Germanium (Free Phase)

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: $Ge_1 = (0, 0, 0)$; $Ge_2 = (a/4, a/4, a/4)$
Isotopes: 20.38% ^{70}Ge (69.924 amu); 27.31% ^{72}Ge (71.922 amu); 7.76% ^{73}Ge (72.923 amu); 36.72% ^{74}Ge (73.921 amu); 7.83% ^{76}Ge (75.921 amu)

DFT: Use VASP, QE, or both. PBEsol PAW, no *d* states in valence
(QE: Ge.pbesol-n-kjpaw_psl.1.0.0.UPF; VASP: standard version with sol flag)
Warning: DFT may give metallic. May need to check convergence with electronic smearing.

Checklist (*all data should be reported for the 2-atom primitive cell*)

Structure

- Converged relaxed 'temperature (T)=0' lattice constant *a* (target accuracy < 0.005 Å)
  - Single value with 4 significant figures: X.XXX
- Methods / convergence criteria
  - Energy/force thresholds
- Other notes / cpu hours (*e.g., multiple relaxations, compilers, hardware*)
- All input files to run fully converged calculations (*e.g., qe.sc.in, POSCAR*)

Electrons

- Converged electron band dispersion (target accuracy < 0.1 eV for Γ/X/L frequencies)
  - Numerical data: normalized wavevectors (q) and band energies (E): 4 valence bands and 6 conduction bands (excel or text file)
  - q in units of $2\pi/a$ and f in eV
  - 3 segments: Γ→X, Γ→K→X, and Γ→L evenly divided with ~100 q points per segment
  - 3 files, one for each segment. For each scaled q from 0 to 1 list (~100 rows): q, E_1, E_2, E_3, E_4, E_5, E_6, E_7, E_8, E_9, E_{10}
- Methods / convergence criteria
  - Thresholds/ Integration mesh / grid shifting
- Evidence of converged band structure
  - Band structure with varying integration meshes
- Other notes / cpu hours
- All input files to run fully converged calculations

Harmonic

- Converged dispersion (target accuracy < 0.1 THz for Γ/X/L frequencies)
  - Numerical data: normalized wavevectors (q) and frequencies (f) for 6 polarizations (*j*) (excel or text file)
  - q in units of $2\pi/a$ and f in THz ($f=\omega/2\pi$)
  - 3 segments: Γ→X, Γ→K→X, and Γ→L evenly divided with ~100 q points per

segment
  - 3 files, one for each segment. For each scaled q from 0 to 1 list (~100 rows): q, f_1, f_2, f_3, f_4, f_5, f_6
- Converged harmonic interatomic force constants (IFCs)
  - Standard format for code used (*e.g.*, QE, Phonopy)
  - Will be supplied as supplemental material upon publication
- Methods / convergence criteria
  - Thresholds
  - Supercell size / integration mesh
  - Symmetries / irreducibility / number of calculations (*linked to cpu hours below*)
  - Post-processing (*e.g., enforce invariance constraints*)
- Evidence of converged dispersion
  - Dispersions with varying supercell sizes and integration meshes
- Other notes / cpu hours (*e.g., accuracy vs cpu cost, shifted meshes*)
- All input files to run fully converged calculations

Anharmonic thermal transport

- Four converged T-dependent thermal conductivities (k): natural isotopes with full BTE solution ($k_{nat,full}$), natural isotopes with the relaxation time approximation (RTA) ($k_{nat,RTA}$), isotopically pure (100% ^{70}Ge (69.924 amu)) with full BTE solution ($k_{pure,full}$), and isotopically pure with RTA ($k_{pure,RTA}$). If only RTA available, then only $k_{nat,RTA}$ and $k_{pure,RTA}$
  - Do not include boundary scattering, even at low T. We want to see how the codes behave at low T without this extrinsic scattering.
  - Numerical data: T (K) and k (W/m/K) in range 10K < T < 1000K (excel or text file)
  - For 10K ≤ T ≤ 50K increments of 10K (5 data points); for 50K < T ≤ 300K increments of 25K (10 data points); for 300K < T ≤ 1000K increments of 100K (7 data points).
  - 1 file with T from 10K to 1000K list (22 rows): T, $k_{nat,full}$, $k_{nat,RTA}$, $k_{pure,full}$, $k_{pure,RTA}$
- Accumulated T=300K k_{acc} vs frequency and k_{acc} vs mean free path (mfp) for converged $k_{nat,RTA}$ value
  - Numerical data for each mode (q, j) sampled in the Brillouin zone integration: f (THz), mfp=|sqrt(v_x^2+v_y^2+v_z^2)×lifetime| (nm), mode contribution to k (W/m/K) for $k_{nat,RTA}$
  - 1 file (excel or text) with row for each mode (q, j): f, mfp, mode contribution to k
- RTA T=300K three-phonon scattering rates ($1/\tau_{3ph}$) and phonon-isotope scattering rates for natural abundance ($1/\tau_{iso}$)
  - Numerical data: f (THz), $1/\tau_{3ph}$ (THz=1/ps), and $1/\tau_{iso}$ (THz)
  - 1 file (excel or text) with row for each mode (q, j): f, $1/\tau_{3ph}$, $1/\tau_{iso}$
- Converged third-order anharmonic IFCs
  - Standard format for code used
  - Will be supplied as supplemental material upon publication
- Methods / convergence criteria: thermal conductivity
  - Delta function representation (*with details; e.g., adaptive smearing, cutoff*)
  - Integration grid

    - Symmetries used
- Methods / convergence criteria: anharmonic IFCs
    - Cutoff radius, supercell size, integration mesh, thresholds, displacement parameter for supercell derivatives
    - Post-processing
- Evidence of converged *k* at T=300K
    - Varying integration meshes
- Other notes / cpu hours
- All input files to run fully converged calculations