Monolayer $MoSe_2$

Structure: hexagonal, P-6m2, layer group 78
Lattice vectors: $R_1 = (a, 0, 0)$; $R_2 = (-a/2, a\times\text{sqrt}(3)/2, 0)$ (2D structure – define vacuum space with third lattice vector)
Atom positions: Mo = $(0, a/\text{sqrt}(3), 0)$; $Se_1 = (a/2, a/(2\times\text{sqrt}(3)), \Delta)$; $Se_2 = (a/2, a/(2\times\text{sqrt}(3)), -\Delta)$
Atom positions crystallographic: Mo = (1/3, 2/3, 0); Se_1 = (1/3, 2/3, Δ); Se_2 = (1/3, 2/3, -Δ)
Isotopes: 14.649% ^{92}Mo (91.907 amu); 9.187% ^{94}Mo (93.905 amu); 15.873% ^{95}Mo (94.906 amu); 16.673% ^{96}Mo (95.905 amu); 9.582% ^{97}Mo (96.906 amu); 24.292% ^{98}Mo (97.905 amu); 9.744% ^{100}Mo (99.907 amu); 0.89% ^{74}Se (73.922 amu); 9.37% ^{76}Se (75.919 amu); 7.63% ^{77}Se (76.920 amu); 23.77% ^{78}Se (77.917 amu); 49.61% ^{80}Se (79.917 amu); 8.73% ^{82}Se (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: X.XX
- Methods / convergence criteria
  - Energy/force thresholds
  - Vacuum layer distance
- Other notes / cpu hours (*e.g., multiple relaxations, compilers, hardware*)
- All input files to run fully converged calculations (*e.g., qe.scf.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): top 7 valence bands and 4 conduction bands (excel or text file)
  - q in units of $2\pi/a$ 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, E_1, E_2, E_3, E_4, E_5, E_6, E_7, E_8, E_9, E_{10}, E_{11}
- 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/M frequencies)
  - Numerical data: normalized wavevectors (q) and frequencies (f) for 9 polarizations (j) (excel or text file)
  - q in units of $2\pi/a$ and f in THz ($f=\omega/2\pi$)
  - 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, f_1, f_2, f_3, f_4, f_5, f_6, f_7, f_8, f_9 (Note that 3 more bands with respect to previous materials)
- Converged harmonic interatomic force constants (IFCs)
  - Standard format for code used (*e.g.*, QE, Phonopy)
  - Will be supplied as supplemental material upon publication
- Long range Coulomb corrections
  - Dielectric matrix (1 matrix)
  - Born effective charge matrices (3 matrices)
  - Method of long-range Coulomb corrections
- 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

- **Use thickness of 6.470 Å** (measured $c/2$ of bulk $MoSe_2$)
- 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 ($k_{nat,full}$), natural isotopes with the relaxation time approximation (RTA) ($k_{nat,RTA}$), isotopically pure (*e.g.*, 100% ^{98}Mo and 100% ^{80}Se) 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 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, $k_{nat,full}$, $k_{nat,RTA}$, $k_{pure,full}$, $k_{pure,RTA}$
  - Masses used for pure and natural calculations
- 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