**Germanium**

*Space group*: Fd-3m, 227

*Lattice vectors*: *R*1 = (-*a*/2, 0, *a*/2); *R*2 = (0, *a*/2, *a*/2); *R*3 = (-*a*/2, *a*/2, 0)

*Atom positions*: Ge1 = (0, 0, 0); Ge2 = (*a*/4, *a*/4, *a*/4)

*Isotopes*: 20.38% 70Ge (69.924 amu); 27.31% 72Ge (71.922 amu); 7.76% 73Ge (72.923 amu); 36.72% 74Ge (73.921 amu); 7.83% 76Ge (75.921 amu)

Pseudopotential: Ge.pbesol-n-kjpaw\_psl.1.0.0.UPF

DFT & Harmonic: QE 6.7MaX

Phonon thermal conductivity: ShengBTE v1.1.1

***Structure***

* Converged relaxed lattice constant is 5.686 Å.
* Methods / convergence criteria

- The ‘vc-relax’ method is used for lattice constant optimization.

- The total energy threshold is 10-4 a.u.

- The convergence threshold on force is 10-3 a.u.

* Two ‘vc-relax’ loops are performed, and the total time consumed is 4.6 cpu hours.
* Hardware used: Intel(R) Xeon(R) Gold 6248 CPU @ 2.50GHz,
* Compiler used: Intel-19.1.1 compiler.
* All input files to run fully converged calculations are listed in ./structure/input.

***Electrons***

* Electron band dispersions for the three segments (Γ→X, Γ→K→X, and Γ→L) are listed in ./electrons/band.xlsx. Fermi energy is 5.5890 eV.
* Methods / convergence criteria

- Kinetic energy cutoff for wavefunctions is 80 Ry.

- Kinetic energy cutoff for charge density is 400 Ry.

- Convergence threshold for self-consistency is 10-10 Ry.

- The integration mesh is 161616 with no grid shifting.

* Evidence of converged band structure

- Band dispersions with integration meshes of 141414, 161616, 181818

are plotted in the following figure (The band dispersions for these three meshes

are overlapped):



(Fermi energy: 5.5890 eV)

* The total time to run the fully converged calculations is 0.97 cpu hours.
* All input files to run fully converged calculations are listed in ./electrons/input.

***Harmonic***

* Converged dispersions for the three segments (Γ→X, Γ→K→X, and Γ→L) are listed in ./harmonic/phonon.xlsx.
* Converged harmonic interatomic force constants (IFCs) are contained in ./harmonic/Ge.fc with QE standard format.
* Methods / convergence criteria

- Threshold for self-consistency in DFPT calculation is 10-15 Ry.

- Integration mesh for phonon is 777 uniform mesh.

- Irreducibility: 20 irreducible q-points.

- Post-processing: Acoustic Sum Rule (asr) imposed by the simple method.

* Evidence of converged dispersion

- Dispersions with integration meshes of 666, 777, 888 are plotted in the

following figure (most of these three phonon dispersions are overlapped):



* The total time to run the fully converged calculations is 48.67 cpu hours.
* All input files to run fully converged calculations are contained in ./harmonic/input.

***Anharmonic thermal transport***

* The values of *knat,full* , *knat,RTA*, *kpure,full*, *kpure,RTA*  are contained in ./anharmonic thermal transport/Thermal conductivity vs Temperature.xlsx.
* The file with row for each mode (q, *j*): f, mfp, mode contribution to *k* is listed in ./anharmonic thermal transport/Accumulated contribution.xlsx.
* Numerical data for f (THz), 1/*τ3ph* (THz=1/ps), and 1/*τiso* (THz) is contained in ./anharmonic thermal transport/Scattering rate.xlsx.
* Converged third-order anharmonic IFCs are listed in ./anharmonic thermal transport/FORCE\_CONSTANTS\_3RD.
* Methods / convergence criteria: thermal conductivity

- Scale parameter for Gaussian smearing is 1.0 for adaptive smearing.

- Integration grid is 303030 mesh for converged thermal conductivity.

- Symmetries used:

In the BTE calculation, we consider point-group symmetries, which means only

those q points lying inside the irreducible wedge of the BZ.

* Converged criteria for iterative thermal conductivity: relative change in the thermal conductivity tensor is less than 10-5 W/mK.
* Group velocity is obtained perturbatively (implemented in ShengBTE).
* Methods / convergence criteria: anharmonic IFCs

- Kinetic energy cutoff for wavefunctions is 80 Ry.

- Kinetic energy cutoff for charge density is 400 Ry.

- Convergence threshold for self-consistency is 10-12 Ry.

- The integration mesh is 222 with no grid shifting.

- Supercell size is 444.

- The force cutoff distance is the maximum distance among 5th neighbor

Interactions.

* The displacement parameter is default value of 2.1210-3 nm.

- For anharmonic IFCs, we use both Fd-3m point-group symmetries and the

translational symmetries. Total 124 self-consistent calculations are needed to

calculate the third-order anharmonic force constant for 444 supercell with

cutoff up to 5th neighbor interactions.

* Evidence of converged *k* at T=300K

- The thermal conductivity of natural isotopes with full BTE solution at 300 K is

calculated for 333, 444 supercells with variant force cutoff and BTE

integration meshes, which are listed in the following table.

* **Thermal conductivity with 333 supercell**

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| nth neighbor interactions | BTE integration meshes | | | | | | | | | | |
| 202020 | | 252525 | | 303030 | | 333333 | 343434 | | | 353535 |
| 1 | 35.951 | 36.87 | | 37.18 | | 37.48 | | | 37.53 | 37.55 | |
| 2 | 44.26 | 45.08 | | 46.08 | | 46.42 | | | 46.43 | 46.40 | |
| 3 | 44.88 | 45.92 | | 47.09 | | 47.55 | | | 47.60 | 47.63 | |
| 4 | 47.25 | 48.45 | | 49.63 | | 50.12 | | | 50.18 | 50.26 | |
| 5 | 36.11 | 35.87 | | 36.13 | |  | | |  |  | |
| 6 | 37.76 |  | |  | |  | | |  |  | |

1 Unit of the thermal conductivity is W/mK.

* **Thermal conductivity with 444 supercell**

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **nth neighbor interactions** | **BTE integration meshes** | | | | | |
| **202020** | **252525** | **303030** | **323232** | **333333** | **353535** |
| 1 | 36.761 | 37.79 | 38.20 | 38.47 | 38.53 | 38.61 |
| 2 | 47.93 | 49.02 | 50.15 | 50.48 | 50.58 | 50.65 |
| 3 | 47.27 | 48.49 | 49.62 | 49.97 | 50.08 | 50.23 |
| 4 | 48.63 | 49.96 | 51.11 | 51.48 | 51.60 | 51.75 |
| 5 | 50.58 | 53.00 | **54.342** | 54.79 | 54.89 | 55.10 |
| 6 | 53.94 | 56.47 | 57.76 | 58.15 | 58.25 | 58.47 |

1 Unit of the thermal conductivity is W/mK.

2 Thermal conductivity is considered to be converged value at this condition.

* The time to run fully converged calculations of anharmonic IFCs (highlighted in the above table) is 3024 cpu hours; the time to run fully converged calculations of BTE calculations for 300 K is 13.3 cpu hours.
* All input files to run fully converged calculation of thermal conductivity with natural isotopes and full BTE solution at 300 K (highlighted in the above table) are contained in ./anharmonic thermal transport/input.