Supplementary Information for

Thermoelectric Properties of Single-layer SnSe Sheet

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To get accurate lattice thermal conductivity κ_l, the convergence of three important parameters: cutoff radius (R_{cutoff}), scalebroad, and k-point grid density are examined carefully by using the ShengBTE code.[1] All interactions between atomic triplets at distances larger than R_{cutoff} can be discarded when the radius exceeds the range of physically relevant anharmonic interactions in the crystal. Considering both the symmetry of single-layer SnSe and calculation expense, R_{cutoff} is set as 6.5 Å (taking into account interactions up to the 21st coordination shell) to ensure the accuracy.

The convergence of κ_l with respect to k-points and scalebroad is well testified. We fixed k-points equal to 10×10×1, then κ_l gets convergence when scalebroad reaches 1.0; next scalebroad was fixed equal to 1.0, and we found κ_l converges when the k-point mesh grid reaches 30×30×1.

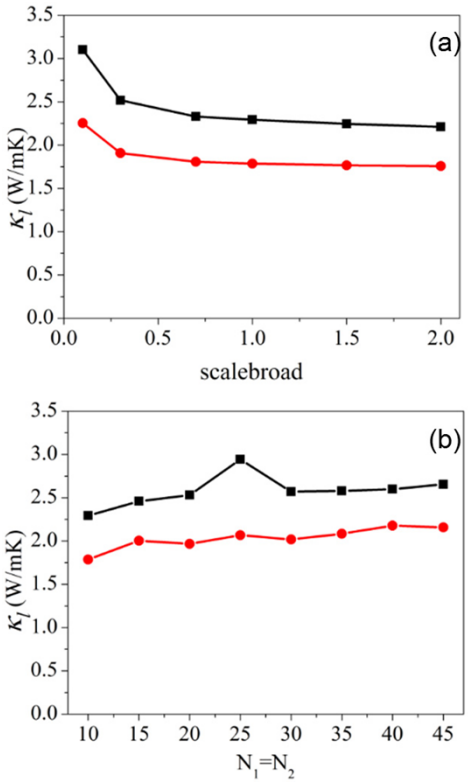


Fig. S1 Lattice thermal conductivity (κ_l) as a function of (a) scalebroad and (b) the grid density of k-points ($N_1 \times N_2 \times 1$) along the zigzag (black dots) and the armchair (red dots) direction.

To study the variance of the Sn-Se bond length with temperature, we classified the Sn-Se bonds into two types: namely the in plane Sn-Se bonds, labeled as d_1, and the Sn-Se bonds in the direction nearly perpendicular to the plane, labeled as d_2, as shown in Fig. S2. We have looked at the bond length variance carefully during the simulations at the different temperatures, and found that the bond lengths (d_1 and d_2) oscillate around their equilibrium bond lengths at 0 K. For comparison, the average bond lengths of d_1 and d_2 are summarized in Table S1, which shows that the change of average bond lengths $\bar{d}_1$ is less than 5% while that of $\bar{d}_2$ about 1%, as compared to their equilibrium bond lengths at 0 K, respectively.

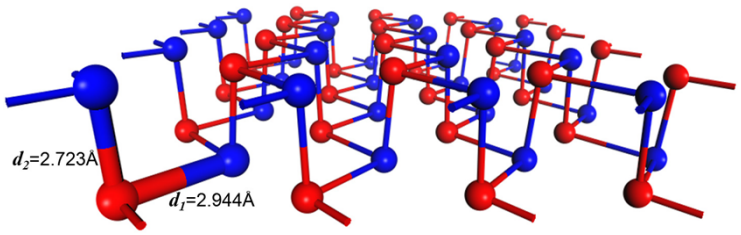
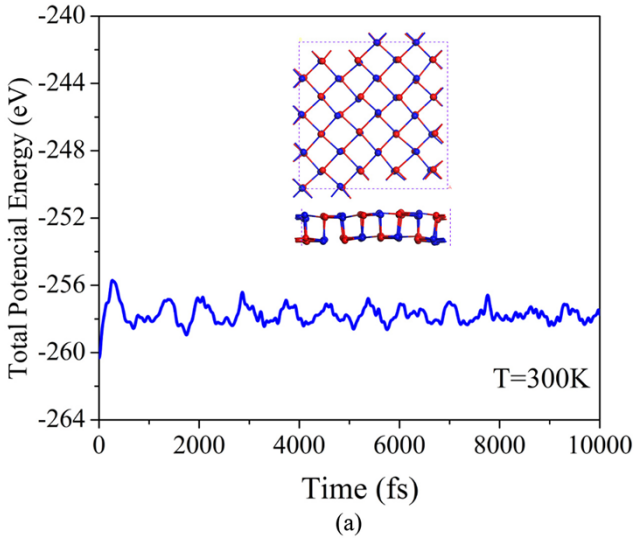


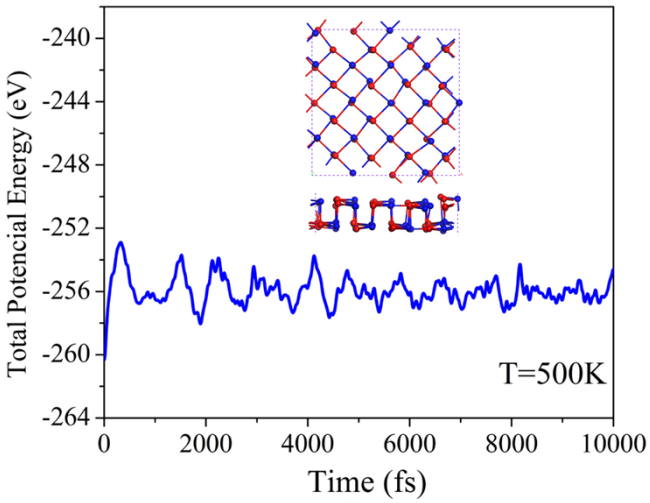
Fig. S2 Initial structure (0K) of single-layer SnSe from perspective view. Two types of bonds are bolded and labeled as d_1 and d_2. The typical bond length of d_1 and d_2 are 2.944Å and 2.723Å, respectively.

Table S1. The average bond length of single-layer SnSe at different temperatures (in Å)

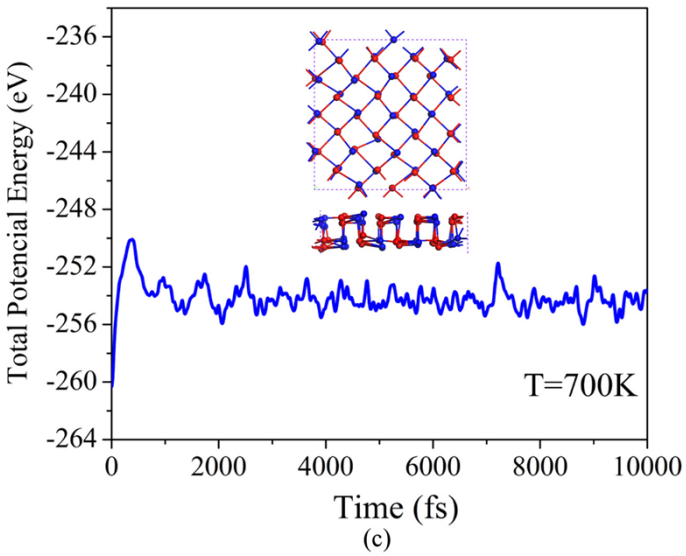
	0 K	**300 K**	**500 K**	**700 K**	**900 K**
$\bar{d}_1$	2.944	2.981	3.004	3.087	3.116
$\bar{d}_2$	2.723	2.709	2.777	2.733	2.752



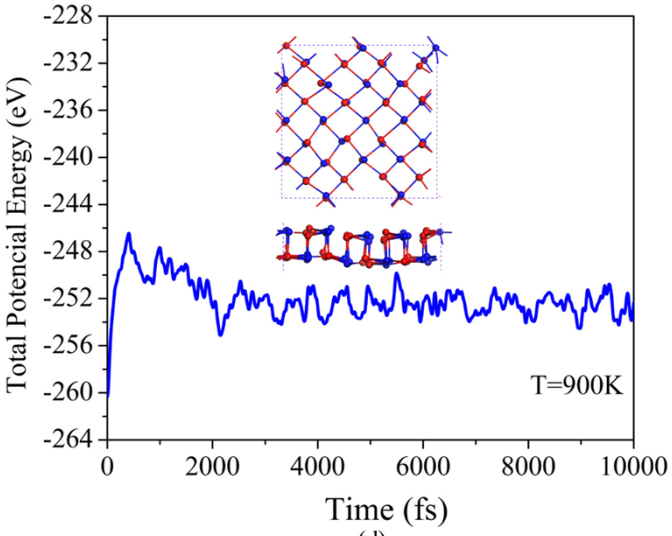
(a)


Total Potencial Energy (eV)
-240
-244
-248
-252
-256
-260
-264
0
2000
4000
6000
8000
10000
Time (fs)
T=500K

(b)


Total Potencial Energy (eV)
-236
-240
-244
-248
-252
-256
-260
-264
0
2000
4000
6000
8000
10000
Time (fs)
T=700K

(c)


Total Potencial Energy (eV)
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-232
-236
-240
-244
-248
-252
-256
-260
-264
0
2000
4000
6000
8000
10000
Time (fs)
T=900K

(d)

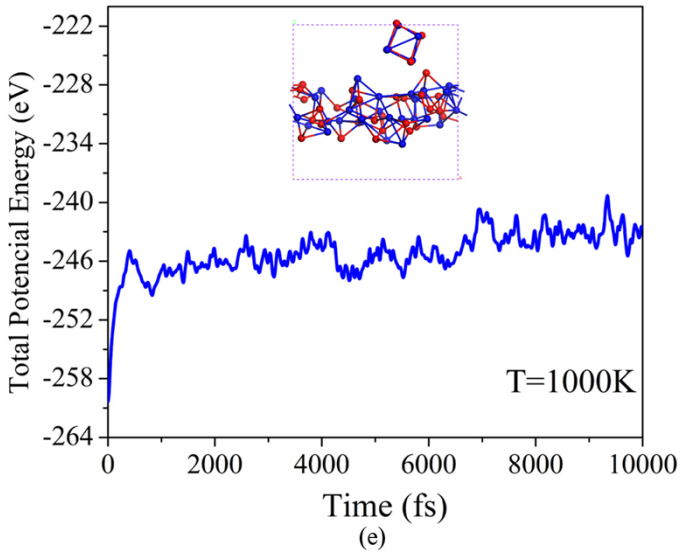

Fig. S3 Total potential energy fluctuation during AIMD simulation of single-layer SnSe at different temperatures, namely (a) 300 K, (b) 500K, (c) 700K, (d) 900K, and (e) 1000K. The insets show the snapshots of atomic configuration of single-layer SnSe at the end of AIMD simulation.

We calculated the thermal conductivity by solving the linearized phonon BTE with an iterative approach using ShengBTE code, which focuses on the anharmonic scattering and isotopic impurity scattering. In the isotopic impurity scattering process, higher frequency governs the thermal conductivity in the form of $A\omega^{-4}$, where A is a physical parameter depending on the nature of the point defects.[2] That means isotopic impurity scattering rates only has relationship with high frequency optical modes. In anharmonic phonon-phonon process we assume that only the acoustic phonon modes participate in the heat conduction process, the thermal conductivity can be expressed as $\kappa = B\frac{\overline{M}\theta_a^3\delta}{\gamma^2 n^{\frac{2}{3}}T}$,[3] where $\overline{M}$ is the average mass of an atom in the crystal (in atomic mass unit), θ_a is the Debye temperature of acoustic phonons, δ^3 is the volume per atom (δ in Å), n is the number of atoms in the

primitive unit cell, γ is the high temperature limit of the acoustic phonon mode Grüneisen parameter, and B≈3.1×10^{-6} is a collection of physical constants. When the anharmonic phonon-phonon interactions are dominant in phonon scattering mechanism, the lattice thermal conductivity is subject to the relationship $\kappa_l \propto \frac{1}{T}$. We fitted the κ_l-1/T curves using the least square fitting method. The curves exhibit perfect linearity is as shown in Fig. 5 (d), from where we obtain the scale factor to be 770.56 and 604.90 W/m for the zigzag and armchair directions, respectively.

The mode Grüneisen parameter is given as $\gamma_\lambda = -\frac{A}{\omega_\lambda}\frac{\partial\omega_\lambda}{\partial A}$, where A is the area of a unit cell of single-layer SnSe, and ω_λ is the angular frequency of a specific phonon mode. γ of the three acoustic modes is calculated using the Phonopy Package[4] and shown in Figure S4. To obtain reliable Grüneisen parameters, phonon calculations are performed in three individual lattices respectively, namely, the equilibrium lattice, as well as in the slightly stretched (+0.3%) and compressed lattice (-0.3%). The ZA mode diverges when its momentum approaches Γ point because of the quadratic band dispersion, similar to the cases in graphene and phosphorene.[5]

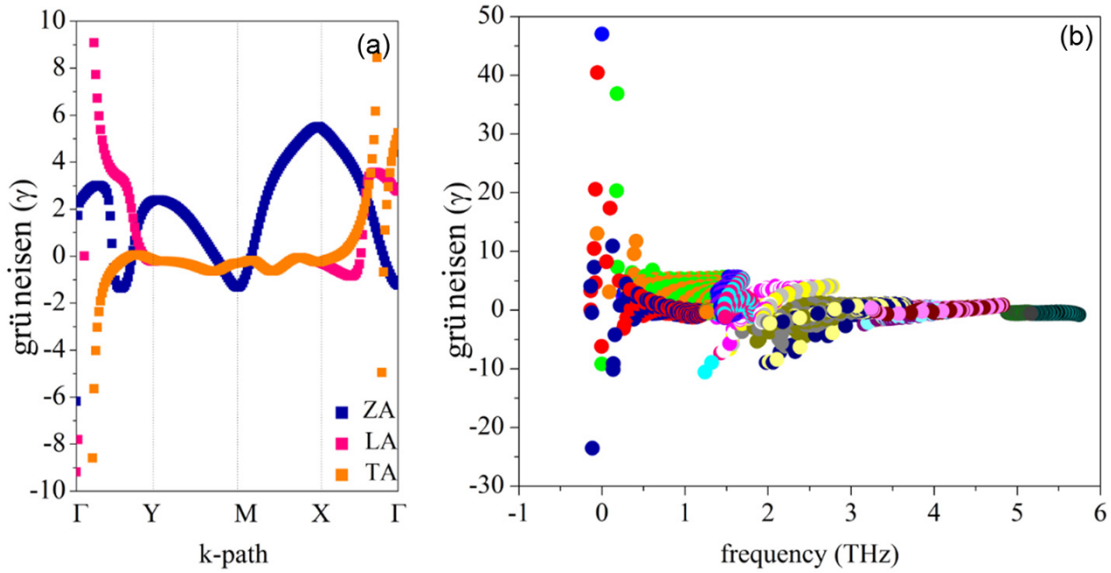

Fig. S4 Mode Grüneisen parameter (γ) of the acoustic modes of single-layer SnSe (a) along high symmetry point paths in the first Brillouin zone. (b) Frequency distribution of the mode Grüneisen parameter, calculated using a reciprocal mesh (31×31×1).

1. W. Li, J. Carrete, N. A. Katcho and N. Mingo, *Comput. Phys. Commun.*, 2014, **185**, 1747-1758.
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3. D. Morelli, V. Jovovic and J. Heremans, *Phys. Rev. Lett.*, 2008, **101**, 035901.
4. A. Togo, F. Oba and I. Tanaka, *Phys. Rev. B*, 2008, **78**, 134106.
5. R. Fei, A. Faghaninia, R. Soklaski, J.-A. Yan, C. Lo and L. Yang, *Nano. Lett.*, 2014, **14**, 6393-6399.