Dielectric function and excitons with QSGŴ

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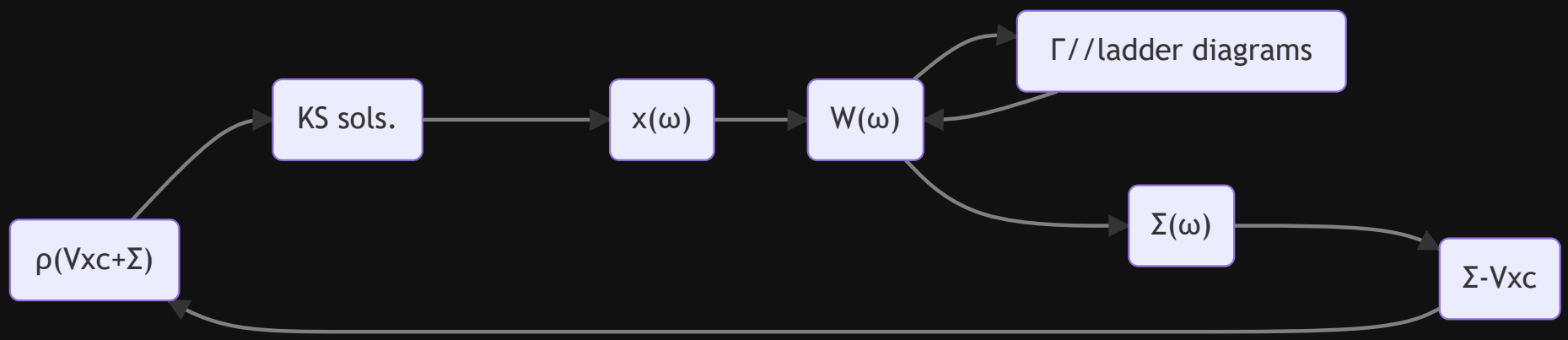
QSGŴ

All-electron quasi-particle self-consistent GW.

Constructs a *starting point independent*, *ab-initio*, static, non-local, effective potential from GW Σ iteratively



Basic prep

We will walk through a LiF calculation using 4x4x4 k-mesh, screened basis, ladder diagram corrected QSGW (a.k.a. QSGŴ a.k.a QSGW~ ...) with active space of 4 occupied and 4 unoccupied states.

Let's copy the input data

```
DATA="/apps20/contrib/questaal/d/tutorial/xct/LiF/4s"

rsync -av $DATA/i/ $DATA/qsgw-bsw/{rst.lm,sig*} qsgw-bsw-bse-eps/
cd qsgw-bsw-bse-eps/

# For the curious, the input calculation was done with

# m1="env OMP_NUM_THREADS=32 MKL_NUM_THREADS=32 mpirun -n 1 -ppn 1"
# mn="env OMP_NUM_THREADS=4 MKL_NUM_THREADS=4 mpirun -n 8 -ppn 8"
# mb="env OMP_NUM_THREADS=2 MKL_NUM_THREADS=2 mpirun -n 11 -ppn 11" # 11 because it matches the ibz+oft
# lmgw.sh --lm-flags='--v8' --mpirun-1 "$m1" --mpirun-n "$mn" --cmd-bs "$mb" --bsw ctrl.lm
```

Basic prep

The following command for copying files from/to the HPC facility:

```
rsync -Pavzz scarf:w/...../file ./
```

Alternatively you can `sshfs mount` the work directory to your local machine with the following commands (or similar) executed on your machine:

```
cd path/to/school/related/directory/on/your/laptop/
mkdir remote
sshfs scarf:/path/to/rundir remote -o idmap=user
```

then you can open files locally (though it may be laggy). Relies on uninterrupted connection, otherwise it can hang.

BSE Dielectric function

This is done with the `lmgw.sh` script and the `--bse` option with some 'familiar' boilerplate options:

```
m1="env OMP_NUM_THREADS=32 MKL_NUM_THREADS=32 mpirun -n 1 -ppn 1"
mn="env OMP_NUM_THREADS=4 MKL_NUM_THREADS=4 mpirun -n 8 -ppn 8"
lmgw.sh --lm-flags='--v8' --mpirun-1 "$m1" --mpirun-n "$mn" --cmd-bs "$m1" --bse ctrl.lm
```

The `--cmd-bs` option specifies the launcher command to be used for the `bse` executable, in the optics mode, contrary to the self-consistent mode, it only deals with q=(0 0 0) and while it can be run in MPI multiprocessing more it does well in a single process with many threads too.

This step also generates an eigensolution in file `w2q0e.h5` which can be reused in later steps.

BSE Dielectric function

The following output is expected:

```
+ lmgw.sh --lm-flags=--v8 --mpirun-1 '$m1' --mpirun-n '$mn' --cmd-bs '$m1' --bse ctrl.lm
recording execution environment to file env
06:11:46 env > env
    starting iteration 1 of 100
06:11:46 env $m1 lmfgwd --v8 --job=0 --lmqp~rdgwin --gwcode=2 --no-iactive ctrl.lm &> llmfgw00  ... 1s
06:11:47 env $mn lmfgwd --v8 --job=1 --gwcode=2 --no-iactive ctrl.lm &> llmfgw01  ... 1s
06:11:48 env $m1 rdata4gw_v2 --job=0 --efmode=1 --chknwmode=1 --v8 ctrl.lm &> lrdata4gw
06:11:48 env $m1 hbasfp0 --job=0 --v8 ctrl.lm &> lbas
06:11:48 env $mn hvccfp0 --v8 ctrl.lm &> lvcc  ... 1s
06:11:49 env $mn lmsig --wrw-only --om0 --v8 ctrl.lm &> lx0  ... 2s
06:11:51 env $mn lmf --revec:gw --opt:woptmc,permqp --v8 ctrl.lm &> llmf02  ... 1s
06:11:52 env $m1 bse --optdfl=optdatac.h5 --v8 ctrl.lm &> lbse  ... 1s
```

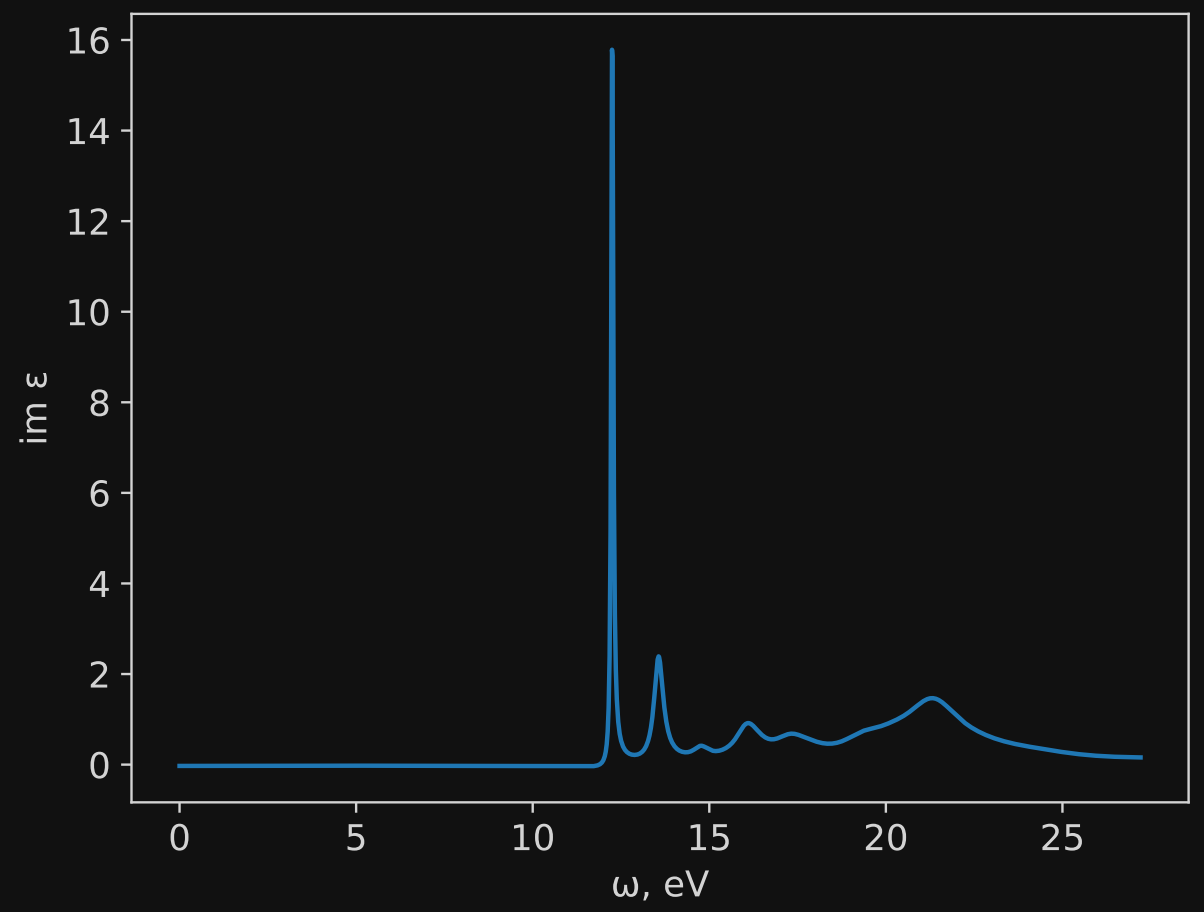
Let's save the last command from the output above because it will be useful starting point later on:

```
bsc="env OMP_NUM_THREADS=12 mpirun -n 1 bse --optdfl=optdatac.lm --v8 ctrl.lm"
```

BSE Dielectric function

Without further ado, the macroscopic dielectric function is saved in file `eps-plot.txt`. It is trivially plottable, for example:

```
    python -c 'from pylab import *; d=loadtxt("qsgw-bsw-bse-eps/eps-plot.txt");
plot(d[:,0], d[:,4]); xlabel("ω, eV"); ylabel("im ε"); savefig("LiF-qsgww-bse.pdf")'
```



Exciton listing

The stdout saved in file "lbse" has the list of the bound excitons found, together with some basic info to guide further steps, here is an excerpt:

```
...
qvecs
1.00000000   0.00000000   0.00000000
0.00000000   1.00000000   0.00000000
0.00000000   0.00000000   1.00000000
spin 1 evals in [-3.7177, -.9005] Ry
writing restart data to file w2q0e.h5 ... done

344 evals in range: [0.000,1.658], grouped by degeneracy tol 0.1000E-2ry
isp  hidx  group    ev, ry     ev, ev      oscillator  strengths along qvecs
0   1023     0    0.90048319   12.251698      5.527      0.1697      0.1441
0   1022     0    0.90048390   12.251708     0.2582      0.7724       4.810
0   1021     0    0.90048402   12.251709     0.5561E-01   4.898      0.8865
0   1020     1    0.99749445   13.571603      3.398      0.2108      0.2094
0   1019     1    0.99749501   13.571611     0.3136      0.1875       3.317
0   1018     1    0.99749514   13.571613     0.1066       3.420      0.2916
0   1017     2     1.0376747   14.118284     0.1441E-20  0.5664E-20  0.2912E-20
0   1016     2     1.0376748   14.118285     0.4443E-21  0.3197E-20  0.3377E-21
0   1015     2     1.0376749   14.118286     0.6919E-20  0.2083E-20  0.2692E-21
0   1014     3     1.0419070   14.175867     0.3710E-21  0.5438E-20  0.2938E-20
0   1013     3     1.0419071   14.175869     0.1794E-21  0.5925E-21  0.3888E-20
0   1012     4     1.0431189   14.192355     0.1237E-05  0.2528E-06  0.4511E-06
0   1011     4     1.0431190   14.192357     0.1008E-06  0.6227E-06  0.1268E-05
0   1010     4     1.0431192   14.192360     0.6638E-06  0.1118E-05  0.2825E-06
...
```

Excitonic band weights / projection

The lowest exciton is at ~12.25eV (group 0) and it is not dark as attested by the oscillator strengths.

We can try to quantify the level to which the active states participate in a particular exciton by integrating the 2 particle weights for a chosen energy range and decorate a band plot with them.

Let's start with the weights for energy window 12-13ev and record the output to a file named exbw-12-13ev.h5:

```
$bsc --wexbw~ewin=12,13eV~fn=exbw-12-13ev.h5
```

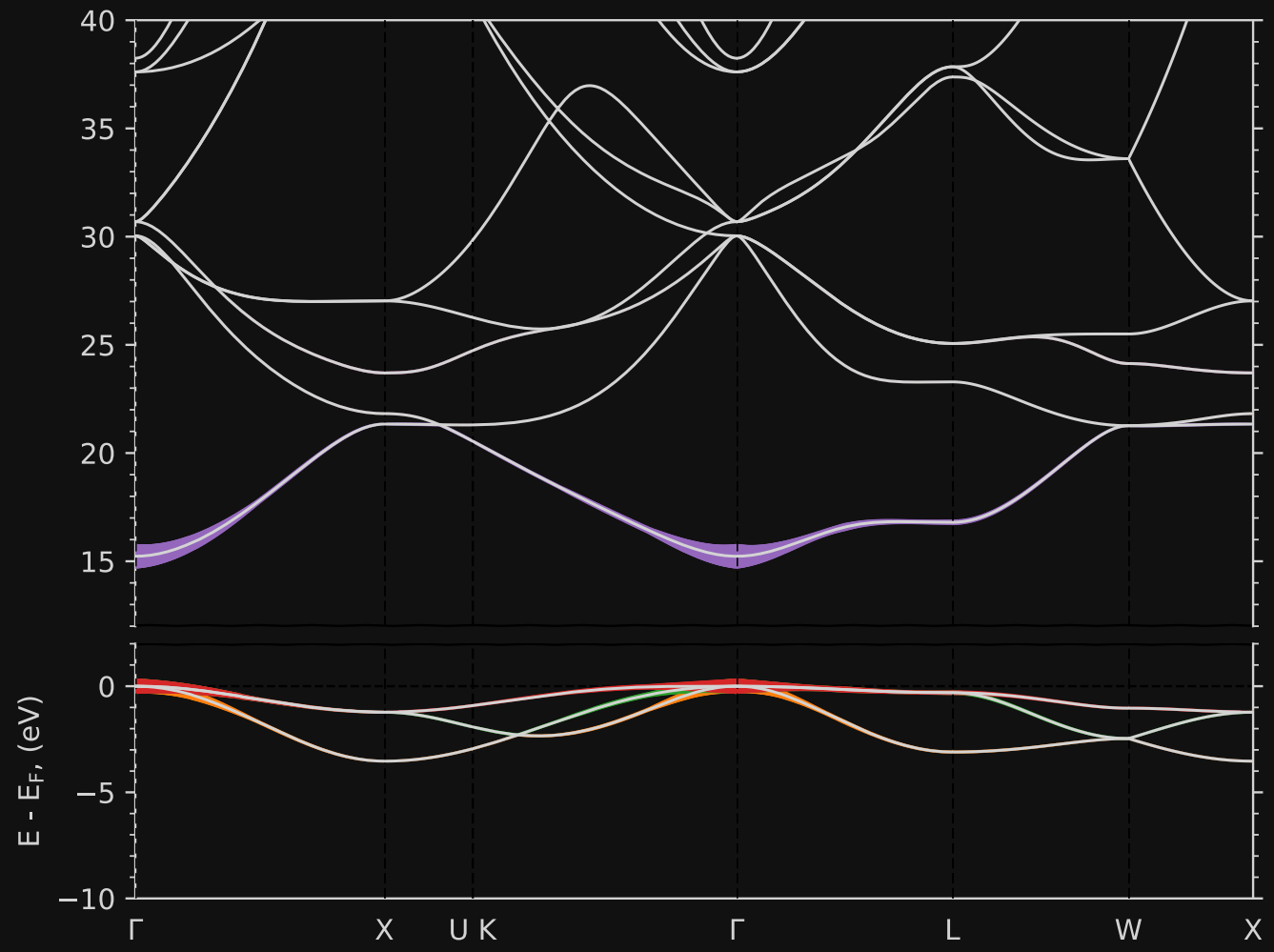
As a sidestep get the energy bands on which to plot the weights in the usual way:

```
$mn lmf --v8 ctrl.lm --quit=rho # this is to find Ef
$mn lmf --v8 ctrl.lm --band~fn=syml
```

now that all ingredients are in place, there is only plotting left:

```
band-plot syml.lm bnds.lm -e -10,2,12,40 --exbw exbw-12-13ev.h5 -o bands-exbw-12-13.pdf
```

Excitonic band weights / projection



There are two immediate observations in this case:

- while the occupied states have similar contributions, only the lowest unoccupied band seems to participate in a meaningful way
- the exciton is of Wannier type, localised in q, likely spread out in r (to be seen).

Three shells and a pea / here it is, here is it

We can see how (de)localised an exciton is in r/direct space by plotting Ψ(r_h, r_e) or its square modulus. Since covering the full (r_h, r_e) space for a reasonably dense r-mesh would require quite a bit of storage we fix one of the particles and let the other cover a supercell of size nk because this is the limit of the periodicity of the constituent single particle wavefunctions (Born-von Karman).

Two popular examples are to either fix the hole on the F atom or the electron on Li, however it is not always this obvious and there sometimes can be very many choices available.

Exciton atomic weights / projection

To narrow it down to a set of physically significant choices we can project to orbitals (calculate orbital weights) for selected exciton.

```
    $bsc --wexow~ewin=12,13eV~fn=exow-12-13ev.h5
```

```
isp: 0 ei: 1023 ev: 0.90059398ry (12.253205ev) oscillator strengths: 5.527 0.1699 0.1441 0.832 MKL_NUM_THRE
...
site class onsite:
     Li       F
    0.0193   0.0479
site class intersite:
v\c       Li⁻      F⁻
    Li⁺  0.2101   0.0909
    F⁺   0.4890   0.1430
... same stuff repeats from the other 2 degenerate states
averaged site distribution from selected excitons:
site class onsite:
    Li       F
   0.0192   0.0479
site class intersite:
v\c       Li⁻      F⁻
    Li⁺  0.2101   0.0909
    F⁺   0.4890   0.1429
```

It is now clear the largest weight is indeed on the $Li^{-}F^{+}$ pair and this supports the intuitive choice.

Exciton atomic weights / projection

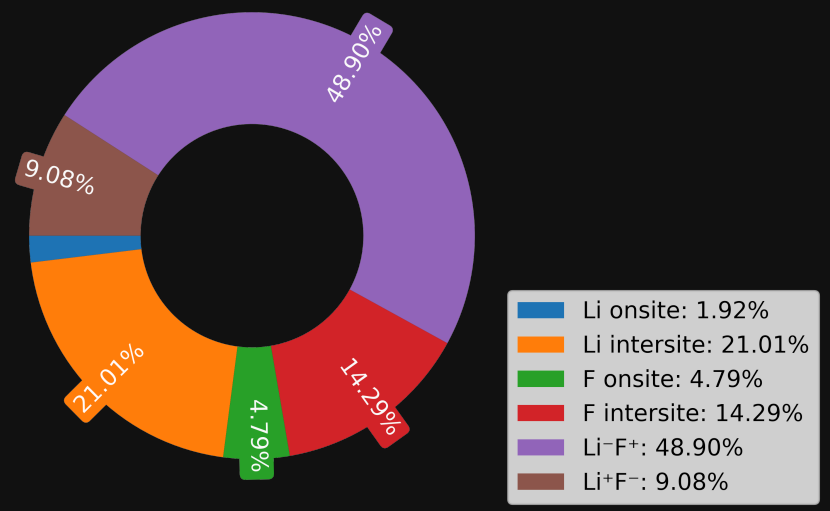
The projections can be visualised by a pie chart, line plot etc... one way is with:

```
exow-plot -s Li:0 -s F:1 exow-12-13.log
```

The output files are `exow-12-13.log.pdf` and `exow-12-13.log.png`

`-s` defines the specie name with its associated indexes in the matrix above in case there are multiple of the same due to disabled symmetry.

There are a few potentially useful options available, try `--help` for a description.



Excitonic probability density

To plot the 3d probability density for F^+ case fix the hole (r+) at cartesian coordinate [-0.5,0,0] (the F atom),

units of alat (see ctrl file for its value), swapping the c suffix with p switches to units of plat,

useful if your input is in the same units (using xpos instead of pos in ctrl file):

```
$bsc --wexpsi~ewin=12,13eV~r+=-0.5,0.0,0.0c~fn=xcfh-f-12-13ev.xsf | tee xcfh-f-12-13ev.log
```

for the Li^- case fix the electron (r-) at cartesian coortinate [0,0,0] (the Li atom):

```
$bsc --wexpsi~ewin=12,13eV~r-=0.0,0.0,0.0c~fn=xcfe-li-12-13ev.xsf | tee xcfe-li-12-13ev.log
# might want to increase gmax for better resolution 3d images
```

Snippet from the xcfe-li-12-13ev.log file:

```
integrating excitonic eigenstates over energy window [.8820, .9555] Ry
 ws  1021  wn     3  ngmx: 1892 gmax: 10.0 fmsh: 14 14 14
e- at: .0000 .0000 .0000 alat, .0000 .0000 .0000 plat,
snap to ftmesh point: .0000 .0000 .0000 plat, deviation: .0000 .0000 .0000 alat
r range: [ -2.0000 -2.0000 -2.0000] to [ 2.0000 2.0000 2.0000]
 vs 0 nv 4 cs 4 nc 4 nr: 175616
 isp: 0 ei: 1023 ev: 0.90048319ry (12.251698ev) oscillator strengths: 5.527 0.1697 0.1441
v\c
   0.7348   0.0004   0.0013   0.0001
   0.1944   0.0007   0.0009   0.0002
   0.0665   0.0005   0.0004   0.0000
   0.0000   0.0000   0.0000   0.0000
...
```

Note band projections table

Excitonic probability density, atom weighed

The resulting volume data is saved in the specified xsf files which can be opened with various visualisation tools

What may yield better results is offsetting the fixed point slightly away from the atom centre.

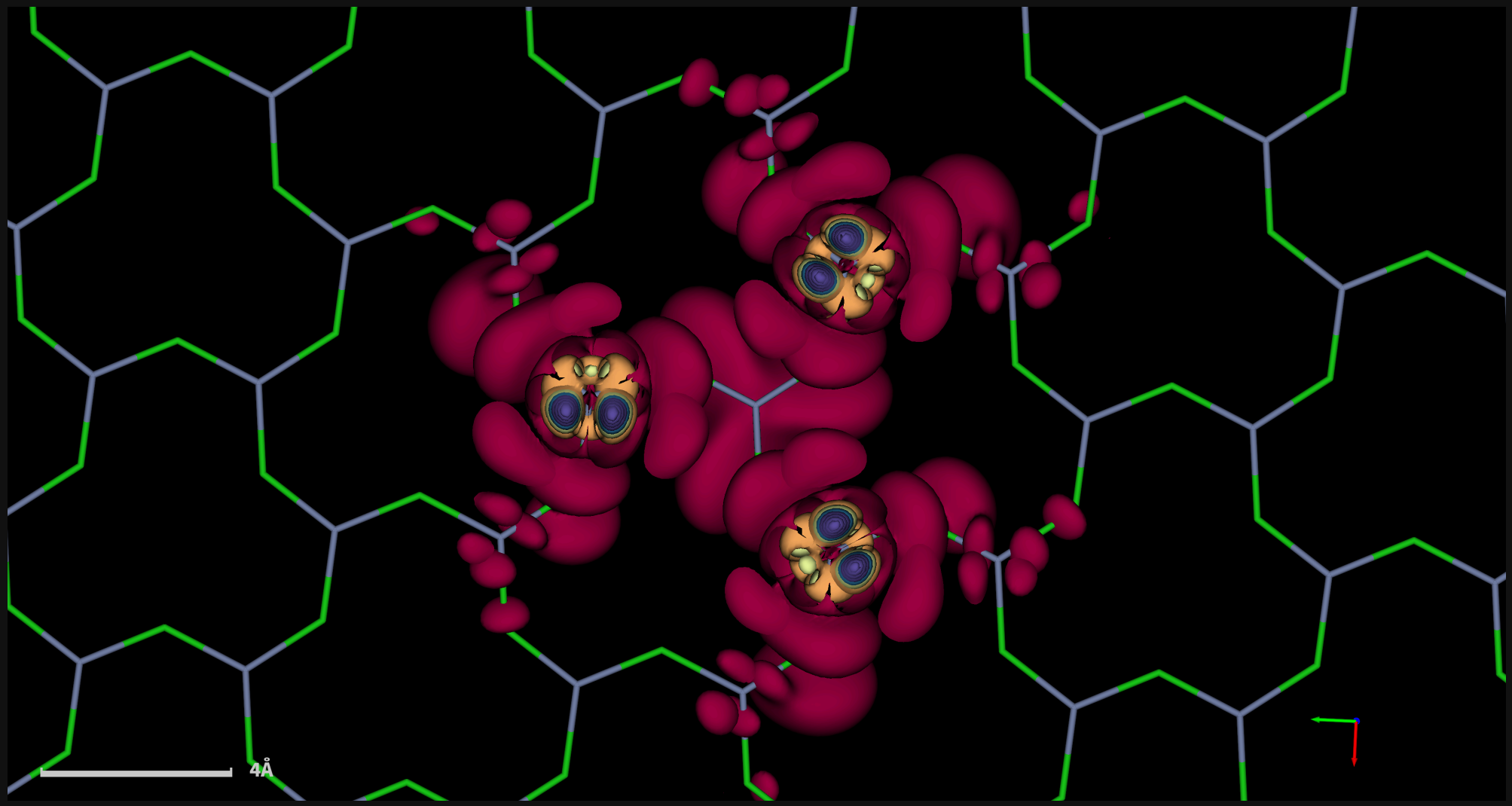
```
$bsc --wexpsi~ewin=12,13eV~r+=-0.5,0.0,0.1c~fn=xcfh-f-z0.1-12-13ev.h5
$bsc --wexpsi~ewin=12,13eV~r-=0.0,0.0,0.1c~fn=xcfe-li-z0.1-12-13ev.h5
```

Choosing an offset point is somewhat arbitrary, very different pictures can emerge depending on the direction of the point, also a point not on the ftmesh can yield complex projections. To spare some trouble and avoid ambiguity, one may wish to integrate all posisble r point weighted by a chosen set of atomic orbitals, for example the one associated with an atom. For the examples above this can be done with:

```
$bsc --wexrow~ewin=12,13eV~atom-=1~fn=exrow-f-e-12-13ev.h5
$bsc --wexrow~ewin=12,13eV~atom+=2~fn=exrow-li-h-12-13ev.h5
```

instead of ewin can use eidx to supply a list of excitation eigenstates add ~orth=1 to project on orthogonalised states.

Thanks for your attention!



Questions?