Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Electronic Supplementary Information for:

WSe_2 Nanoribbons: New High-performance Thermoelectric Materials

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1. Atom-projected density of states

First we plot the atom-projected density of states of the AWNR-6 with H-passivation as an example to show the W-Se bond information in **Figure S1**. We can observe that the highest occupied valence subbands and the lowest unoccupied conduction subbands are mainly attributed to the p orbitals from Se atoms and the d orbitals from W atoms. In addition, in order to study the bond covalency of W-Se pair, the energy range of [-10.0, 10.0] eV would be sufficient since the density of electronic states of the above two projected orbitals outside this area is almost ignorable.

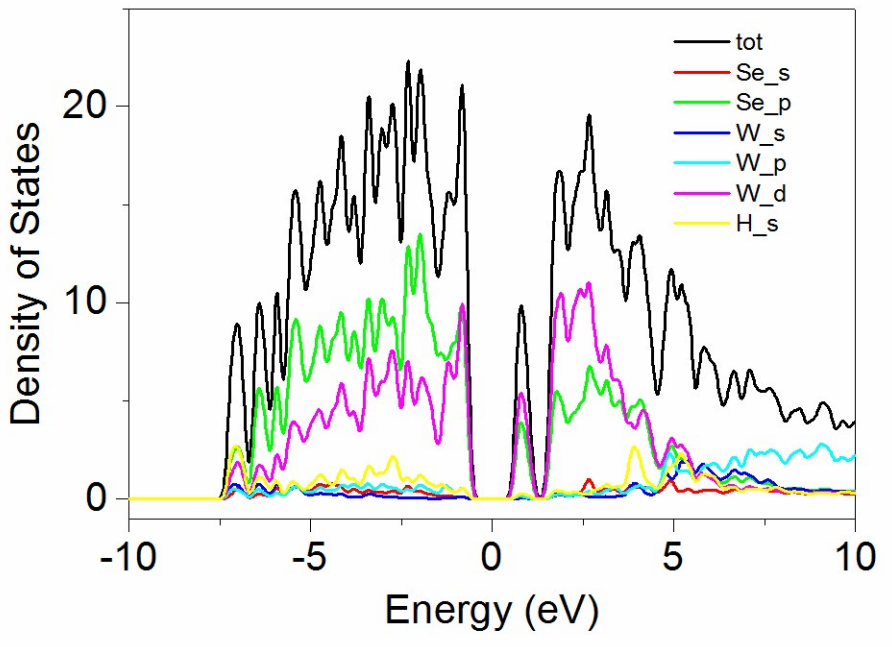

Figure S1. The atom-projected density of states of AWNR-6 with H-passivation

2. Phonon dispersion of zigzag WSe_2 nanoribbons

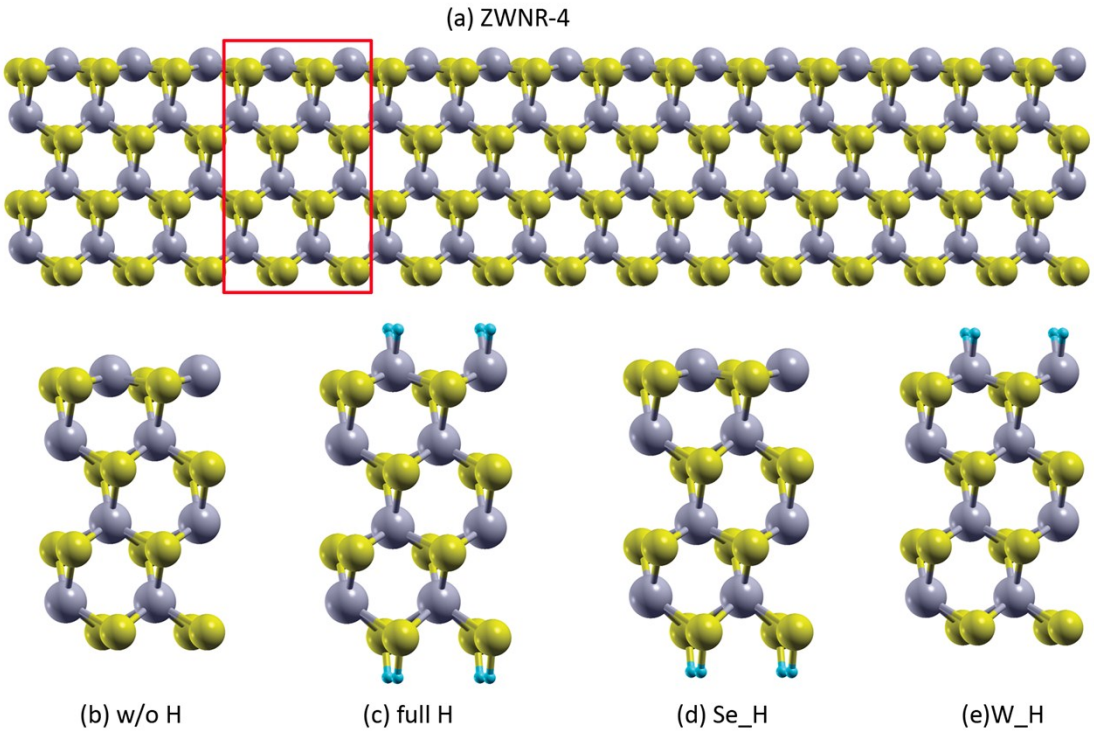

Figure S2. (a) The nanoribbon model of ZWNR-4 with a twofold supercell denoted in a red solid box. The structure of ZWNR-4 in the case of (b) without H-passivation; (c) with full H-passivation; with partial H-passivation on (d) Se atoms and (e) W atoms.

When it comes to zigzag WSe_2 nanoribbons, stability seems to be a tough problem. To start with, unlike the armchair nanoribbons, the pristine zigzag WSe_2 nanoribbons without H-passivation are thermodynamically unstable, and so are the cases with full H-passivation. ZWNR-4 is taken as an example with their structure shown in **Figure S2**. The low frequency part of phonon dispersion of ZWNR-4 without and with full H-passivation is shown in **Figure S3**a and b. It can be clearly observed that there is hugh imaginary frequency at the Z point (0, 0, 0.5). This suggests that neither unpassivated nor full passivated nanoribbons are supposed to be investigated in our work due to the instability, which means that only the partial H-passivation cases are left. There are two types of terminated atoms at each edge of zigzag nanoribbons, i.e. Se atoms and W atoms, leading to two kinds of partial H-passivation cases, denoted as Se_H and

W_H. **Figure S3**c and d show the corresponding low frequency part of phonon dispersions of these two partial passivated zigzag nanoribbons. It turns out that only the zigzag WSe_2 nanoribbons with partial H-passivation on W terminated edge are stable thermodynamically. That is why in the study of zigzag nanoribbons, we only consider the case of partial H-passivation on W atoms, which is denoted as "part H" for convenience in the paper. The zigzag nanoribbons of ZWNR-4, 5, 6, 7 with partial H-passivation are investigated, with their phonon dispersion shown in **Figure S4**. The imaginary frequency would be negligible.

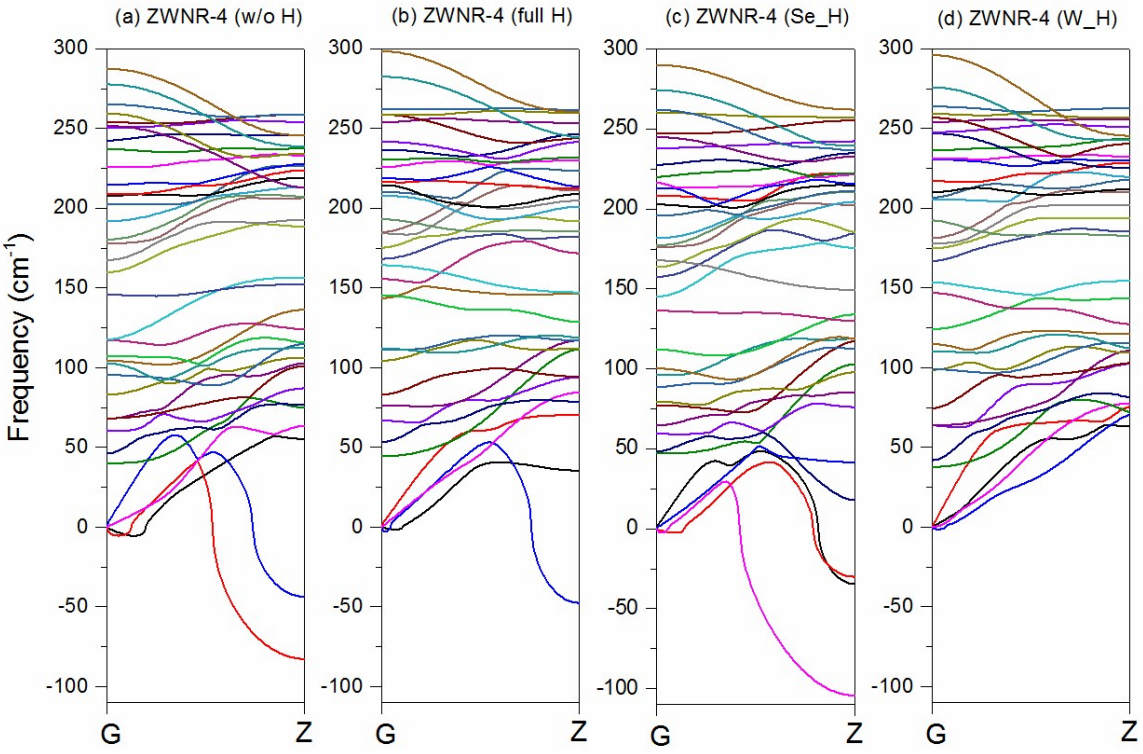

Figure S3. The low frequency part of phonon dispersion of ZWNR-4: (a) without H-passivation; (b) with full H-passivation; (c) with partial H-passivation on Se atoms; (d) with partial H-passivation on W atoms.

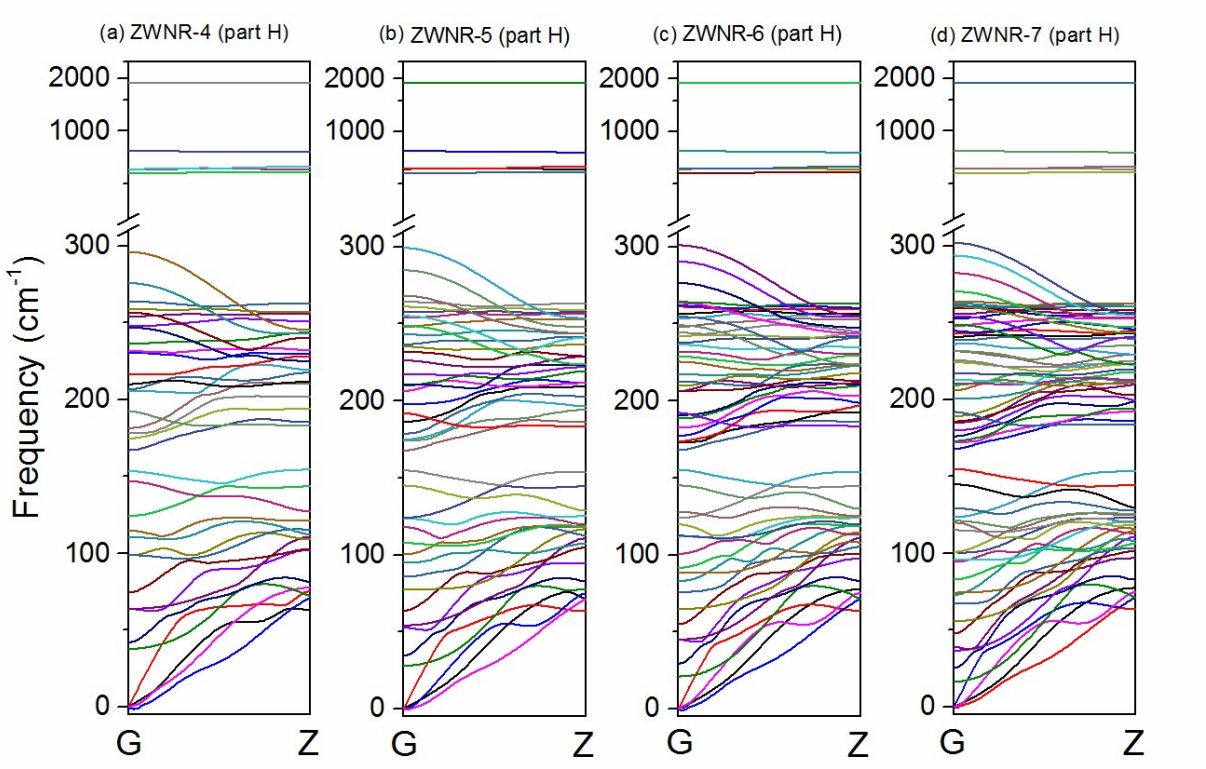

Figure S4. The phonon dispersion of zigzag WSe_2 nanoribbons with partial H-passivation for (a) ZWNR-4: (b) ZWNR-5; (c) ZWNR-6; (d) ZWNR-7.