Phono3py calculation of Ge (Fd-3m)

Crsytal structure

- Space-group type: Fd-3m
- Experimental lattice parameter of conventional unit cell at 300K: $a = 5.65791$Å

Computational method

- Supercell approach
- Finite displacements

Tools

- Phono3py
- Phonopy
- ALM (https://github.com/ttadano/ALM)
- VASP
- SeeK-path

Computational procedure

1. Relax crystal structure.
2. Generate supercells with displacements using phonopy code for fc2 and fc3.
3. Run force supercell calculations using VASP code.
4. Check electronic occupancies of KS eigenstates not being fractional.
5. Calculate force constants (fc2 and fc3) using ALM code via phono3py code with least squared fitting.
6. Calculate lattice thermal conductivity.

Computational details of force constants calculations

Unit cell structure relaxation

- Input structure: Conventional unit cell
- Cutoff energy: 240 eV
- Off-Gamma uniform k-point sampling mesh of $8 \times 8 \times 8$
- Gaussian smearing of 0.01 eV width
- In relaxation, all bands are filled or empty.
- `PREC = Accurate` (https://www.vasp.at/wiki/index.php/PREC)
- PBEsol
- Ge PAW dataset in which d-electrons are not treated as core, `Ge`
- Energy convergence criteria of `1e-8` eV.
- Iterate 5 relaxation VASP runs.

Third-order force constants (fc3)

- $2 \times 2 \times 2$ supercell of conventional unit cell

- 200 supercells with random-directional displacements of all atoms with 0.03 Å.
- $4 \times 4 \times 4$ off-Gamma-centred k-point uniform sampling mesh with respect to reciprocal of the supercell basis vectors.
- Cutoff energy: 240 eV
- Gaussian smearing of 0.01 eV width
- All electronic bands are filled or empty.
- `PREC = Accurate` (https://www.vasp.at/wiki/index.php/PREC)
- PBEsol
- Ge PAW dataset in which d-electrons are treated as core, `Ge`
- Energy convergence criteria of `1e-8` eV.

Second-order force constants (fc2)

- $4 \times 4 \times 4$ supercell of conventional unit cell for second-order force constants (fc2)
- 4 supercells with random-directional displacements of all atoms with 0.03 Å.
- $2 \times 2 \times 2$ off-Gamma-centred k-point uniform sampling mesh with respect to reciprocal of the supercell basis vectors.
- Cutoff energy: 240 eV
- Gaussian smearing of 0.01 eV width
- All electronic bands are filled or empty.
- `PREC = Accurate` (https://www.vasp.at/wiki/index.php/PREC)
- PBEsol
- Ge PAW dataset in which d-electrons are not treated as core, `Ge`
- Energy convergence criteria of `1e-8` eV.

Lattice thermal conductivity (RTA and direct solution)

- Linear tetrahedron method to calculate scattering rate (imaginary part of self-energy or collision matrix)
- Linear tetrahedron method to calculate scattering rate between phonons and isotopes according to Tamura's paper (1983)
- No cutoff distance in real space was used.
- Gamma-centre uniform sampling mesh with respect to reciprocal of basis vectors of primitive cell
- Group velocity was calculated by analytical derivative of dynamical matrix.
- Accumulated thermal conductivities were calculated using linear tetrahedron method.

Results

Obtained lattice parameter and electronic band structure

- Lattice parameter of conventional unit cell: 5.702Å
- Electronic band structure (Figure. 1)

Phonon band structure

- Phonon band structure (Figure. 2)

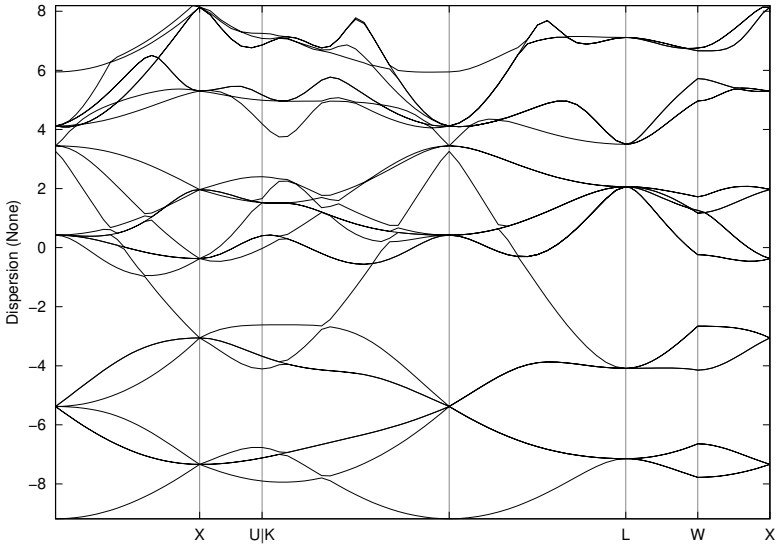

Figure 1: Electronic band structure

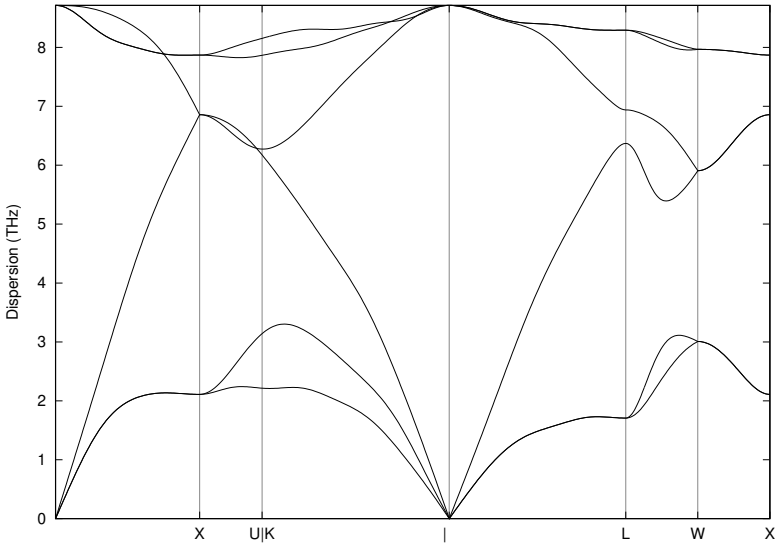

Figure 2: Phonon band structure

Lattice thermal conductivity without isotope scattering at 300K

RTA

```
W/m-k  mesh
51.55 [30 30 30]
51.85 [36 36 36]
52.03 [43 43 43]
52.14 [49 49 49]
52.18 [55 55 55]
52.24 [61 61 61]
52.29 [67 67 67]
52.30 [73 73 73]
52.33 [79 79 79]
```

RTA + isotope (RTA-iso)

```
W/m-k  mesh
44.93 [30 30 30]
45.20 [36 36 36]
45.48 [43 43 43]
45.58 [49 49 49]
45.62 [55 55 55]
45.69 [61 61 61]
```

Direct solution (LBTE)

```
W/m-k  mesh
54.48 [30 30 30]
54.84 [36 36 36]
55.11 [43 43 43]
55.23 [49 49 49]
55.23 [49 49 49]
55.29 [55 55 55]
55.37 [61 61 61]
```

Direct solution + isotope (LBTE-iso)

```
W/m-k  mesh
46.55 [30 30 30]
46.86 [36 36 36]
47.21 [43 43 43]
47.31 [49 49 49]
47.36 [55 55 55]
47.45 [61 61 61]
```

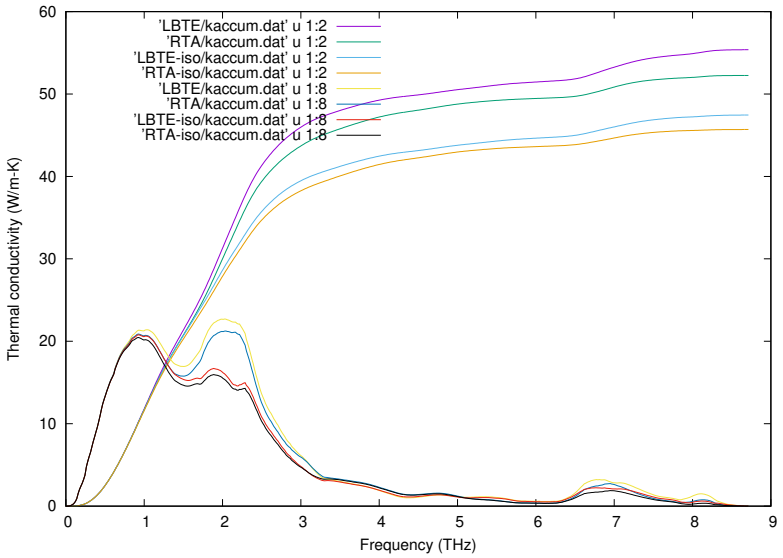

Figure 3: Accumulated lattice thermal conductivities with respect to frequency.

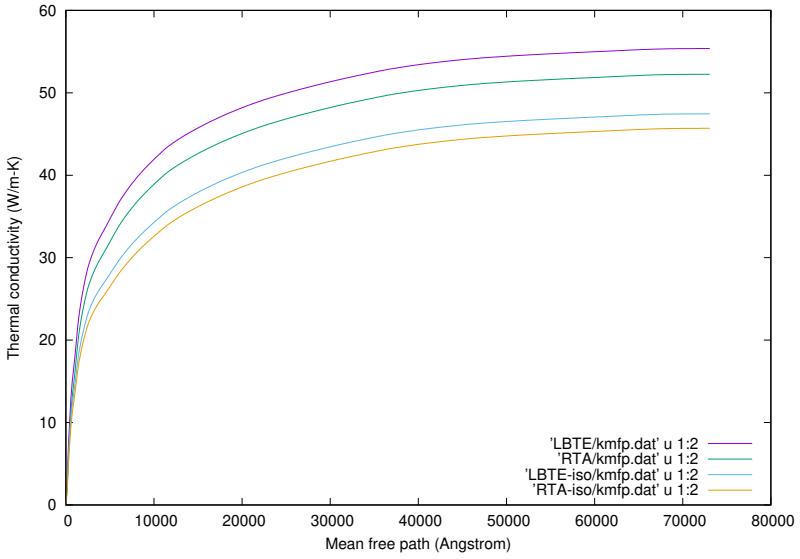

Figure 4: Accumulated lattice thermal conductivities with respect to mean free path