***Structure***

1. Converged relaxed lattice constant is 5.687 Å.
2. Threshold for energy is 10-7 Ryd; for force is 10-7 Ryd/Bohr.
3. CPU hours for scf calculation with 16\*16\*16 kmesh is 2.52s on two nodes (each has 24 processors) and total 48 processors. Each node equipped with E5-2680v3 and 125 Gb mem. QE version 6.6 compiled with Intel compiler version 2019.1.144 and OpenMPI version 4.0.4.
4. Input files are attached.

***Electrons (\*I cannot get a DFT-level band structure without band inversion)***

1. Numerical data are attached, and named as seg1.csv, seg2.csv, and seg3.csv corresponding to the three different paths.
2. Thresholds for energy is 10-10 Ryd. Well converged integration k-mesh is 12\*12\*12 without grid-shifting. The maximum eigenenergy difference between k-mesh 12\*12\*12 and 14\*14\*14 is 0.0001 eV. Actually, the maximum eigenenergy difference between k-mesh 4\*4\*4 and 6\*6\*6 is 0.0367 eV, which is already hard to capture in Figure 1 from the item 3 in this section.
3. The converged band structure is shown in Figure 1, also attached.

Chart, diagram

Description automatically generatedFigure 1. Band structure calculated with different k-meshes. The left, middle and right panels are for paths along Γ→X, Γ→K→X, and Γ→L, respectively. Y axis is the eigenenergy, which is in eV.

1. N/A
2. Input files are attached.

***Harmonic***

1. Numerical data are attached, and named as seg1\_phonon.csv, seg2\_phonon.csv, and seg3\_phonon.csv corresponding to the three different paths.
2. Converged harmonic interatomic force constants are attached (espresso.fc).
3. Thresholds for phonon calculation is 10-22 Ryd2. Well converged integration q-mesh for density functional perturbation theory calculation is 6\*6\*6. The maximum eigen-frequency difference between q-mesh 6\*6\*6 and 7\*7\*7 is 0.0967 THz.
4. The converged phonon dispersion is shown in Figure 2, also attached.

Chart

Description automatically generated

Figure 2. Phonon dispersion calculated with different q-meshes. The left, middle and right panels are for paths along Γ→X, Γ→K→X, and Γ→L, respectively. Y axis is the eigen-frequency, which is in THz.

1. CPU hours for phonon calculation with 6\*6\*6 q-mesh is 57m32.87s on two nodes (each has 24 processors) and total 48 processors.
2. All input files are attached.

***Anharmonic thermal transport***

1. The files containing the lattice thermal conductivities is attached. The lattice thermal conductivity is in W/(mK) and temperature in K.
2. The files containing the accumulated lattice thermal conductivity at RTA level with isotope scattering v.s. mean free path (frequency) are attached. The lattice thermal conductivity is in W/(mK), mean free path in nm and frequency in THz.
3. File containing three-phonon scattering rates (1/*τ3ph*) and phonon-isotope scattering rates (1/*τiso*) is attached.
4. The file containing converged third order IFCs is attached.
5. Delta function was dealt with adaptive smearing. The scalebroad was set to 0.1, which was well converged. For example, the percentage difference on the lattice thermal conductivity obtained from exact solution with isotopes scattering between scalebroad=0.1 and scalebroad=0.2 is 0.649%. And the percentage difference between scalebroad=0.1 and scalebroad=1 is 1.33%. Integration grid was chosen as 32\*32\*32, whose convergence was also tested. For example, the percentage difference on the lattice thermal conductivity obtained from exact solution with isotopes scattering between k-grid 32\*32\*32 and 34\*34\*34 is 1.73%. And the percent difference is 0.618% between k-grid 32\*32\*32 and 40\*40\*40.
6. Up to the 4th nearest neighbors was used to calculate the anharmonic IFCs with 5\*5\*5 supercell. The convergence of the cutoff radius was well tested. For example, the percentage difference on the lattice thermal conductivity obtained from exact solution with isotopes scattering between 4th and 5th nearest neighbors is 1.93%. And the percent difference is 1.18% between the 4th and 9th nearest neighbors. When calculating the force on after displacement in the supercell, only Gamma point was used. Energy threshold was set as 10-16 Ryd. Default displacement parameter as implemented in thirdorder\_v1.1.1 when constructing displaced supercells.
7. Convergence test for lattice thermal conductivity obtained from iterative solution with isotope scattering with respect to k-meshes is shown in Figure 3, which is also attached.

Chart, line chart

Description automatically generatedFigure 3 Convergence test for lattice thermal conductivity

1. CPU hours for 10-50K (6 points) is 4m4s; for 50-300K (11 points) is 7m3s; for 300-1000K (8 points) is 6m0s with two nodes (each has 24 processors) and total 48 processors. 50 K and 300 K are calculated twice. These data are for the calculations with isotope scattering. RTA and full solution can be obtained simultaneously. Each scf calculation on the displaced supercell will take around 30m with one node (with 24 processors).
2. Input files are attached.