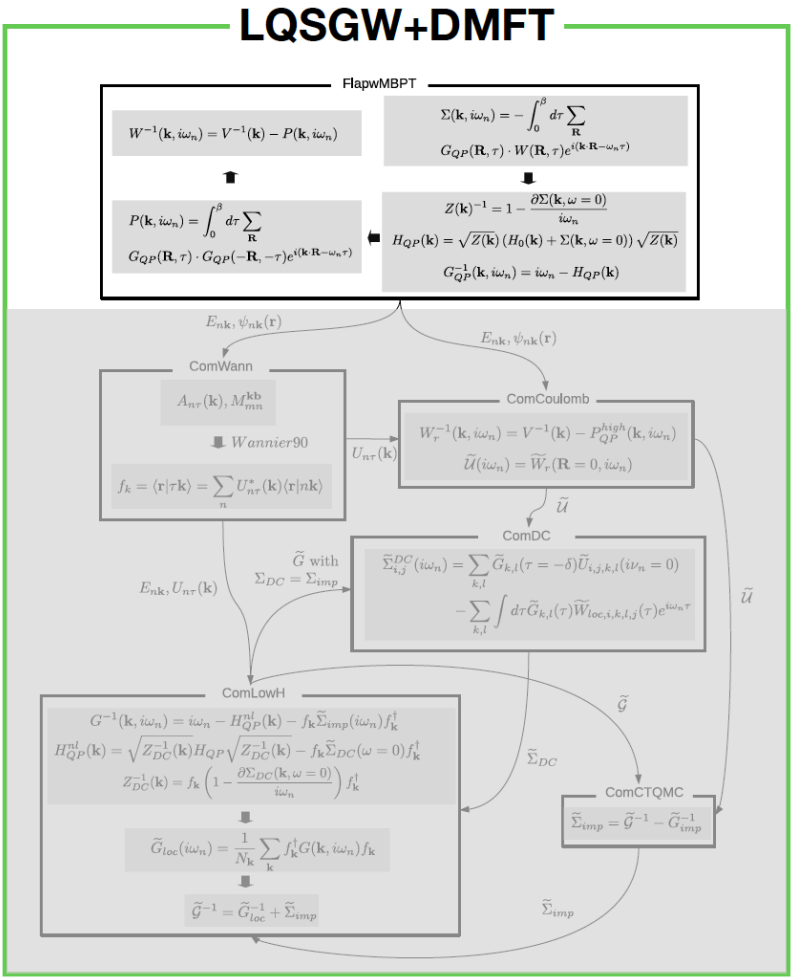
These components along with Wannier90 library are located at install_directory ('install_directory' is described in 'Build and Install' section in Installation of COMSUITE page). To access COMSUITE executables, you should export the above bin path in your startup shell script, i.e.,

```
export COMSUITE_BIN=install_directory/bin
```

This tutorial consists of mainly three parts: i) LQSGW prerun, ii) LQSGW+DMFT run, and finally iii) Analysis.

MnO LQSGW prerun

To run LQSGW+DMFT, the first step is the LQSGW prerun (this prerun is described in the unshaded part of the following figure):



To proceed with the LQSGW calculation, please create a directory named “lqsgw” in your work directory. It is possible to give this directory another name. However it is important that this name be specified in ‘comdmft.ini’ (as will be explained in the next section). After doing so, cd to this directory

```
$ mkdir lqsgw
$ cd lqsgw
```

Now create an input file “ini” using MnO cif file by executing cif2matdelab.py:

```
$ $COMSUITE_BIN/cif2matdelab.py –m qp –k 2.5 MnO.cif
```

The generated ini file of MnO looks as follows:

```
TEXT band structure calculation
CONTROL   iter_dft= 150  iter_hf=  0  iter_gw=  0 iter_qp= 20
          admix=0.100  adspin=0.600 adm_gw=0.100 acc_it_gw=0.15
          iexch=005 scal_spin= 1.0000
          nproc_tau=  24 nproc_k=   16
          irel=1 clight=274.074e+00 rel_interst=F irel_core=1
          temperature=   1000.00  restart=F
FILES
  allfile=mdl
SYM symgen=I_R4[0.0,1.0,0.0]_R2[1.0,0.0,0.0]_R3[1.0,-1.0,1.0]_
STRUCTURE  par=  1.0000000  natom=  2 nsort=  2 istruct= -3
      is=  1  2
      b/a= 1.000000  c/a= 1.000000
      a=   0.0000000000000000  -4.2005777179643129  -4.2005777179643129
      b=   4.2005777179643129   0.0000000000000000  -4.2005777179643129
      c=   4.2005777179643129  -4.2005777179643129   0.0000000000000000
    tau=   0.0000000000000000   0.0000000000000000   0.0000000000000000
    tau=   0.5000000000000000   0.5000000000000000   0.5000000000000000
REAL SPACE MESHES mdiv=  14   14   14
                  nrdiv=  10   10   10
BASIS  cut_lapw_ratio=0.610 cut_pb_ratio=0.980
       eps_pb=1.e-03
ZONES nbndf=   0
DOS   emindos=-15.000  emaxdos= 15.000   ndos=  800
      n_cont_frac=  30 e_small=2.e-02
      dos= T           bandstructure= T
K_POINT  ndiv=   6   6   6  metal=T n_k_div=  9 k_line=010
MULTI_SCF vv0=  1.00
MAGNET  b_extval=   0.000000 iter_h_ext=0000100
        b_ext=  0.000  0.000  1.000
TAU MESH n_tau=   46 n_tau_int=  900
OMEGA MESH n_omega_exa=   29 n_omega_asy=   18 omega_max=  200.00
           interp_omega_d= 2
NU MESH n_nu_exa=   29 n_nu_asy=   18 nu_max=  200.00
        interp_nu_d= 2
ATOMIC DATA ------------------------------------------------------------
  txtel=mn    z= 25.0 magn_shift= 0.000
  smt= 2.32800 h= 0.0120 nrad= 1216 z_dop=0.000
  lmb= 6  lmpb= 6
  lim_pb_mt= 30 30 30 30 30 30 30
  ntle=  4  3  3  2  2  2  2
  l  augm  atocc   ptnl  corr idmd
  0   LOC  2.000  3.800     N    0
  0   APW  2.000  4.800     N    0
  0   LOC  0.000  5.800     N    1
  0   LOC  0.000  6.800     N    1
  1   LOC  6.000  3.800     N    0
  1   APW  0.000  4.800     N    0
  1   LOC  0.000  5.800     N    1
  2   APW  5.000  3.800     N    0
  2   LOC  0.000  4.800     N    1
  2   LOC  0.000  5.800     N    1
  3   APW  0.000  4.800     N    0
  3   LOC  0.000  5.800     N    1
  4   APW  0.000  5.800     N    0
  4   LOC  0.000  6.800     N    1
  5   APW  0.000  6.800     N    0
  5   LOC  0.000  7.800     N    1
  6   APW  0.000  7.800     N    0
  6   LOC  0.000  8.800     N    1
  txtel=o     z=  8.0 magn_shift= 0.000
  smt= 1.74600 h= 0.0120 nrad= 1216 z_dop=0.000
  lmb= 6  lmpb= 6
  lim_pb_mt= 30 30 30 30 30 30 30
  ntle=  3  3  2  2  2  2  2
  l  augm  atocc   ptnl  corr idmd
  0   APW  2.000  2.800     N    0
  0   LOC  0.000  3.800     N    1
  0   LOC  0.000  4.800     N    1
  1   APW  4.000  2.800     N    0
  1   LOC  0.000  3.800     N    1
  1   LOC  0.000  4.800     N    1
  2   APW  0.000  3.800     N    0
  2   LOC  0.000  4.800     N    1
  3   APW  0.000  4.800     N    0
  3   LOC  0.000  5.800     N    1
  4   APW  0.000  5.800     N    0
  4   LOC  0.000  6.800     N    1
  5   APW  0.000  6.800     N    0
  5   LOC  0.000  7.800     N    1
  6   APW  0.000  7.800     N    0
  6   LOC  0.000  8.800     N    1
```

Now modify several of the input keywords as follows:

- Iter_dft: The number of DFT iterations. Set to 150.
- iter_qp: The number of LQSGW iterations. Set to 20.
- nproc_tau: The number of MPI processes associated with imaginary time and frequency. Set to 24. For the details on the MPI parallization in FlapwMBPT, please goto FlapwMBPT homepage (https://www.bnl.gov/cmpmsd/flapwmbpt).
- nproc_k: The number of MPI processes associated with parallelization over the Brillouin zone k-points. For the present case, set to 16. For the details on the MPI parallization in FlapwMBPT, please go to FlapwMBPT homepage (https://www.bnl.gov/cmpmsd/flapwmbpt)
- ndiv: The number of k-mesh points in Brillouin zone. $6 \times 6 \times 6$ k-mesh for the present case.
- We also added more manganese and oxygen muffin-tin basis set for a better convergence. For the muffin-tin basis set format, please go to FlapwMBPT homepage (https://www.bnl.gov/cmpmsd/flapwmbpt). Note also that the total number of MPI processes is nproc_tau $\times$ nproc_k = 384 for the present case.

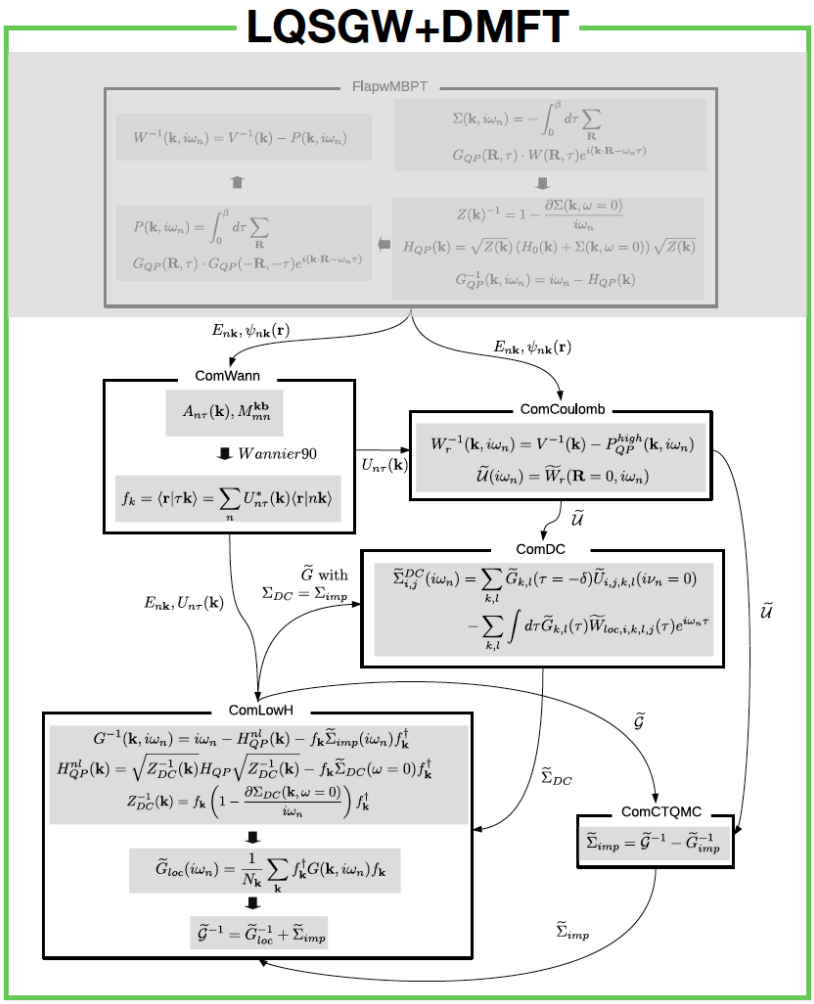
The next step is to run the LQSGW calculation by executing rspflapw.exe. An example of a job script to run rspflapw.exe using SLURM is

```
#!/bin/bash -l
#SBATCH -J temp
#SBATCH -p regular
#SBATCH -N 32
#SBATCH -e temp.%j.err
#SBATCH -o temp.%j.out
#SBATCH -L SCRATCH
#SBATCH -C haswell
#SBATCH -t 05:00:00
srun -n 384 $COMSUITE_BIN/rspflapw.exe
```

The total number of processes is 384 as specified in ini file. Note that rspflapw will redistribute the number of processes for double parallelization if the number of MPI process requested by the job submission script doesn't match the number of process you declared by using nproc_k and nproc_tau in the "ini" .

MnO LQSGW+DMFT run

Once the prerun is finished successfully, the next step is the LQSGW+DMFT calculation (see the unshaded part of following figure). To run LQSGW+DMFT, an input file named "comdmft.ini" is needed (see input file section below). The calculation of MnO within LQSFW+DMFT reads output data from the LQSGW prerun. If you specify the prerun path in comdmft.ini (e.g., '../lqsgw' in this case) correctly, it will read the necessary data automatically.



To run LQSGW+DMFT, move back to the work directory, create a lqsgw_dmft directory (you can name this what you want) and then cd to it:

```
$ cd ..
$ mkdir lqsgw_dmft
```

```
$ cd lqsgw_dmft
```

We now create comdmft.ini (see input file section below) file for the LQSGW+DMFT calculation. Having done so, execute ‘comdmft.py’ python file in $COMSUITE_BIN. An example of a job script using SLURM that does so is

```
#!/bin/bash -l
#SBATCH -J temp
#SBATCH -q regular
#SBATCH -N 12
#SBATCH -e temp.%j.err
#SBATCH -o temp.%j.out
#SBATCH -L SCRATCH
#SBATCH -C haswell
#SBATCH -t 05:00:00
$COMSUITE_BIN/comdmft.py
```

Here ‘comdmft.py’ is a python script which controls the LQSGW+DMFT simulation. Based on the ‘comdmft.ini’ input file, ‘comdmft.py’ generates all the necessary input files for the individual programs (ComWann, ComCoulomb,ComDC, ComLowH and ComCTQMC) to run and execute jobs.

Input file (comdmft.ini)

In order to perform the LQSGW+DMFT calculation, we need only a single input file ‘comdmft.ini’. This input file should be written in python dictionary format. All dictionary keys are in small letters. ‘comdfmt.ini’ is composed of three python dictionaries of ‘control’, ‘wan_hmat’ and ‘imp’:

```
control={'initial_lattice_dir'          : '../lqsgw',
         'method'                 : 'lqsgw+dmft',
         'spin_orbit'             : False,
         'mpi_prefix': "srun -n 384",
         ‘max_iter_nu_impurity’: 200,
         'impurity_problem':[[1, 'd']],
         'impurity_problem_equivalence':[1],
        }

wan_hmat={
        'kgrid': [15, 15, 15],
        'froz_win_min': -15.0,
        'froz_win_max': 7.0,
                    }

imp={'temperature'            : 300,
     '1':
     {
     'impurity_matrix': [
         [1,0,0,0,0],
         [0,1,0,0,0],
         [0,0,2,0,0],
         [0,0,0,1,0],
         [0,0,0,0,2]
         ],
     ‘thermalization_time': 1,
     'measurement_time': 10,
     'green_cutoff':  50,
     'coulomb': 'full',
     }
     }
```

Here we note that convergence of the *ab initio* LQSGW+DMFT for MnO is particularly slow and takes almost 200 iterations in Cori at NERSC, for example. You should converge the calculation until chemical potential (mu) written in “convergence.log” is below -1.0 eV.

■ In Control

These fields contain the basic parameters which control the LQSGW+DMFT run.

- 'initial_lattice_dir' : '../lqsgw'
  Enter the path which contains the LQSGW output such as quasi-particle eigenvalues and eigenfunctions. This output is in the lqsgw prerun folder.

- 'method' : 'lqsgw+dmft'
  Either lda+dmft or lqsgw+dmft. Currently COMSUITE has these two options. Choose 'lqsgw+dmft' for the present work (LQSGW+DMFT approximation).

- 'spin_orbit': False
  Enter True of False. If False, correlated orbitals correspond to cubic spherical harmonics

$$Y_{lm} = \begin{cases} \frac{i}{\sqrt{2}}\left(Y_l^{-|m|} - (-1)^m Y_l^{|m|}\right), & m < 0 \\ Y_l^0, & m = 0 \\ \frac{1}{\sqrt{2}}\left(Y_l^{-|m|} + (-1)^m Y_l^{|m|}\right), & m > 0 \end{cases}$$

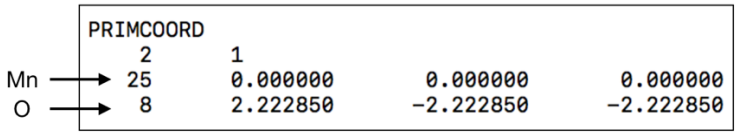
  where Y_l^m is a spherical harmonics.
  If True, correlated orbitals chosen at each correlated atom correspond to spin-angular functions |l,i,m>

$$\Omega_{l,i=\pm\frac{1}{2},m} = \sum_{s\pm 1/2} C_{i,s}^{l,m} Y_l^{m-s}(\hat{r}) u_s$$

  where u_s is a spinor, and $C_{i,s}^{l,m} = \langle l, m-s, \frac{1}{2}, s | l+i, m \rangle$.

- 'mpi_prefix': 'srun -n 384'
  MPI prefix used for ComCoulomb, ComDC, ComLowH, ComWann, and ComCTQMC programs. If a different MPI prefix is necessary for an individual program, use 'mpi_prefix_coulomb', 'mpi_prefix_lowh', 'mpi_prefix_dc', 'mpi_prefix_wannier', and 'mpi_prefix_impurity'. Note that 384 is the number of total MPI processes we want to use for the present case.
  You can however set any number you want. Here we note that, for ComCoulomb, comdmft.py will adjust the number of processes and distribute them on a two-dimensional MPI grid.

- 'impurity_problem': [[1,'d']]
  A python list to specify the correlated orbitals. The first and second indices indicates the atom index and subshell type, respectively. Atom index gives the atoms in the order listed in the file "../lqsgw/coord.xsf"



  Shell index is either "d" or "f".

- 'impurity_problem_equivalence': [1]
  Equivalence of each impurity problem. The value is identified by a positive integer starting from 1. If an impurity has the same value the one for another impurity, they are equivalent..

- 'restart': False
  Enter True or False. If True, it will resume the calculation from the previous LQSGW+DMFT run. The default value is False.

- 'mpi_prefix_lowh':
  MPI prefix for ComLowH. The default value is the one specified in control['mpi_prefix']

- 'mpi_prefix_impurity':
  MPI prefix for the impurity solver. The default value is the one specified in control['mpi_prefix']

- 'mpi_prefix_wannier':
  MPI prefix for ComWann. The default value is the one specified in control['mpi_prefix'].

- 'mpi_prefix_coulomb':

MPI prefix for ComCoulomb. The default value is the one specified in control['mpi_prefix'].

- 'mpi_prefix_dc':
  MPI prefix for ComDC. The default value is the one specified in control['mpi_prefix'].

- 'sigma_mix_ratio':
  Self-energy linear mixing ratio. You can specify any number within 0.0 – 1.0. The default value is 0.5.

- 'max_iter_num_impurity':
  Maximum iteration for the DMFT self-consistent loop. The default value is 50.

- 'proj_win_min':
  Low-energy cutoff to renormalize the projectors. The default value is the one specified in wan_hmat['dis_win_min']

- 'proj_win_max':
  High-energy cutoff to renormalize the projectors. The default value is the one specified in wan_hmat['dis_win_max']

■ **In wan_hmat**:
These fields define the frozen window, disentanglement window, and ab initio calculation from which maximally localized Wannier functions (MLFWs) are constructed.

- 'kgrid': [15,15,15]
  Crystal momentum grid for the Wannier interpolation of the LQSGW band structure.

- 'froz_win_min': -15.0 eV
  Lower boundary of the inner (frozen) window in eV.

- 'froz_win_max': 7.0 eV
  Upper boundary of the inner (frozen) window in eV.

- 'dis_win_min':
  Lower boundary of the outer (disentanglement) window in eV. The default value is same with wan_hmat['froz_win_min']

- 'dis_win_max':
  Upper boundary of the outer (disentanglement) window in eV. The default value is wan_hmat['froz_win_max'] +40.0

- 'num_iter':
  The number of minimization steps for the wannierization process (gauge dependent part of total spreading). The default value is 0.

- 'dis_num_iter':
  The number of minimization steps for the disentanglement process (gauge independent part of total spreading). The default value is 100.

■ **In imp**:
These fields are related to the Monte-Carlo algorithm and sampling of observables.

- 'temperature': 300
  Simulation temperature in K

- For each distinct impurity problem indexed by the value in control ["impurity_problem_equivalence"],
  - 'impurity_matrix': [
    [1,0,0,0,0],
    [0,1,0,0,0], 1:t_{2g}
    [0,0,2,0,0], 2:e_g
    [0,0,0,1,0],
    [0,0,0,0,2]
    ],
    Equivalence of the matrix element of the hybridization function and impurity self-energy. Starting from "1", you can set any positive number. If these values are

the same, the corresponding hybridization functions and impurity self-energies will be identical. If an element in the matrix is zero, then it will not be sampled by the impurity solver. Each column and row correspond to the Wannier orbitals in the following order|xy>,|yz>,|z^2>,|xz>, |x^2-y^2> if control['spin_orbit']==False. If control['spin_orbit']==True, the most rapidly changing index is "m" with the next one "i". They are sorted in ascending order. For the case of "f" shell, for example, they are ordered as:|3,-0.5, -2.5>,|3,-0.5, -1.5>,|3,-0.5, -0.5>, |3,-0.5, 0.5>,|3,-0.5, 1.5>,|3,-0.5, 2.5>, |3,0.5, -3.5>,|3,0.5, -2.5>,|3,0.5,-1.5>,|3,0.5, -0.5>,|3,0.5, 0.5>,|3,0.5, 1.5>,|3,0.5, 2.5>, |3,0.5, 3.5>,

- 'thermalization_time': 1,
  Wall time for thermalization in minutes.

- 'measurement_time': 10,
  Wall time for measurement in minutes.

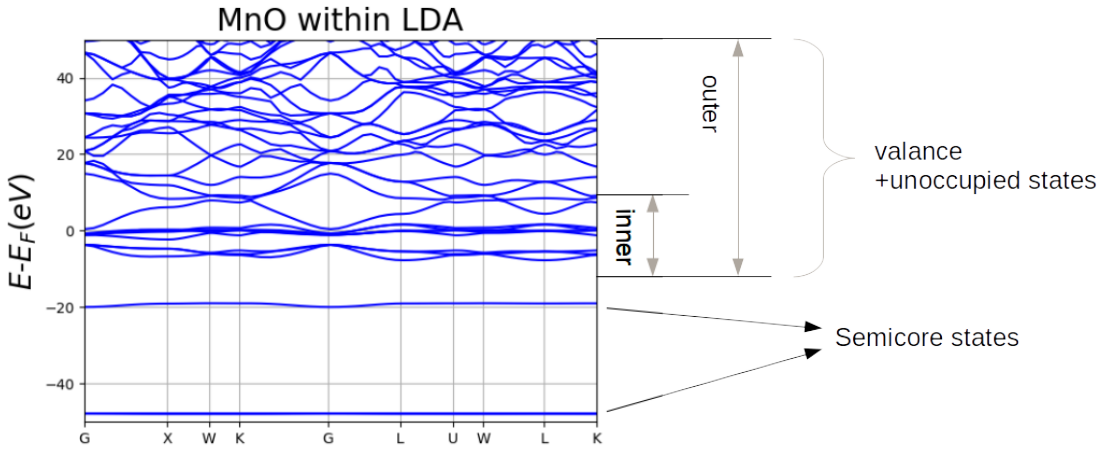
- 'green_cutoff': 50,
  Cutoff-energy in eV to sample Green's function and self-energy. The values beyond this energy will be provided by analytic extrapolations.

- 'Coulomb': 'full',
  'full' or 'ising' are available. We construct the Coulomb matrix in the following way:
  $$U_{m_1,m_2,m_3,m_4} = \sum_{k=0}^{2l,even} \frac{4\pi}{2k+1} F_l^k \sum_{q=-k}^{k} \langle Y_l^{m_1} | Y_k^q Y_l^{m_4} \rangle \langle Y_l^{m_2} Y_k^q | Y_l^{m_3} \rangle$$
  If 'full', no additional approximation is considered. If 'ising', only U_{abba} or U_{abab} are non-zero.

- 'susceptibility_cutoff':
  Cutoff-energy to sample susceptibility. The default value is 300 eV.

Input file (comdmft.ini)-Important concepts for wan_hmat

COMSUITE uses localized orbitals such as Wannier functions to represent the low-energy Hilbert space. To construct the Wannier functions, the inner (frozen) energy window can be set to range from E_F-15eV to E_F+7eV, and the outer (disentanglement) energy window can be E_F-15eV to E_F+47eV; see the figure below (to illustrate these concepts we employ an LDA band structure).

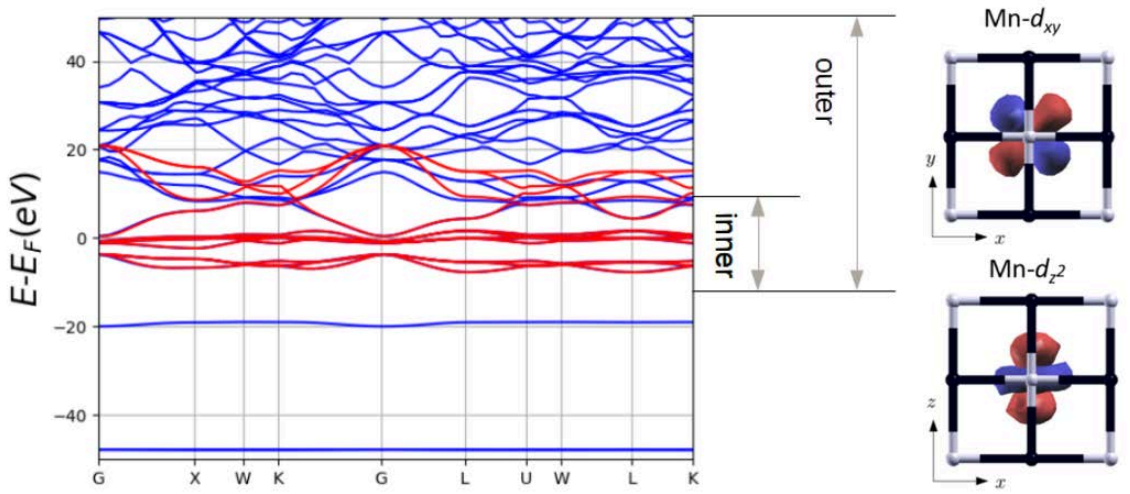


We choose initial trial orbitals $|\tau\mathbf{R}=0\rangle_t$ using muffintin orbitals with the desired angular momentum character. The radial functions of $|\tau\mathbf{R}=0\rangle_t$ are chosen in such a way to maximize

$$\frac{1}{N_{\mathbf{k}}} \sum_{n\mathbf{k}}^{E_{min}^{inner} < E_{n\mathbf{k}} < E_{min}^{inner}} |\langle n\mathbf{k}|\tau\mathbf{k}\rangle_t|^2$$

, where $|\tau\mathbf{k}\rangle_t = \frac{1}{N_\mathbf{k}} \sum_\mathbf{R} |\tau\mathbf{R}\rangle_t e^{i\mathbf{k}\cdot\mathbf{R}}$. Among the MT orbitals above, we have chosen ones which are larger than 0.15. For correlated orbitals, final Wannier functions $|\tau\mathbf{R}=0\rangle_f$ usually satisfy a condition of ${}_f\langle\tau\mathbf{R}=0|\tau\mathbf{R}=0\rangle_t > 0.95$. This means that $|\tau\mathbf{R}=0\rangle_f$ are strongly localized and are regarded as atom-like wavefunction.

The figure below shows Wannier functions and the interpolated band structure of MnO in comparison with the LDA band structure for MnO. The number of bands in the inner window is 10, while the number of bands in the outer window is 28. The number of trial orbitals is 12 (Mn-*s*, Mn-*p*, Mn-*d*, O-*p*).



Output files

COMSUITE locates important output files generated from individual program in the work directory ("lqsgw_dmft" in this example). The list of files is

cmd.log
convergence.log : convergence log file
delta.dat : hybridization function
sig.dat : impurity self-energy
sig_dc.dat : double-counting self-energy
sig_dc_hf.dat : the high-frequency limit of the double-counting self-energy
u_slater.dat : Slater-Condon parameterization of the bosonic Weiss-field
v_slater.dat : Slater-Condon parameterization of the bare Coulomb interaction (V_{loc})
w_slater.dat : Slater-Condon parameterization of the screened Coulomb interaction (W_{loc})

The format of each file and meaning of the fields are introduced below. The results of MnO LQSGW+DMFT calculation are presented with illustrative plots.

■ convergence.log

step	i_imp	causality	static_f0	w_sp_min	w_sp_max	mu	std_sig	n_imp	histo_1	histo_2
wannier				0.36291687	1.96593249					
coulomb_1			5.57603655552							
dc_1		good								
delta	1	good				0.007953387249				
impurity_1	1	good					1.90602117407	5.12777	146.776843399	163.76035554
delta	2	good				-0.205779536435				
impurity_1	2	good					1.074732719	5.09779	133.682320701	139.31567829
delta	3	good				-0.221765990535				
impurity_1	3	good					0.57247138017	5.0934	130.367736799	134.873739184
delta	4	good				-0.234693855509				
impurity_1	4	good					0.329673531991	5.09233	129.142363164	132.780730538
delta	5	good				-0.244398460158				
impurity_1	5	good					0.203407795362	5.0919	128.58334606	132.345209648

- Keeps track of the convergence of selected quantities at each iteration
- i_imp: The number of impurity problem being solved through ComLowH + ComCTQMC + ComDC
- causality: causality of the hybridization function/self-energy
- w_sp_min: minimum spreading of the Wannier functions
- w_sp_max: maximum spreading of the Wannier functions
- mu: LQSGW+DMFT chemical potential w.r.t. LQSGW chemical potential
- std_sig:

$$\sqrt{\frac{\sum_i (\Sigma_i^j(i\omega_n) - \Sigma_i^{j-1}(i\omega_n))^2}{n_\omega n_{orb}}}$$

- n_imp: occupation in the impurity orbitals
- histo_1: the first moment of the perturbation order histogram in the CTQMC solver
- histo_2: the second moment of the perturbation order histogram in the CTQMC solver
- ctqmc_sign: CTQMC sign

■ **u_slater.dat** and **w_slater.dat**

Data format in u_slater.dat and W_{loc} in w_slater.dat:

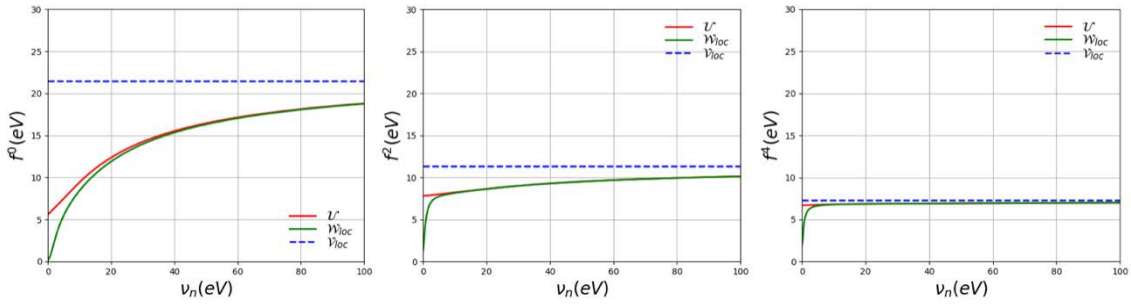
# nu(eV)	1:f0(eV)	1:f2(eV)	1:f4(eV)
0.000000000000	5.576036555520	7.794254788970	6.684458696734
0.162432849384	5.641351035972	7.800224004715	6.686831611341
0.324865698768	5.694491750277	7.804804733794	6.688329606083
0.487298548153	5.743311411165	7.808892622369	6.689517049807
0.649731397537	5.804658939529	7.814321347531	6.691507367040
0.812164246921	5.874472111574	7.820719809124	6.694110735352
0.974597096305	5.944466800415	7.827262771812	6.696864247113
1.137029945690	6.012119490988	7.833728852638	6.699641434646
1.299462795074	6.076897725008	7.840097632156	6.702396415820
1.461895644458	6.138300463962	7.846333893446	6.705067311804
1.624328493842	6.198131929126	7.852647124254	6.707697093537
1.786761343226	6.258094458145	7.859218786594	6.710324803345
1.949194192611	6.318133493012	7.865899565037	6.712936906491
2.111627041995	6.378094314763	7.872512446961	6.715515991596

The first column is the bosonic Matsubara frequencies and the second, third, and fourth column are the Slater's integrals F0, F2, and F4, respectively.

Data format of Bare Coulomb interaction in v_slater.dat is as follows:

# 1:f0(eV)	1:f2(eV)	1:f4(eV)
21.461484629544	11.311647977228	7.289678446990

Plots of dynamical U using u(v,w)_slater.dat files :



Slater's integrals of partially-screened Coulomb interactions associated with Mn-*d* orbitals are marked by red lines. For comparison, Slater's integrals of bare Coulomb interactions and fully-screened Coulomb interaction are shown by blue dashed lines and green full lines respectively. By excluding the polarizability between correlated bands, dielectric screening is suppressed, and Slater's integrals of a partially screened Coulomb interaction are larger than those of fully-screened Coulomb interactions in the low-energy regions. In addition, the monopole integral (F^0) shows much stronger frequency dependence than F^2 and F^4. As evidence of this, notice that F^0 of a partially screened Coulomb interaction varies from 5.6 eV to 21.4 eV but F^4 varies from 6.7 eV to 7.3 eV.

■ **sig_dc.dat**

'sig_dc.dat' contains impurity self-energies within the local GW approximation. The first column lists Matsubara frequencies and the next columns are the real and imaginary parts of the self-energies of the "t_{2g}" (Sig_{1,1}) and "e_g" (Sig_{1,2}) of the Mn d-orbitals.

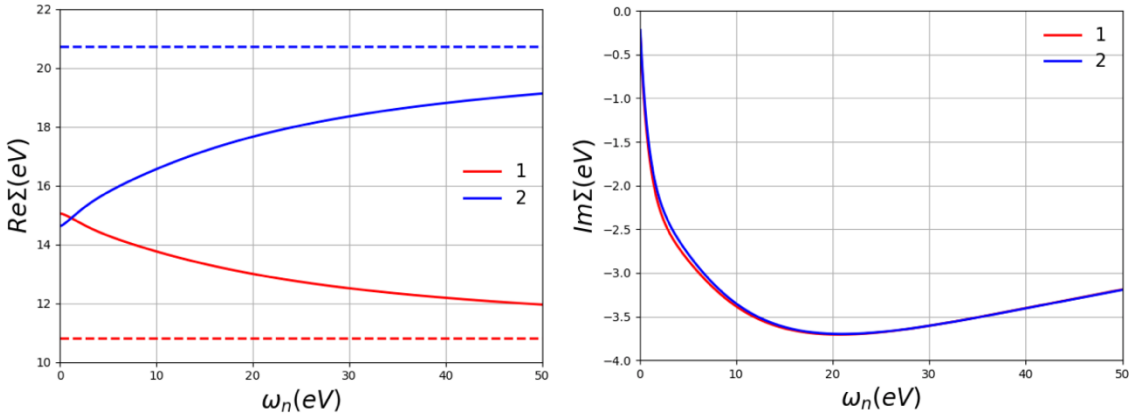
# omega(eV)	Re Sig_{1,1}(eV)	Im Sig_{1,1}(eV)	Re Sig_{1,2}(eV)	Im Sig_{1,2}(eV)
0.081216424692	15.044076161147	-0.254431733878	14.627736226076	-0.219963325953
0.243649274076	15.033658776174	-0.636741828609	14.650535806749	-0.568848707879
0.406082123461	15.015654293474	-0.937765823766	14.684590433428	-0.854035231347
0.568514972845	14.993152229741	-1.183011278893	14.724757327000	-1.091330969868
0.730947822229	14.968012719982	-1.386834232521	14.768235370279	-1.290511262610
0.893380671613	14.941263039099	-1.558458567565	14.813383610335	-1.458818811089
1.055813520997	14.913486331800	-1.704387724152	14.859237626192	-1.602016615185
1.218246370382	14.885042414197	-1.829462464146	14.905232007019	-1.724751087765
1.380679219766	14.856181238232	-1.937416181317	14.951027308022	-1.830763832337
1.543112069150	14.827096825677	-2.031214051280	14.996408753848	-1.923070642317
1.705544918534	14.797950835459	-2.113245128371	15.041230749043	-2.004098680820
1.867977767919	14.768882637313	-2.185447991002	15.085385513214	-2.075800841781

■ sig_dc_hf.dat

'sig_dc_hf.dat' contains real and imaginary parts of the Hartree-Fock contribution to the impurity self-energy within the local GW approximation.

# Re Sig_{1,1}(eV)	Im Sig_{1,1}(eV)	Re Sig_{1,2}(eV)	Im Sig_{1,2}(eV)
10.806021000000	0.000000000000	20.710902000000	0.000000000000

Plots of the local-GW impurity self-energy using sig_dc.dat and sig_hf_dc.dat files:



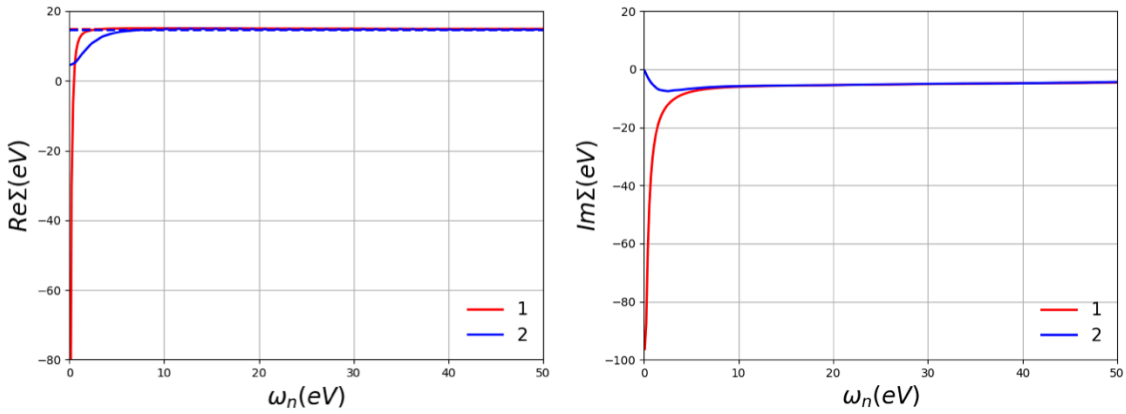
The double-counted electronic self-energy within the local GW approximation associated with the five Mn-*d* orbitals on the imaginary frequency axis. Red and blue colors represent the Mn-t_{2g} (d_{xy}, d_{yz}, and d_{zx}) and Mn-e_g (d_{z2}, and d_{x2-y2}) orbitals, respectively. Both the real and imaginary parts of self-energy do not show divergent behaviors near the Fermi energy, and the imaginary part is even linear along the imaginary frequency axis. However, if all the Feynman diagrams associated with the five Mn-*d* orbitals are summed, then, as we will see below, the self-energy shows a qualitatively different behavior.

■ sig.dat

'sig.dat' contains impurity self-energies obtained from ComCTQMC. The first column lists Matsubara frequencies and the next columns are the real and imaginary parts of the self-energies for "t_{2g}" (Sig_{1,1}) and "e_g" (Sig_{1,2}) of the Mn d-orbital.

# omega(eV)	Re Sig_{1,1}(eV)	Im Sig_{1,1}(eV)	Re Sig_{1,2}(eV)	Im Sig_{1,2}(eV)
0.081216424692	-135.590108173649	-96.332687524268	4.585573020796	-0.595745615898
0.243649274076	-30.220993388417	-87.388450496445	4.686916082241	-1.759850324803
0.406082123461	-6.259112581702	-62.546264060041	4.890100729310	-2.817851602824
0.568514972845	4.263460298568	-46.814969309441	5.175733137444	-3.744372026473
0.730947822229	8.642230589745	-37.146338378088	5.573539994318	-4.513295225373
0.893380671613	10.845625747838	-30.790228571536	6.087855157576	-5.154608569693
1.055813520997	12.101661764398	-26.339363030797	6.681014680018	-5.713577487684
1.218246370382	12.881497415855	-23.066808539907	7.288033364541	-6.213130200106
1.380679219766	13.396523076467	-20.568437827195	7.852025496793	-6.639615176511
1.543112069150	13.753056200846	-18.603932750236	8.358434592510	-6.958436287242

Plots of impurity self-energy using the sig.dat file:

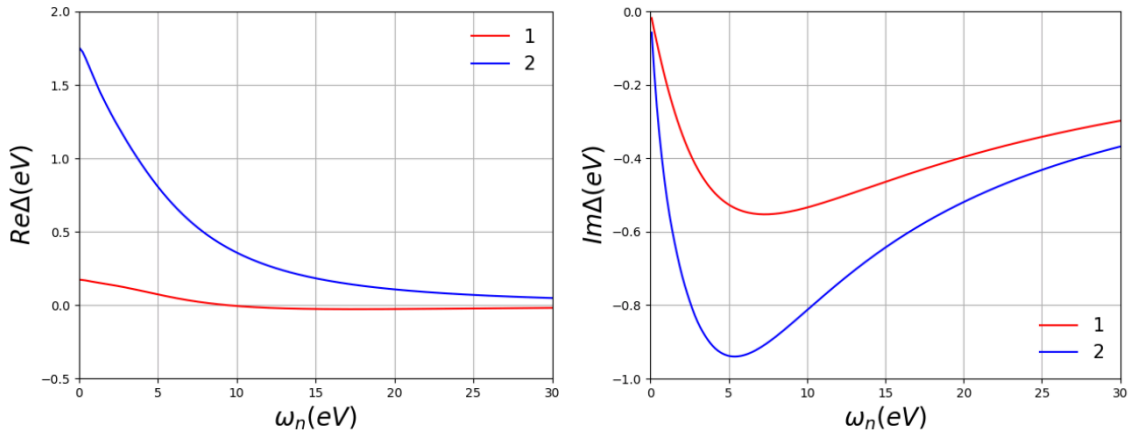


The impurity self-energy from ComCTQMC on the imaginary axis. In contrast to the electronic self-energy within the local GW approximation, both real and imaginary part of self-energy on the imaginary frequency axis show divergent behaviors near the Fermi level. In addition, electron occupations in the Mn-t2g and Mn-eg orbitals within DMFT are 0.50 and 0.52, which are much closer to half filling than those within the LQSGW approximation.

■ delta.dat

'delta.dat' is in the same format with 'sig.dat'

Plots of the real and imaginary parts of the hybridization function using the delta.dat file:



The hybridization function associated with Mn-d orbitals on the imaginary frequency axis. The red and blue lines represent Mn-t_{2g} and Mn-e_g characters, respectively. The Mn-t_{2g} orbitals experience less hybridization with the rest of the electrons than the Mn-e_g orbitals. The imaginary part of the hybridization approaches zero at the Fermi level, implying a gap opening in MnO.

Analytical continuation of self-energy

To obtain the density of states (DOS), we need to perform an analytic continuation of 'sig.dat' to produce the impurity self-energy on real frequency axis. To do this, we will adopt maximum entropy (maxent) method. Any publicly available maxent code can be employed. For the purposes of this tutorial, however, we will use K. Haule's maxent code (freely available at http://www.physics.rutgers.edu/~haule).

To access the maxent code, you should export the path to the executable in your startup shell script.

export WIEN_DMFT_ROOT=[path to Haule's code bin directory where maxent_run.py is located]

To run the maxent code, move to your work directory ("lqsgw_dmft"), create the maxent directory (you can name it however you would like) and then cd to it:

$ mkdir maxent
$ cd maxent

By executing ‘maxent_wrapper.py’, we can obtain the self-energy on the real axis by automatically calling maxent_run.py:

$ $COMSUITE_BIN/maxent_wrapper.py ../sig.dat 0.02

You can see its options with -h option:

$COMSUITE_BIN/maxent_wrapper.py –h

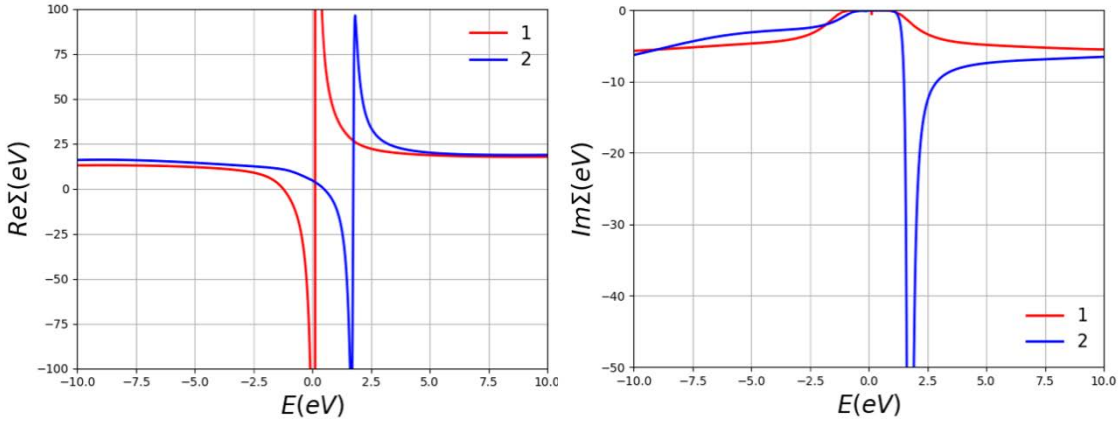
```
usage: maxent_wrapper.py [-h] sig [error]

call maxent_run.py from EDMFTF package and return self-energy on real axis. If
maxent_params.dat is not present in the directory, it generates one. The name
of the output file will be sig_realaxis.dat

positional arguments:
  sig         self-energy file on imaginary axis
  error       Errors for the maxent.Optional. Default value=0.05

optional arguments:
  -h, --help  show this help message and exit
```

Here note that in order to perform maxent, `maxent_params.dat’ file must be in the same directory as ‘sig.dat’. This file should be written in python dictionary format as follows:

```
params={'statistics': 'fermi', # fermi/bose
        'Ntau'      : 3000,     # Number of time points
        'L'         : 70.0,     # cutoff frequency on real axis
        'Nw'        : 501,      # number of frequency points on real axis
        'gwidth'    : 140.0,  # width of gaussian
        'idg'       : 1,        # error scheme: idg=1 -> sigma=deltag ; idg=0 -> sigma=deltag*G(tau)
        'deltag'    : 0.05,   # error
        'Asteps'    : 4000,     # anealing steps
        'alpha0'    : 1000,     # starting alpha
        'x0'        : 0.01,     # low energy cutoff
        'min_ratio' : 0.001,    # condition to finish, what should be the ratio
        'iflat'     : 1,        # iflat=0 : constant model, iflat=1 : gaussian of width gwidth, iflat=2 : input using file model.dat
        'Nitt'      : 1000,     # maximum number of outside iterations
        'Nr'        : 0,        # number of smoothing runs
        'bwdth'     : 0.03,     # smoothing width
        'Nf'        : 5,      # to perform inverse Fourier, high frequency limit is computed from the last Nf points
         }
```

If ‘maxent_params.dat’ file is not present in the directory, maxent_wrapper.py will automatically generate the file with default options and execute the subsequent jobs. If maxent is finished successfully, you will obtain a file of analytically continued self-energies, ‘sig_realaxis.dat’. ‘sig_realaxis.dat’ is in the same format as ‘sig.dat’ but for the first column which is now has real frequency values.

Plots of the impurity self-energy on the real frequency axis



The impurity self-energy on the real frequency axis as obtained using the maximum entropy method. Electronic self-energy for both Mn-t_{2g} and Mn-e_g has a pole near the Fermi level indicative of the paramagnetic Mott gap in MnO.

LGSGW+DMFT density of states

To compute the DOS, we need to post-process by executing ComLowH again. First, create a directory for the DOS calculation in the “lqsgw_dmft” directory and then cd to this directory, i.e.

$ mkdir realgrid
$ cd realgrid

Copy the necessary files to calculate the DOS and partial DOS by using prepare_realaxis.py which is in the $COMSUITE_BIN directory:

```
$ $COMSUITE_BIN/prepare_realaxis.py 0.1 ../lowh/ ../wannier/ ../maxent/sig_realaxis.dat 30 30 30 -m 2
```

You can see its options using the -h option.

```
$COMSUITE_BIN/prepare_realaxis.py -h
```

```
usage: prepare_realaxis.py [-h] [-m MODE]
                           broadening lowh_directory wan_directory self_energy
                           [kmesh_b1_for_dos] [kmesh_b2_for_dos]
                           [kmesh_b3_for_dos]

prepare inputs of comlowh calculation on real axis

positional arguments:
  broadening            broadening
  lowh_directory        lowh directory
  wan_directory         wannier directory
  self_energy           real-axis self-energy
  kmesh_b1_for_dos      finer kmesh along b1 axis for the DOS. Optional
  kmesh_b2_for_dos      finer kmesh along b2 axis for the DOS. Optional
  kmesh_b3_for_dos      finer kmesh along b3 axis for the DOS. Optional

optional arguments:
  -h, --help            show this help message and exit
  -m MODE, --mode MODE  If 3, code calculates spectral function along the high
                        symmetry line defined in 'kpath.dat'. If it is 2, it
                        calculates projected density of states. Default: 3
```

Run ComLowH with a job submission script. An example of such a script using SLURM is

```
#!/bin/bash -l
#SBATCH -J temp
#SBATCH -q debug
#SBATCH -N 1
#SBATCH -e temp.%j.err
#SBATCH -o temp.%j.out
#SBATCH -L SCRATCH
#SBATCH -C haswell
#SBATCH -t 00:30:00
srun -n 30 $COMSUITE_BIN/ComLowH
```

Doing this, you will obtain two files, tdos.dat and pdos.dat.

The format of tdos.dat file is:

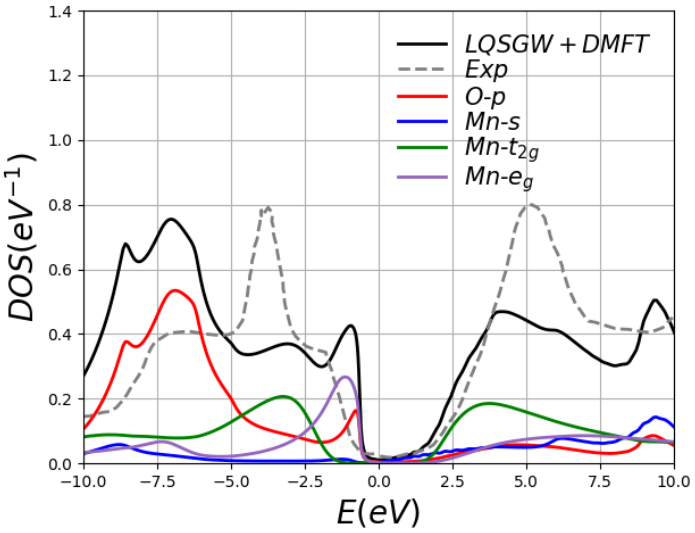
```
#       omega (eV)          DOS (1/eV)
   -70.000000000000     0.000073938864
   -65.733023713200     0.000084092948
   -61.955278682000     0.000094974529
   -58.587128546700     0.000106621041
   -55.565331497900     0.000119059367
   -52.839027896300     0.000132325897
   -50.366851286200     0.000146490590
   -48.114812211200     0.000161718115
   -46.054723948000     0.000178323962
   -44.163014879300     0.000197166674
   -42.419821079200     0.000219936353
```

while the format of the pdos.dat file is:

```
#     omega (eV)              (1,0,0)              (1,1,-1)              (1,1,0)              (1,1,1)              (1,2,-2)
   -70.000000000000     0.000005944444     0.000005002443     0.000005002443     0.000005002443     0.000006217714
   -65.733023713200     0.000006720094     0.000005599524     0.000005599524     0.000005599524     0.000007099839
   -61.955278682000     0.000007543398     0.000006224409     0.000006224409     0.000006224409     0.000008046122
   -58.587128546700     0.000008414898     0.000006876753     0.000006876753     0.000006876753     0.000009068535
   -55.565331497900     0.000009335247     0.000007556307     0.000007556307     0.000007556307     0.000010167696
   -52.839027896300     0.000010305230     0.000008262924     0.000008262924     0.000008262924     0.000011346072
   -50.366851286200     0.000011325775     0.000008996580     0.000008996580     0.000008996580     0.000012613715
   -48.114812211200     0.000012397982     0.000009757389     0.000009757389     0.000009757389     0.000013993314
```

- (atom index, l, m) if spin_orbit==False and (atom index, l, i,m) if spin_orbit==True

A plot of pdos.dat along with experiment data:



MnO DOS within LQSGW+DMFT. For comparison, experimental data is marked by a gray dashed line. LQSGW+DMFT reproduces the experimentally observed four peak structure at 5eV, -2eV, -4eV and -7eV relative to the Fermi energy reasonably well. A projected density of state calculation attributes each peak to Mn-t_{2g}, Mn-e_g, Mn-t_{2g}, and O-p orbitals, respectively. A substantial contribution of O-p to the top of the valence band is a manifestation of strong hybridization between O-p and Mn-e_g, which is consistent with the Zhang-Rice picture.

LGSGW+DMFT spectral function

To obtain the spectral function, we need to post-process by executing ComLowH again. First, create a directory for the spectral function in the “lqsgw_dmft” directory and then cd to it:

```
$ mkdir realaxis
$ cd realaxis
```

Copy the necessary files to calculate the spectral function by using prepare_realaxis.py which is in the $COMSUITE_BIT directory. Note that the option should be ‘-m 3’ which is different from the DOS calculation.

```
$ $COMSUITE_BIN/prepare_realaxis.py 0.1 ../lowh/ ../wannier/ ../maxent/sig_realaxis.dat -m 3
```

Then you should create a k-path file (kpath.dat). In the first line, put the number of k points. And in the following lines, k points are specified in terms of the reciprocal lattice vector.

```
186   # The number of k points
 0.00000000  0.00000000  0.00000000
 0.01666667  0.00000000  0.01666667
 0.03333333  0.00000000  0.03333333
 0.05000000  0.00000000  0.05000000
 0.06666667  0.00000000  0.06666667
 0.08333333  0.00000000  0.08333333
 0.10000000  0.00000000  0.10000000
 0.11666667  0.00000000  0.11666667
 0.13333333  0.00000000  0.13333333
 0.15000000  0.00000000  0.15000000
 0.16666667  0.00000000  0.16666667
 0.18333333  0.00000000  0.18333333
 0.20000000  0.00000000  0.20000000
 0.21666667  0.00000000  0.21666667
 0.23333333  0.00000000  0.23333333
 0.25000000  0.00000000  0.25000000
 0.26666667  0.00000000  0.26666667
 0.28333333  0.00000000  0.28333333
 0.30000000  0.00000000  0.30000000
 0.31666667  0.00000000  0.31666667
```

Run ComLowH with a job submission script. An example of such a job script using SLURM is

```
#!/bin/bash -l
#SBATCH -J temp
#SBATCH -q debug
#SBATCH -N 1
```

```
#SBATCH -e temp.%j.err
#SBATCH -o temp.%j.out
#SBATCH -L SCRATCH
#SBATCH -C haswell
#SBATCH -t 00:30:00
srun -n 30 $COMSUITE_BIN/ComLowH
```

Having done so you will have obtained the file, spectral.dat. The format of this file is

# kpoint	E (eV)	A (1/eV)
1	-70.000000000000	0.000071879936
2	-70.000000000000	0.000071887786
3	-70.000000000000	0.000071911184
4	-70.000000000000	0.000071949699
5	-70.000000000000	0.000072002704
6	-70.000000000000	0.000072069475
7	-70.000000000000	0.000072149260
8	-70.000000000000	0.000072241286
9	-70.000000000000	0.000072344689
10	-70.000000000000	0.000072458368
11	-70.000000000000	0.000072580824
12	-70.000000000000	0.000072710036

Here is a plot of the spectral.dat file along a high symmetry line in the first Brillouin zone: