BerkeleyGW at NERSC

Jack Deslippe

BERKELEY LAB
Lawrence Berkeley National Laboratory

Part 1: Intro to GW/BSE

DFT Kohn-Sham Formulation

Minimize Energy Functional By Solving Kohn Sham Eqns

Kohn, W.; Sham, L. J. Phys. Rev. A 1965, 140, 1133.

$$\rho(r) = \sum_{nk}^{occ} |\psi_{nk}(r)| \qquad \left[-\frac{\nabla^2}{2} + V_{ion} + V_{Hartree} + V_{xc\,KS} \right] \psi_{nk} = E_{nk\,KS}\, \psi_{nk}$$

Total energy is exact so long as approximation for V_{xc} is good. Commonly use Local Density Approximation (LDA) and Gradient Approximations (GGA) – Hybrid functionals etc...

Interpretation of KS Eigenvalues (states)

Ordinary Kohn-Sham eigenvalues under-estimate the electronic gap

"Band Gap Problem"

Silicon Indirect Gap

50% off in LDA

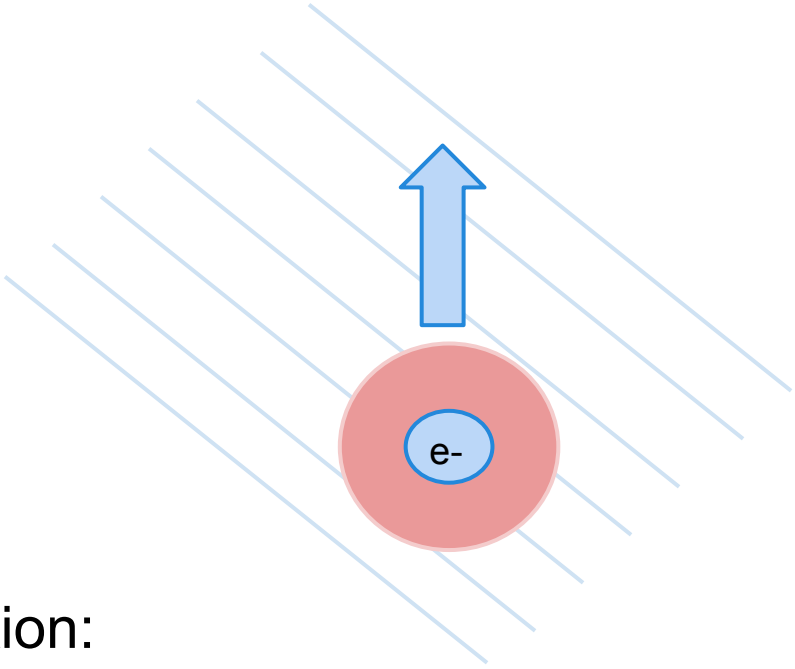
	LDA	Present	Expt.[d]
Diamond			
E_g	3.9	5.6	5.48
$\Gamma_{1v} \rightarrow \Gamma_{25v}$	21.6	23.0	24.2±1
$\Gamma'_{25v} \rightarrow \Gamma_{15c}$	5.5	7.5	7.3
$X_{4v} \rightarrow X_{1c}$	10.8	12.9	12.5
Silicon			
E_g	0.52	1.29	1.17
$\Gamma_{1v} \rightarrow \Gamma'_{25v}$	11.93	12.04	12.5±0.6
$\Gamma'_{25v} \rightarrow \Gamma_{15c}$	2.57	3.35	3.4
$L'_{3v} \rightarrow L_{1c}$	2.73	3.53	3.54
$L'_{3v} \rightarrow L_{3c}$	4.58	5.50	5.51

*Hybertsen, M. S.; Louie, S. G. Phys. Rev. B 1986, 34, 5390.

[17] *Zahlenwerte und Funktionen aus Naturwissenschaften und Technik*, in Vol. III of Landolt-Bornstein (Springer, New York, 1982), pt. 17a.

Quasiparticles

In many systems the Green's Function takes the form of an independent particle with complex Energy E



The Dyson equation:

$$\left[E_{n\mathbf{k}} - H_0(\mathbf{r}) - V_H(\mathbf{r})\right]\psi_{n\mathbf{k}}(\mathbf{r}) - \int \Sigma(\mathbf{r},\mathbf{r}',E_{n,\mathbf{k}})\psi_{n\mathbf{k}}(\mathbf{r}')d\mathbf{r}' = 0$$

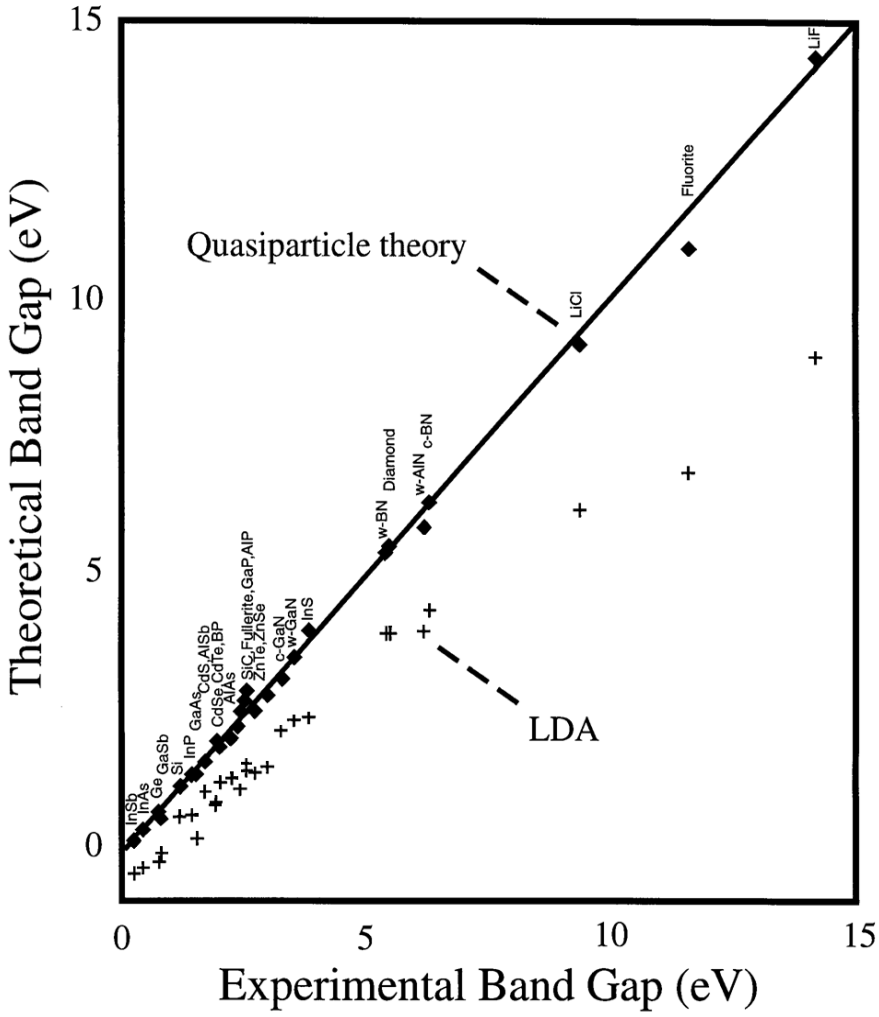
GW Approximation

$$\Sigma = iGW$$

$$W(\mathbf{q},\mathbf{G},\mathbf{G}') = \varepsilon^{-1}(\mathbf{q},\mathbf{G},\mathbf{G}') \cdot V(\mathbf{q}+\mathbf{G}')$$

(L. Hedin. Phys. Rev. 139, A796 (1965))
(L. Hedin, S. Lundquist. Solid State Physics 23, 1 (1969))
M. S. Hybertsen, S. G. Louie, Phys. Rev. Lett. 55 (1985) 1418.

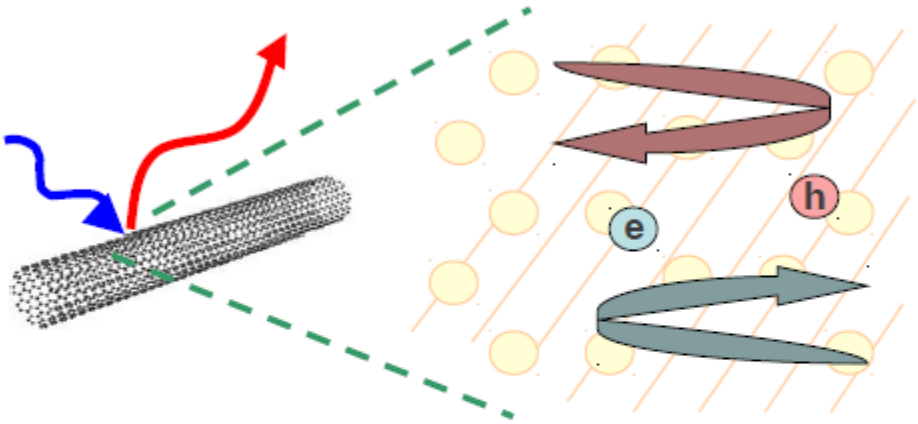
Materials:
InSb, InAs, Ge, GaSb, Si, InP, GaAs, CdS
AlSb, AlAs, CdSe, CdTe, BP, SiC, C_{60}, GaP
AlP, ZnTe, ZnSe, c-GaN, w-GaN, InS, w-BN, c-BN
Diamond, w-AlN, LiCl, Fluorite, LiF

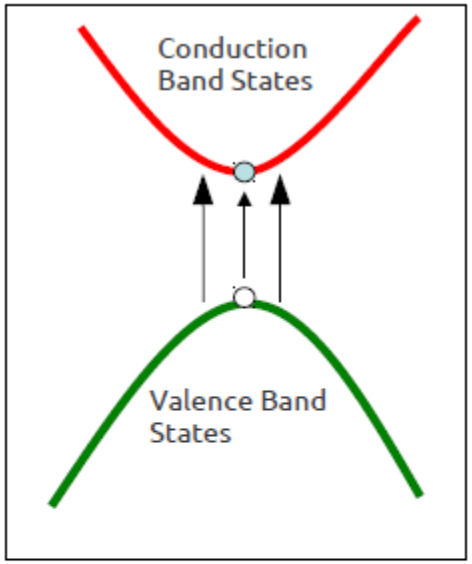


After S.G. Louie in *Topics in Computational Materials Science* (World Scientific, 1997)

Excitons and the Bethe-Salpeter Equation

$$|N,S\rangle = \sum_{v}^{hole} \sum_{c}^{elec} A_{vc}^{S} a_{v}^{+} b_{c}^{+} |N,0\rangle + \ldots$$





$$\left(E_{ck}^{QP} - E_{vk}^{QP}\right) A^{S}{}_{vck} + \sum_{k'v'c'} \langle vck | K^{eh} | v'c'k' \rangle A^{S}{}_{v'c'k'} = \Omega^{S} A^{S}{}_{vck}$$

$$\varepsilon_2(\omega) = \frac{16\pi^2 e^2}{\omega^2} \sum_{S} |\langle N,0 | e \cdot v | N,S \rangle|^2 \delta(\Omega_S - \hbar\omega)$$

GW/BSE Method

DFT Kohn/Sham
$\{\varphi^{DFT}_{nk}(\mathbf{r}), \varepsilon^{DFT}_{nk}\}$

↓

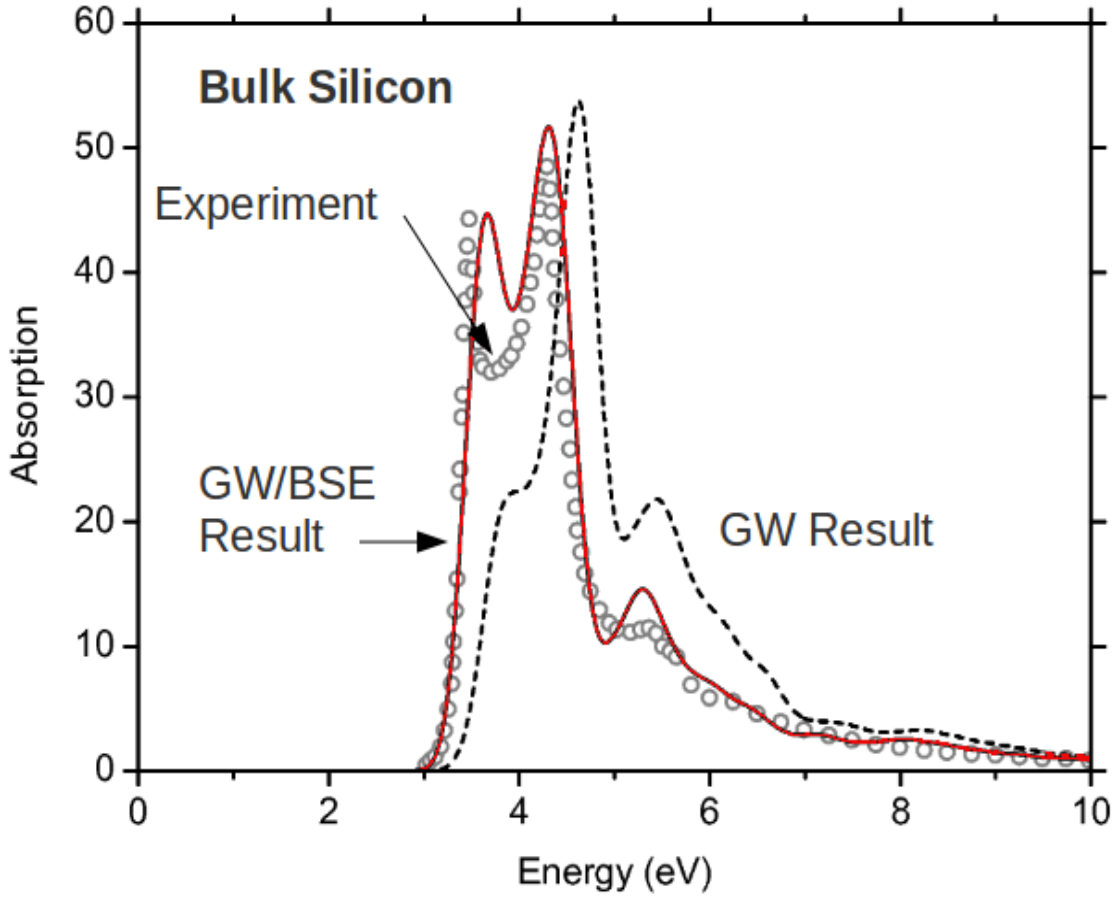
Σ $\{\varphi^{QP}_{nk}(\mathbf{r}), \varepsilon^{QP}_{nk}\}$

↓

Construct Bethe-Salpeter
Kernel - K(k,c,v,k',c',v')

↓

Diagonalize BSE
$\{A^{s}_{cvk}, E^{s}_{cvk}\}$



Expt. G.E. Jellison, M.F. Chisholm, S.M. Gorbatkin, Appl. Phys. Lett. 62, 3348 (1993).

GW Reputation

The Good:

-Quantitatively accurate for quasiparticle properties in a wide variety of systems.

The Bad:

-Prohibitively slow for large systems. Usually thought to cost orders of magnitude more time that DFT.

-Memory intensive and scales badly. Limited ~50 atoms.

GW Reputation

The Good:

-Quantitatively accurate for quasiparticle properties in a wide variety of systems.

The Bad:

-Prohibitively slow for large systems. Usually thought to cost orders of magnitude more time that DFT.

-Memory intensive and scales badly. Limited ~50 atoms.

BerkeleyGW on NERSC resources scales to 10'000+ cores and can handle systems with hundreds of atoms

Part 2: Using BerkeleyGW at NERSC

Resources and Getting Started

Resources:

http://www.berkeleygw.org

http://arxiv.org/abs/1111.4429

https://www.nersc.gov/users/software/applications/materials-science/berkeleygw

NERSC BGW Example:

```
% module load berkeleygw/1.0.3

% ls $BGW_EXAMPLES/NERSC_silicon
```

NERSC Example

There is a NERSC ready silicon example in the berkeleygw module on hopper.

```
% module load berkeleygw/1.0.3

% ls $BGW_EXAMPLES/NERSC_silicon

07-epsilon  08-sigma  09-kernel  10-absorption  ESPRESSO  README  script_3  test_bgw.pbs


% cp -r $BGW_EXAMPLES/NERSC_silicon $SCRATCH/

% cd $SCRATCH/NERSC_silicon

% qsub test_bgw.pbs
```

The GW Starting Point

$$[E_{n\mathbf{k}} - H_0(\mathbf{r}) - V_H(\mathbf{r})]\,\psi_{n\mathbf{k}}(\mathbf{r}) - \int \Sigma(\mathbf{r},\mathbf{r}',E_{n,\mathbf{k}})\psi_{n\mathbf{k}}(\mathbf{r}')d\mathbf{r}' = 0$$

When solved self-consistently, Dyson's equation independent of starting point.

Diagonal approximation

$$\varepsilon^n_{GW} \approx \varepsilon^n_{MF} + \langle \phi^n_{MF} | \Sigma - V_{xc} | \phi^n_{MF} \rangle$$

Accuracy depends MF wavefunction and operator.

Quantum ESPRESSO

Typically use
DFT as the Mean-Field
starting point for BGW.

Supported codes include PARATEC, SIESTA, Octopus, **Quantum ESPRESSO**

+more coming soon

Step 1: QE SCF

Every GW calculations starts with the calculation of the charge density through an SCF calculation (e.g. DFT+LDA)

```
% ls $SCRATCH/NERSC_silicon/ESRESSO/01-scf

in  out  reference  silicon.save  Si.UPF
```

Inputs: pseudopotential in ("calculation = 'scf'")

Rel. Outputs: silicon.save/charge-density.dat silicon.save/data-file.xml

Considerations for QE SCF

By default QE divides up the Plane Waves among processors.

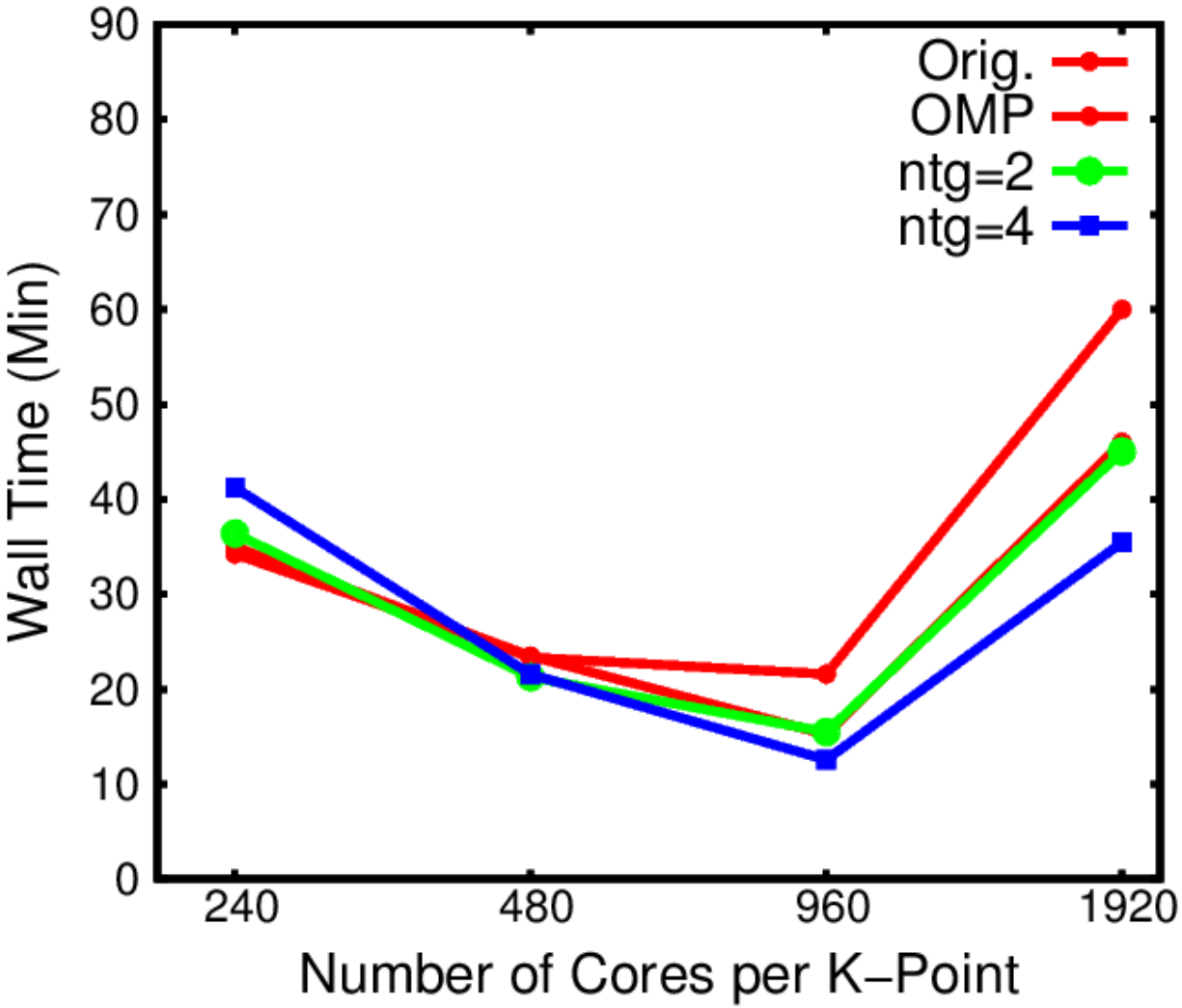
(ncores = # planes in the FFT Grid is limit of good scaling). To go beyond that:

-npools - Divide processors into pools over kpts

-ntg - Divide processors into pools for doing multiple FFTs at once.

-OpenMP - Add's more scalability

QE Performance on Hopper (1000 atom Supercell)



Step 2: QE NSCF

We need to generate wavefunction files with many empty bands ~ 10-100 x (# of occupied bands) on a few different grids.

```
% ls $SCRATCH/NERSC_silicon/ESRESSO/04-wfn-co

in  out  pp_in  pp_out  reference  rho.real  silicon.save  Si.UPF  vxc.dat  wfn.real
```

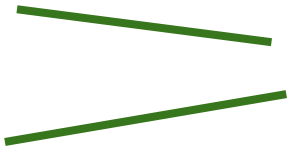
Inputs: pseudopotential charge-density.dat data-file.xml in ("calculation = 'bands'") pp.in

Outputs: wfn.real rho.real vxc.dat

Considerations for QE NSCF

Need Wavefunctions on 4 K-Grids:

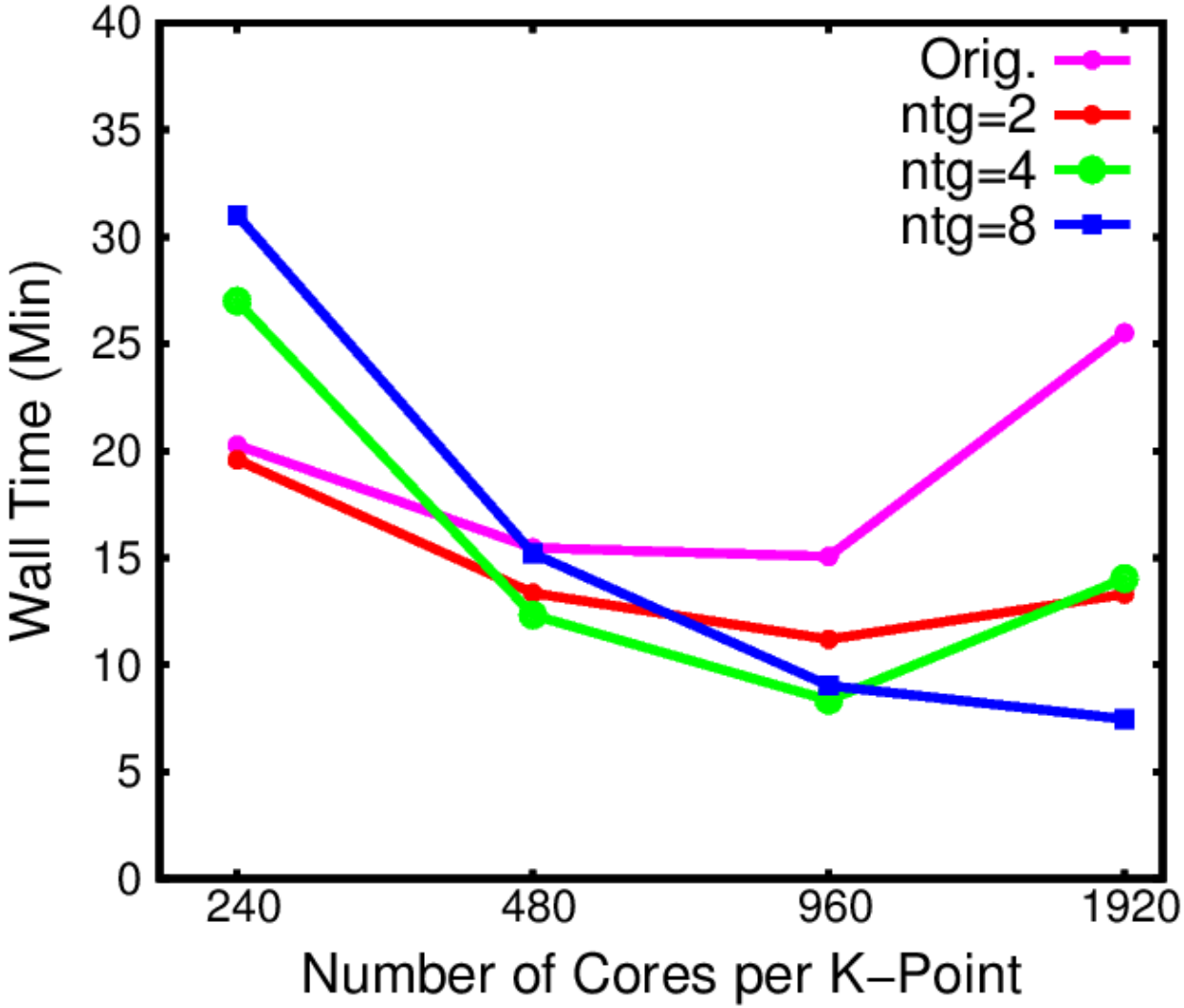
1. Unshifted Coarse Grid
2. Shifted Coarse Grid
3. Unshifted Fine Grid
4. Shifted Fine Grid
   (velocity Op. Only)



For BSE (optics)

Shifted Grids are required to compute the the q = k - k' ~ 0 limit of various quantities.

QE NSCF (1000 Bands) Performance on Hopper



Step 3 BGW Epsilon

After generating the required wavefunctions from Quantum ESPRESSO, we calculate the dielectric matrix for the material.

```
% ls $SCRATCH/NERSC_silicon/07-epsilon

chi_converge.dat  eps0mat  epsilon.inp  epsilon.log  epsmat  OUT.eps  reference  WFN  WFNq
```

Inputs: WFN WFNq epsilon.inp

Rel. Outputs: epsmat, eps0mat, epsilon.log chi_converge.dat

Step 3 BGW Epsilon

epsilon.log:

q= 0.0000 0.0000 0.0010 nmtx= 137 nind= 0 nrq=110

independent matrix elements of chi spin index= 1 1

g			g**2	gp			gp**2	chi(g,gp)
0	0	0	0.00000	0	0	0	0.00000	-0.0000005825
0	0	0	0.00000	-1	-1	-1	1.12482	-0.0000114117
0	0	0	0.00000	-1	0	0	1.12482	-0.0000116730

....

g			gp			inverse epsilon	nspin= 1
0	0	0	0	0	0	0.07959121	
-1	-1	-1	0	0	0	-0.00001097	
-1	-1	-1	-1	-1	-1	0.59728014	
-1	0	0	-1	-1	-1	0.00426155	

Step 3 BGW Epsilon

$$\chi_{\mathbf{GG'}}(\mathbf{q};0) = \sum_{n}^{\text{occ}} \sum_{n'}^{\text{emp}} \sum_{\mathbf{k}} M_{n,n'}(\mathbf{k},\mathbf{q},\mathbf{G}) \cdot M^{*}_{n',n}(\mathbf{k},\mathbf{q},\mathbf{G'}) \cdot \ldots$$

---

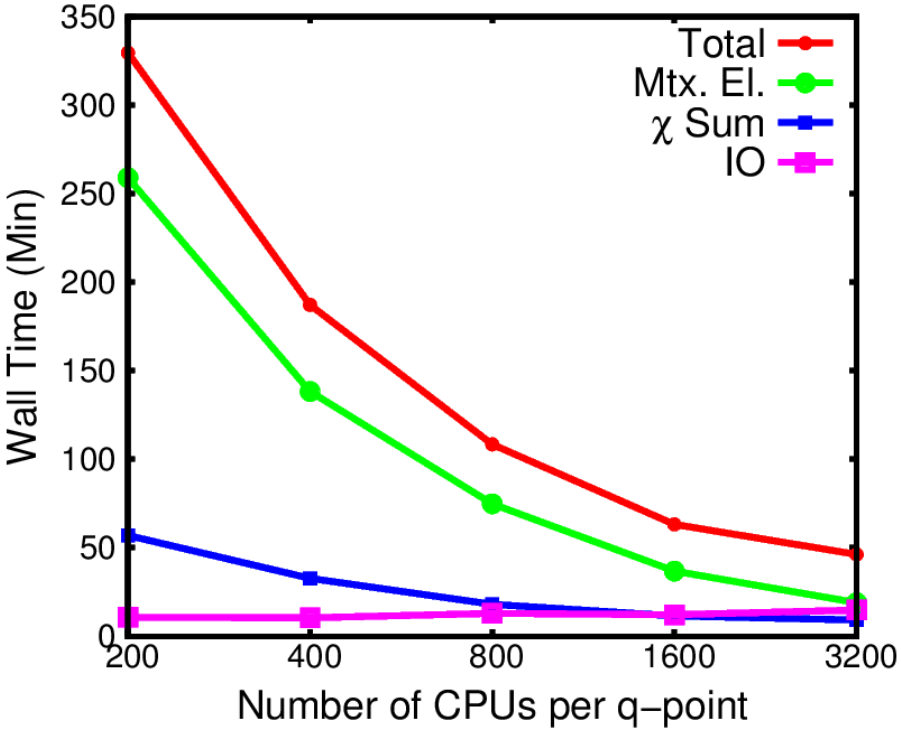
1. $M_{n,n'}(\mathbf{k},\mathbf{q},\mathbf{G}) = \langle n\mathbf{k}+\mathbf{q}| \, e^{i(\mathbf{q}+\mathbf{G})\cdot\mathbf{r}} \, |n'\mathbf{k}\rangle$

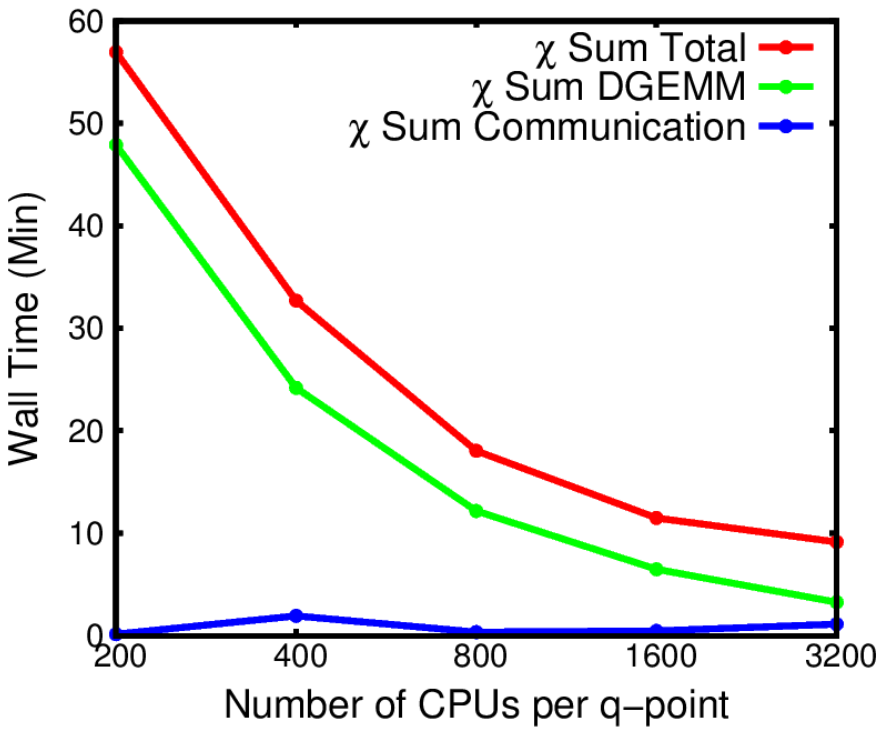
FFTW Scaling – N^3 ln(N)
Parallel scaling – N x ln (N) (parallel over n,n')

2. Sum: $\chi_{\mathbf{GG'}} = \mathbf{M}(\mathbf{G},(n,n',\mathbf{k})) * \mathbf{M}^{T}((n,n',\mathbf{k}),\mathbf{G'})$

Distributed over **G**,**G'**
Scales N^4 - Smaller prefactor
Level 3 Blas

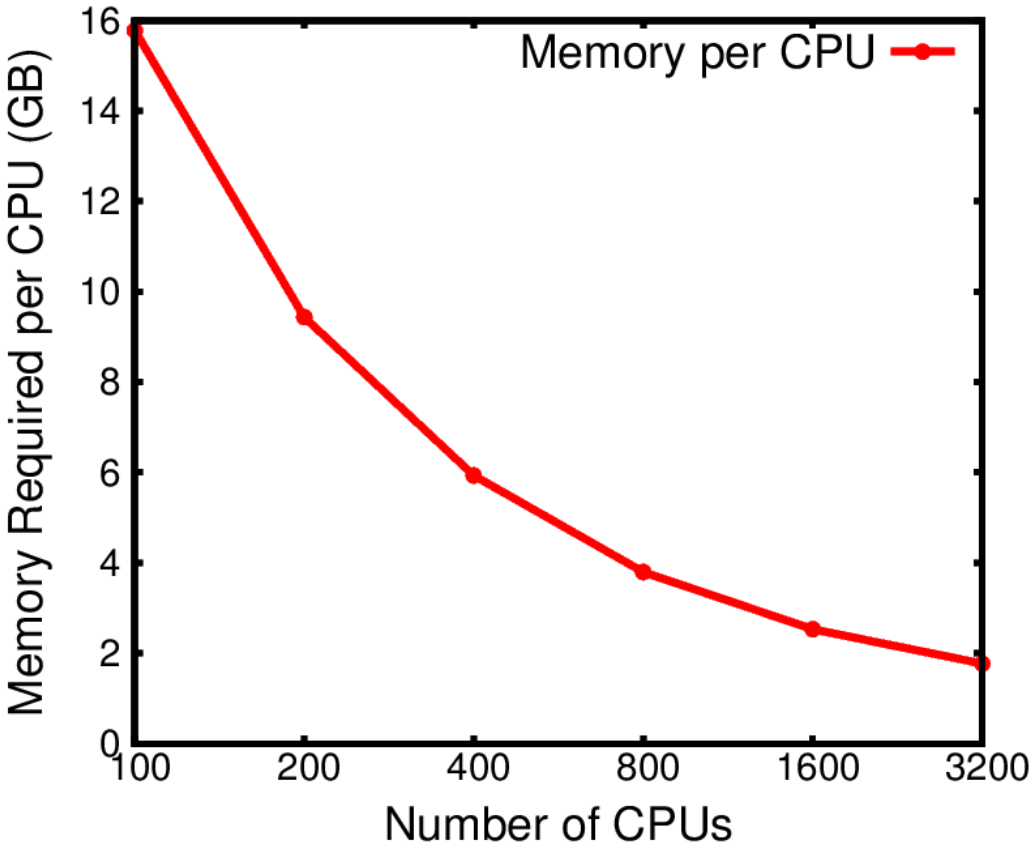
Epsilon Performance Considerations





For best use of resources. Choose a number of cores that evenly divides NC*NV.
NC = # of conduction bands ; NV = # of valence bands

Epsilon Memory Considerations



Trick:

Use less than 24 MPI tasks per node on Hopper to give each task more memory

BGW 1.1 Allows use of OpenMP Threading - so all cores on node can be used for computation, while still giving each MPI task a large amount of memory.

Release date "soon". Contact me if you have an application that needs this.

Step 4 BGW Sigma

We compute diagonal (optionally off-diagonal) elements of the self-energy operator.

$$\varepsilon^{n}_{GW} \approx \varepsilon^{n}_{MF} + \langle \phi^{n}_{MF} | \Sigma - V_{xc} | \phi^{n}_{MF} \rangle$$

```
% ls $SCRATCH/NERSC_silicon/08-sigma

ch_converge.dat  eps0mat  epsmat  OUT.sig  reference  RHO  sigma_hp.log  sigma.inp  sigma.log
vxc.dat  WFN_inner  x.dat
```

Inputs: WFN epsmat eps0mat vxc.dat RHO

Outputs: sigma.log ch_converge.dat

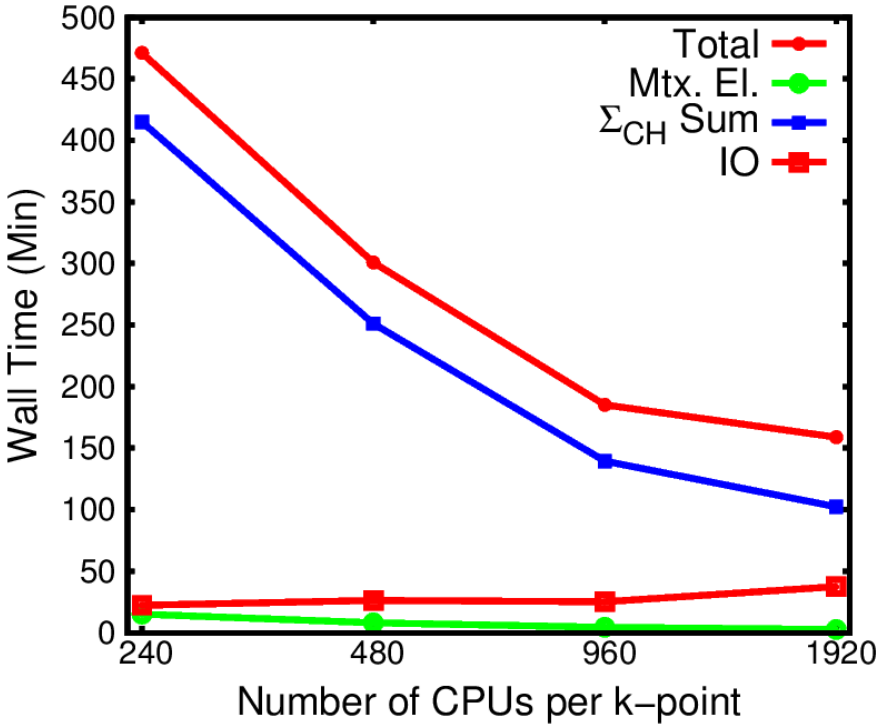
Step 4 BGW Sigma

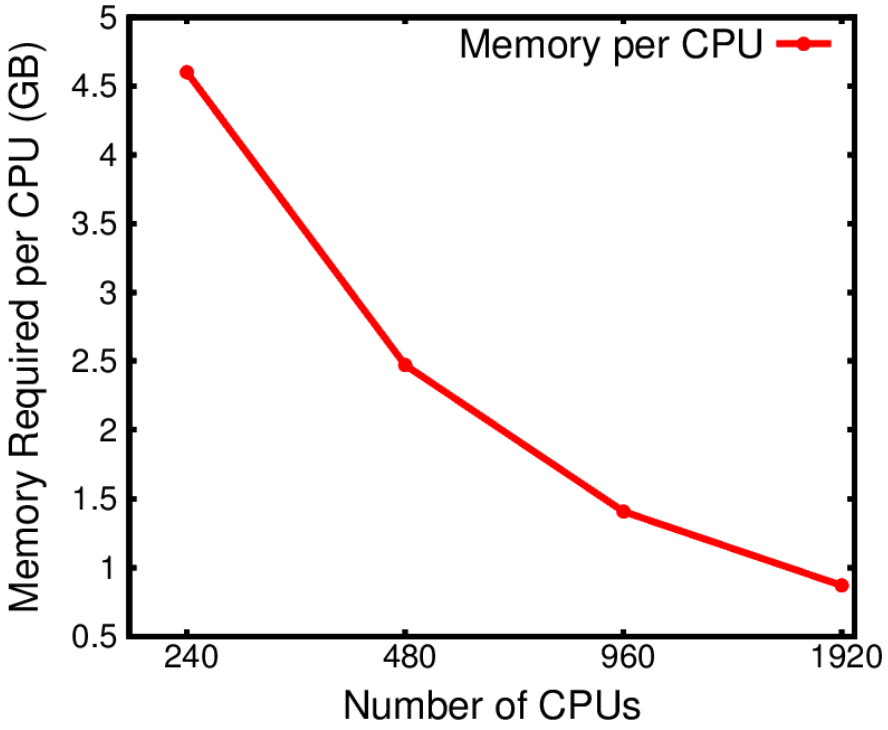
sigma.log

k = 0.000000 0.000000 0.000000 ik = 1 spin = 1

n	elda	ecor	x	sx-x	ch	sig	vxc	eqp0	eqp1	Znk
1	-5.896	-5.896	-17.221	13.356	-7.061	-10.926	-10.447	-6.375	-6.220	0.677
2	6.007	6.007	-12.604	9.015	-8.038	-11.627	-11.250	5.630	5.707	0.794
3	6.007	6.007	-12.604	9.015	-8.038	-11.627	-11.250	5.630	5.707	0.794
4	6.007	6.007	-12.604	9.015	-8.038	-11.627	-11.250	5.630	5.707	0.794
5	8.567	8.567	-5.746	3.941	-7.701	-9.505	-10.047	9.109	8.993	0.786
6	8.567	8.567	-5.746	3.941	-7.701	-9.505	-10.047	9.109	8.993	0.786
7	8.567	8.567	-5.746	3.941	-7.701	-9.505	-10.047	9.109	8.993	0.786
8	9.299	9.299	-5.943	4.393	-8.766	-10.317	-10.833	9.814	9.701	0.780
9	13.710	13.710	-2.686	1.996	-6.963	-7.653	-8.089	14.146	14.049	0.778
10	13.797	13.797	-4.294	3.045	-7.817	-9.066	-10.063	14.794	14.566	0.772
11	13.797	13.797	-4.294	3.045	-7.817	-9.066	-10.063	14.794	14.566	0.772
12	17.165	17.165	-2.300	1.859	-7.517	-7.958	-8.537	17.744	17.592	0.738
13	17.165	17.165	-2.300	1.859	-7.517	-7.958	-8.537	17.744	17.592	0.738
14	17.165	17.165	-2.300	1.859	-7.517	-7.958	-8.537	17.744	17.592	0.738

Sigma Performance Considerations





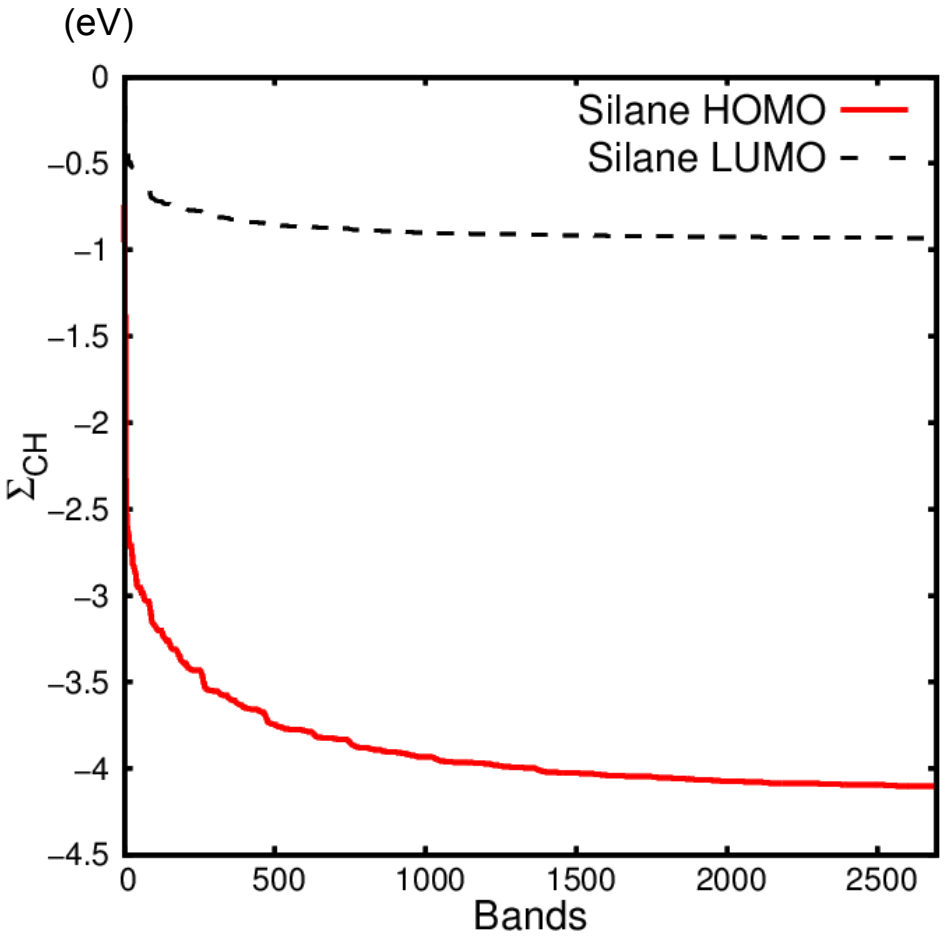
Choose number of cores that divides the total number of occupied and unoccupied bands multiplied by the number of bands at which you wish to compute sigma.

Sigma Convergence

Sigma may converge very slowly with respect to the number of empty orbitals provided.

It is very important to look at the following file to test convergence:

ch_converge.dat



Step 5 BGW Kernel

Compute the Electron Hole Kernel Matrix

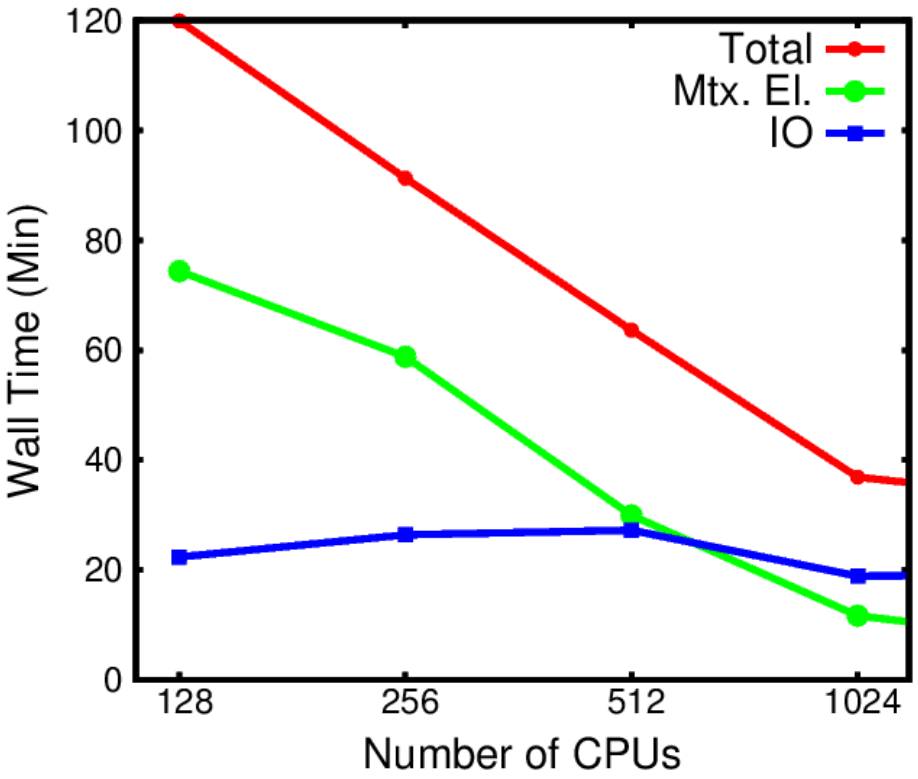
$$\left(E_{ck}^{QP} - E_{vk}^{QP}\right) A^{S}_{vck} + \sum_{k'v'c'} \langle vck | K^{eh} | v'c'k' \rangle A^{S}_{v'c'k'} = \Omega^{S} A^{S}_{vck}$$

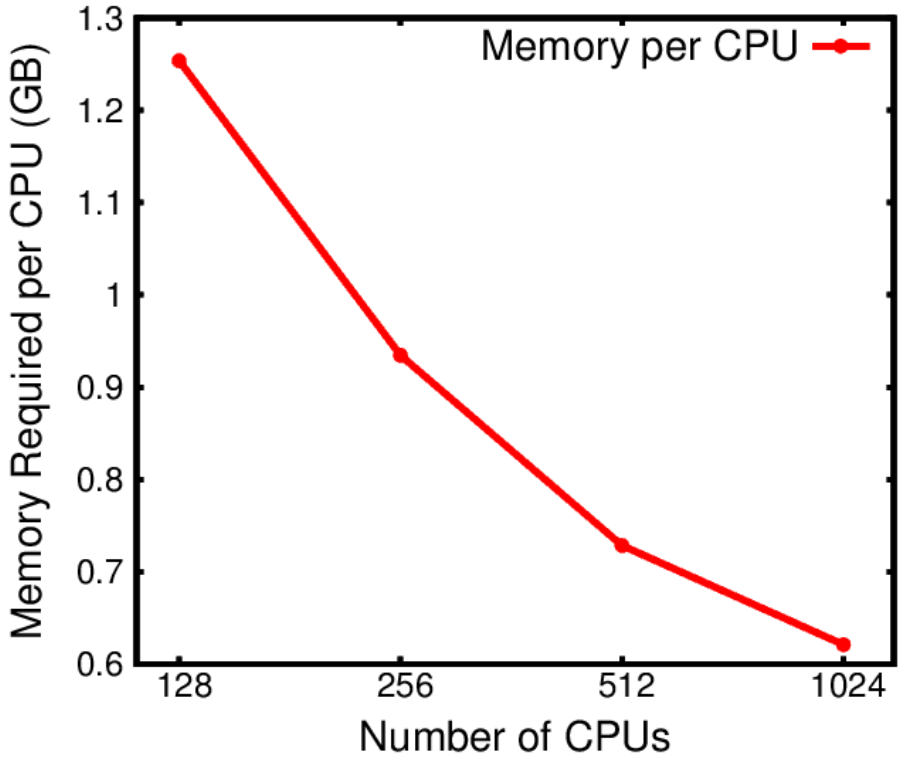
```
% ls $SCRATCH/NERSC_silicon/09-kernel

bsedmat  bsexmat  eps0mat  epsmat  kernel.inp  OUT.krn  reference  WFN_co
```

Inputs: epsmat eps0mat WFN_co

Outputs: bsedmat bsexmat

Step 5 Kernel Considerations





For best performance: choose a number of cores = nk^2 or nk^2 * nc^2 and provide low_comm in kernel.inp

Changes in BGW 1.1: Parallel HDF5 I/O. Threads. Better scaling beyond nproc = nk^2

Step 6 Absorption

Calculate the absorption spectra with and without the electron-hole interaction included.

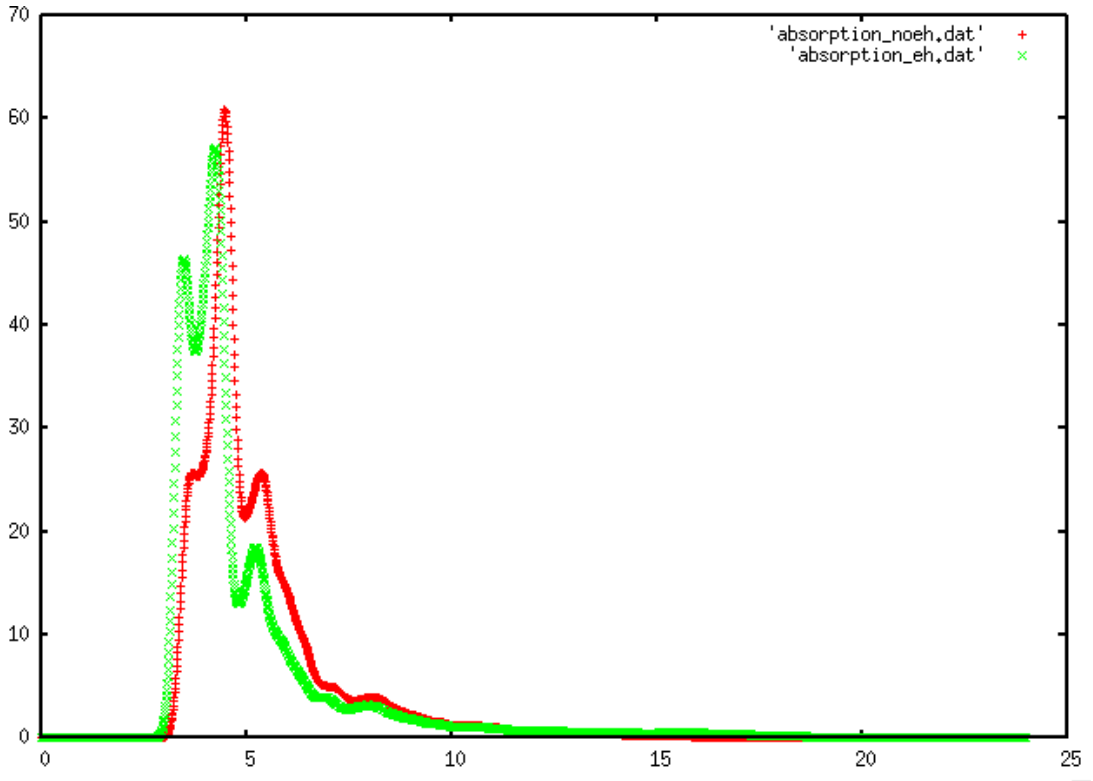
```
% ls $SCRATCH/NERSC_silicon/10-absorption

absorption_eh.dat    bandstructure.dat  dcmat_norm.dat  eigenvalues.dat  epsmat      OUT.abs
WFN_co absorption.inp       bsedmat           dtmat           eps0mat          eqp_co.dat  reference
WFN_fi absorption_noeh.dat  bsexmat           dvmat_norm.dat  epsdiag.dat      eqp.dat     vmtxel
```

Inputs: eps0mat epsmat bsedmat bsexmat WFN_co WFN_fi

Outputs: absorption_eh.dat absorption_noeh.dat eivenvalues.dat eigenvectors.dat dvmat_norm.dat

Step 6 Absorption

```
% module load gnuplot
% gnuplot
gnuplot> plot 'absorption_noeh.dat', 'absorption_eh.dat'
```



Absorption Considerations

Check norms from interpolation (dvmat_norm.dat)

By default, absorption uses scalapack to do exact diagonalization. Choose a large square number of cores.

For larger system choose the haydock iterative scheme.

More Information:

Example Input Files for All Codes:

```
% module load berkeleygw/1.0.3

% ls $BGW_DOCS/NERSC_silicon

absorption.inp  epsilon.inp      epsmat_merge.inp  kernel.inp          plotxct.inp  sigma.inp
wfnmix_QSGW.inp epsconv.inp      epsmat_intp.inp  inteqp.inp          nonlinearoptics.inp  sig2wan.inp
summarize_eigenvectors.inp
```

View the Source Code and READMEs

```
% ls /usr/common/usg/berkeleygw/1.0.3/source
```

Visit - **http://www.berkeleygw.org**

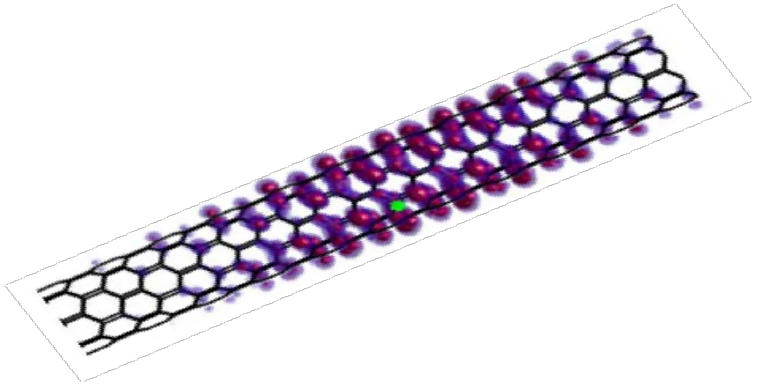
Extra Slides

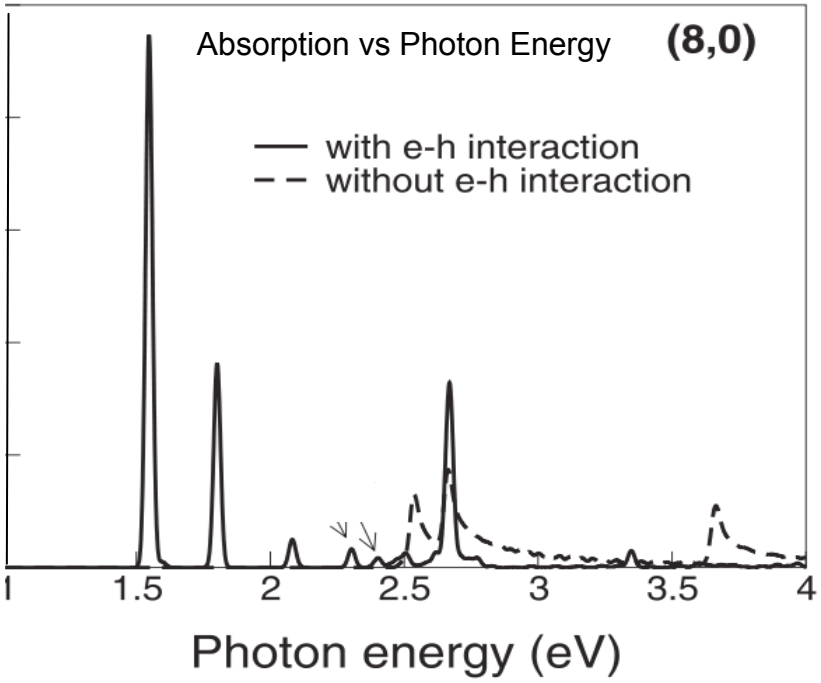
BERKELEY LAB
Lawrence Berkeley National Laboratory

GW-BSE in Nano.

Example – SWCNT: GW-BSE predicts exciton binding energies as large as 1 eV in semiconducting tubes. 100 meV in metallic tubes*

Each interband transition gives rise to exciton complex 1u, 2g, 3u ...





***C.D. Spataru, S. Ismail-Beigi, L.X. Benedict, S.G. Louie. PRL 077402 (2004)**

***J. Deslippe, C.D. Spataru, D. Prendergast, S.G Louie. Nano Letters. 7 1626 (2007)**