**Monolayer MoSe2**

*Structure*: hexagonal, P-6m2, layer group 78

*Lattice vectors*: *R*1 = (*a*, 0, 0); *R*2 = (-*a*/2, *a×*sqrt(3)/2, 0) (2D structure – define vacuum space with third lattice vector)

*Atom positions*: Mo= (0, *a*/sqrt(3), 0); Se1 = (*a*/2, *a*/(2*×*sqrt(3)), Δ); Se2 = (*a*/2, *a*/(2*×*sqrt(3)), -Δ)

*Atom positions crystallographic*: Mo= (1/3, 2/3, 0); Se1 = (1/3, 2/3, Δ); Se2 = (1/3, 2/3, -Δ)

*Isotopes*: 14.649% 92Mo (91.907 amu); 9.187% 94Mo (93.905 amu); 15.873% 95Mo (94.906 amu); 16.673% 96Mo (95.905 amu); 9.582% 97Mo (96.906 amu); 24.292% 98Mo (97.905 amu); 9.744% 100Mo (99.907 amu); 0.89% 74Se (73.922 amu); 9.37% 76Se (75.919 amu); 7.63% 77Se (76.920 amu); 23.77% 78Se (77.917 amu); 49.61% 80Se (79.917 amu); 8.73% 82Se (81.917 amu)

*DFT*: Use VASP, QE, or both. PBEsol PAW  
(QE: Mo.pbesol-spn-kjpaw\_psl.1.0.0.UPF; Se.pbesol-dn-kjpaw\_psl.1.0.0.UPF

VASP: standard version with sol flag; Mo\_sv)

**Checklist** (*all data should be reported for the 3-atom primitive cell*)

* Please provide all computational costs in cores\*hours

***Structure***

* Converged relaxed ‘temperature (T)=0’ lattice constant *a* and internal degree of freedom Δ related to distance between Se atoms in separate layers (2Δ) (target accuracy < 0.05 Å)

- Two values with 3 significant figures: **a=3.27, Δ=1.66**

* Methods / convergence criteria

- Energy/force thresholds: **etot\_conv\_thr=1e-5 Ry, force\_conv\_thr =1e-4 Ry/Bohr**

- Vacuum layer distance: **14 Å**

* Other notes / cpu hours (*e.g., multiple relaxations, compilers, hardware*)
* **36 cores × 2399 minutes**
* All input files to run fully converged calculations (*e.g., qe.scf.in, POSCAR*)
* **pw.MoSe2.in**

***Electrons***

* Converged electron band dispersion (target accuracy < 0.1 eV for Γ/X/L frequencies)

- Numerical data: normalized wavevectors (q) and band energies (E): top 7 valence bands and 4 conduction bands (excel or text file)

- q in units of 2π/*a* and f in eV

- 2 segments: Γ→M and Γ→K→M 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, E1, E2, E3, E4, E5, E6, E7, E8, E9, E10, E11

- **Band\_GM.txt, Band\_GKM.txt**

* Methods / convergence criteria

- Thresholds/ Integration mesh / grid shifting

- **threshold = 1e-10 Ry / Integration mesh = 6×6×6 / grid shifting: No**

* Evidence of converged band structure

- Band structure with varying integration meshes

- **Convergence\_check\_hamakawa\_mose2.xlsx**

* Other notes / cpu hours
* **36 cores × 10 minutes**
* All input files to run fully converged calculations
* **submit\_ele.sh**

***Harmonic***

* Converged dispersion (target accuracy < 0.1 THz for Γ/M frequencies)

- Numerical data: normalized wavevectors (q) and frequencies (f) for 9 polarizations (*j*) (excel or text file)

- q in units of 2π/*a* and f in THz (f=ω/2π)

- 3 segments: Γ→M and Γ→K→M 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, f1, f2, f3, f4, f5, f6, f7, f8, f9 (Note that 3 more bands with respect to previous materials)

- **Harmonic\_GM.txt, Harmonic\_GKM.txt**

* Converged harmonic interatomic force constants (IFCs)

- Standard format for code used (*e.g.*, QE, Phonopy)

- Will be supplied as supplemental material upon publication

- **fcs\_harmonic.xml**

* Long range Coulomb corrections
* Dielectric matrix (1 matrix)
  + (For c-axis cell size = 14)
* Born effective charge matrices (3 matrices)
  + Mo (For c-axis cell size = 14)
  + Se (For c-axis cell size = 14)
  + Se (For c-axis cell size = 14)
* Method of long-range Coulomb corrections: **Ewald method**
* Methods / convergence criteria

- Thresholds: **Check target accuracy < 0.1 THz for Γ/M/K frequencies**

- Supercell size / integration mesh: **5×3×1 / Gamma Point only**

- Symmetries / irreducibility / number of calculations (*linked to cpu hours below*): number of calculations: **9**

- Post-processing (*e.g., enforce invariance constraints*): **Translational Invariance**

* Evidence of converged dispersion→**Convergence\_Check\_hamakawa\_mose2.xlsx**

- Dispersions with varying supercell sizes and integration meshes

* Other notes / cpu hours (*e.g., accuracy vs cpu cost, shifted meshes*)
* **36 core × 23332 minutes**
* All input files to run fully converged calculations
* **submit\_all.sh**

***Anharmonic thermal transport***

* **Use thickness of 6.470 Å** (measured *c*/2 of bulk MoSe2)
* Four converged T-dependent thermal conductivities (*k*) (target accuracy <2% difference between successive grids - please contact us if a problem): natural isotopes with full BTE solution (*knat,full*), natural isotopes with the relaxation time approximation (RTA) (*knat,RTA*), isotopically pure (*e.g.*, 100% 98Mo and 100% 80Se) with full BTE solution (*kpure,full*), and isotopically pure with RTA (*kpure,RTA*). If only RTA available, then only *knat,RTA* and *kpure,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 50K < T < 1000K (excel or text file)

- For 50K ≤ T ≤ 300K increments of 25K (11 data points); for 300K < T ≤ 1000K increments of 100K (7 data points). Note: not sampling T<50K range here.

- 1 file with T from 50K to 1000K list (18 rows): T, *knat,full*, *knat,RTA*, *kpure,full*, *kpure,RTA*

- Masses used for pure and natural calculations: **pure Mo:97.905 Se:79.917, nat Mo:** **95.94873241 Se:** **78.9595608**

**- kappa\_temp.txt**

* Accumulated T=300K *kacc* vs frequency and *kacc* vs mean free path (mfp) for converged *knat,RTA* value: **accum\_freq.txt, accum\_mfp.dat**

- Numerical data for each mode (q, *j*) sampled in the Brillouin zone integration: f (THz), mfp=|sqrt(*vx*2+*vy*2+*vz*2)×lifetime| (nm), mode contribution to *k* (W/m/K) for *knat,RTA*

- 1 file (excel or text) with row for each mode (q, *j*): f, mfp, mode contribution to *k*

- **mode\_kappa.txt**

* RTA T=300K three-phonon scattering rates (1/*τ3ph*) and phonon-isotope scattering rates for natural abundance (1/*τiso*)

- Numerical data: f (THz), 1/*τ3ph* (THz=1/ps), and 1/*τiso* (THz)

- 1 file (excel or text) with row for each mode (q, j): f, 1/*τ3ph*, 1/*τiso*

-**tau\_all.txt**

* Converged third-order anharmonic IFCs

- Standard format for code used

- Will be supplied as supplemental material upon publication

- **fcs\_anharmonic.xml**

* Methods / convergence criteria: thermal conductivity

- Delta function representation (*with details; e.g., adaptive smearing, cutoff*) : **Tetrahedron**

- Integration grid: **300×300×1**

- Symmetries used

* Methods / convergence criteria: anharmonic IFCs→**Convergence\_Check\_hamakawa\_mose2.xlsx**

- Cutoff radius, supercell size, integration mesh, thresholds, displacement parameter for supercell derivatives

- Post-processing

* Evidence of converged *k* at T=300K→**Convergence\_Check\_hamakawa\_mose2.xlsx**

- Varying integration meshes

* Other notes / cpu hours
* **36 core × 108252 minutes**
* All input files to run fully converged calculations
* **submit\_anharm.sh, submit\_thermal.sh**