2023 Virtual School on Many-Body Calculations using EPW and BerkeleyGW

June 5-9 2023

GW and BSE Calculations With BerkeleyGW

Jack Deslippe
NERSC

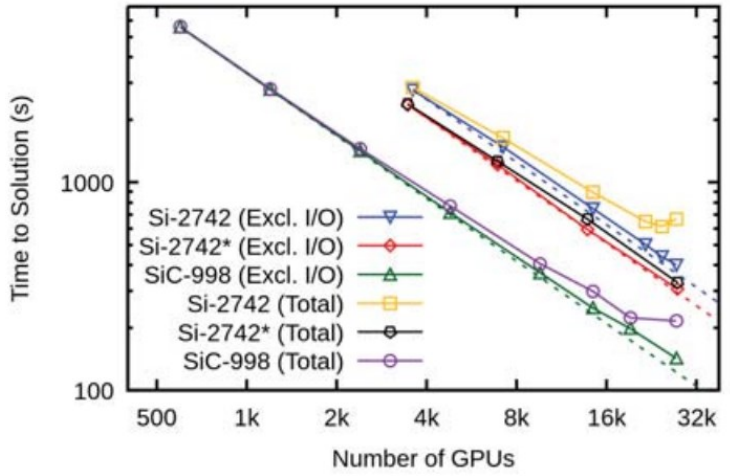
Felipe H. da Jornada
Stanford University

Diana Qiu
Yale

Why BerkeleyGW?

- **Versatile**: supports **3D**, **2D**, **1D** and molecular systems, with **Coulomb truncation** and **efficient k-point sampling** algorithms.
- **Agnostic**: supports a **large set of mean-field codes**: Quantum ESPRESSO, ABINIT, PARATEC, Octopus, PARSEC, SIESTA, JDFTx, RMGDFT, EPM, INQ (to be released).
- **General**: supports **semiconductor**, **metallic** and **semi-metallic** systems.
- **Massively parallel**: scales to **512,000 CPU cores**, supports distributed memory and hybrid architectures. High-performance GPU support (NVIDIA, AMD, Intel). Can handle large systems containing several **thousands of atoms**.
- **Free & open source.**

Del Ben, Yang, Li, da Jornada, Louie, Deslippe, SC '20 4, 1 (2020), ACM Gordon-Bell Finalist



~10 mins for 11k electrons
53% of peak performance!

Outline

1. **Introduction**
2. K-point vs. Q-point grids
3. Screening models for ε(**q**)
4. Frequency dependence of ε(ω)
5. Symmetry and degeneracy
6. Solving Dyson's equation
7. BSE

1. Introduction: GW calculations – Theory

Electronic self-energy Σ within the GW approximation: 2

$$\Sigma(\mathbf{r},\mathbf{r}';t) \approx iG_0(\mathbf{r},\mathbf{r}';t)W_0(\mathbf{r},\mathbf{r}';t)$$

Screened Coulomb interaction W:

$$W^0_{\mathbf{GG}'}(\mathbf{q};\omega) \equiv \varepsilon^{-1}_{\mathbf{GG}'}(\mathbf{q};\omega)v(\mathbf{q}+\mathbf{G}')$$

Sum over wavevectors **q**!

RPA dielectric matrix: 1

$$\varepsilon_{\mathbf{GG}'}(\mathbf{q},\omega) = \delta_{\mathbf{GG}'} - v(\mathbf{q}+\mathbf{G})\chi^0_{\mathbf{GG}'}(\mathbf{q},\omega)$$

Noninteracting polarizability matrix:

$$\chi^0(\mathbf{r},\mathbf{r}';t) = G_0(\mathbf{r},\mathbf{r}';t)G_0(\mathbf{r}',\mathbf{r};-t)$$

Noninteracting Green's function (spectral representation) 0

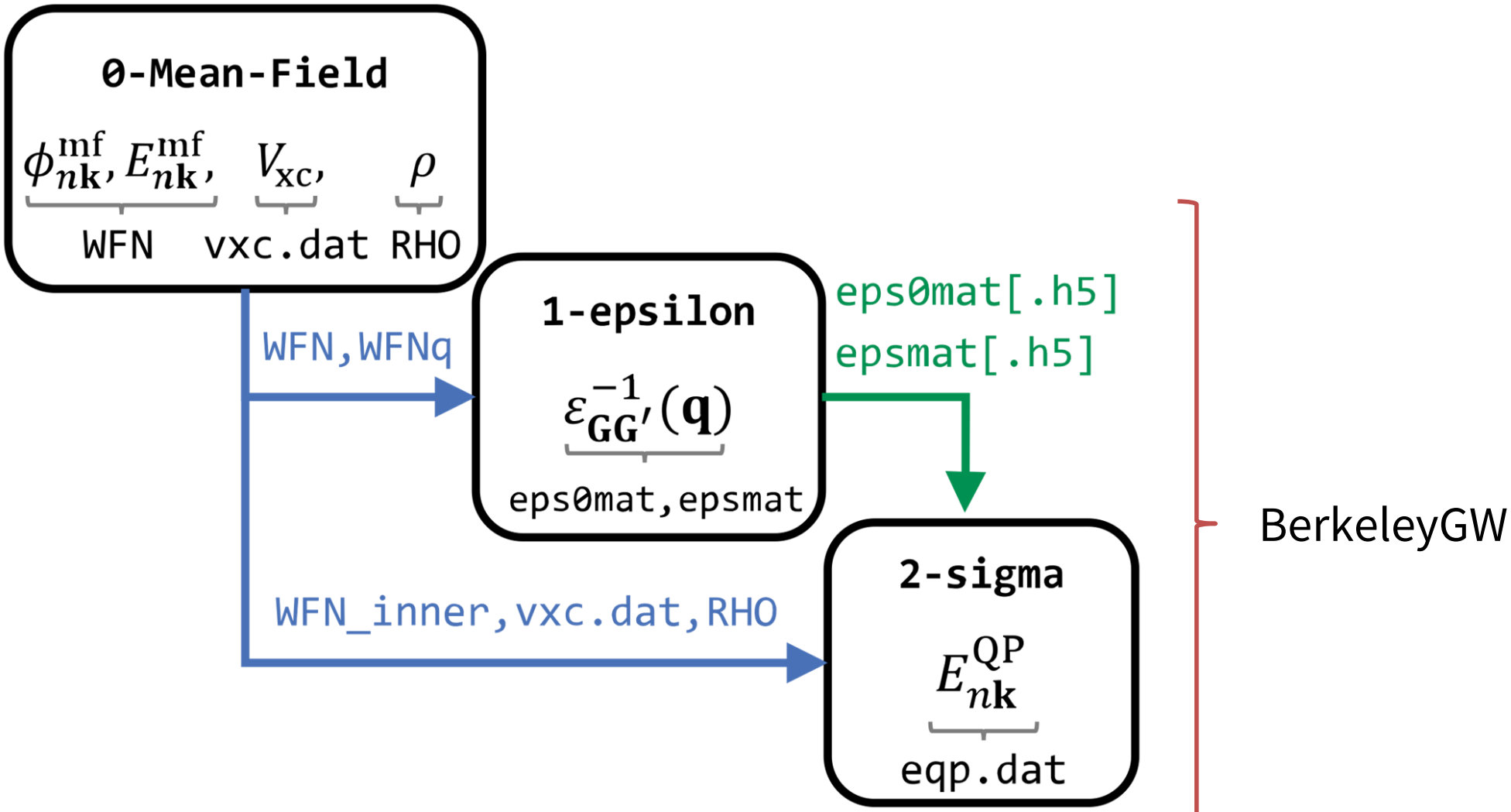
$$G_0(\mathbf{r},\mathbf{r}';\omega) = \sum_{n\mathbf{k}} \frac{\phi_{n\mathbf{k}}(\mathbf{r})\phi^*_{n\mathbf{k}}(\mathbf{r}')}{\omega - E^{\mathrm{mf}}_{n\mathbf{k}} \pm i\eta}$$

Sum over all occupied and unoccupied states *n*!

1. Introduction: GW calculations – Practice

DFT codes:

- Quantum Espresso
- Abinit
- Paratec
- Octopus
- Parsec
- RMGDFT
- JDFTx
- Parabands



Outline

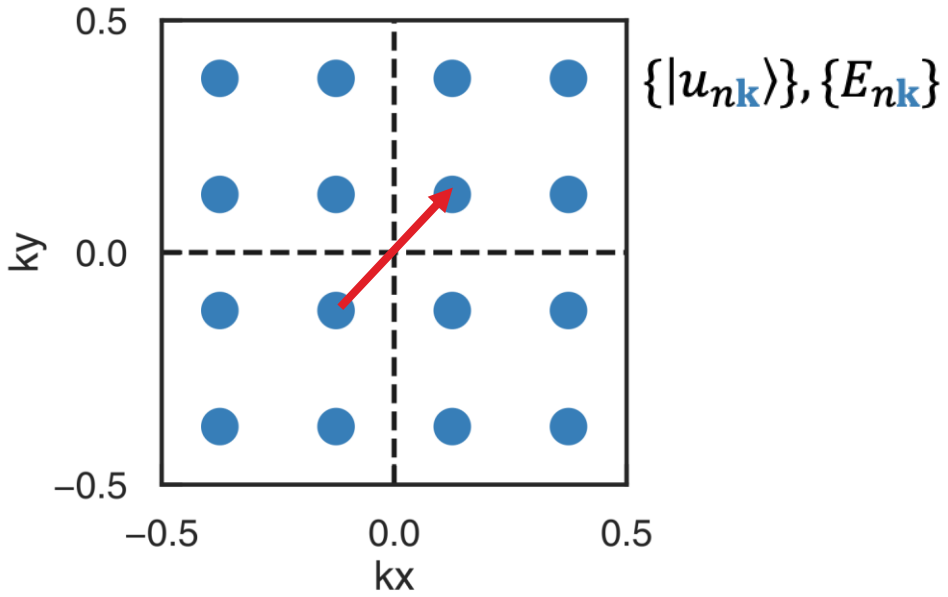
1. Introduction
2. **K-point vs. Q-point grids**
3. Screening models for ε(**q**)
4. Frequency dependence of ε(ω)
5. Symmetry and degeneracy
6. Solving Dyson's equation
7. BSE

2. k-grids and q-grids

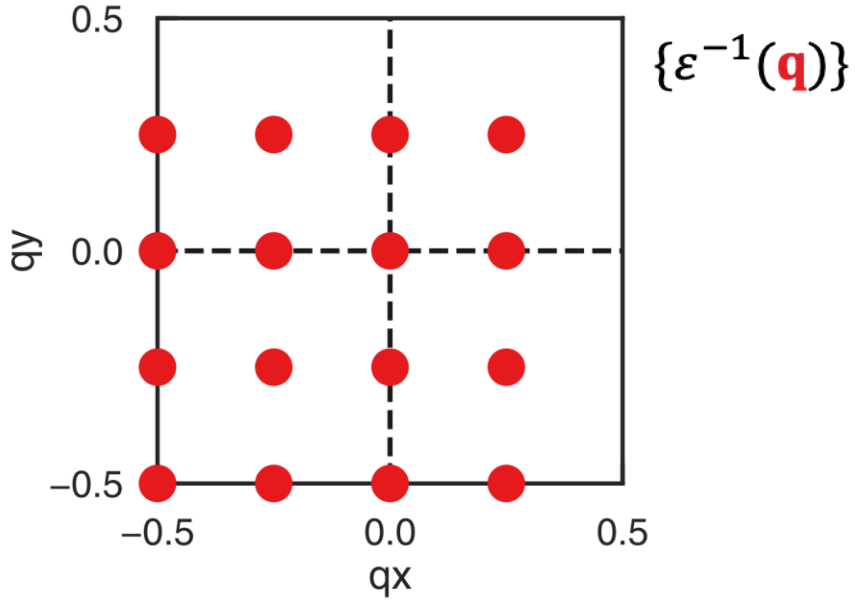
Non-interacting polarizability:

$$\chi^0_{\mathbf{00}}(\mathbf{q}, \omega = 0) \sim \sum_{v\,c\,\mathbf{k}} \frac{\left|\langle u_{v\mathbf{k}+\mathbf{q}} | u_{c\mathbf{k}} \rangle\right|^2}{E_{v\mathbf{k}+\mathbf{q}} - E_{c\mathbf{k}}}$$

- Mean-field quantities: computed in any regular k-point grid (does not need to be Γ centered)



- Polarizability & dielectric matrices: computed in a regular, Γ-centered q-point grid



q-point grid: regular, Γ-centered grid obtained from all possible transfer vectors $\mathbf{q} = \mathbf{k}' - \mathbf{k}$

Getting started: use the **same Γ-centered grid** for both k and q points (esp. bulk systems)

2. **k**-grids and **q**-grids: **q**=0 point

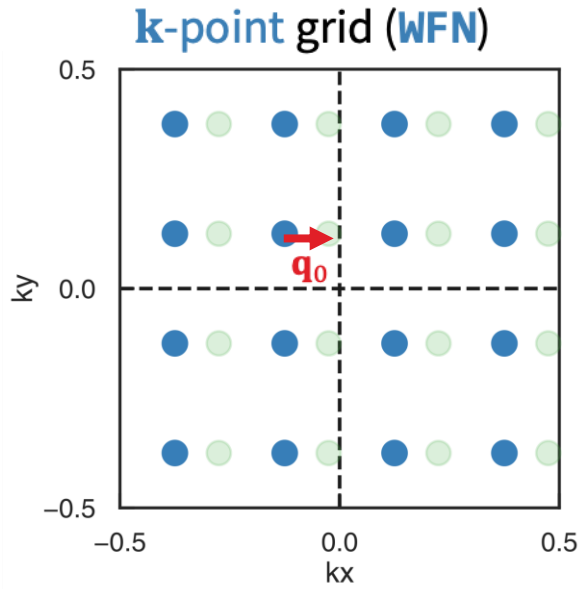
Dielectric matrix:

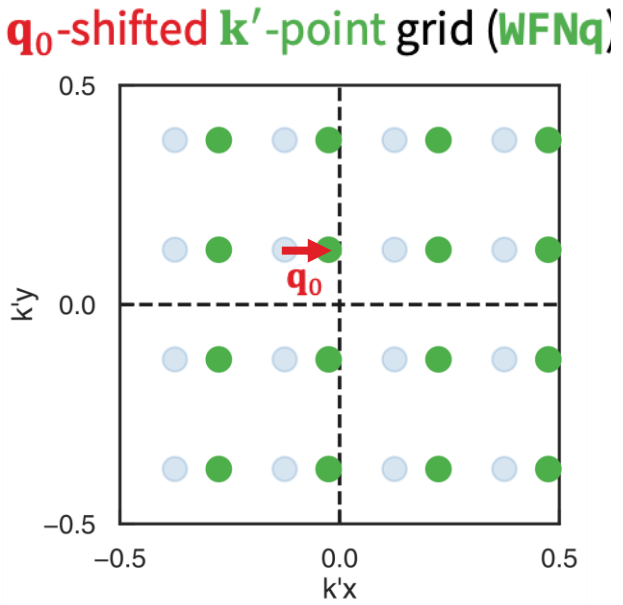
$$\varepsilon_{\mathbf{00}}(\mathbf{q}, \omega = 0) = 1 - \frac{4\pi e^2}{|\mathbf{q}|^2} \chi^0_{\mathbf{00}}(\mathbf{q}, 0)$$

- Cannot directly compute $\varepsilon(\mathbf{q} = \mathbf{0})$!

- For gapped systems:
  - Compute $\varepsilon(\mathbf{q_0})$ at a small but finite $\mathbf{q_0} \sim 0.001$.
  - Use two sets of **k**-point grids and wave functions for valence/conduction states.

- **WFN**: provides **conduction states** for $\varepsilon(\mathbf{q} = \mathbf{q_0})$ + **all states** for $\mathbf{q} \neq \mathbf{q_0}$.
- **WFNq**: provides **valence states** for $\varepsilon(\mathbf{q} = \mathbf{q_0})$.

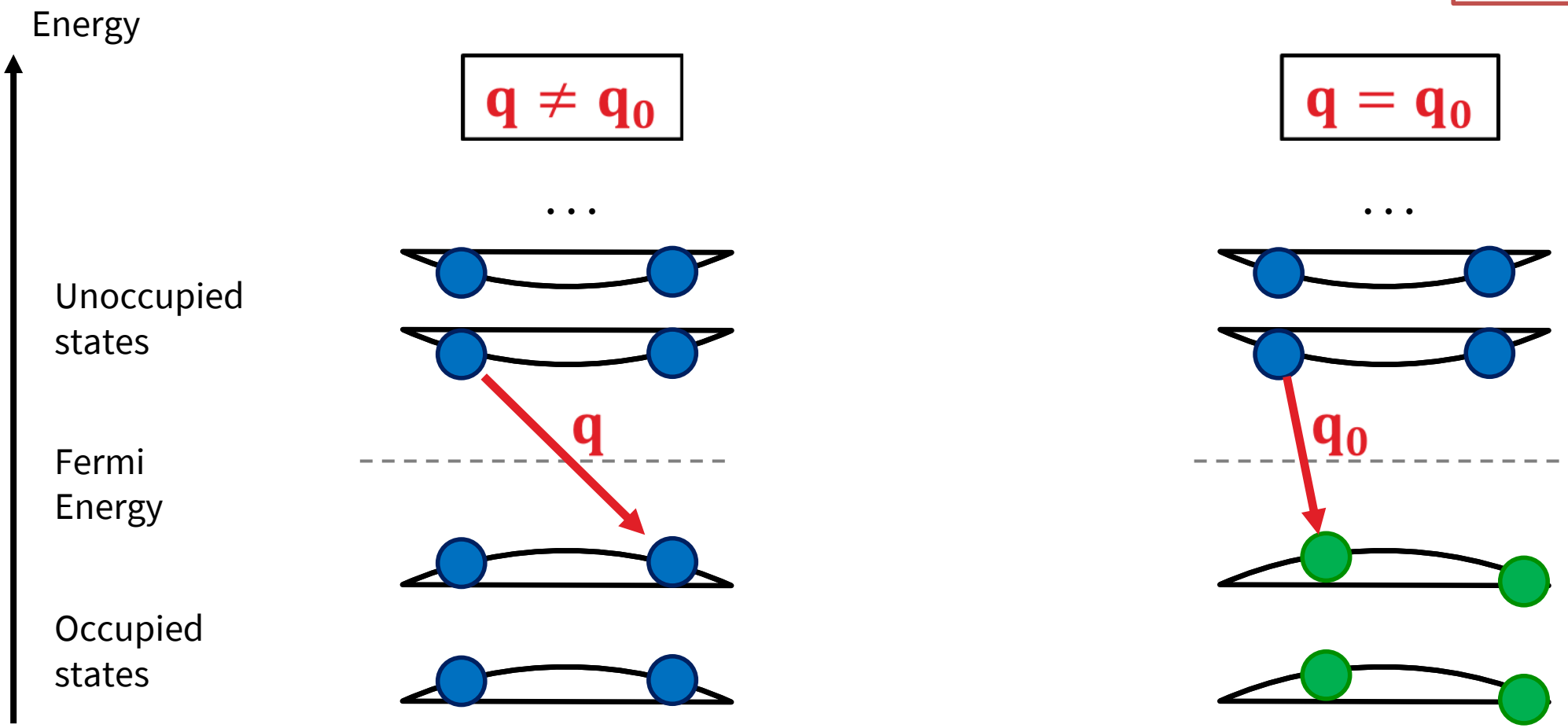




2. **k**-grids and **q**-grids

$$\chi^0_{\mathbf{00}}(\mathbf{q}, \omega = 0) \sim \sum_{v\,c\,\mathbf{k}} \frac{\left|\langle u_{v\mathbf{k}+\mathbf{q}} | u_{c\mathbf{k}} \rangle\right|^2}{E_{v\mathbf{k}+\mathbf{q}} - E_{c\mathbf{k}}}$$

Q-point shifted wavefunctions only work for semiconductors!



Files:
- WFN
- WFNq

2. Specification of **q**-points in epsilon.inp

❖ Semiconductors (**epsilon.inp**)

```
begin qpoints
  0.000000    0.000000    0.001000   1.0   1
  0.000000    0.000000    0.062500   1.0   0
  0.000000    0.000000    0.125000   1.0   0
  0.000000    0.000000    0.187500   1.0   0
  ...
end
```

eps0mat.h5: $\epsilon^{-1}_{\mathbf{G}\mathbf{G}'}(\mathbf{q}_0)$

epsmat.h5: $\epsilon^{-1}_{\mathbf{G}\mathbf{G}'}(\mathbf{q} \neq \mathbf{q}_0)$

❖ Metals

- Screening depends critically on sampling DOS at Fermi surface for intraband transitions. **Cannot use shifted grid**!

❖ Sigma, Kernel, Absorption use **q**-grid defined by eps0mat.h5 / epsmat.h5

2. Sample output from the epsilon code (`epsilon.out`)

```
==============================================================================
 19:13:53   Dealing with q =  0.000000  0.000000  0.001000                1 / 8
==============================================================================

 This is the special q->0 point.
 Rank of the polarizability matrix (nmtx): 137

 BLACS processor grid:   4 x   8; BLOCKSIZE =   17

 Number of k-points in the irreducible BZ(q) (nrk): 20

 ...

 q-pt      1: Head of Epsilon          =    2.5499723692159 74E+001
```

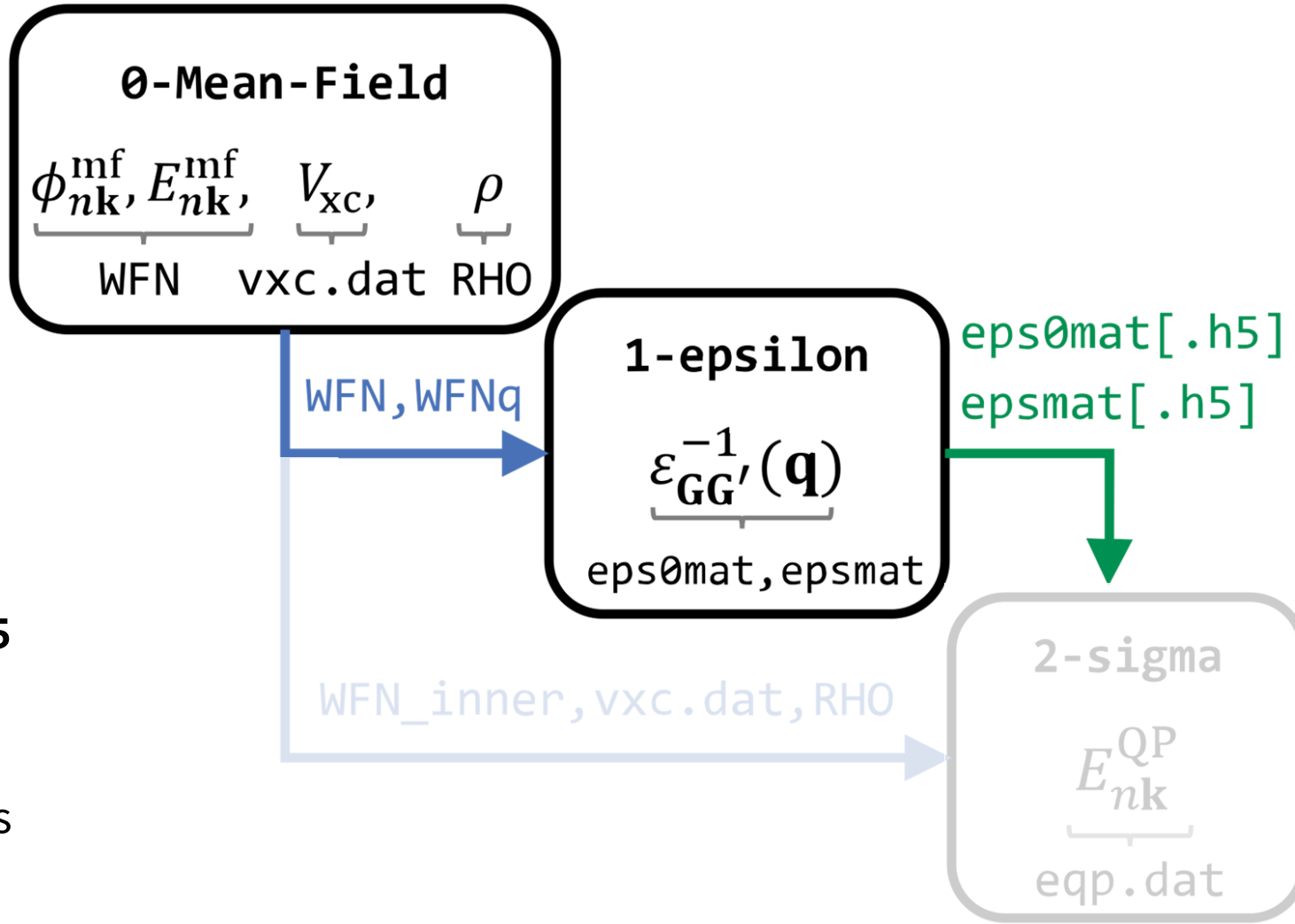
2. Sample output from the epsilon code (`epsilon.out`)

$$\varepsilon_{GG'}(\mathbf{q},\omega) \sim \chi^0_{GG'}(\mathbf{q},\omega) \sim \sum_{v\,c\,\mathbf{k}} \frac{\langle u_{c\mathbf{k}}|e^{i\mathbf{G}\cdot\mathbf{r}}|u_{v\mathbf{k}+\mathbf{q}}\rangle\langle u_{v\mathbf{k}+\mathbf{q}}|e^{i\mathbf{G}'\cdot\mathbf{r}}|u_{c\mathbf{k}}\rangle}{E_{v\mathbf{k}+\mathbf{q}} - E_{c\mathbf{k}}}$$

```
================================================================================
 19:13:53   Dealing with q =  0.000000  0.000000  0.001000                 1 / 8
================================================================================

 This is the special q->0 point.
 Rank of the polarizability matrix (nmtx): 137

 BLACS processor grid:   4 x   8; BLOCKSIZE =   17

 Number of k-points in the irreducible BZ(q) (nrk): 20

 ...

 q-pt      1: Head of Epsilon          =    2.5499723692159 74E+001
```

$\varepsilon_{00}(\mathbf{q},0)$

2. Anatomy of an **epsilon** calculation



Our discussion explains:

- Why **epsilon** outputs both **epsmat.h5** & **eps0mat.h5**
- Why **epsilon** requires 2 input WFN files (**WFN** & **WFNq**).

2. **k**-, **q**-grids and bands

For reference: simplified approach for tutorial

	k-grid	# bands	Comments
SCF	uniform, no shift	occupied	
WFN	uniform, no shift	many	
WFNq	WFN + **q**-shift	occupied	
epsilon.inp **q**-points	WFN but $\mathbf{q}_0$ instead of 0	many	bands to sum over
WFN_inner	WFN	many	bands to sum over
sigma.inp **k**-points	subset of WFN_inner	few	can choose to calculate Sigma just for bands of interest
WFN_co	WFN_inner	few	
WFN_fi (absorption)	uniform, random shift	few	
WFNq_fi	WFN_fi + **q**-shift	occupied	
WFN_fi (inteqp)	anything	few	whatever is of interest

Outline

1. Introduction
2. K-point vs. Q-point grids
3. Screening models for ε(q)
4. Frequency dependence of ε(ω)
5. Symmetry and degeneracy
6. Solving Dyson's equation
7. BSE

3. How do we use ε to compute Σ?

Self-energy Σ depends on the integral of $W(\mathbf{q})$

$$\langle n\mathbf{k}|\Sigma|n\mathbf{k}\rangle \sim \sum_{m\mathbf{G}\mathbf{G}'} \int d^3q \; f_{\mathbf{q}} \, W_{\mathbf{G}\mathbf{G}'}(\mathbf{q}) \sim \sum_{\mathbf{q}} f_{\mathbf{q}} \, W_{\mathbf{G}\mathbf{G}'}(\mathbf{q})$$

Smooth function

$$W_{\mathbf{G}\mathbf{G}'}(\mathbf{q};\omega) = \varepsilon^{-1}_{\mathbf{G}\mathbf{G}'}(\mathbf{q};\omega)\, v(\mathbf{q}+\mathbf{G})$$

Fast variations around $\mathbf{q} = \mathbf{0}$.

Divergence at $\mathbf{q} = 0$

Slow convergence in sum over $\mathbf{q}$, since $W(\mathbf{q})$ may present:

- Divergences for gapped systems.
- Fast variations of $\varepsilon^{-1}_{\mathbf{G}\mathbf{G}'}(\mathbf{q};\omega)$ around $\mathbf{q} = \mathbf{0}$.

3. Problem 1: Divergent behavior around **q** = 0

Head: $\mathbf{G} = 0, \mathbf{G}' = 0$
Wing: $\mathbf{G} = 0, \mathbf{G}' \neq 0$
Wing′: $\mathbf{G} \neq 0, \mathbf{G}' = 0$
Body: $\mathbf{G} \neq 0, \mathbf{G}' \neq 0$

$$W_{\mathbf{GG}'}(\mathbf{q};\omega) = \varepsilon^{-1}_{\mathbf{GG}'}(\mathbf{q};\omega)\, v(\mathbf{q}+\mathbf{G})$$

Sigma code needs "hint" from the user to efficiently compute the integral:

$$\int d^3q\, f_{\mathbf{q}}\, W_{\mathbf{GG}'}(\mathbf{q})$$

$\varepsilon^{-1}_{\mathbf{GG}'}(\mathbf{q}\to 0;0)$	head	wing	wing′	body
Semiconductor	const	$\mathbf{q}$	$\mathbf{q}/q^2$	const
Metal	q^2	q^2	const	const

$W_{\mathbf{GG}'}$	head	wing	wing′	body
Semiconductor	$1/q^2$	$\mathbf{q}/q^2$	$\mathbf{q}/q^2$	const
Metal	const	const	const	const

See BerkeleyGW paper: arXiv:1111.4429.

3. Solution 1: screening models

Use calculation of $\varepsilon(\mathbf{q}_0 \sim 0.001)$ along the periodic direction to **parametrize screening model** for **q** ≈ 0.

📌 The calculation is still *ab initio*! The screening model is just a "hint" the the user give to BerkeleyGW to improve w.r.t. k-point sampling!

Input file `epsilon.inp`:

```
screening_semiconductor
#screening_graphene
#screening_metal
```

See BerkeleyGW paper arXiv:1111.4429 and manual.

Sigma: Use model to perform Monte Carlo integration $\int d^3\mathrm{q}\, M(\mathbf{q}) W_{\mathbf{GG}'}(\mathbf{q})$ over region around **q** = 0

Absorption: interpolate kernel

head wing, wing' body

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

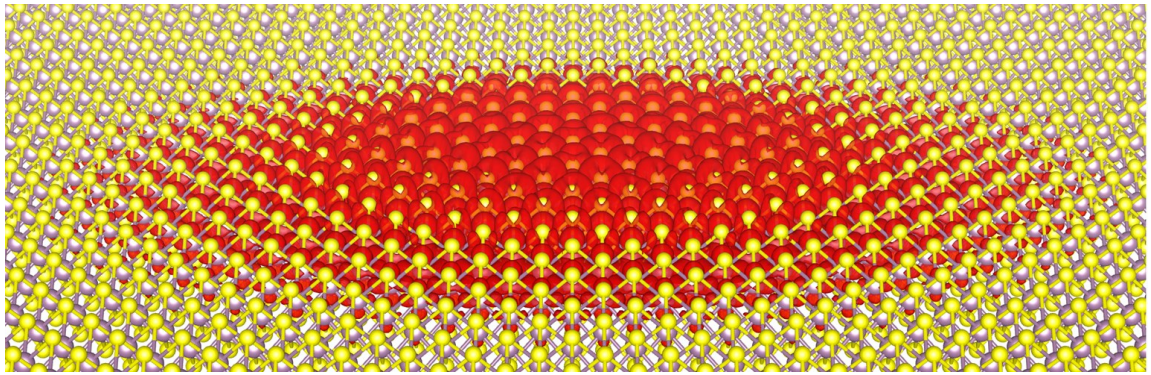
Note: anisotropic materials need to use direction such that $\epsilon^{-1}(\mathbf{q}_0) = \langle \epsilon^{-1}(\mathbf{q}) \rangle$

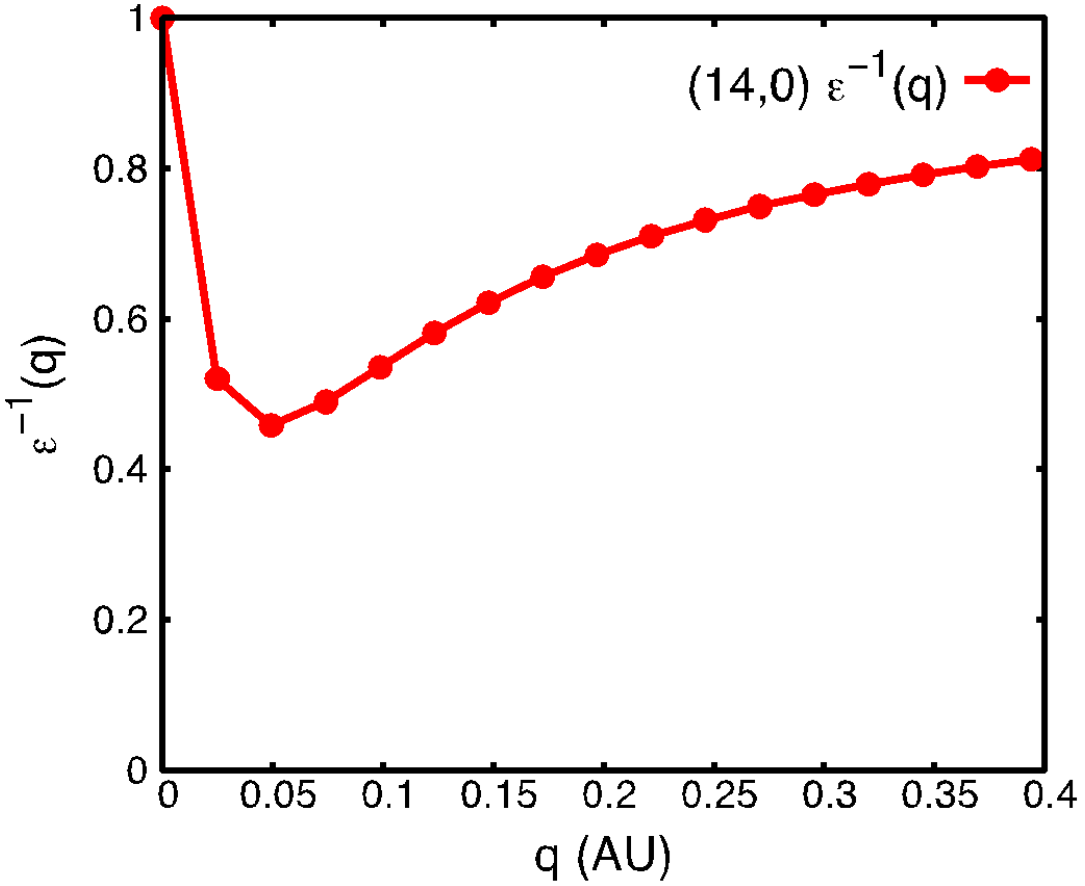
3. Problem 2: Fast variations of $\varepsilon^{-1}_{GG'}(\mathbf{q};\omega)$ around $\mathbf{q} = 0$

$$W_{GG'}(\mathbf{q};\omega) = \varepsilon^{-1}_{GG'}(\mathbf{q};\omega)\, v(\mathbf{q}+\mathbf{G})$$

(14, 0) carbon nanotube wire truncation

General for systems with **reduced dimensionality.**

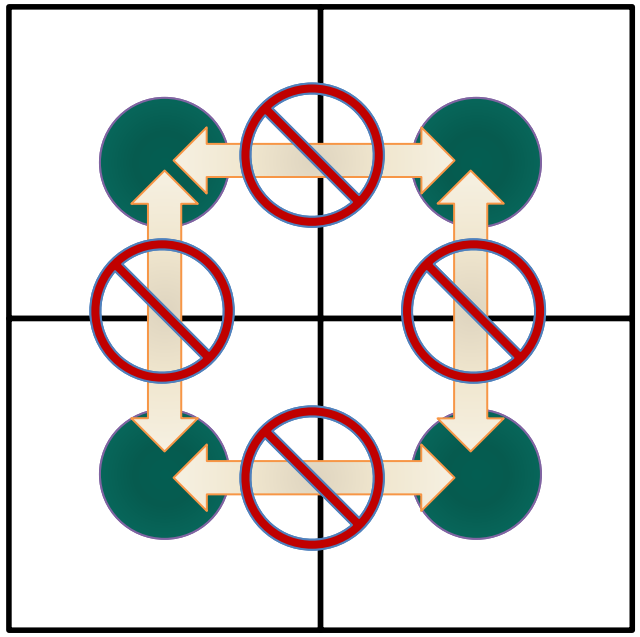




See BerkeleyGW paper: arXiv:1111.4429.

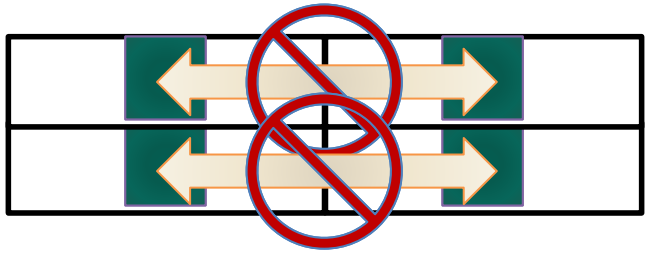
3. Coulomb truncation: different screening models $v_t = \frac{\Theta(f(\mathbf{r}))}{r}$

0D (e.g.: molecule)
fully confined



cell_box_truncation

1D (e.g.: nanotube)
periodic along z

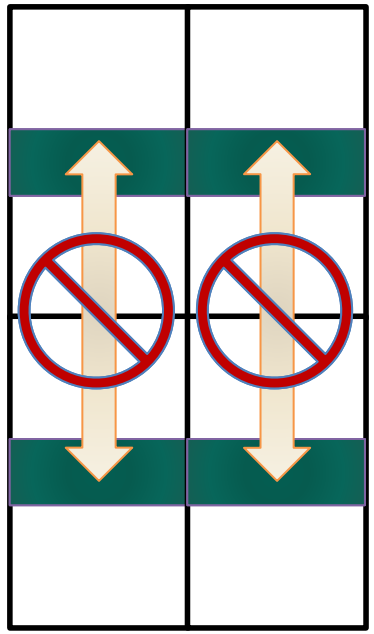


cell_wire_truncation

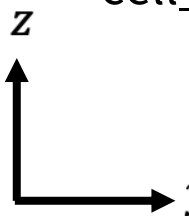
Input file `epsilon.inp`:

```
#cell_slab_truncation
#cell_wire_truncation
#cell_box_truncation
```

2D (e.g.: graphene)
periodic along x,y



cell_slab_truncation



See BerkeleyGW paper arXiv:1111.4429 and manual.

Outline

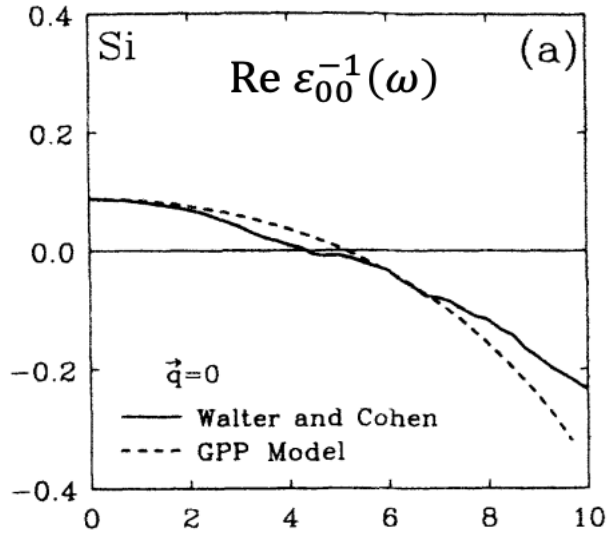
1. Introduction
2. K-point vs. Q-point grids
3. Screening models for ε(**q**)
4. Frequency dependence of ε(ω)
5. Symmetry and degeneracy
6. Solving Dyson's equation
7. Real and complex versions

4. Frequency dependence of ε(ω)

$$\Sigma(\omega) = \frac{i}{2\pi} \int_{-\infty}^{\infty} d\omega' \, G(\omega - \omega') W(\omega')$$

$$W(\omega) = \varepsilon^{-1}(\omega) \, v$$

- Needs accurate sampling of $\varepsilon(\omega)$
- Extra convergence parameters, storage, etc.



Hybertsen & Louie, PRB (1986)

Simplification: plasmon-pole model (PPM)

- **Compute** $\varepsilon(\omega = 0)$, use physical models & constraints to **obtain** $\varepsilon(\omega \neq 0)$: charge density **RHO**.
- PPM is the default option – and a good idea for a first calculation.

"**full-frequency**" vs. "**plasmon-pole**"

Input file `epsilon.inp`:

```
#frequency_dependence 0
```

See BerkeleyGW paper arXiv:1111.4429 and manual.

Outline

1. Introduction
2. K-point vs. Q-point grids
3. Screening models for ε(**q**)
4. Frequency dependence of ε(ω)
5. Symmetry and degeneracy
6. Solving Dyson's equation
7. BSE

5. Degeneracy check utility

- Choice of bands can break symmetry of degenerate states and lead to arbitrary results:

  ERROR: Selected number of bands breaks degenerate subspace.

- Use **degeneracy_check.x** utility to find number of bands that does not break degeneracies.

```
$ degeneracy_check.x WFN

Reading eigenvalues from file WFN
Number of spins:                  1
Number of bands:                 35
Number of k-points:               8

== Degeneracy-allowed numbers of bands (for epsilon and sigma) ==
              4
              8
             14
             18
             20
             32
Note: cannot assess whether or not highest band 35 is degenerate.
...
```

- So, could use **number_bands 32** in Epsilon.

- Can also turn off degeneracy enforcement (**degeneracy_check_override** flag).
  - Ok if include many unoccupied bands (error from breaking deg. subspace vanishes)

Outline

1. Introduction
2. K-point vs. Q-point grids
3. Screening models for ε(**q**)
4. Frequency dependence of ε(ω)
5. Symmetry and degeneracy
6. Solving Dyson's equation
7. BSE

6. Solving Dyson's equation in Sigma

$$E_{n\mathbf{k}}^{\mathrm{QP}} = E_{n\mathbf{k}}^{\mathrm{MF}} + \langle \psi_{n\mathbf{k}} | \Sigma(E_{n\mathbf{k}}^{\mathrm{QP}}) - \Sigma^{\mathrm{MF}} | \psi_{n\mathbf{k}} \rangle$$

How can we solve when we don't know E^{QP} yet?

(1) **`eqp0`**: evaluate at E^{MF}. $E_{n\mathbf{k}}^{\mathrm{QP0}} = E_{n\mathbf{k}}^{\mathrm{MF}} + \langle \psi_{n\mathbf{k}} | \Sigma(E_{n\mathbf{k}}^{\mathrm{MF}}) - \Sigma^{\mathrm{MF}} | \psi_{n\mathbf{k}} \rangle$

(2) **`eqp1`**: solve linearized approximation (Newton's Method)

$$E_{n\mathbf{k}}^{\mathrm{QP1}} = E_{n\mathbf{k}}^{\mathrm{QP0}} + \frac{d\Sigma_{n\mathbf{k}}/dE}{1 - d\Sigma_{n\mathbf{k}}/dE} \left(E_{n\mathbf{k}}^{\mathrm{QP0}} - E_{n\mathbf{k}}^{\mathrm{MF}} \right)$$

Available as columns in **`sigma_hp.log`**, and **`eqp0.dat`** and **`eqp1.dat`** files

📌 Note: for full frequency calculations, **`eqp1`** reports the full numerical solution of Dyson's equation.

6. Mean-field exchange-correlation functional

$$E_{n\mathbf{k}}^{\mathrm{QP}} = E_{n\mathbf{k}}^{\mathrm{MF}} + \langle n\mathbf{k}|\Sigma(E_{n\mathbf{k}}^{\mathrm{QP}}) - \Sigma^{\mathrm{MF}}|n\mathbf{k}\rangle = E_{n\mathbf{k}}^{\mathrm{MF}} + \langle n\mathbf{k}|\Sigma^{\mathrm{GW}}(E_{n\mathbf{k}}^{\mathrm{QP}}) - V_{XC}|n\mathbf{k}\rangle$$

BerkeleyGW accepts two forms of V_{XC} :

- **VXC**: Binary file containing the operator in G space: $V_{XC}(\mathbf{r}) \rightarrow V_{XC}(\mathbf{G})$
  - BerkeleyGW can compute arbitrary matrix elements given the operator.
  - Only works for (semi)-local functionals

- **`vxc.dat`**: ASCII file containing the matrix elements in a KS orbitals: $\langle n\mathbf{k}|V_{XC}|n'\mathbf{k}\rangle$.
  - DFT code must compute all matrix elements for the relevant states.
  - Supports arbitrary functionals.

📌 BerkeleyGW also natively supports some hybrid functionals even if the DFT wrapper cannot write $\langle n\mathbf{k}|\Sigma_x|n'\mathbf{k}\rangle$. See flag **`bare_exchange_fraction`** for the `Sigma` code.

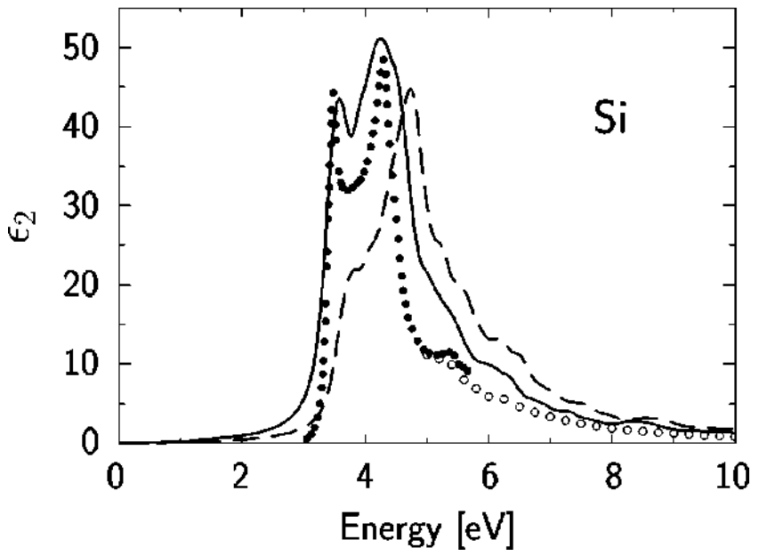
Outline

1. Introduction
2. K-point vs. Q-point grids
3. Screening models for ε(**q**)
4. Frequency dependence of ε(ω)
5. Symmetry and degeneracy
6. Solving Dyson's equation
7. BSE

Optical Absorption Spectra

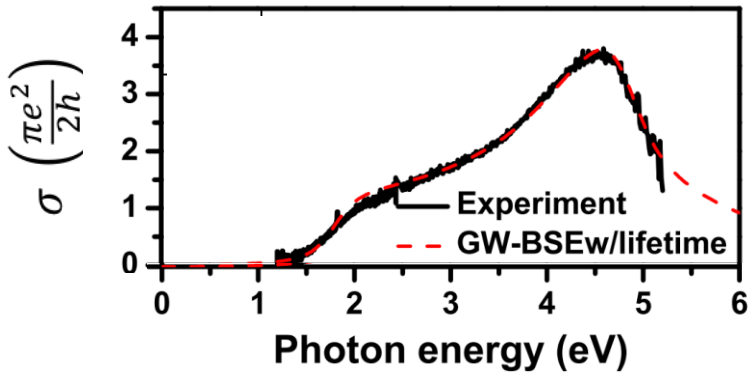
- The solution of the BSE can be used to calculate optical absorption spectra
- BerkeleyGW can handle calculations on metals, semiconductors, and insulators with varying degrees of confinement

Silicon – Bulk Semiconductor



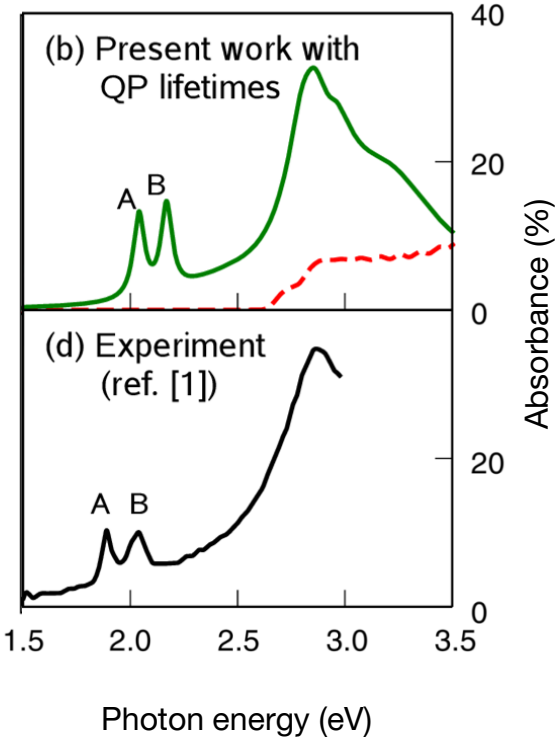
M. Rohlfing, S. G. Louie, PRB 62, 8 (2000).

Graphene – 2D Metal



K. F. Mak, F. H. da Jornada, et al., PRL 112, 207401 (2014).

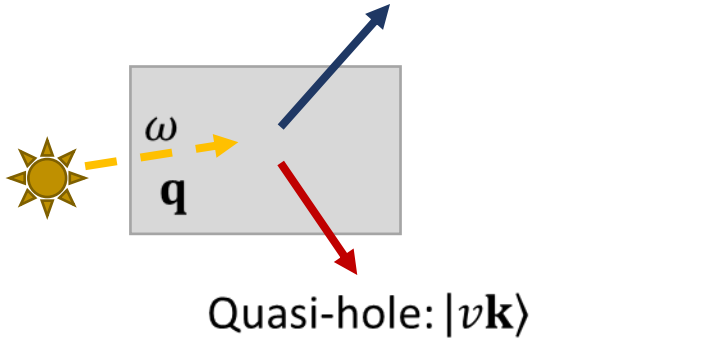
Monolayer MoS_2 – 2D Semiconductor



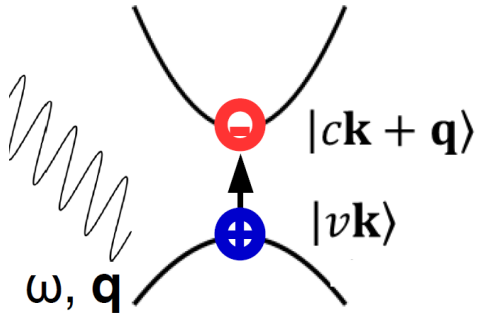
D. Y. Qiu, F. H. da Jornada, S. G. Louie, PRL 111, 216805 (2013).

Theory Overview – Optical Absorption (IP)

- What happens during optical absorption?
- An incoming photon excites an electron from the ground state to a conduction state, leaving behind a hole.



Band picture

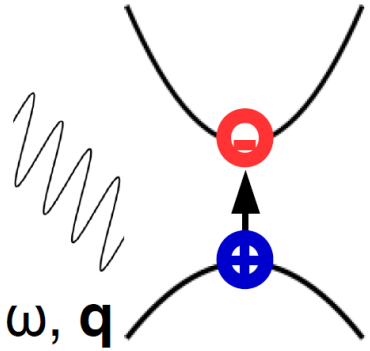


Absorption:

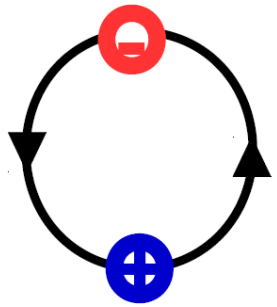
$$\varepsilon_2(-\mathbf{q},\omega) \propto \sum_{vc\mathbf{k}} |\langle v\mathbf{k}+\mathbf{q}|\hat{v}|c\mathbf{k}\rangle|^2 \delta\left[\omega-\left(E_{c\mathbf{k}}-E_{v\mathbf{k}+\mathbf{q}}\right)\right]$$

- In this picture, we are neglecting the interaction between the excited quasi-electron and quasi-hole
- This is known as the independent-particle (or GW-RPA) picture

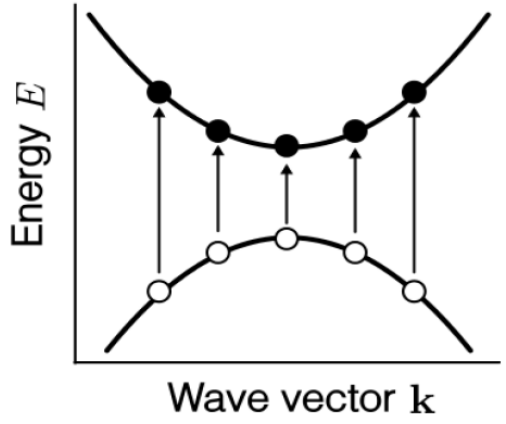
Theory Overview – Excitons



Incoming photon excites quasi-electron and quasi-hole

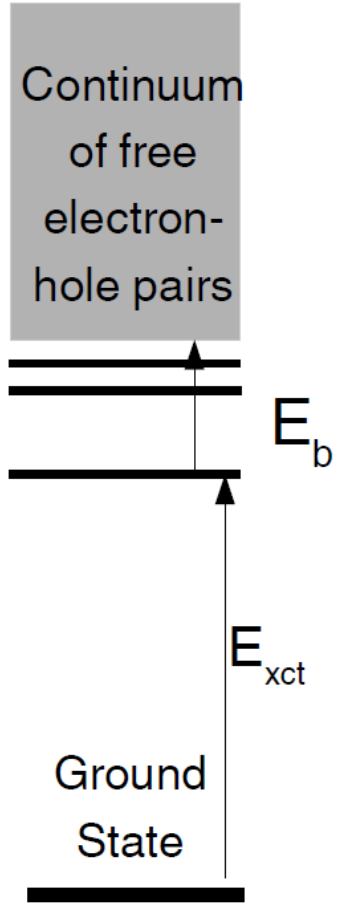


Electron and hole interact forming an **exciton**



Excited-state:

$$\Psi_S(\mathbf{r}_e, \mathbf{r}_h) = \sum_{vck} A^S_{vck} \psi^*_{vk}(\boldsymbol{r}_h) \psi_{ck}(\boldsymbol{r}_e)$$



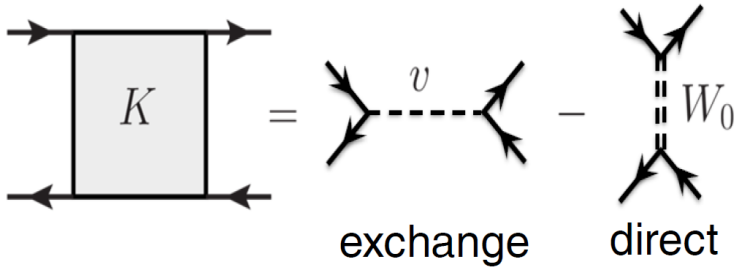
Theory Overview – GW-BSE

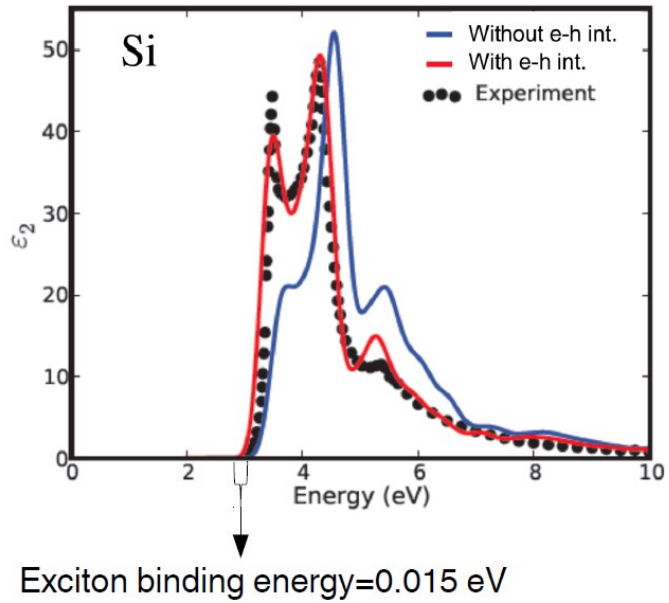
- The Bethe Salpeter equation can be written as an effective Hamiltonian in the electron-hole basis

$$H^{\mathrm{BSE}}\Psi_S(\mathbf{r}_e,\mathbf{r}_h) = \Omega_S\Psi_S(\mathbf{r}_e,\mathbf{r}_h)$$

$$H^{\mathrm{BSE}}_{vc\mathbf{k},v'c'\mathbf{k}'} = (E_{c\mathbf{k}} - E_{v\mathbf{k}})\delta_{\mathbf{k},\mathbf{k}'} + K_{vc\mathbf{k},v'c'\mathbf{k}'}$$

- Interaction Kernel





- Absorption

$$\epsilon_2 \propto \sum_S |\langle 0|\mathbf{v}|S\rangle|^2\delta(\omega - \Omega_S)$$

NERSC

Rohlfing and Louie, *PRB* 62, 4947 (2000). Deslippe et al, *Comput. Phys. Commun.* 183, 1269 (2012).

BSE in BerkeleyGW — What is the basis?

$$H^{BSE}_{vc\mathbf{k},v'c'\mathbf{k}'} = (E_{c\mathbf{k}} - E_{v\mathbf{k}})\delta_{vv'}\delta_{cc'}\delta_{v\mathbf{k}\mathbf{k}} + K_{vc\mathbf{k},v'c'\mathbf{k}'}$$

diagonal

dense

- Challenge: Quasiparticle corrections and Kernel matrix elements must be computed on a very fine k-point grid
  - E.g. Monolayer MoS_2 requires 300x300 k-grid to converge!
  - We already know how to **interpolate** the Quasiparticle corrections. Can we do something similar for the Kernel?

NERSC

BSE in BerkeleyGW — Interpolation

- Computing the Kernel matrix elements is expensive

$$\langle vc\mathbf{k}_{\mathrm{fi}}|K|v'c'\mathbf{k}'_{\mathrm{fi}}\rangle$$

"fine (fi)" grid refers to the dense k-grid needed to converge the solution of the BSE

- We want to compute the kernel on a "coarse" grid and interpolate the "fine" grid

$$\langle vc\mathbf{k}_{\mathrm{co}}|K|v'c'\mathbf{k}'_{\mathrm{co}}\rangle \longrightarrow \langle vc\mathbf{k}_{\mathrm{fi}}|K|v'c'\mathbf{k}'_{\mathrm{fi}}\rangle$$

- Linear interpolation? Wannier interpolations?

No complex phases Hard to operate/unstable

- BerkeleyGW: Projection interpolation/ dual grid interpolation
  - Explicitly generate coarse- and fine-grid WFNs
  - Expand fine-grid WFNs in term of coarse-grid WFNs.
  - Captures band crossing, etc.

BSE in BerkeleyGW — Interpolation Scheme

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

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

$$u_{n\mathbf{k}_{\mathrm{fi}}} = \sum_{n'} C^{\mathbf{k}_{\mathrm{co}}}_{n,n'} u_{n'\mathbf{k}_{\mathrm{co}}} \qquad C^{\mathbf{k}_{\mathrm{co}}}_{n,n'} = \int d\mathbf{r}\, u_{n\mathbf{k}_{\mathrm{fi}}}(\mathbf{r}) u^*_{n'\mathbf{k}_{\mathrm{co}}}(\mathbf{r})$$

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

$$E^{\mathrm{QP}}_n(\mathbf{k}_{\mathrm{fi}}) = \quad E^{\mathrm{MF}}_n(\mathbf{k}_{\mathrm{fi}}) + \left\langle \sum_{n'} \left|C^{\mathbf{k}_{\mathrm{co}}}_{n,n'}\right|^2 \left(E^{\mathrm{QP}}_{n'}(\mathbf{k}_{\mathrm{co}}) - E^{\mathrm{MF}}_{n'}(\mathbf{k}_{\mathrm{co}})\right) \right\rangle_{\mathbf{k}_{\mathrm{co}}}$$

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

$$\langle vc\mathbf{k}_{\mathrm{fi}}|K|v'c'\mathbf{k}'_{\mathrm{fi}}\rangle = \sum_{n_1,n_2,n_3,n_4} C^{\mathbf{k}_{\mathrm{co}}}_{c,n_1} C^{*\mathbf{k}_{\mathrm{co}}}_{v,n_2} C^{*\mathbf{k}'_{\mathrm{co}}}_{c',n_3} C^{\mathbf{k}'_{\mathrm{co}}}_{v',n_4} \langle n_2 n_1 \mathbf{k}_{\mathrm{co}}|K|n_4 n_3 \mathbf{k}'_{\mathrm{co}}\rangle$$

NERSC

BSE in BerkeleyGW — Interpolation Scheme

- Note: a direct interpolation of K is not very accurate
  - Explicit dependence on $\mathbf{q}_{\mathrm{fi}} = \mathbf{k}_{\mathrm{fi}} - \mathbf{k}'_{\mathrm{fi}} \neq \mathbf{q}_{\mathrm{co}} = \mathbf{k}_{\mathrm{co}} - \mathbf{k}'_{\mathrm{co}}$
- 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:

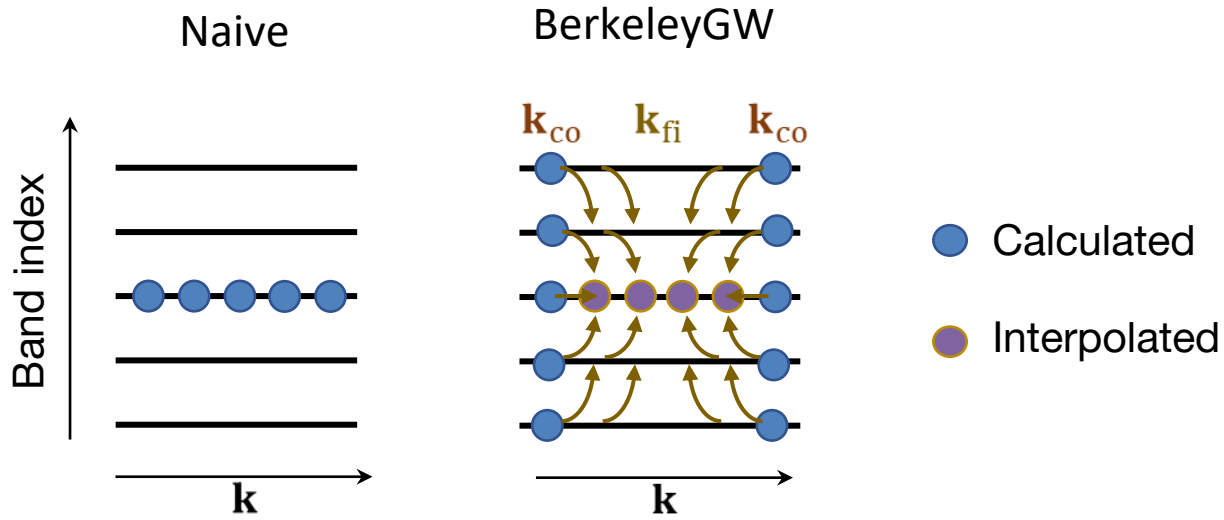
$$A(\mathbf{q}) = q^2, \qquad B(\mathbf{q}) = q, \qquad C(\mathbf{q}) = 1$$

- Important flags: `screening_*` and `*truncation`

BSE in BerkeleyGW — Interpolation Scheme

- In practice: trading bands for k-points

$$\langle vc\mathbf{k}_{\text{fi}}|K|v'c'\mathbf{k}'_{\text{fi}}\rangle = \sum_{n_1,n_2,n_3,n_4} C^{\mathbf{k}_{\text{co}}}_{c,n_1} C^{*\mathbf{k}_{\text{co}}}_{v,n_2} C^{*\mathbf{k}'_{\text{co}}}_{c',n_3} C^{\mathbf{k}'_{\text{co}}}_{v',n_4} \langle n_2 n_1 \mathbf{k}_{\text{co}}|K|n_4 n_3 \mathbf{k}'_{\text{co}}\rangle$$

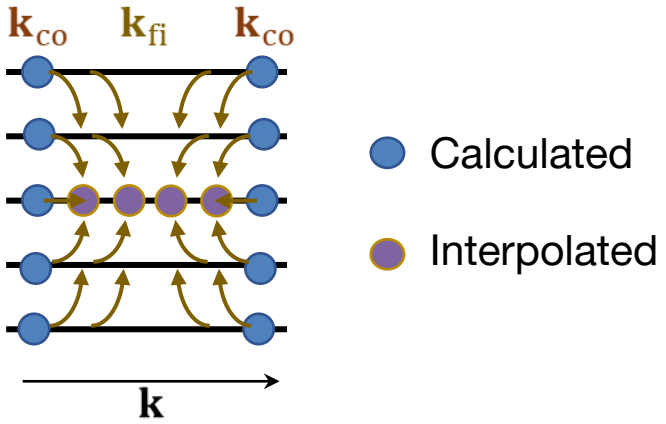


- How to get a good interpolation?
  - Include a large number of bands from the coarse grid and start from a coarse grid that is not too coarse.

BSE in BerkeleyGW — Interpolation Scheme

- How do I know if I included enough bands?

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



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

absorption.out / inteqp.out

BerkeleyGW Workflow

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

`epsmat.h5`, $\{E_c\}_{\text{co}}$, $\{E_v\}_{\text{co}}$

`kernel.x`

Step 1: Calculate BSE kernel on the same coarse grid

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

`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

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

NERSC

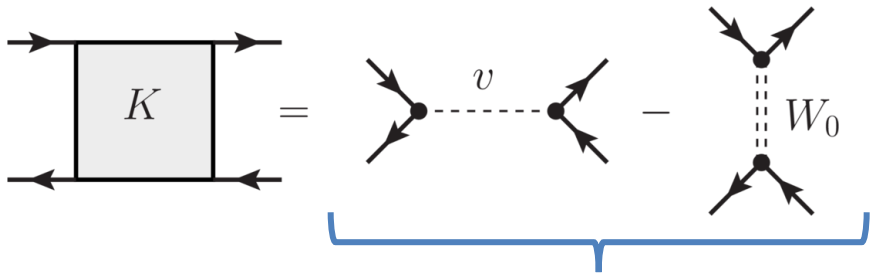
1. Kernel

`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
- Input: epsmat.h5, eps0mat.h5, WFN_co

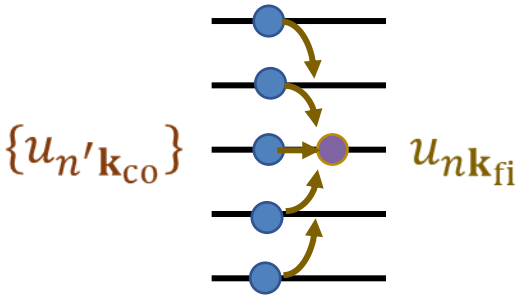


`bsemat.h5`

1. Kernel

Sample **kernel.inp**

```
number_val_bands <?>
number_cond_bands <?>

<?>_symmetries_coarse_grid
screening_<?>
```



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

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!)

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

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

- Absorption needs same coarse WFN_co from Kernel
- Two fine WFN files are needed

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

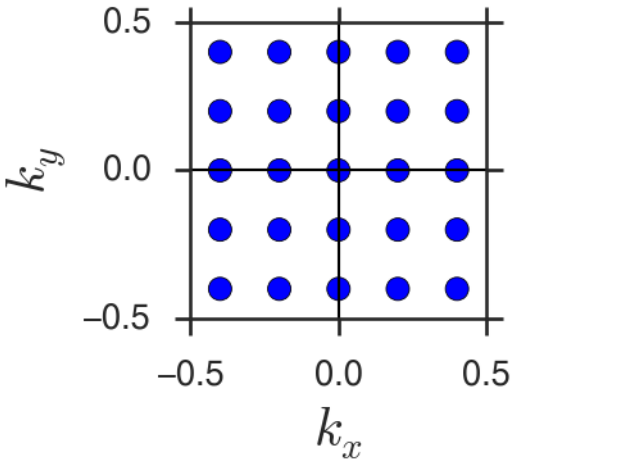
- Typically q is 10^{-3}
- Fine WFN files:
  - WFN_fi: for conduction states $|c\mathbf{k}\rangle$
  - WFNq_fi: for q-shifted valence states $|v\mathbf{k}+\mathbf{q}\rangle$

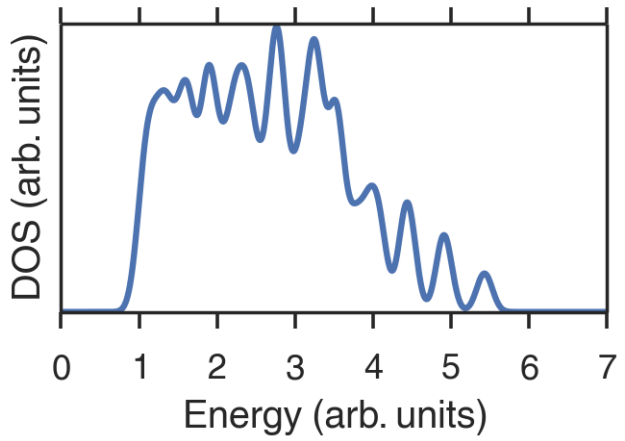
Direction of q = polarization of light= difference between $\{\mathbf{k}\}$ in WFNq and $\{\mathbf{k}\}$ in WFN

NERSC

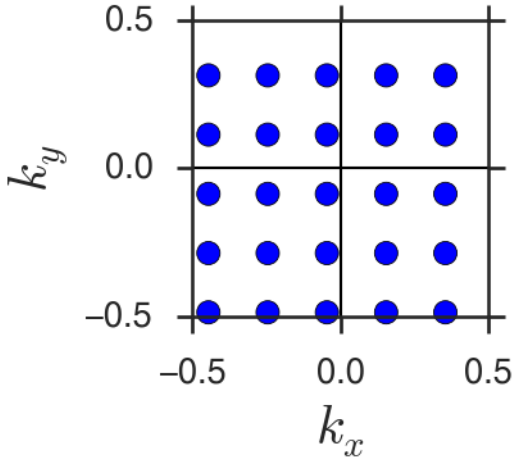
2. Absorption — Randomly Shifted k-grids

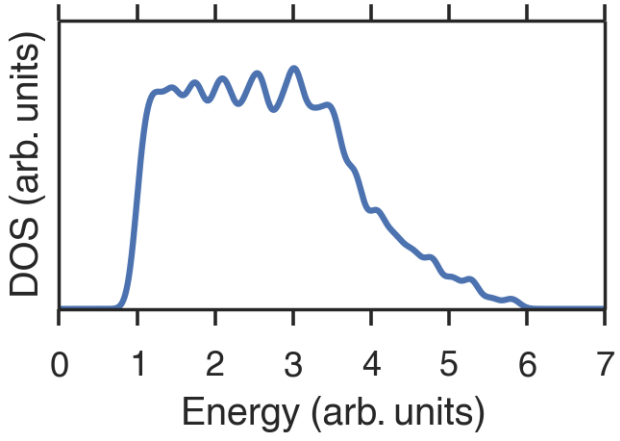
WFN_fi, no k-shift





WFN_fi, random k-shift





2. Absorption — Randomly Shifted k-grids

Sample **absorption.inp**

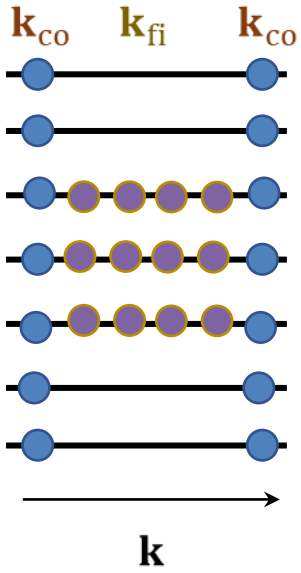
```
diagonalization

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



Summary

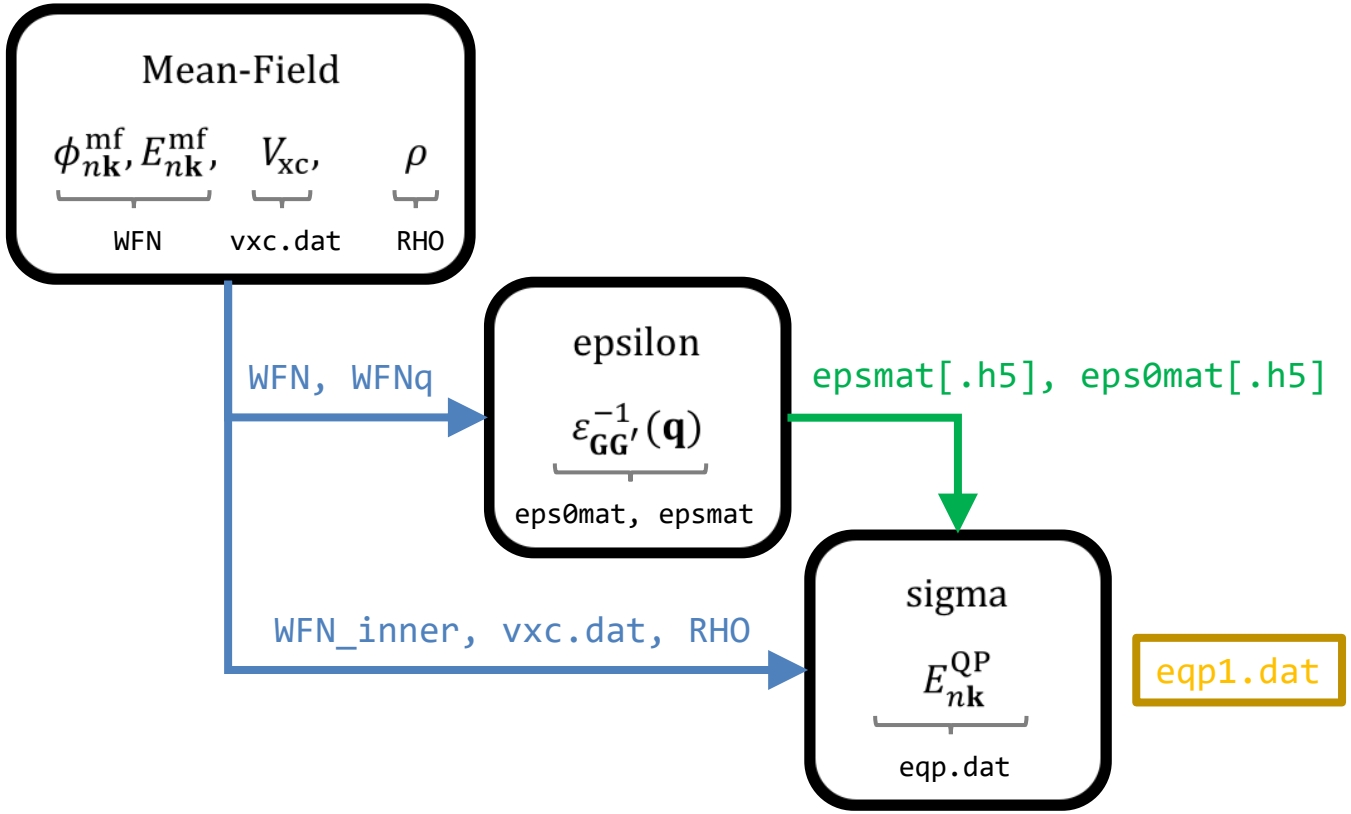
- BSE needs to be solved on fine k-grid

- BGW interp.: projection of fine WFNs onto coarse WFNs
  - Need to *include more bands* in kernel calculation

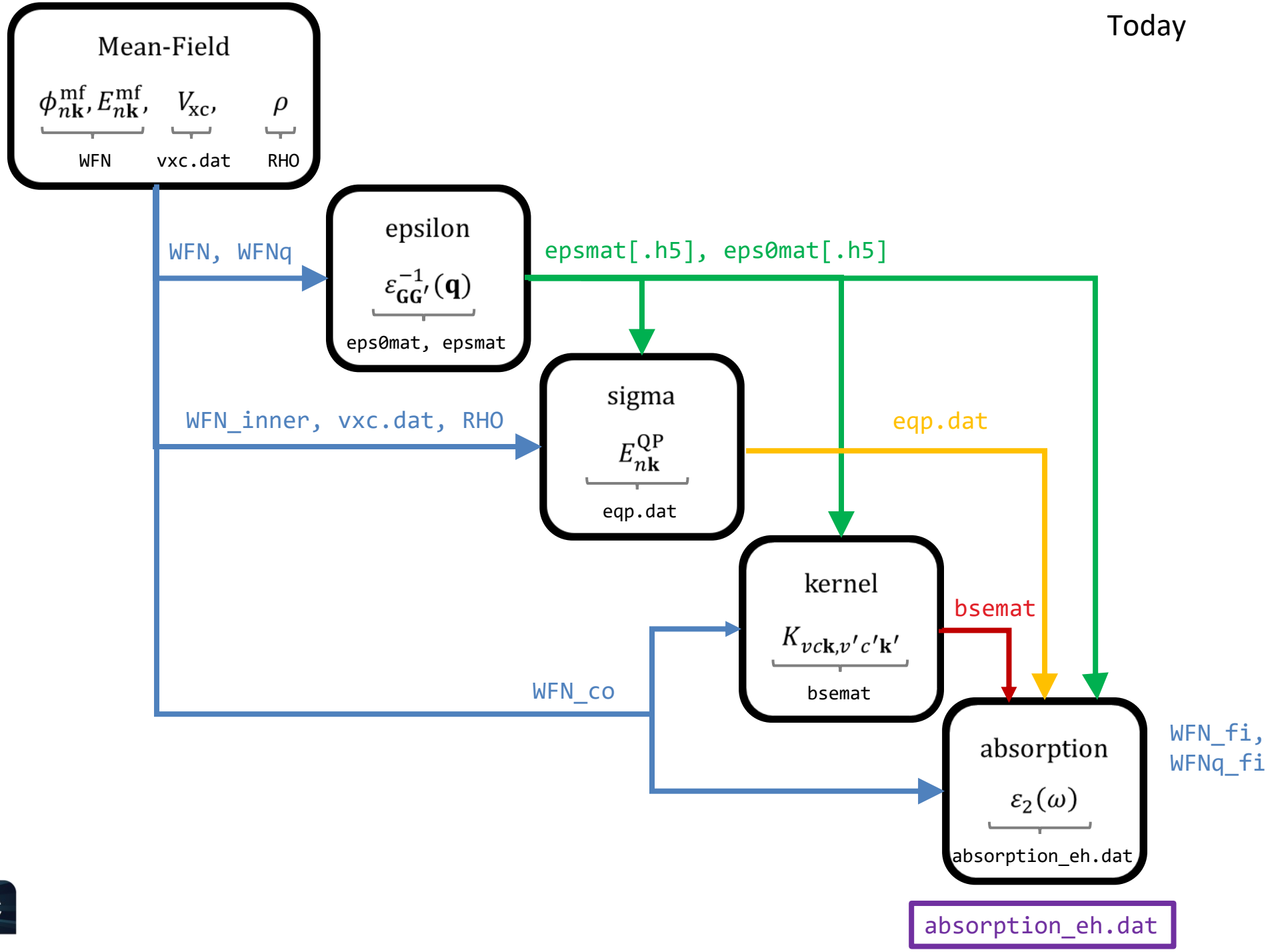
- WFNs:
  - Kernel: WFN_co
  - Absorption: WFN_co, WFN_fi, WFNq_fi
  - WFN_co: unshifted
  - WFN_fi: random k-shift
  - WFNq_fi: random k-shift + q-shift (dir. = pol. of light)

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

Yesterday


Mean-Field
$\phi^{\mathrm{mf}}_{n\mathbf{k}}, E^{\mathrm{mf}}_{n\mathbf{k}}$, V_{xc}, ρ
WFN
vxc.dat
RHO
WFN, WFNq
epsilon
$\varepsilon^{-1}_{\mathbf{GG'}}(\mathbf{q})$
eps0mat, epsmat
epsmat[.h5], eps0mat[.h5]
WFN_inner, vxc.dat, RHO
sigma
$E^{\mathrm{QP}}_{n\mathbf{k}}$
eqp.dat
eqp1.dat

Today


Mean-Field
$\phi^{mf}_{n\mathbf{k}}, E^{mf}_{n\mathbf{k}}$, V_{xc}, ρ
WFN
vxc.dat
RHO
WFN, WFNq
epsilon
$\varepsilon^{-1}_{GG'}(\mathbf{q})$
eps0mat, epsmat
epsmat[.h5], eps0mat[.h5]
WFN_inner, vxc.dat, RHO
sigma
$E^{QP}_{n\mathbf{k}}$
eqp.dat
eqp.dat
kernel
$K_{vc\mathbf{k},v'c'\mathbf{k}'}$
bsemat
bsemat
WFN_co
absorption
$\varepsilon_2(\omega)$
absorption_eh.dat
WFN_fi,
WFNq_fi
absorption_eh.dat

Extra Slides

k-, q-grids and bands

recommended approach (using half-shifted grids)

	k-grid	# bands	Comments
SCF	Uniform, 0.5 shift	occupied	as usual in DFT
WFN	Uniform, 0.5 shift	many	
WFNq	WFN + **q**-shift	occupied	
epsilon.inp **q**-points	WFN but no shift, $\mathbf{q}_0$	many	bands to sum over
WFN_inner	WFN but no shift	many	bands to sum over
sigma.inp **k**-points	subset of WFN_inner	few	can choose to calculate Sigma just for bands of interest
WFN_co	WFN_inner	few	
WFN_fi (absorption)	Uniform, random shift	few	
WFNq_fi	WFN_fi + **q**-shift	occupied	
WFN_fi (inteqp)	anything	few	whatever is of interest

5. Choice of bands can break symmetry in GW/BSE

Symmetry operations are only defined for non-degenerate states!

Example:

- 2D system with mirror symmetry $\hat{\sigma}_x$ about the x=0 plane.
- Consider two degenerate KS states of $|p_x\rangle$ and $|p_y\rangle$ characters:
  - $\hat{\sigma}_x|p_x\rangle = -|p_x\rangle$
  - $\hat{\sigma}_x|p_y\rangle = |p_y\rangle$
- If $|p_x\rangle$ and $|p_y\rangle$ degenerate, the DFT code generates arbitrary linear combination, e.g. $|p_x\rangle \pm i|p_y\rangle$, which are not an eigenstate of $\hat{\sigma}_x$.

➢ Including only some of a degenerate space will break symmetry.

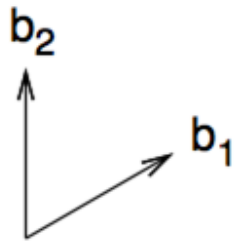
➢ Results depends on arbitrary linear combinations in mean-field. Not reproducible!

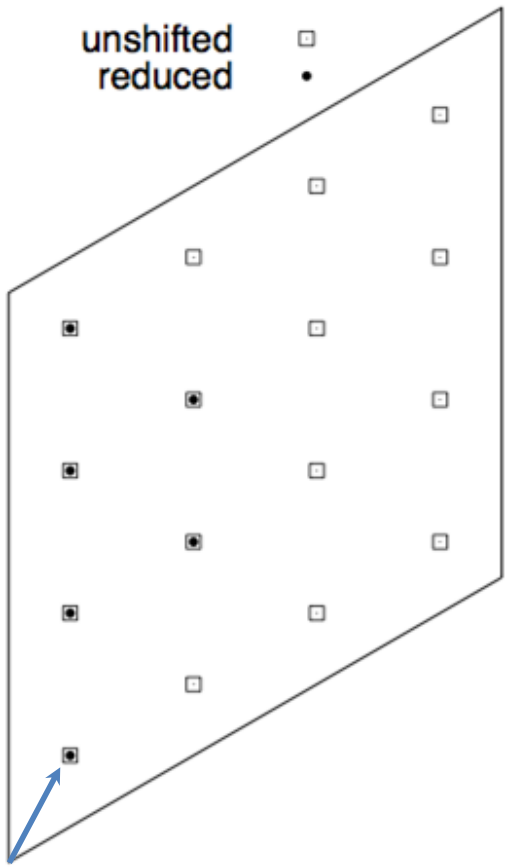
k-grid construction: 4×4 shifted grid for graphene

(0.5, 0.5) Monkhorst-Pack shift

`kgrid.x`

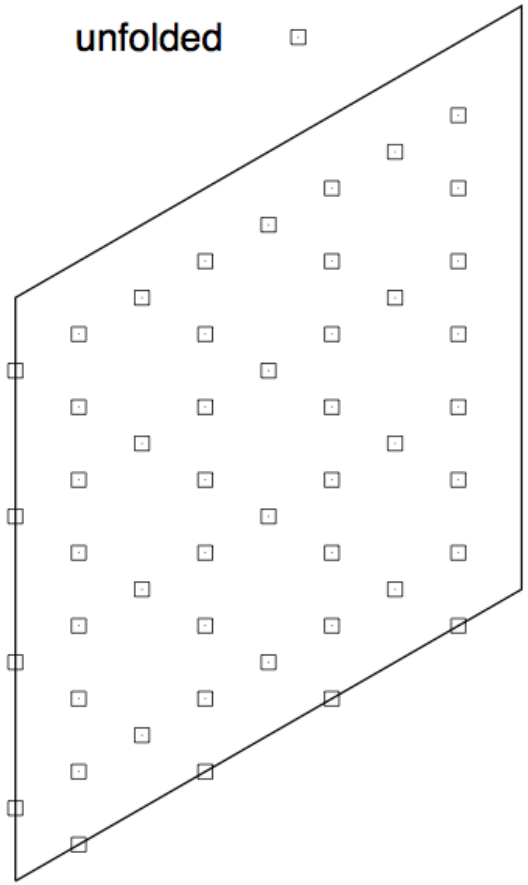
Uniform -> unfold -> shift with **q** -> reduce





(0.5, 0.5)

Main grid (WFN)
16 in full BZ
Reduced to 6



0.05)

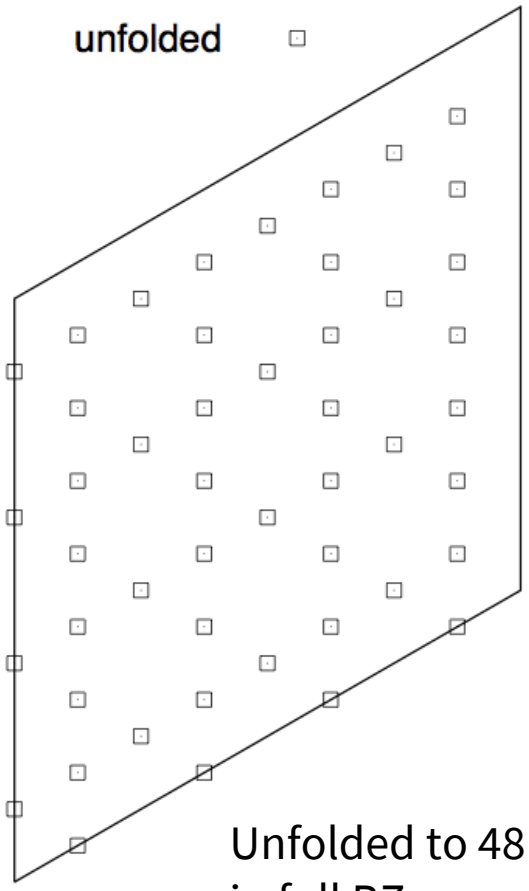
Unfolded to 48
in full BZ

Unfolding gives more points!

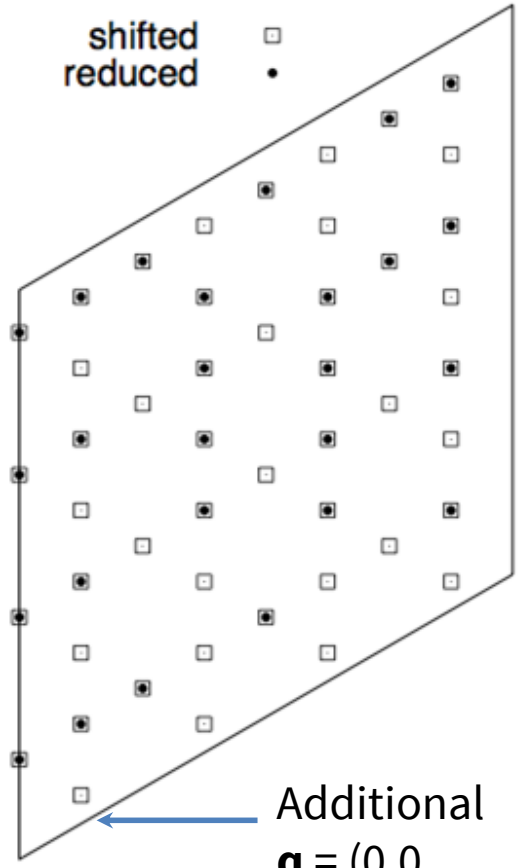
k-grid construction: 4×4 shifted grid for graphene

`kgrid.x`

Uniform -> unfold -> shift with **q** -> reduce



Unfolded to 48 in full BZ



Additional **q** = (0.0, 0.05)

Shifted grid (WFNq)
48 in full BZ
Reduced to 26

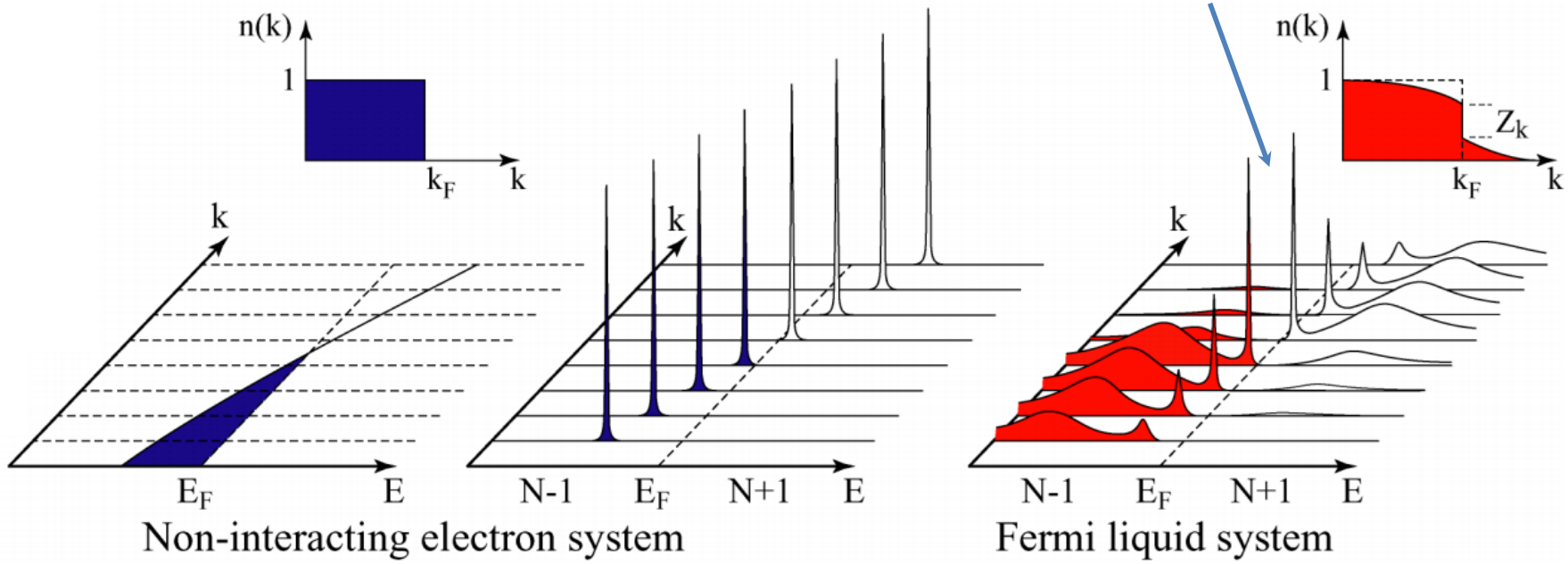
Unfolding and breaking symmetry gives more points!

Quasiparticle renormalization factor Z

$$E_{n\mathbf{k}}^{\mathrm{QP1}} = E_{n\mathbf{k}}^{\mathrm{QP0}} + (Z_{n\mathbf{k}} - 1)\left(E_{n\mathbf{k}}^{\mathrm{QP0}} - E_{n\mathbf{k}}^{\mathrm{MF}}\right)$$

$$Z_{n\mathbf{k}} = \frac{1}{1 - d\Sigma_{n\mathbf{k}}/dE}$$

Between 0 and 1
Weight in QP peak



A. Damascelli, Z. Hussain, Z.-X Shen, Rev. Mod. Phys. **75**, 473 (2003)

Special treatment for metals

Coefficients depend critically on sampling DOS at Fermi surface for intraband transitions.

Two separate runs of Epsilon

eps0mat: $\epsilon^{-1}_{\mathbf{G}\mathbf{G}'}(\mathbf{q}_0)$

WFN = WFNq
only a small number of bands for intra-band transitions around Fermi surface
very fine: grid spacing is $\mathbf{q}_0$ *e.g.* grid = 32 × 32 × 32 unshifted, $\mathbf{q}_0$ = (0, 0, 1/32)

epsmat: $\epsilon^{-1}_{\mathbf{G}\mathbf{G}'}(\mathbf{q} \neq \mathbf{q}_0)$

$\epsilon^{-1}_{\mathbf{G}\mathbf{G}'}$	head	wing	wing′	body
Semiconductor	const	$\mathbf{q}$	$\mathbf{q}/q^2$	const
Metal	q^2	q^2	const	const
$W_{\mathbf{G}\mathbf{G}'}$	head	wing	wing′	body
Semiconductor	$1/q^2$	$\mathbf{q}/q^2$	$\mathbf{q}/q^2$	const
Metal	const	const	const	const

WFN = WFNq. unshifted, many bands, ordinary fineness. *e.g.* grid = 12 × 12 × 12.

epsilon.inp for metals

```
begin qpoints
  0.000000000  0.000000000  0.031250000 1.0 2
end
```

eps0mat:

$\epsilon^{-1}_{\mathbf{G}\mathbf{G}'}(\mathbf{q}_0)$

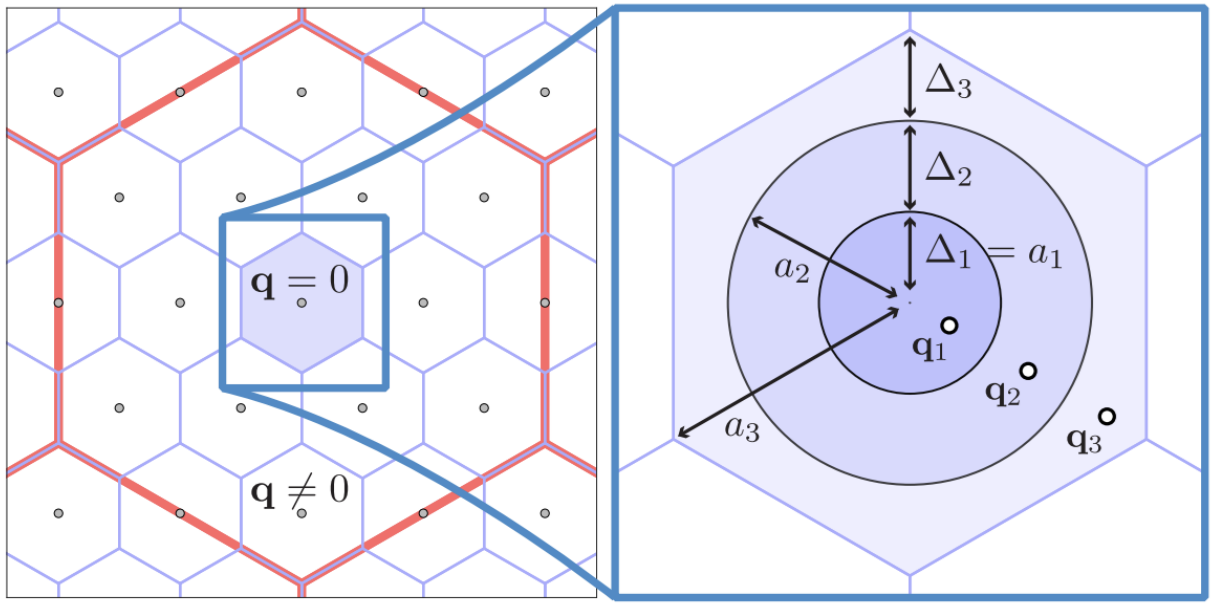
```
begin qpoints
  0.000000000  0.000000000  0.083333333 1.0 0
  0.000000000  0.000000000  0.166666667 1.0 0
  0.000000000  0.000000000  0.250000000 1.0 0
…
end
```

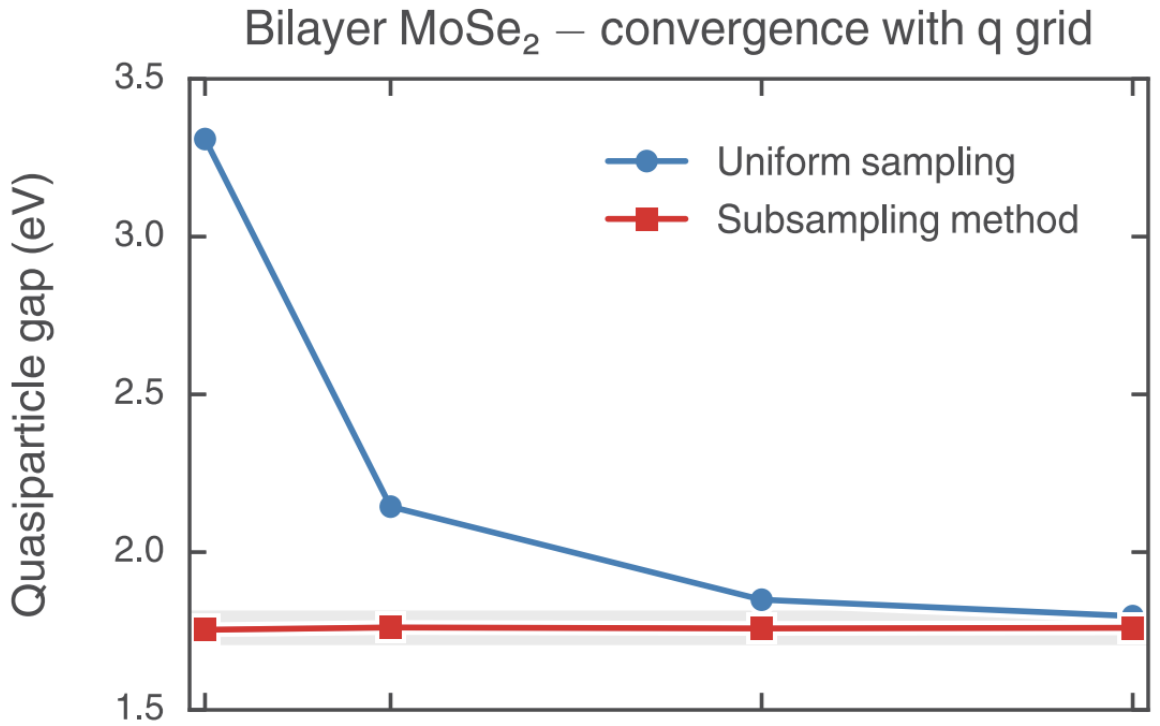
epsmat:

$\epsilon^{-1}_{\mathbf{G}\mathbf{G}'}(\mathbf{q} \neq \mathbf{q}_0)$

2. Advanced **k**-grid construction

- For advanced users, BGW can use different k- and q-point grids.
- BerkeleyGW pioneered **nonuniform sampling** schemes of the BZ for optimally dealing with **k**- and **q**-grids in metals and low-D materials.





Jornada, Qiu, Louie, PRB 95, 035109 (2017).

7. Real or complex flavor?

e.g. `epsilon.real.x, epsilon.cplx.x`

Complex is general, but real is faster, uses less memory and disk space

Real: only with inversion symmetry about the origin **and** time-reversal symmetry

$$u(-\mathbf{r}) = a u(\mathbf{r})$$
$$u^*(\mathbf{r}) = b u(\mathbf{r})$$
$$a, b \text{ each equal to } \pm 1$$

Plane-wave expansion:

$$u(\mathbf{r}) = \sum_{\mathbf{G}} u_{\mathbf{G}} e^{i\mathbf{G}\cdot\mathbf{r}} \rightarrow u_{\mathbf{G}} = c u^*_{-\mathbf{G}}$$

- Can choose $c = 1$ for real coefficients

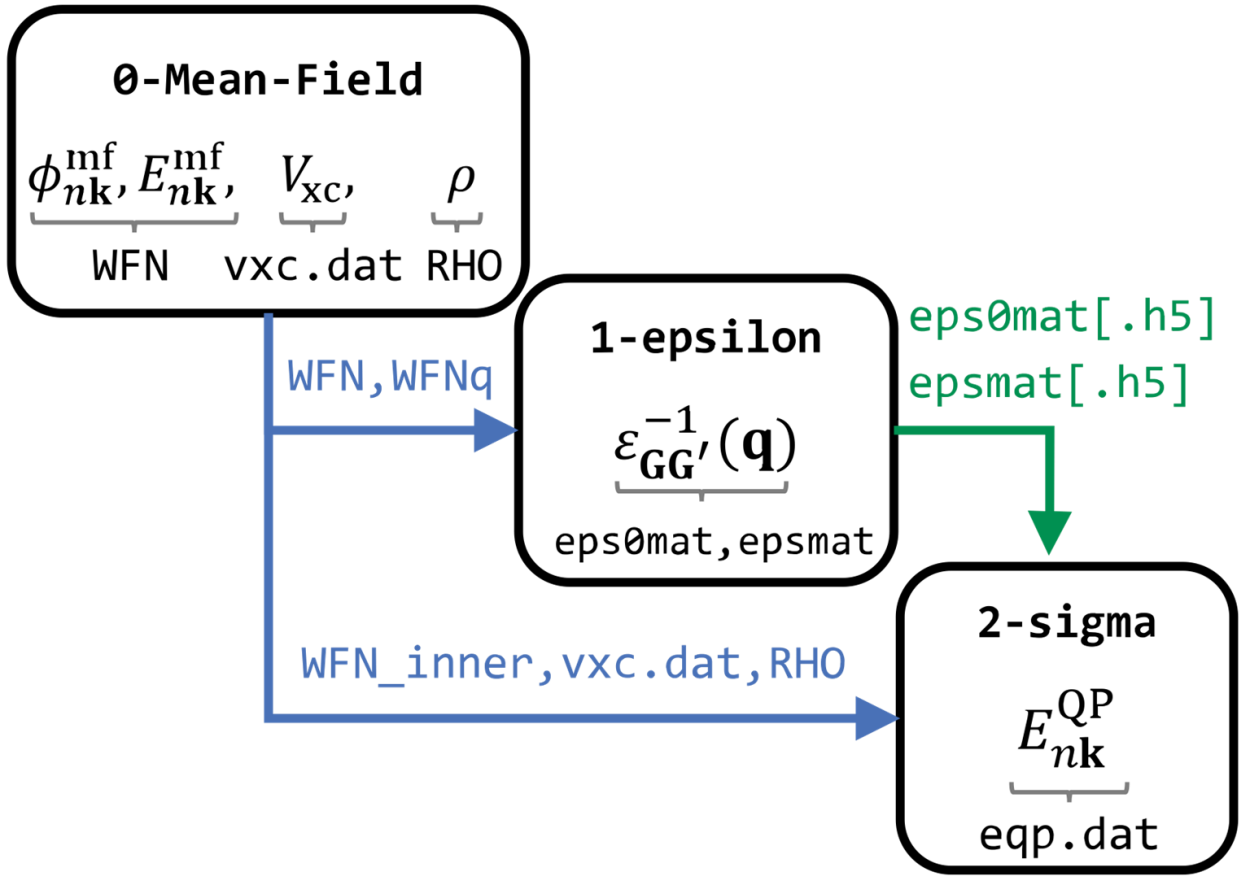
Same for density and V_{xc}, except no need for time-reversal

$$\rho(\mathbf{r}) = \rho^*(\mathbf{r})$$

- What breaks time-reversal? External magnetic field, spin-polarization (FM).
- Plane-wave codes generally just use complex wavefunctions.
- Conditions for reality depends on the basis!

Questions?

1. Introduction
2. K-point vs. Q-point grids
3. Screening models for ε(**q**)
4. Frequency dependence of ε(ω)
5. Symmetry and degeneracy
6. Solving Dyson's equation
7. Real and complex versions



Extra slides:
discussion of half-shifted grids,
calculations for metals

2. Example **k**-grid construction: 4×4 grid for graphene

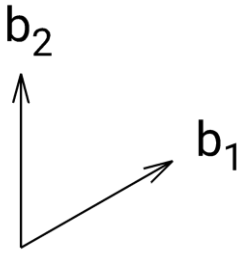
How to construct k-point grid (**WFN**) and q-shifted k-point grids (**WFNq**)?

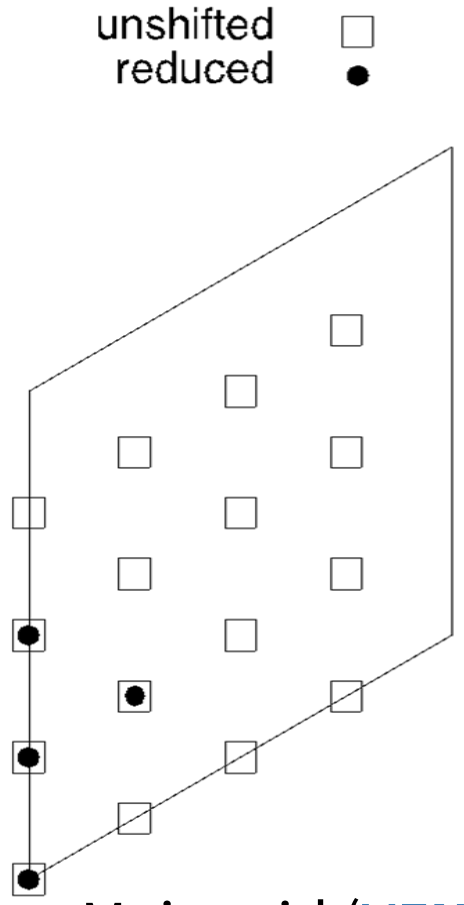
➢ **`kgrid.x`** utility!

Example on the right:

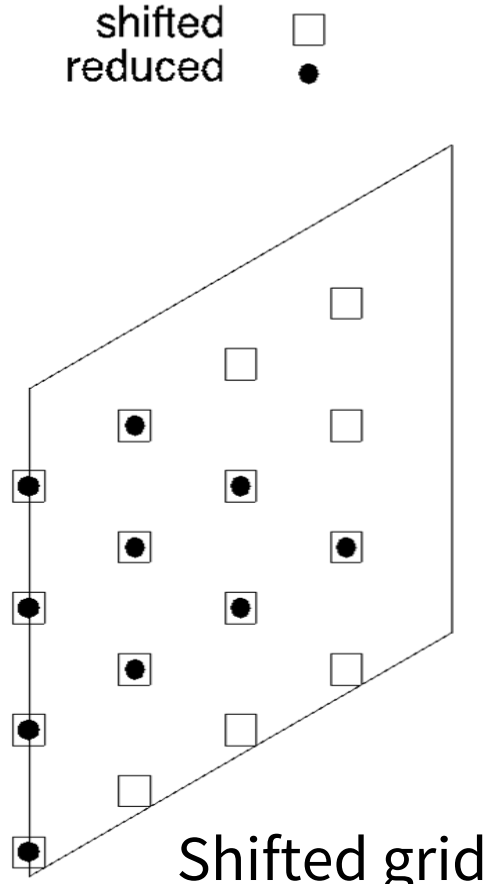
- Graphene
- 4x4x1 Monkhorst-Pack grid
- $\mathbf{q}_0$= (0.0,0.05,0.0)

$\mathbf{q}_0$ shift breaks symmetry and gives more points.





Main grid (**WFN**)
16 in full BZ
Reduced to 4



Shifted grid (**WFNq**)
16 in full BZ
Reduced to 10