General overview

The design of ComRISB follows ComDMFT, with a newly-written, python-based Wannier interface to the established CyGutz code. The overview is shown in Fig.1. The gold colored blocks stands for software modules that are shared with ComDMFT, while the blue colored blocks appear only in ComRISB. The typical ComRISB calculation starts with the DFT module, FlapwMBPT. It produces the DFT band energies and wave functions and passes them to the ComWann module. ComWann generates a wannier90-based tight-binding model which faithfully represents the DFT band structures within a specified energy window, which usually covers the occupied space up to several eV above the Fermi level. The Wannier interface prepares all the necessary input files for CyGutz according to this model. Then, CyGutz solves the generated generic Kohn-Sham-Hubbard (KSH) Hamiltonian. CyGutz essentially solves a set of coupled eigenvalue problems self-consistently. The solution, which is coded in the 2-dimensional arrays of the variational parameters $\mathcal{R}$ λ are then fed back to the Wannier interface. Then, the Wannier interface prepares a renormalized electron density matrix which is compatible

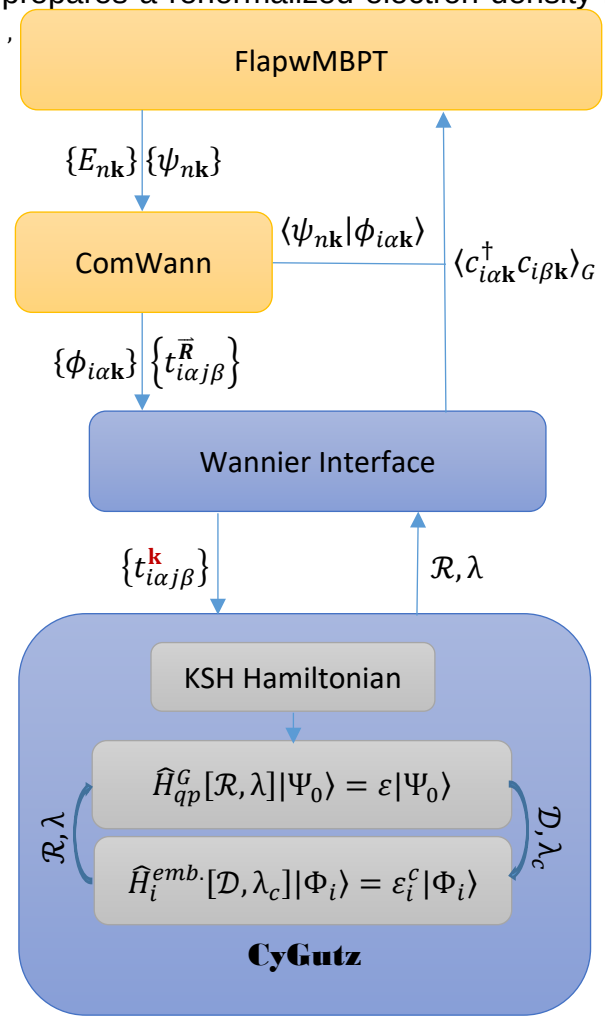


Fig. 1 Overall flowchart of ComRISB package.

with ComDMFT and returns it to FlapwMBPT. This triggers another iteration until the electron density self-consistency is reached. The overall workflow is conveniently managed by the *comrisb.py* script, which is itself based on *comdmft.py*.

Tutorial 1: MnO

We present here a DFT+G-RISB calculation of MnO using the comsuite package. At ambient conditions, MnO is in a rock salt crystal structure and characterized as a Mott charge transfer insulator. The Mn atom has roughly half-filled 3d high-spin states, where the intra-shell strong electron correlations will be treated at the G-RISB level. The steps of the computation follow closely the DFT+DMFT calculations in *comsuite*.

Initial DFT calculation

The first step is to perform a DFT-LDA calculation of MnO using the all-electron DFT code *rspflapw.exe* with the provided input file (*ini*), where we specify a k-grid of $10 \times 10 \times 10$. In DFT+G-RISB calculations, we prefer to use the same k-grid for both the DFT and G-RISB calculations, since the DFT calculations are quite efficient, especially with k-point parallelization. We store the DFT result in a subdirectory of the current working directory, ${work_dir}/dft.

Input file

Next, we perform the DFT+G-RISB calculation in the directory ${work_dir}/dftg. The only required input file for the managing script of DFT+G-RISB, *comrisb.py*, is the same as DFT+DMFT, *i.e.*, *comdmft.ini*. The input file includes three blocks.

```
control={
   'initial_dft_dir'       : '../dft/', # directory storing the initial dft calculation
   'method'                : 'lda+risb', # either lda+dmft, lda+risb or lqsgw+dmft.
   'spin_orbit'            : False,  # True or False. spin-orbit coupling is included or not
   'mpi_prefix': "mpirun -n 8", # parallel execution parameters
   'impurity_problem':[[2, 'd']], # correlated atom and shell (Mn-d)
   'impurity_problem_equivalence':[1], # equivalence index
   'proj_win_min': -80.0,
   'proj_win_max': 80.0,
   'max_iter_num_outer': 50,
   }
wan_hmat={
   'kgrid': [15, 15, 15], # not used by comrisb.
   'froz_win_min': -10.0,
   'froz_win_max': 10.0, # energy window within which states to be exactly reproduced.
   }
imp={'temperature'           : 300, # temperature (in K)
   '1':
   {
```

```
    'f0': 9.00, # Coulomb parameters, can be obtained from constrained RPA calculation.
    'f2': 9.82,
    'f4': 6.13,
    'nominal_n': 5.0, # fixed double counting with nominal occupancy of the ionic state
    'impurity_matrix': [ # equivalent orbital index matrix. starting from 1, not used by comrisb.
       [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': 2,
   'green_cutoff':  10,     # How often to record measurements
   'coulomb': 'full',
  }}
```

The input file specifies that the corelated atom and shell is Mn d-orbitals ('impurity_problem':[[2, 'd']]), the Coulomb parameters (f0, f2, f4), and the nominal double counting. The design philosophy behind *comrisb.py* is to use the same input files as *comdmft.py*. Nevertheless, since DFT+G-RISB calculation is much more efficient than DFT+DMFT, it is advisable to use less resources, e.g., fewer cores. In this example, we use 8 cores for the parallel execution.

Job submission script

We prepare a job file as below to be submitted in HPC using Slurm, which may require some customization for specific environments.

```
#!/bin/bash

#SBATCH --mail-user=ykent@iastate.edu
#SBATCH --mail-type=end
#SBATCH --partition=normal
#SBATCH --nodes=1
#SBATCH --ntasks=1
#SBATCH --cpus-per-task=8
#SBATCH --time=0-10
#SBATCH --job-name=MnO
#SBATCH --account=ykent
```

```
# record start date and time and host
date
hostname
python ${COMSUITE_BIN}/comrisb.py >& comrisb.log
# record end date and time
Date
```

Monitoring job

The job can be monitored by several log files. First, *cmd.log* file records the execution sequence of several modules of the *comrisb* package.

```
**********
        ComRISB
**********

top_dir /home/ykent/GitHub/DMFT_MatDeLab/example/lda_dmft/MnO/dftg
lattice_directory
/home/ykent/GitHub/DMFT_MatDeLab/example/lda_dmft/MnO/dftg/lattice
wannier_directory
/home/ykent/GitHub/DMFT_MatDeLab/example/lda_dmft/MnO/dftg/wannier
dc_directory /home/ykent/GitHub/DMFT_MatDeLab/example/lda_dmft/MnO/dftg/dc
impurity_directory
/home/ykent/GitHub/DMFT_MatDeLab/example/lda_dmft/MnO/dftg/impurity
lowh_directory /home/ykent/GitHub/DMFT_MatDeLab/example/lda_dmft/MnO/dftg/lowh

**********
iteration: 1
**********
initial dft directory setup done.
mpirun -n 8 /home/ykent//GitHub/DMFT_MatDeLab/bin/ComWann
comwann start at 16/08/2018 00:20:04 end at 16/08/2018 00:22:36
reading wannier.inip to get basis information.
control['impurity_wan']: [[7, 8, 9, 10, 11]]
mpirun -n 8 /home/ykent//GitHub/DMFT_MatDeLab/bin/gwannier.py
gwannier start at 16/08/2018 00:22:39 end at 16/08/2018 00:22:41
mpirun -n 8 /home/ykent//GitHub/DMFT_MatDeLab/bin/CyGutz
cygutz start at 16/08/2018 00:22:47 end at 16/08/2018 00:24:02
mpirun -n 8 /home/ykent//GitHub/DMFT_MatDeLab/bin/gwannden.py
gwannden start at 16/08/2018 00:24:02 end at 16/08/2018 00:24:06
prepare_dft_input done.
dft calculation done.
**********
iteration: 2
**********
mpirun -n 8 /home/ykent//GitHub/DMFT_MatDeLab/bin/ComWann
```

comwann start at 16/08/2018 00:24:29 end at 16/08/2018 00:27:05
…

After the initial setup of the working subdirectories, *ComWann* is called to get the maximally localized wannier function (MLWF) representation of the low energy Hamiltonian. *gwannier.py* is subsequently called to prepare the input files for the G-RISB calculation, which is carried out by *CyGutz*. *gwannden.py* updates the renormalized quasiparticle density matrix, which is fed back to *rspflapw.exe* to update the DFT band dispersions. New iteration starts with the update of MLWF and continues until the electron density self-consistency or maximal iteration is reached. The convergence information of each iteration is stored in *convergence.log* file.

i_outer	delta_rho	etot	mu	err_risb	min_z
1	0.00545234	-2462.37780376	1.46740078	0.00000075	0.46703504
2	0.00198489	-2462.38845685	1.46740065	0.00000092	0.46703516
3	0.00048840	-2462.39441458	1.58022329	0.00000006	0.46219550
4	0.00019862	-2462.39630649	1.66512558	0.00000060	0.44089061
5	0.00012258	-2462.39708467	1.69271594	0.00000046	0.44196886
6	0.00008794	-2462.39745149	1.70480973	0.00000004	0.44280585
7	0.00006436	-2462.39767512	1.70975698	0.00000001	0.44338582
8	0.00004745	-2462.39784080	1.71205084	0.00000000	0.44376551
9	0.00003535	-2462.39797546	1.71345594	0.00000064	0.44400615
10	0.00002662	-2462.39808840	1.71455214	0.00000046	0.44415529
11	0.00002022	-2462.39818390	1.71550627	0.00000036	0.44424342
12	0.00001546	-2462.39826478	1.71636294	0.00000026	0.44429134
13	0.00001188	-2462.39833331	1.71713668	0.00000014	0.44431294
14	0.00000916	-2462.39839145	1.71783001	0.00000006	0.44431721
15	0.00000708	-2462.39844082	1.71845177	0.00000014	0.44431072
16	0.00000549	-2462.39848281	1.71900875	0.00000009	0.44429765
17	0.00000426	-2462.39851859	1.71950512	0.00000008	0.44428087
18	0.00000331	-2462.39854913	1.71994748	0.00000002	0.44426231
19	0.00000258	-2462.39857526	1.72034005	0.00000002	0.44424324
20	0.00000201	-2462.39859765	1.72068833	0.00000001	0.44422450

It prints the information on the electron density difference, total energy (for reference only, not exactly coded yet), fermi level, G-RISB convergence and the minimal quasiparticle weight, which will alert the user to the possibility of Mott solutions. A well-known issue of G-RISB method is that the quasi-particle weight is generally overestimated, compared to the DMFT approach. This is already manifested in the current calculation. For the full Mott solution of G-RISB, it is expected that the quasiparticle weights will all be zero; however, the minimal quasiparticle weight is shown to be 0.44 in the example above. Within the G-RISB method, somewhat larger Coulomb parameters should be used to reach the Mott phase. Nevertheless, we will proceed with the given parameters to show how a typical *comrisb* calculation proceeds. At the end of this tutorial, we will address the issue of Mott solutions.

G-RISB results

The main text output file of G-RISB calculation is *GUTZ.LOG*. It contains information like the one-body part of the local Hamiltonian,

```
************       h1e-sym  ************
 imp=  1
 real part
-0.4235   0.0000   0.0000   0.0000  -0.0000   0.0000  -0.0000   0.0000   0.0000   0.0000
   0.0000  -0.4235   0.0000   0.0000   0.0000  -0.0000   0.0000  -0.0000   0.0000
0.0000
   0.0000   0.0000  -0.4235   0.0000  -0.0000   0.0000  -0.0000   0.0000   0.0000
0.0000
   0.0000   0.0000   0.0000  -0.4235   0.0000  -0.0000   0.0000  -0.0000   0.0000
0.0000
  -0.0000   0.0000  -0.0000   0.0000  -0.4235   0.0000  -0.0000   0.0000  -0.0000
0.0000
   0.0000  -0.0000   0.0000  -0.0000   0.0000  -0.4235   0.0000  -0.0000   0.0000  -
0.0000
  -0.0000   0.0000  -0.0000   0.0000  -0.0000   0.0000  -0.2199   0.0000   0.0000
0.0000
   0.0000  -0.0000   0.0000  -0.0000   0.0000  -0.0000   0.0000  -0.2199   0.0000
0.0000
   0.0000   0.0000   0.0000   0.0000  -0.0000   0.0000   0.0000   0.0000  -0.2199
0.0000
   0.0000   0.0000   0.0000   0.0000   0.0000  -0.0000   0.0000   0.0000   0.0000  -
0.2199
   orbital splitting:
   0.00000   0.00000  -0.00000  -0.00000  -0.00000  -0.00000   0.20359   0.20359
0.20359   0.20359
max error due to symmetrization =      0.000001
```

It shows the expected t_{2g} and e_g splitting of the Mn-d orbital with cubic symmetry. The bare DFT crystal field splitting is found to be $\Delta_{cf} = \varepsilon_{t_{2g}} - \varepsilon_{e_g} = 0.20\,\mathrm{eV}$. The last line shows the symmetrization error of the local quantities, which is a good place to check for possible symmetry breaking solutions.

The local one-body density matrix before applying the correlation is shown as the first entry of “nks-sym” in the output file.

```
************       nks-sym  ************
 imp=  1
 real part
   0.6982   0.0000   0.0000   0.0000  -0.0000   0.0000  -0.0000   0.0000  -0.0000   0.0000
   0.0000   0.6982   0.0000   0.0000   0.0000  -0.0000   0.0000  -0.0000   0.0000  -0.0000
   0.0000   0.0000   0.6982   0.0000  -0.0000   0.0000  -0.0000   0.0000  -0.0000   0.0000
```

```
  0.0000   0.0000   0.0000   0.6982   0.0000  -0.0000   0.0000  -0.0000   0.0000  -0.0000
 -0.0000   0.0000  -0.0000   0.0000   0.6982   0.0000   0.0000   0.0000   0.0000   0.0000
  0.0000  -0.0000   0.0000  -0.0000   0.0000   0.6982   0.0000   0.0000   0.0000   0.0000
 -0.0000   0.0000  -0.0000   0.0000   0.0000   0.0000   0.3582   0.0000  -0.0000   0.0000
  0.0000  -0.0000   0.0000  -0.0000   0.0000   0.0000   0.0000   0.3582   0.0000  -0.0000
 -0.0000   0.0000  -0.0000   0.0000   0.0000   0.0000  -0.0000   0.0000   0.3582   0.0000
  0.0000  -0.0000   0.0000  -0.0000   0.0000   0.0000   0.0000  -0.0000   0.0000   0.3582
  sub_tot=  5.622014  0.000000
 max error due to symmetrization =      0.000000
```

It is a 10×10 matrix, under the basis of Mn-d orbitals including spin as the faster index. It shows the bare total d-occupancy is 5.6, which is bigger than the nominal value of Mn^{2+}. The t_{2g} occupancy is almost twice that of the e_g occupancy due to the crystal field splitting. The converged quasiparticle density matrix, which are obtained based on the quasiparticle band structure, is identified by the last "*nks-sym*" entry at the end of the file.

```
************       nks-sym  ************
 imp=  1
 real part
  0.8140   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000
  0.0000   0.8140   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000
  0.0000   0.0000   0.8140   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000
  0.0000   0.0000   0.0000   0.8140   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000
  0.0000   0.0000   0.0000   0.0000   0.8140   0.0000   0.0000   0.0000   0.0000   0.0000
  0.0000   0.0000   0.0000   0.0000   0.0000   0.8140   0.0000   0.0000   0.0000   0.0000
  0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0731   0.0000   0.0000   0.0000
  0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0731   0.0000   0.0000
  0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0731   0.0000
  0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0731
  sub_tot=  5.176403  0.000000
max error due to symmetrization =      0.000000
```

In this example, it shows the d-occupancies are quite polarized and total occupancy is reduced to about 5.2. The physical one-particle density matrix, which is evaluated based on the local many-body density matrix, is identified by the last "*ncp-renorm*" entry at the end of the file.

```
************       ncp-sym  ************
 imp=  1
 real part
  0.7792   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000
  0.0000   0.7792   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000
  0.0000   0.0000   0.7792   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000
  0.0000   0.0000   0.0000   0.7792   0.0000   0.0000   0.0000   0.0000   0.0000   0.0000
  0.0000   0.0000   0.0000   0.0000   0.7792   0.0000   0.0000   0.0000   0.0000   0.0000
  0.0000   0.0000   0.0000   0.0000   0.0000   0.7792   0.0000   0.0000   0.0000   0.0000
```

```
  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.1253  0.0000  0.0000  0.0000
  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.1253  0.0000  0.0000
  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.1253  0.0000
  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.1253
  sub_tot=  5.176403  0.000000
 max error due to symmetrization =      0.000000
```

It clearly shows some deviation from the quasiparticle density matrix due to correlations. One can also see that the quasi-particle renormalization matrix $Z = R^{\dagger}R$ deviates significantly from identity matrix. The Z-matrix is identified by the last "*z-out-sym*" entry at the end of the file.

```
************     z-out-sym  ************
 imp=  1
 real part
  0.4442  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000
  0.0000  0.4442  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000
  0.0000  0.0000  0.4442  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000
  0.0000  0.0000  0.0000  0.4442  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000
  0.0000  0.0000  0.0000  0.0000  0.4442  0.0000  0.0000  0.0000  0.0000  0.0000
  0.0000  0.0000  0.0000  0.0000  0.0000  0.4442  0.0000  0.0000  0.0000  0.0000
  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.8061  0.0000  0.0000  0.0000
  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.8061  0.0000  0.0000
  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.8061  0.0000
  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.0000  0.8061
imp=  1 eigen values of        z:
  0.8061    0.8061    0.8061    0.8061    0.4442    0.4442    0.4442    0.4442    0.4442
0.4442
```

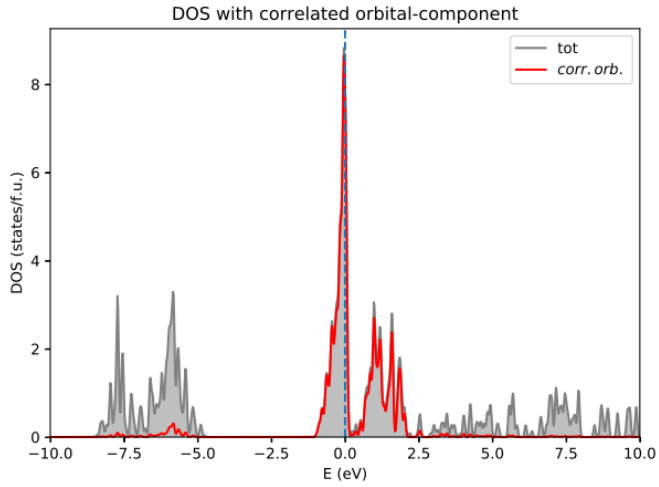
The quasiparticle weight is about 0.44 for t_{2g} orbitals and 0.80 for e_g orbitals. In the Mott phase, the G-RISB method would yield 0 quasi-particle weight.

Density of states

The quasiparticle density of states (dos) with Mn-d orbital component can be conveniently plotted by executing the following command:

```
${COMSUITE_BIN}/plot_dos_tf.py -el -10 -eh 10
```

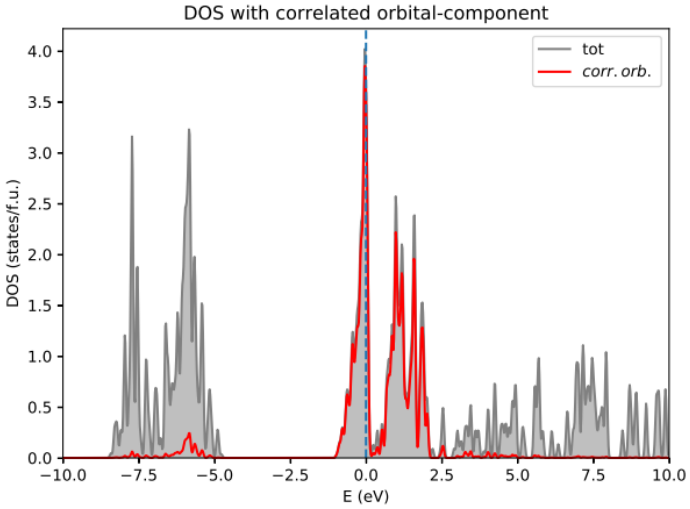
It specifies the energy window for the dos plot to be [-10eV, 10eV] with the Fermi level



pinned at 0.

The Mn-d states are located at the Fermi level, with some hybridization with O-2p states at lower energies. In G-RISB theory, the quasiparticle DOS is simply the renormalized coherent part of the DOS. The noncoherent part of the DOS is unfortunately very hard to evaluate. The coherent DOS can be conveniently plotted by adding an inline argument of -ch:

${COMSUITE_BIN}/plot_dos_tf.py -el -10 -eh 10 -ch



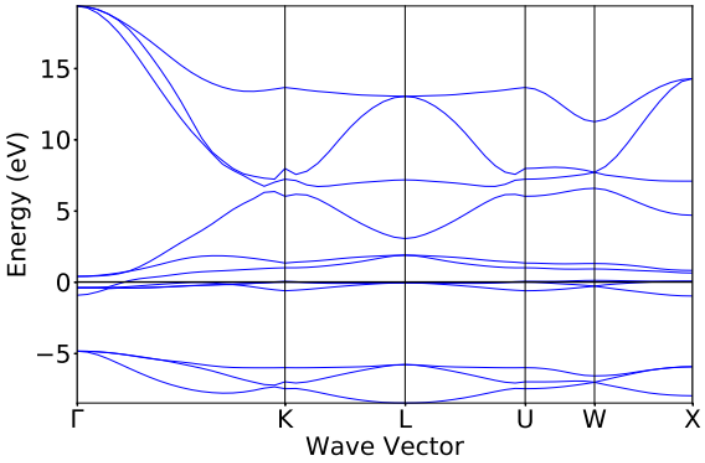
The Mn-d spectral weight is rescaled by the amount corresponding to quasi-particle renormalization matrix Z.

Quasi-particle band structure

A convenient python script, gwannier_plot_bands.py, has been prepared with the high-symmetry k-path automatically chosen based on the symmetry of the lattice. It can also

be executed in parallel via *mpirun*. For example, use the following command to plot DFT+G quasi-particle band structure with 4 cores.

mpirun -np 4 ${COMSUITE_BIN}/gwannier_plot_bands.py -g



The inline argument of -g specifies DFT+G band calculation rather than the default DFT calculation. The obtained band structure is shown below.

Mott phase

As we have discussed, G-RISB overestimates the quasiparticle weight, or equivalently the critical Coulomb parameters for the Mott transition in paramagnetic phase. One straightforward way is to scan the system with gradually increasing Coulomb parameters, which is affordable since G-RISB calculations are generally quite efficient. Nevertheless, the Mott solution involves fractionally occupied flat bands, which deteriorates the numerical stability. Therefore, a specific Mott solver has been implemented, which will stabilize the calculation. The other approach to address the issue of Mott solution is to introduce spin-symmetry breaking. It is essentially a much better version of LSDA+U where the Gutzwiller embedding Hamiltonian is solved exactly rather than at Hartree-Fock level as in LSDA+U. We focus on the first approach in this tutorial.

Special Mott solver

To choose the special Mott solver, first go to the subdirectory of *lowh*, then type the command:

${COMSUITE_BIN}/init_mott.py

We should answer the questions accordingly:

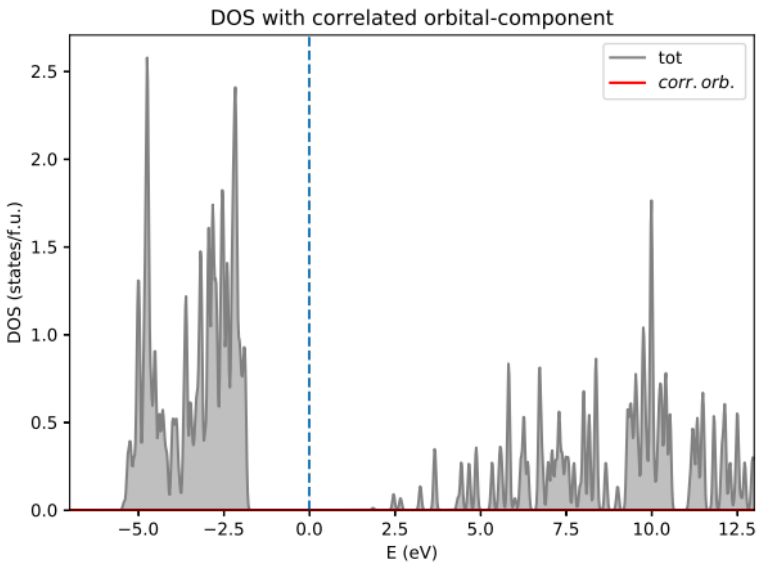
********** Impurity 1 **********
Sigma structure:
index 0 1 2 3 4 5 6 7 8 9

```
  0     1   0   0   0   0   0   0   0   0   0
  1     0   1   0   0   0   0   0   0   0   0
  2     0   0   1   0   0   0   0   0   0   0
  3     0   0   0   1   0   0   0   0   0   0
  4     0   0   0   0   1   0   0   0   0   0
  5     0   0   0   0   0   1   0   0   0   0
  6     0   0   0   0   0   0   2   0   0   0
  7     0   0   0   0   0   0   0   2   0   0
  8     0   0   0   0   0   0   0   0   2   0
  9     0   0   0   0   0   0   0   0   0   2
 Please provide the indices of orbitals to be Mott localized
 (e.g., 0 2 ): 0   1   2   3   4   5   6   7   8   9
 You selected [0   1   2   3   4   5   6   7   8   9]
 to be Mott localized, right? (y/n):y
 Please provide the total number of Mott localized electrons (per unit cell): 5
 Total 5 electrons will be Mott localized, right? (y/n):y
 R structure:
index    0   1   2   3   4   5   6   7   8   9

  0     0   0   0   0   0   0   0   0   0   0
  1     0   0   0   0   0   0   0   0   0   0
  2     0   0   0   0   0   0   0   0   0   0
  3     0   0   0   0   0   0   0   0   0   0
  4     0   0   0   0   0   0   0   0   0   0
  5     0   0   0   0   0   0   0   0   0   0
  6     0   0   0   0   0   0   0   0   0   0
  7     0   0   0   0   0   0   0   0   0   0
  8     0   0   0   0   0   0   0   0   0   0
  9     0   0   0   0   0   0   0   0   0   0
 Lambda structure:
index    0   1   2   3   4   5   6   7   8   9

  0     0   0   0   0   0   0   0   0   0   0
  1     0   0   0   0   0   0   0   0   0   0
  2     0   0   0   0   0   0   0   0   0   0
  3     0   0   0   0   0   0   0   0   0   0
  4     0   0   0   0   0   0   0   0   0   0
  5     0   0   0   0   0   0   0   0   0   0
  6     0   0   0   0   0   0   0   0   0   0
  7     0   0   0   0   0   0   0   0   0   0
  8     0   0   0   0   0   0   0   0   0   0
  9     0   0   0   0   0   0   0   0   0   0

 Please select the method to solve embedding Hamiltonian.
 LDIAG = -2: Valence truncation ED for Mott solution.
         -3: Option (-2) with additional S=0 constraints.
```

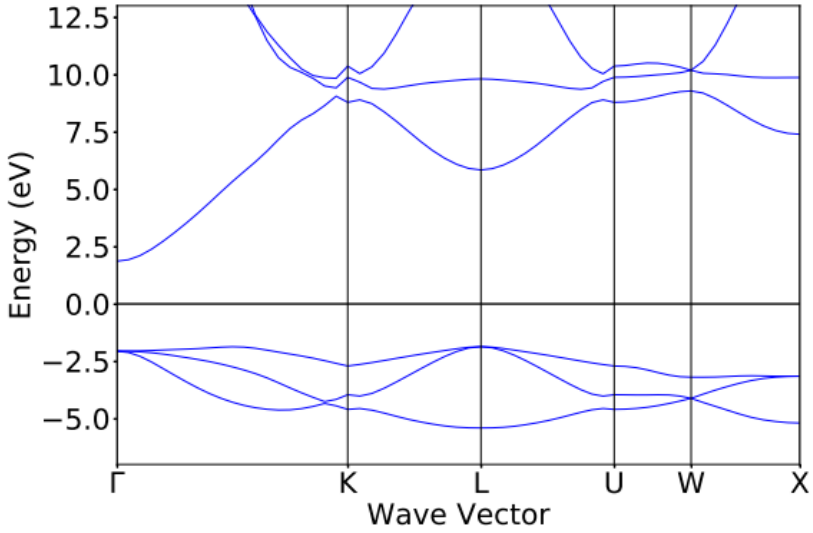
-4: Option (-2) with additional J=0 constraints.
-31: Option (-2) with S=0
and local symmetry constraints.
Please select LDIAG:
Pick one from [-2, -3, -4, -31]...-3

Here we choose the full-Mott solution, *i.e.*, all the d-orbitals (including spin here) are Mott localized. We put occupancy to be 5. The script will then print the structure of R and λ, which are the variables for the G-RISB nonlinear equations to be solved. In the full Mott case, R is the zero matrix, and λ is pinned by the Fermi level. Finally we choose LDIAG=-3 for the embedding Hamiltonian solver in the case without spin-orbit coupling.

In practices, the Mott solution can be computed faster than the normal state calculations, because of the reduced degreed of freedom. The job can be submitted in the same way as before. Finally, we can check the coherent part of the dos, which now appears as below:



The Mn-d states are not present since their quasiparticle weights become zero. The corresponding coherent part of the band structure is shown below.



This concludes the tutorial on the DFT-G-RISB calculation of MnO using the comrisb package.