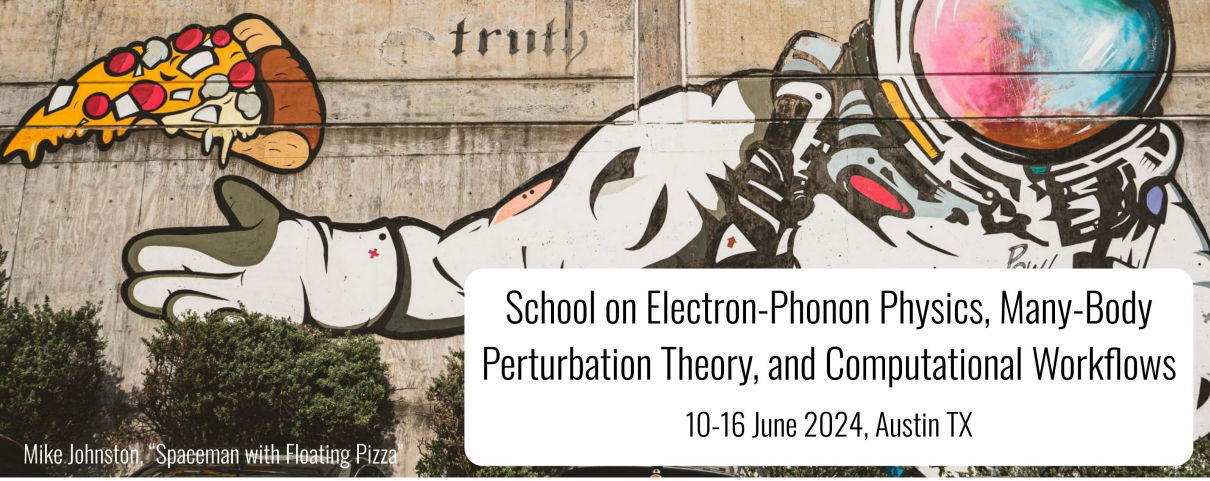

Mike Johnston, "Spaceman with Floating Pizza"

School on Electron-Phonon Physics, Many-Body Perturbation Theory, and Computational Workflows

10-16 June 2024, Austin TX

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Tutorial intro Fri.7

The Special Displacement Method

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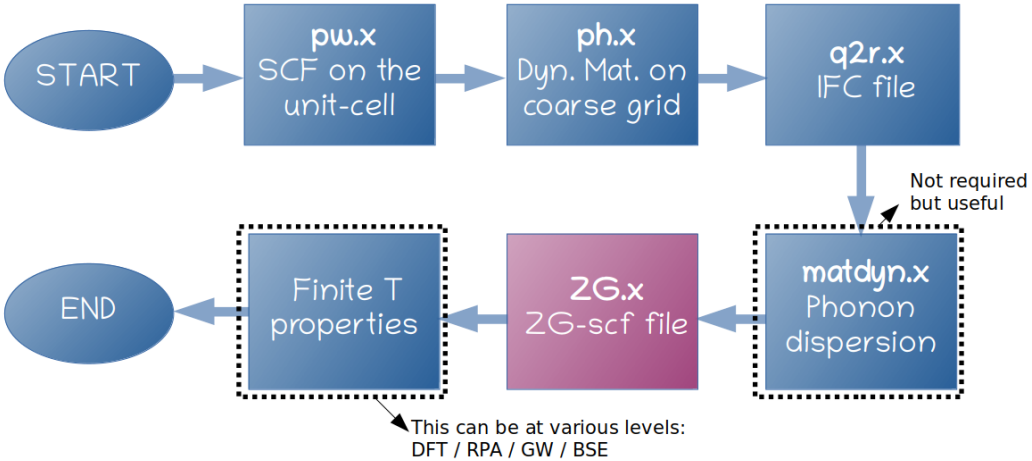
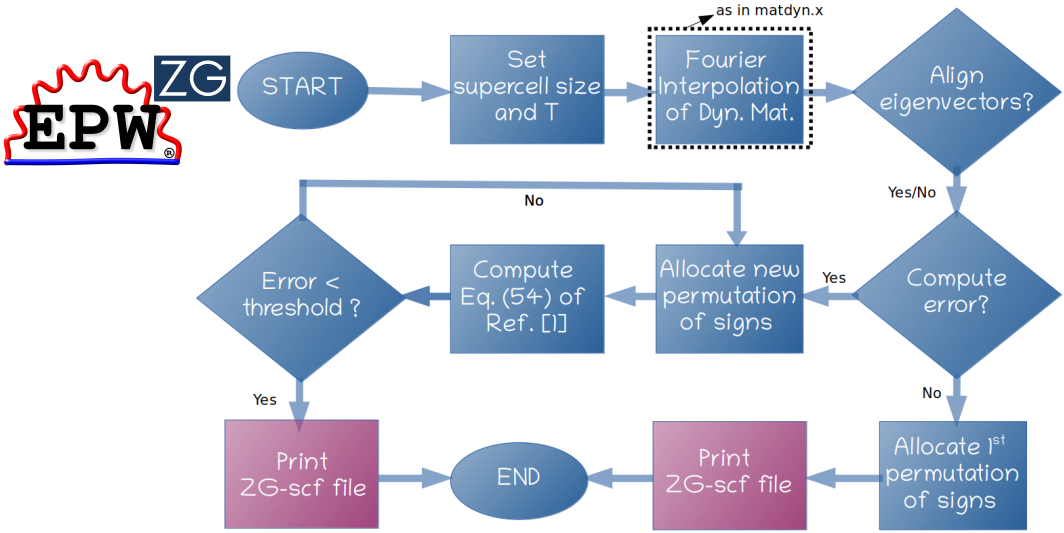
Instructions

- The tutorial is long so you are expected to run the first three exercises.
- If you have used `ZG.x` before, you can go directly to the last three exercises: `exerciseC`, `exercisePbTe`, and `exerciseCsPbBr3`.
- For help please ask the TAs or myself.

Flowchart for ab-initio calculations with ZG configurations



Flowchart for `ZG.x` to generate ZG-configuration



Ref. [1]: M. Zacharias, F. Giustino, Phys. Rev. Research 2, 013357 (2020)

Compute and minimize the function $E(\{S_{\mathbf{q}\nu}\}, T)$

Find the best ZG displacements for a given *supercell size* and *temperature* by setting
`compute_error = .true.`, `error_thresh = 0.2`
so that the function:

$$E(\{S_{\mathbf{q}\nu}\}, T) = \sum_{\substack{\kappa\alpha \\ \kappa'\alpha'}} \frac{\left| \sum_{\substack{\mathbf{q}\in\mathcal{B} \\ \nu<\nu'}} \Re[e^*_{\kappa\alpha,\nu}(\mathbf{q}) e_{\kappa'\alpha',\nu'}(\mathbf{q})] \sigma_{\mathbf{q}\nu,T} \sigma_{\mathbf{q}\nu',T} S_{\mathbf{q}\nu} S_{\mathbf{q}\nu'} \right|}{\left| \sum_{\substack{\mathbf{q}\in\mathcal{B} \\ \nu}} \Re[e^*_{\kappa\alpha,\nu}(\mathbf{q}) e_{\kappa'\alpha',\nu}(\mathbf{q})] \sigma^2_{\mathbf{q}\nu,T} \right|}$$

is lower than `error_thresh` based on the choice of $\{S_{\mathbf{q}\nu}\}$.

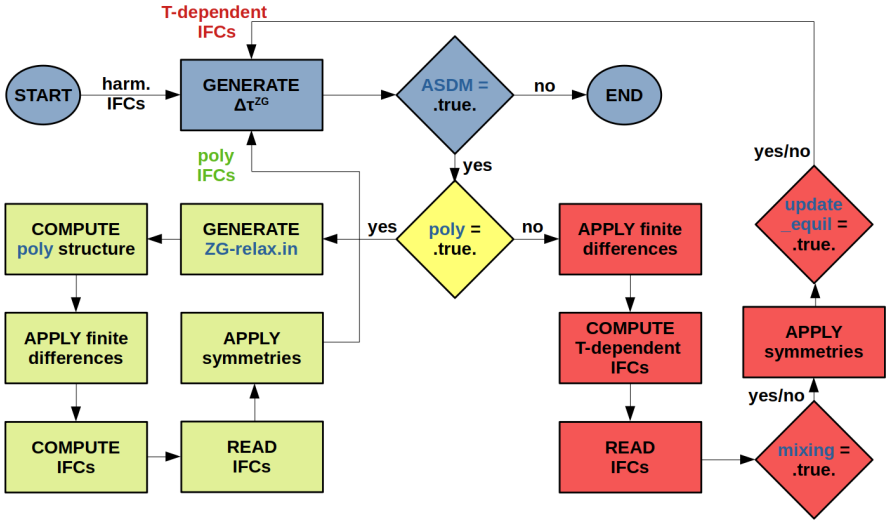
All quantities in $E(\{S_{\mathbf{q}\nu}\})$ can be computed from DFPT;
no extra DFT calculations are required to find the optimum ZG configuration.

Example input file for `ZG.x` (similar structure to `matdyn.x`)

```
--
&input
  flfrc='si.444.fc',
  asr='simple', amass(1)=28.0855, atm_zg(1) = 'Si',
  flscf = 'si.scf.in'
  T = 0.00,
  dim1 = 5, dim2 = 5, dim3 = 5
  compute_error = .true., synch = .true., error_thresh = 0.2
  incl_qA = .false.
/
```

Tutorials and input flags available in `https://docs.epw-code.org/doc/`.

Appendix: Flowchart for the A-SDM in ZG.x



M. Zacharias, G. Volonakis, F. Giustino, J. Even Phys. Rev. B 108, 035155 (2023)

Inputs for ZG.x (A-SDM)

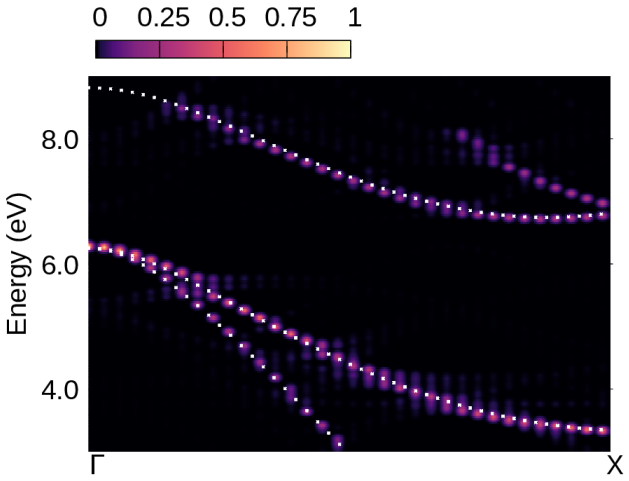
```
&input                                                              ZG_4
    flfrc='poly_iter_00.fc', flscf = 'scf.in', asr='crystal',
    amass(1)=132.90545, amass(2)=207.2, amass(3)= 79.904,
    atm_zg(1) = 'Cs', atm_zg(2) = 'Pb', atm_zg(3) = 'Br'
    T = 380.00,
    dim1 = 2, dim2 = 2, dim3 = 2
    compute_error = .true., synch = .true., error_thresh = 0.5
    ASDM = .true., na_ifc = .true.
/
&A_ZG
   iter_idx = 1
   apply_fd = .true.
   incl_epsil = .true.
/
```

Note: T = 380 K, flfrc = 'poly_iter_00.fc' (computed from iter 0), and na_ifc = .true. for nonanal contrib

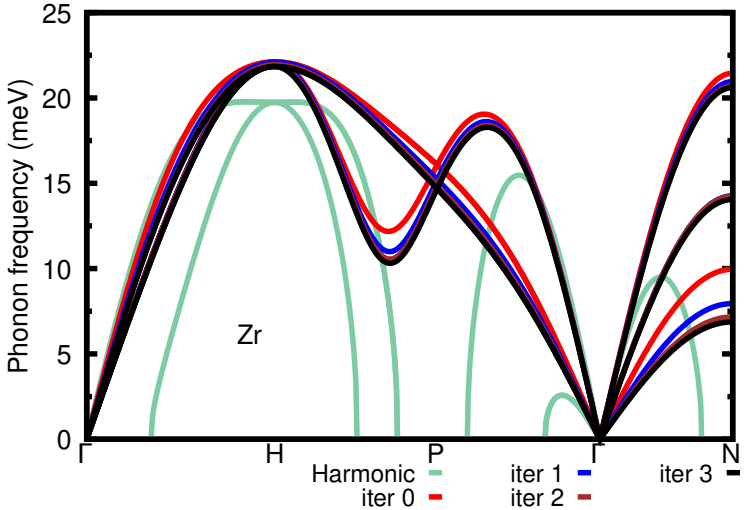
Tutorial Summary

- **Exercise 1**: ZG displacements in silicon and total energy via SDM
- **Exercise 2**: Zero-point band gap renormalization of silicon and temperature dependent band structure via band structure unfolding
- **Exercise 3**: Anharmonic lattice dynamics of Zr via A-SDM
- **Exercise 4**: Phonon-assisted spectra of silicon with ZG displacements
- **Exercise 5**: Phonon-induced diffuse scattering in graphene
- **Exercise 6**: Phonon dispersions of P-doped silicon using phonon unfolding
- **Exercise C**: T-dependent free energy and direct gap of diamond up to 1000 K.
- **Exercise PbTe**: Anharmonic phonons and band gap renormalization of PbTe.
- **Exercise $CsPbBr_3$**: Anharmonic phonons and band gap renormalization of monomorphous and polymorphous $CsPbBr_3$.

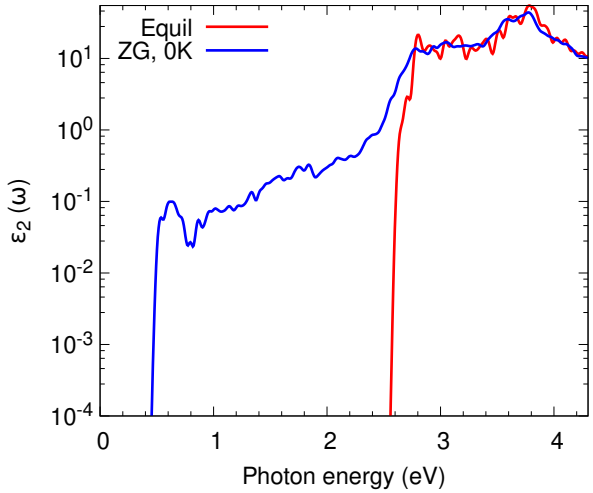
Exercise 2



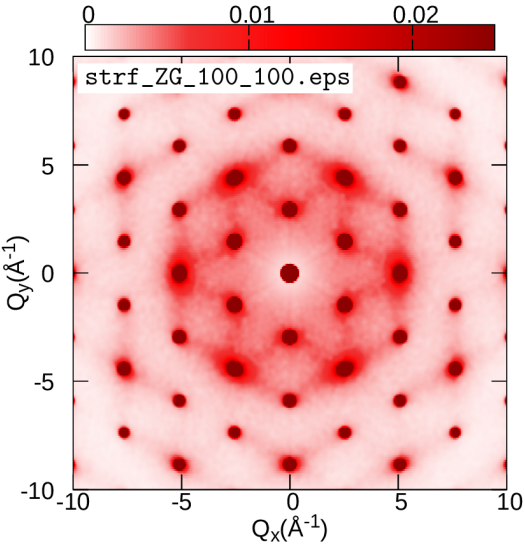
Exercise 3


Phonon frequency (meV)
25
20
15
10
5
0
Γ
H
P
Γ
N
Zr
Harmonic
iter 0
iter 1
iter 2
iter 3

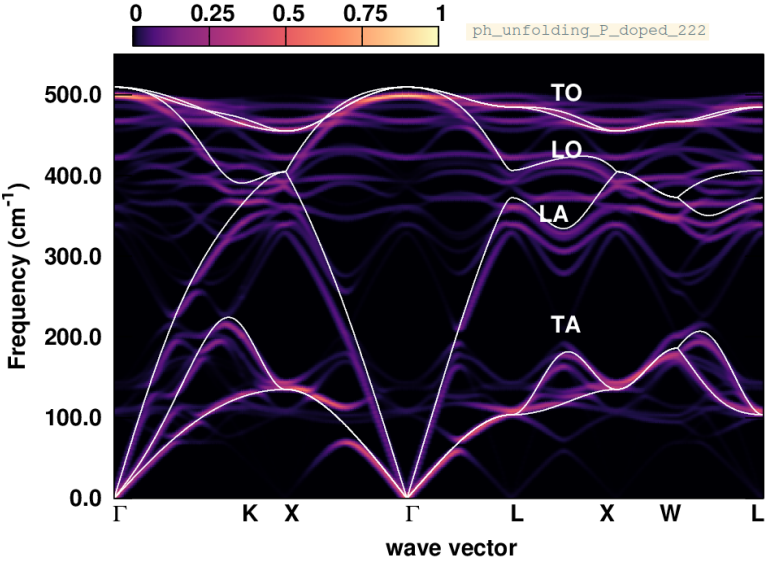
Exercise 4


Equil
ZG, 0K
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
$\varepsilon_2(\omega)$
0
1
2
3
4
Photon energy (eV)

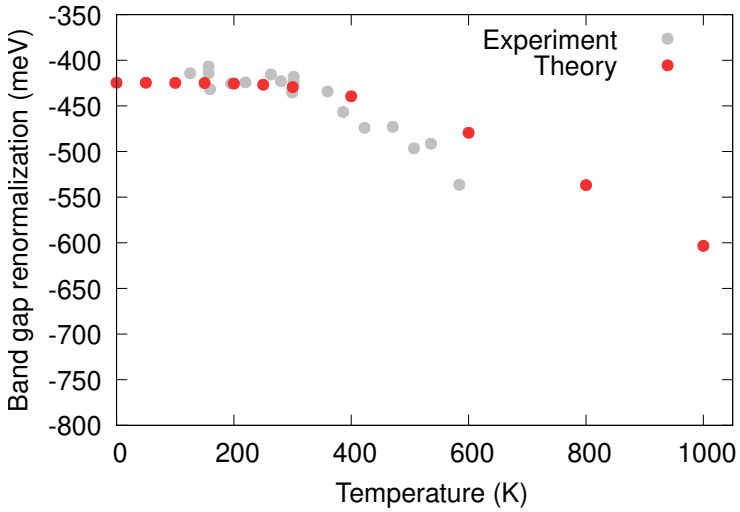
Exercise 5



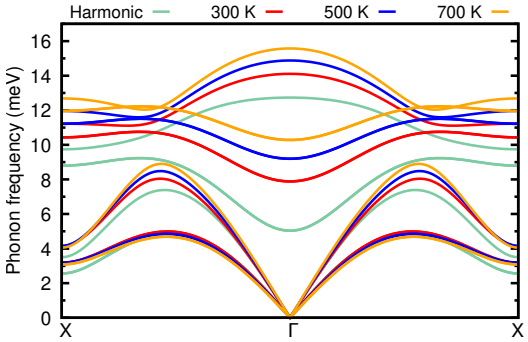
Exercise 5

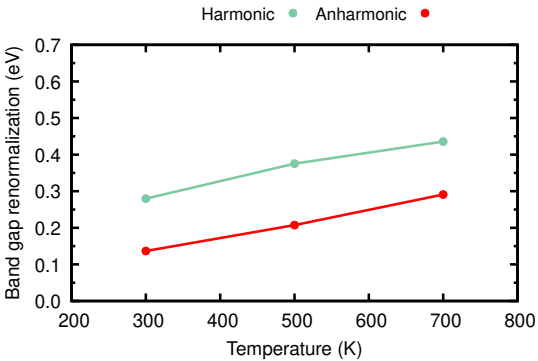

0
0.25
0.5
0.75
1
ph_unfolding_P_doped_222
500.0
400.0
300.0
200.0
100.0
0.0
Frequency (cm-1)
TO
LO
LA
TA
Γ
K
X
Γ
L
X
W
L
wave vector

Exercise C

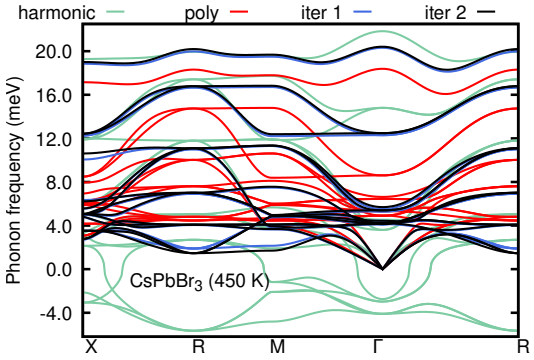

Band gap renormalization (meV)
-350
-400
-450
-500
-550
-600
-650
-700
-750
-800
0
200
400
600
800
1000
Temperature (K)
Experiment
Theory

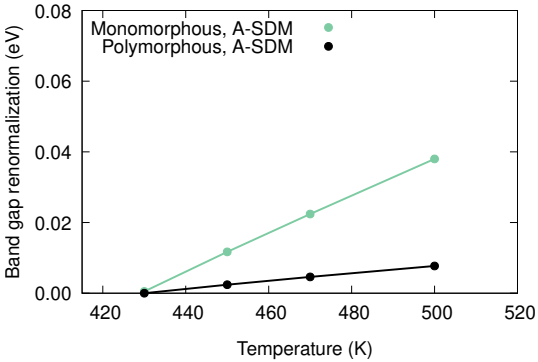
Exercise PbTe





Exercise $CsPbBr_3$





Things to have in mind when applying SDM via `ZG.x`:

- Always make sure that the phonon dispersion has no instabilities. For *anharmonic* systems one can upgrade the IFC file using the A-SDM.
- **q**-grid for phonons should not be necessarily the same with the supercell size. Use a coarse **q**-grid and generate any size of ZG configurations.
- Achieve convergence of the T-dependent observable with the supercell size.
- Set `error_thresh` around < 0.4.
- Check the anisotropic displacement tensor data at the end of the output `ZG_XXX.out` (as in `exercise1`).
- Pointless to minimize the error function for systems with many atoms (> 15) in the unit-cell (set `compute_error = .false.`).
- If you work in a 2×2×2 supercell set `incl_qA = .true.`. Useful for anharmonic phonons with A-SDM. In this case `error_thresh` can be (< 0.6).