Practical BSE Calculations with BerkeleyGW

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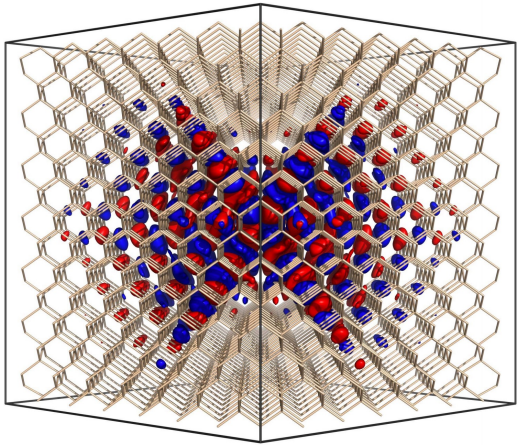
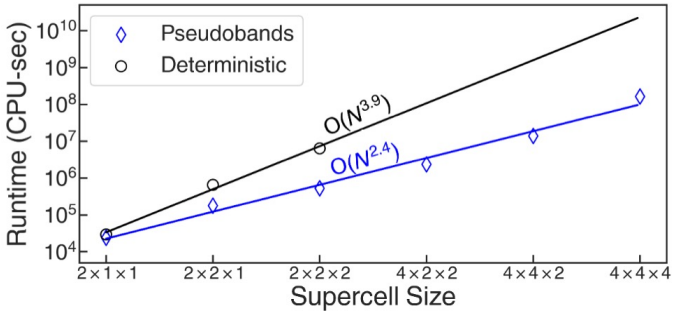
Why BerkeleyGW?

- **General** excited-state phenomena code, supporting:
  - GW & GW-BSE: 3D, 2D, 1D and molecular systems
  - Semiconducting, metallic and semi-metallic systems
  - Large set of DFT codes: PARATEC, Quantum Espresso, PARSEC, Octopus, ABINIT, RMGDFT, jDFTx
- **Massively parallel**, with cross-platform support:
  - Wide support for GPU hardware (NVIDIA, AMD & Intel) + many programming paradigm (MPI, OpenACC, OpenMP, Cuda kernels)
  - Scales linearly up to 100,000's of CPUs and 10,000's of GPUs.
- **Actively developed**, w/ unique algorithms + methods:
  - Efficient accurate solution to the BSE via k-point interpolation
  - Stochastic techniques: GW calc. speedup from $O(N^4)$ to $O(N^{2-3})$
- Example of **recent large-system** studies with BerkeleyGW:
  - Defects in Si and SiC (>2k atoms/cell), LiF (>15k atoms/cell)
  - Time to solution 270 secs at 50% peak performance (12 petaflops)

Divacancy defect in Si

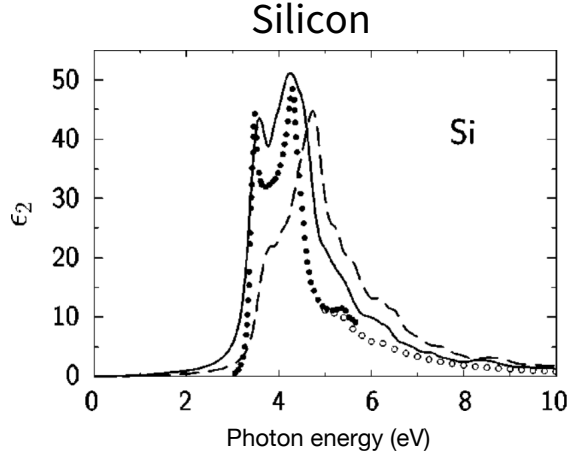


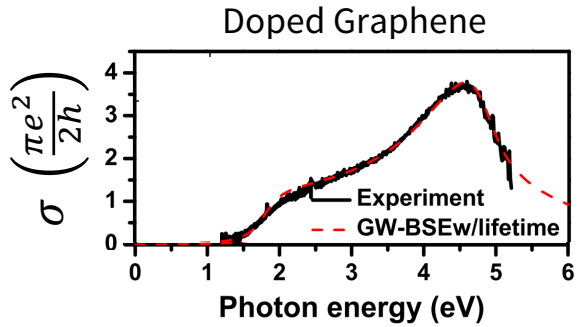
Del Ben, Yang, Li, Jornada, Louie, Deslippe, SC20 (2020).
Del Ben, Jornada, et al, CPC (2019).

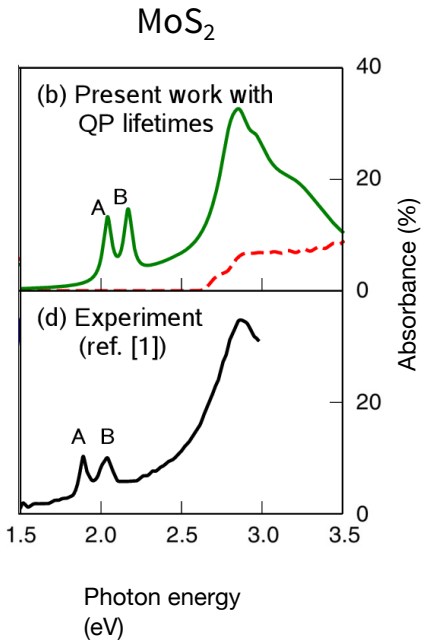


Altman, Kundu, Jornada, PRL 32, 086401 (2024).

Optical absorption spectrum







With eh interactions (GW-BSE)

No eh interactions (GW-RPA)

[1] M. Rohlfing, S. G. Louie, PRB 62, 8 (2000).
[2] D. Qiu, F. H. da Jornada, S. G. Louie, PRL 111, 216805 (2013).
[3] K. F. Mak, F. H. da Jornada, et al., PRL 112, 207401 (2014).

Practical BSE Calculations

- #1 – Theoretical and methodological overview
- #2 – Typical BSE workflow in BerkeleyGW
- #3 – Issues unique to the BSE code

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Bethe-Salpeter equation (BSE)

Challenge: compute quasiparticle corrections and kernel matrix elements on a VERY FINE K-GRID!
E.g.: 300x300 k-grid for MoS_2

- Absorption spectrum with excitonic effects → solve BSE

$$H|S\rangle = \Omega_S|S\rangle$$

- In an orbital basis set:

$$[H] = [E_c - E_v] + [K]$$

$[K]$ ← dense "kernel" ~ potential term

$[E_c - E_v]$ ← diagonal ~ kinetic term

BerkeleyGW Interpolation Scheme

- Expensive to compute kernel matrix elements $\langle vc\mathbf{k}_{\mathrm{fi}}|K|v'c'\mathbf{k}'_{\mathrm{fi}}\rangle$
- Strategy: $\langle vc\mathbf{k}_{\mathrm{co}}|K|v'c'\mathbf{k}'_{\mathrm{co}}\rangle \longrightarrow \langle vc\mathbf{k}_{\mathrm{fi}}|K|v'c'\mathbf{k}'_{\mathrm{fi}}\rangle$
- Linear interpolation? Wannier interpolation? K.p interpolation?

  No complex phases Hard to operate/unstable Unreliable
- BerkeleyGW: **band projection interpolation**
  - Explicitly generate coarse- and fine-grid WFNs
  - Expand of fine-grid WFNs in term of coarse-grid WFNs.
  - No need to perform Wannier interpolation, etc.
  - Captures band crossing, etc.

BerkeleyGW Interpolation Scheme

- Step 0: Obtain WFNs on a coarse and fine grid.

- Step 1: Expand fine WFNs in terms of coarse WFNs

$$\left|u_{n\mathbf{k}_{\mathrm{fi}}}\right\rangle = \sum_m C_{nm}^{\mathbf{k}_{\mathrm{co}}} \left|u_{m\mathbf{k}_{\mathrm{co}}}\right\rangle \qquad C_{nm}^{\mathbf{k}_{\mathrm{co}}} = \left\langle u_{m\mathbf{k}_{\mathrm{co}}} \middle| u_{n\mathbf{k}_{\mathrm{fi}}}\right\rangle$$

- Step 2: Interpolate QP energies (assume Σ is diagonal in $(n\mathbf{k})$):

$$E_n^{\mathrm{QP}}(\mathbf{k}_{\mathrm{fi}}) = E_n^{\mathrm{mf}}(\mathbf{k}_{\mathrm{fi}}) + \left\langle \sum_m \left|C_{nm}^{\mathbf{k}_{\mathrm{co}}}\right|^2 \left(E_m^{\mathrm{QP}}(\mathbf{k}_{\mathrm{co}}) - E_m^{\mathrm{mf}}(\mathbf{k}_{\mathrm{co}})\right)\right\rangle_{\mathbf{k}_{\mathrm{co}}}$$

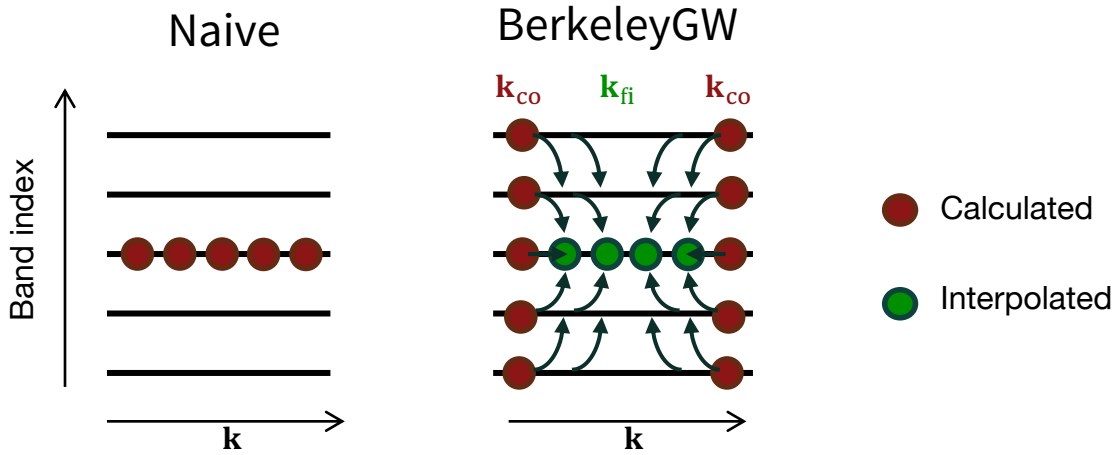
- Step 3: Interpolate BSE kernel matrix elements (head+wings+body)

$$\langle vc\mathbf{k}_{\mathrm{fi}}|K|v'c'\mathbf{k}'_{\mathrm{fi}}\rangle = \sum_{\substack{c_1\,c_2\\ v_1\,v_2}} C_{cc_1}^{\mathbf{k}_{\mathrm{co}}} C_{vv_1}^{*\mathbf{k}_{\mathrm{co}}} C_{c'c_2}^{*\mathbf{k}'_{\mathrm{co}}} C_{v'v_2}^{\mathbf{k}'_{\mathrm{co}}} \times \langle v_1 c_1 \mathbf{k}_{\mathrm{co}}|K|v_2 c_2 \mathbf{k}'_{\mathrm{co}}\rangle$$

BerkeleyGW Interpolation Scheme

$$\langle vc\mathbf{k}_{\mathrm{fi}}|K|v'c'\mathbf{k}'_{\mathrm{fi}}\rangle = \sum_{\substack{c_1\,c_2\\ v_1\,v_2}} C_{cc_1}^{\mathbf{k}_{\mathrm{co}}} C_{vv_1}^{*\mathbf{k}_{\mathrm{co}}} C_{c'c_2}^{*\mathbf{k}'_{\mathrm{co}}} C_{v'v_2}^{\mathbf{k}'_{\mathrm{co}}} \times \langle v_1 c_1 \mathbf{k}_{\mathrm{co}}|K|v_2 c_2 \mathbf{k}'_{\mathrm{co}}\rangle$$

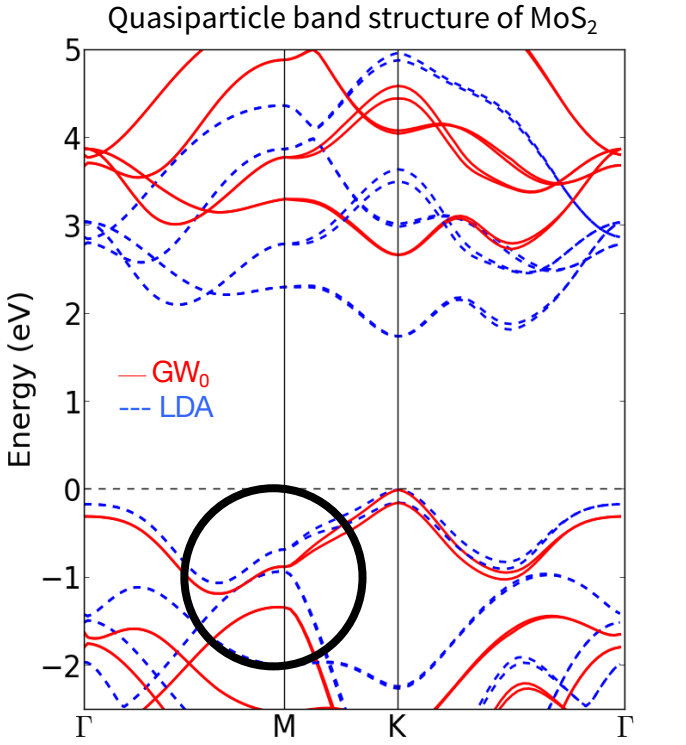
- In practice: trading bands for k-points



- How to get a good interpolation?
  - Include a large number of bands from the coarse grid and start from a coarse grid that is not too coarse.

BerkeleyGW QP Interpolation

- BerkeleyGW also performs a linear interpolation for QP corrections.
- Linear interpolation + expansion over bands:
  - Captures (n**k**)-dependent QP correction and band crossing
  - Smooth interpolation of band structure, few parameters
  - Works with arbitrary non-uniform k-point sampling
- This is how **`inteqp.x`** works!

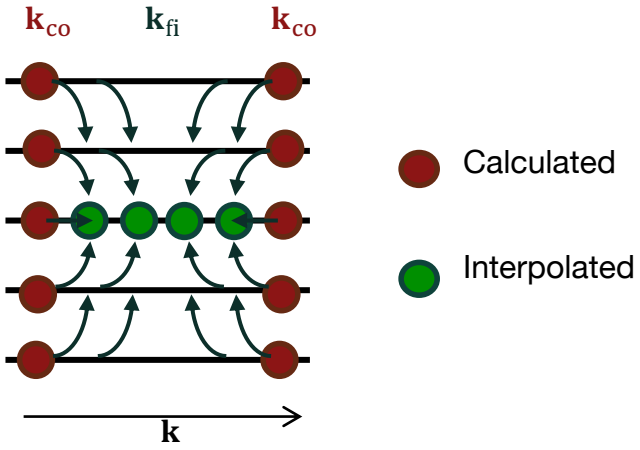


D. Qiu, F. H. da Jornada, S. G. Louie, PRL **111**, 216805 (2013)

BerkeleyGW Interpolation Scheme

- Did I include enough bands?

$$\mathrm{Error}(\mathrm{n}, \mathbf{k}_{\mathrm{fi}}) = 1 - \sum_{m} \left| C_{n,m}^{\mathbf{k}_{\mathrm{fi}}} \right|^2$$



- Detail of “completion relation”:
  - `dvmat_norm.dat`
  - `dcmat_norm.dat`

- `absorption.out / inteqp.out`

Practical BSE Calculations

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BerkeleyGW Workflow

Step 0: Calculate QP–corrected band structure on a coarse grid

`epsmat.h5`, $\{E_c^{\mathrm{QP}}\}_{\mathrm{co}}$, $\{E_v^{\mathrm{QP}}\}_{\mathrm{co}}$

`kernel.x`

Step 1: Calculate BSE kernel on the same coarse grid

$[K]_{\mathrm{co}}$

`absorption.x`

Step 2: Interpolate to a fine k-grid and build BSE Hamiltonian…

$$[H]_{\mathrm{co}} \Rightarrow [H]_{\mathrm{fi}}$$

… and diagonalize BSE Hamiltonian

$$\mathrm{evals}\ [H]_{\mathrm{fi}} \Rightarrow \varepsilon_2$$

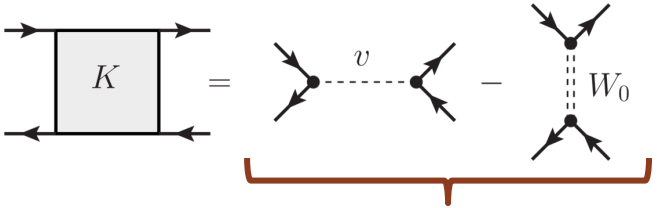
1. Kernel

`kernel.x`

Step 1: Calculate BSE kernel on the same coarse grid

$$[K]_{\mathrm{co}}$$

- Time consuming: Computes $(n_v n_c n_k)^2$ matrix elements
- Uses a coarse-grid WFN_co



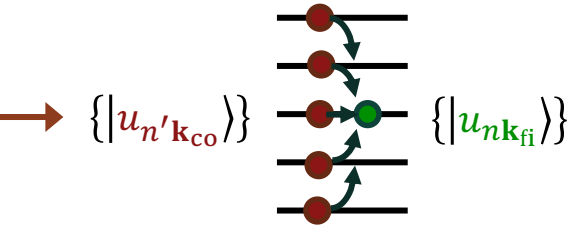
- Recommended:
  - Use same WFN_co as in Sigma (WFN_inner)

bsemat.h5

1. Kernel

Sample **kernel.inp**

```
number_val_bands <?>
number_cond_bands <?>

<?>_symmetries_coarse_grid
screening_<?>
```



Bands counted wrt FE:

- vbm, vbm-1, …
- cbm, cbm+1, …

Remember to calculate Kernel on more bands because of the interpolation!

(# of bands in Sigma can't be less than this number!)

You'll typically want to use symmetries here, so put: `use_symmetries_coarse_grid`

2. Absorption

`absorption.x`

Step 2: Interpolate to a fine k-grid and build BSE Hamiltonian…

$$[H]_{\text{co}} \Rightarrow [H]_{\text{fi}}$$

… and diagonalize BSE Hamiltonian

$$\text{evals } [H]_{\text{fi}} \Rightarrow \varepsilon_2$$

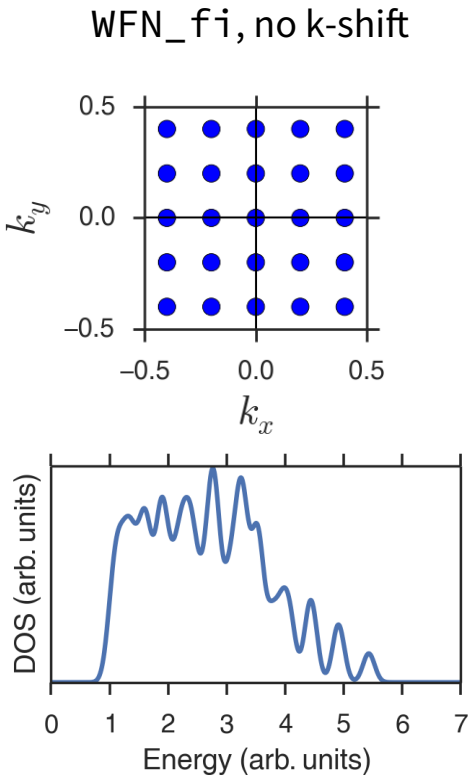
- Compute velocity matrix elements $\hat{v}$ with finite differences (*i.e.*, k.p) on two fine grids

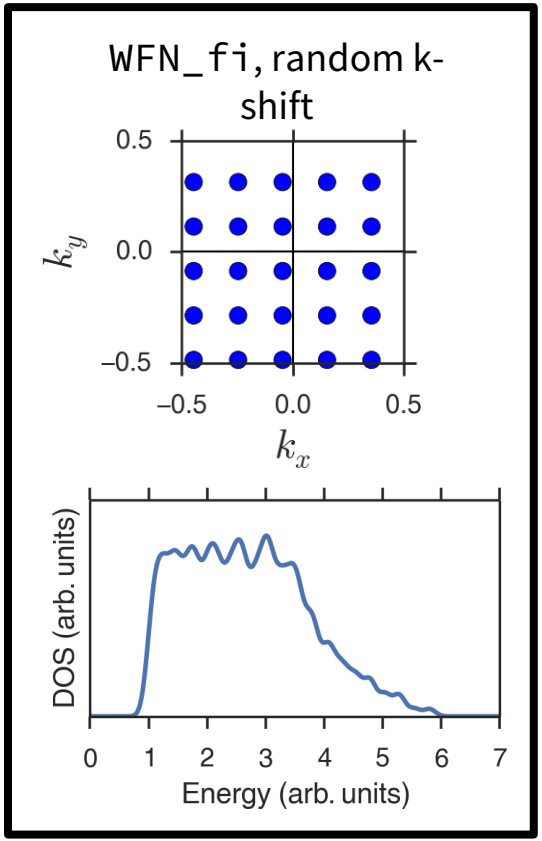
$$\varepsilon_2(\omega) \propto \sum_S |\langle 0|\hat{v}|S\rangle|^2 \delta[\omega - \Omega_S] \qquad \langle 0|\hat{v}|S\rangle = \frac{\Omega_S}{q} \sum_{vc\mathbf{k}} A^S_{vc\mathbf{k}} \langle u_{v\mathbf{k}+\mathbf{q}}|u_{c\mathbf{k}}\rangle$$

- We need two fine WFN files:
  - `WFN_fi`: for conduction states $|u_{c\mathbf{k}}\rangle$
  - `WFNq_fi`: for q-shifted valence states $|u_{v\mathbf{k}+\mathbf{q}}\rangle$

Direction of **q** = polarization of light
= difference between
$\{\mathbf{k}'\}$ in `WFNq` and $\{\mathbf{k}\}$ in `WFN`

2. Absorption – Randomly Shifted K-grids





2. Absorption

Sample **absorption.inp**

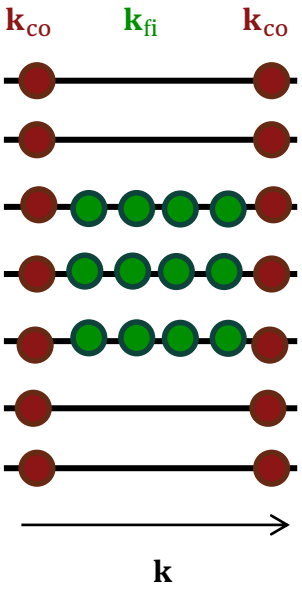
```
diagonalization

number_val_bands_coarse <?>
number_cond_bands_coarse <?>
number_val_bands_fine <?>
number_cond_bands_fine <?>

use_symmetries_coarse_grid
no_symmetries_fine_grid
no_symmetries_shifted_grid

screening_semiconductor

use_velocity

gaussian_broadening
energy_resolution 0.15

eqp_co_corrections
```

Unshifted grid (WFN_co)

Both randomly shifted grids (WFN_fi and WFNq_fi)

Broaden each delta function.

Interpolate eqp_co.dat



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Issues Unique to the BSE Code

1. Convergence
2. Analyzing results
3. Other features

1. Convergence

- There are 4 convergence parameters in a typical BSE calculation:
  - # of k-points in the fine grid ←
  - # of bands in the fine grid
  - # of k-points in the coarse grid ←
  - # of bands in the coarse grid

- Some rules of thumb:
  - # of bands in fine grid: energy window + binding energy (~ EASY)
  - # of bands in the coarse grid: estimate from **`inteqp.x`**
  - # of k-points in coarse grid ~ **`sigma.x`** (FOR BULK SYSTEMS ONLY)

2. Analyzing Excitons

- Optical spectrum $\varepsilon_2(\omega), \varepsilon_1(\omega)$:
  - **absorption_noeh.dat**: GW-RPA
  - **absorption_eh.dat**: GW-BSE

- Eigenvalues of the BSE equation Ω_S:
  - **eigenvalues.dat**: useful to see if there are degeneracies, splitting, etc.

- Where the exciton is coming from:
  - **summarize_eigenvectors.x**
  - Need to set the flag **write_eigenvectors** in **absorption.inp**

3. Other Features

- Some other features:
  - Haydock method: iterative solution for the absorption spectrum. No evecs can be obtained.
  - Unrestricted interpolation: improves the quality of the interpolation by allowing mixtures of conduction and valence states. Important for metals!
  - Momentum operator: allows you not to use WFNq_fi file (but neglects non-local part of PP and Σ)

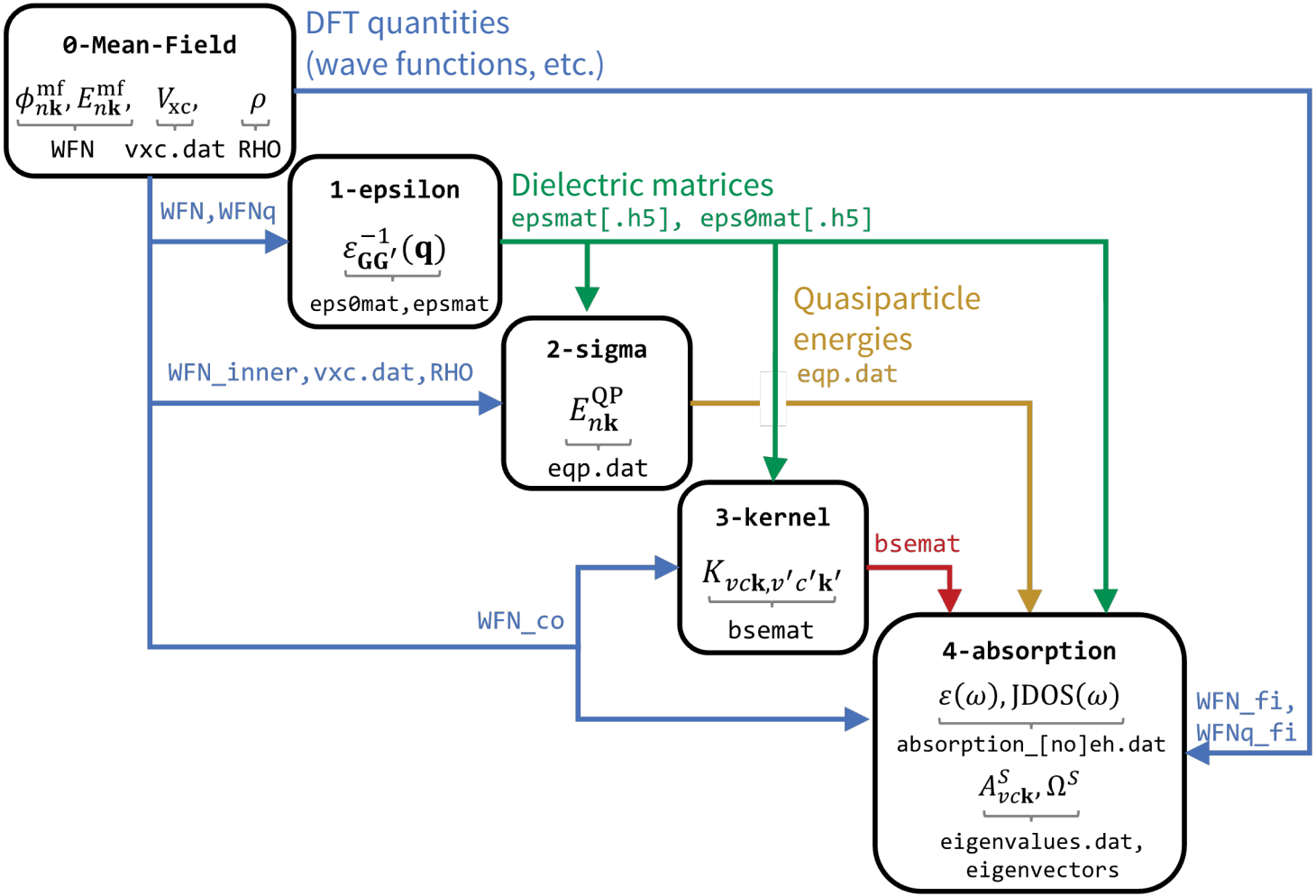
- Approximations:
  - Tamm-Dancoff approximation (not required)
  - Static screening

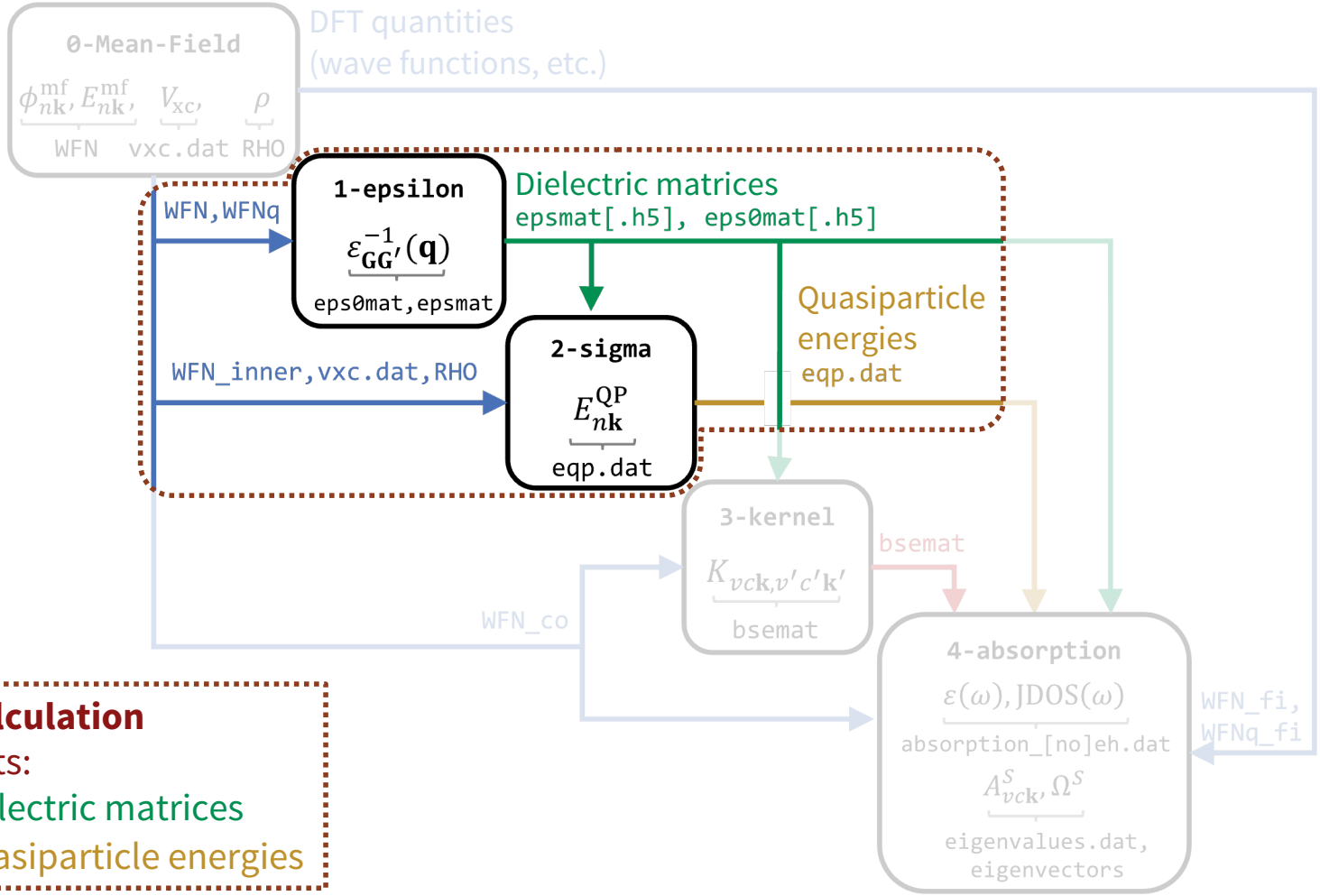
Wrapping Up

- BSE needs to be solved on fine k-grid
- BGW interpolation : projection of fine WFNs onto coarse WFNs
  - Need to *include more bands* in kernel calculation

- WFNs:
  - Kernel: WFN_co
  - Absorption: WFN_co, WFN_fi, WFNq_fi
  - WFN_co: unshifted
  - WFN_fi: random k-shift
  - WFNq_fi: random k-shift + q-shift (dir. = pol. of light)

- 4 convergence parameters: {bands, **kpts**} x {co, **fi**}

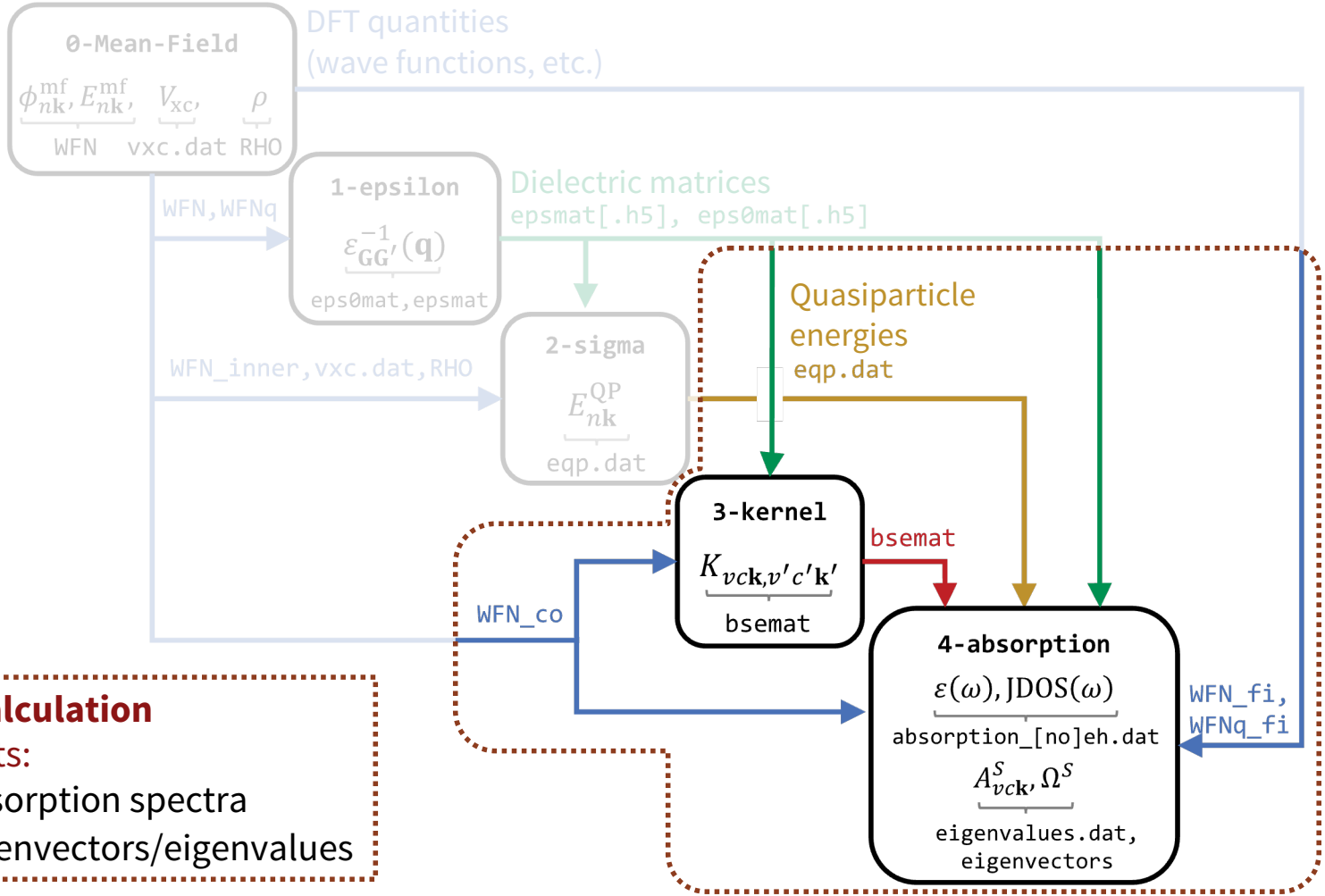

0-Mean-Field
$\phi^{mf}_{n\mathbf{k}}, E^{mf}_{n\mathbf{k}}$, V_{xc}, ρ
WFN vxc.dat RHO
DFT quantities
(wave functions, etc.)
WFN,WFNq
1-epsilon
$\varepsilon^{-1}_{\mathbf{GG'}}(\mathbf{q})$
eps0mat,epsmat
Dielectric matrices
epsmat[.h5], eps0mat[.h5]
Quasiparticle
energies
eqp.dat
WFN_inner,vxc.dat,RHO
2-sigma
$E^{QP}_{n\mathbf{k}}$
eqp.dat
3-kernel
$K_{vc\mathbf{k},v'c'\mathbf{k}'}$
bsemat
bsemat
WFN_co
4-absorption
$\varepsilon(\omega)$, JDOS(ω)
absorption_[no]eh.dat
$A^S_{vc\mathbf{k}}, \Omega^S$
eigenvalues.dat,
eigenvectors
WFN_fi,
WFNq_fi



GW calculation

Outputs:

- Dielectric matrices
- Quasiparticle energies



BSE calculation

Outputs:

- Absorption spectra
- Eigenvectors/eigenvalues

And now… the hands-on session!

BerkeleyGW

EXTRA SLIDES

BerkeleyGW Interpolation Scheme

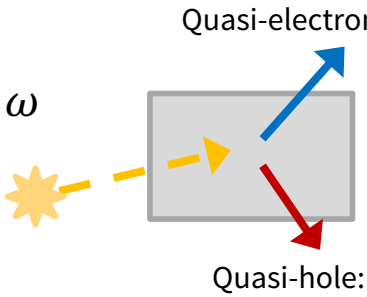
- Note: a direct interpolation of K is not very accurate:
  - Explicit dependence on $\mathbf{q}_{\text{fi}} = \mathbf{k}_{\text{fi}} - \mathbf{k}'_{\text{fi}} \neq \mathbf{q}_{\text{co}} = \mathbf{k}_{\text{co}} - \mathbf{k}'_{\text{co}}$
- We decompose the kernel matrix elements into components having different analytical behavior wrt $\mathbf{k} - \mathbf{k}'$:

$$\langle vc\mathbf{k}|K|v'c'\mathbf{k}'\rangle = \frac{a_{vc\mathbf{k},v'c'\mathbf{k}'}}{A(\mathbf{k}'-\mathbf{k})} + \frac{b_{vc\mathbf{k},v'c'\mathbf{k}'}}{B(\mathbf{k}'-\mathbf{k})} + \frac{c_{vc\mathbf{k},v'c'\mathbf{k}'}}{C(\mathbf{k}'-\mathbf{k})}$$

- We interpolate each component individually.
- Ex: for 3D semiconductor:
  - $A(\mathbf{q}) = q^2, \quad B(\mathbf{q}) = q, \quad C(\mathbf{q}) = 1$
- Important flags: **`screening_*`** and **`*truncation`**

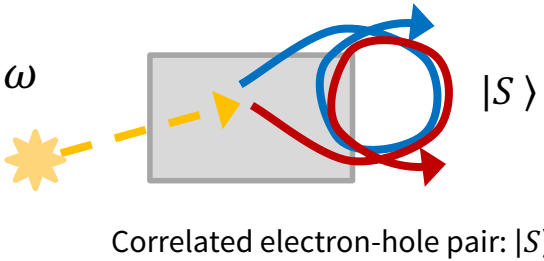
Theory Review: Optical Absorption

No electron-hole interactions (indep. part)



$$\varepsilon_2(\omega) \propto \sum_{vc\mathbf{k}} |\langle v\mathbf{k}|\hat{v}|c\mathbf{k}\rangle|^2 \delta[\omega - (E_{c\mathbf{k}} - E_{v\mathbf{k}})]$$

With electron-hole interactions (BSE)



$$\varepsilon_2(\omega) \propto \sum_{S} |\langle 0|\hat{v}|S\rangle|^2 \delta[\omega - \Omega_S]$$

$$|S\rangle = \sum_{vc\mathbf{k}} A^{S}_{vc\mathbf{k}} \, |v\mathbf{k}\rangle \otimes |c\mathbf{k}\rangle$$

BerkeleyGW Interpolation Scheme

- `dcmat_norm.dat` from yesterday's session:

```
 -------  Norm of dcc matrices : Spins =           1  -------
          k-point           ik_co    c   dist    |dcc|^2
-------------------------------------------------------------
 (  0.500 ,  0.500 ,  0.500 )   13    1  0.000   1.000000
[...]
 (  0.367 ,  0.000 ,  0.367 )   18    9  0.144   0.878516
 (  0.367 ,  0.000 ,  0.367 )   18   10  0.144   0.000000
 (  0.378 ,  0.000 ,  0.378 )   58    1  0.150   0.988656
```

- Band crossing. Look at **`inteqp.inp`**:

```
number_val_bands_coarse 4
number_val_bands_fine 4
number_cond_bands_coarse 10
number_cond_bands_fine 10
```

- #fi bands = # co bands! This is a bad idea is general!!
- But we didn't plot the 10th cond. band, so it's alright ☺

BerkeleyGW Interpolation Scheme

- **`inteqp.out`:**

```
 Max. error in norm of transformation coefficients (1 - \sum_co |d_fi,co|^2):
- For valence states: 2.887E-01
- For conduction states: 1.000E+00

WARNING: there are fine/coarse transformation coefficients with 2-norm < 95%.
To improve convergence, you might want to consider:
- using the "unrestricted_transformation" flag in the input file
- including more bands from the coarse WFN_co file (number_*_bands_coarse)
- using a coarse WFN_co file with a denser k-mesh
```

- Error = 100% for conduction states?!
- Where is it coming from?