Supplementary Information

Formation of Stable Polonium Monolayer with Tunable Semiconducting Property Driven by Strong Quantum Size Effects

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1. Phonon band structures of polonene

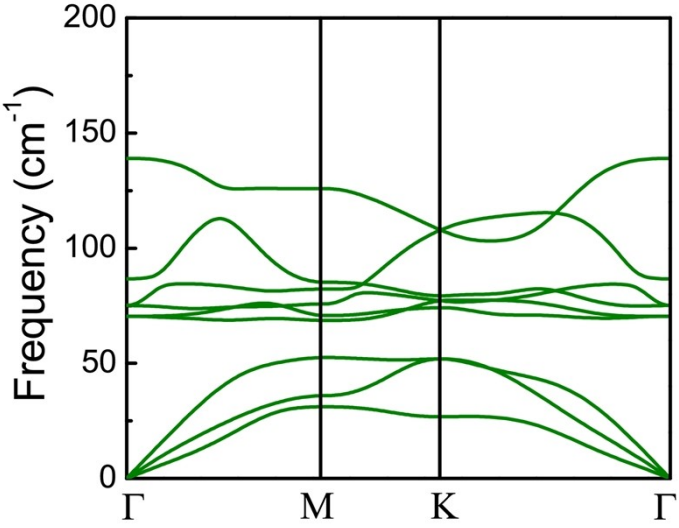

Figure S1. The calculated phonon band structure of polonene monolayer. There are no imaginary frequencies over the entire Brillouin zone (BZ), verifying its dynamic stability.

2. Ab initio molecular dynamics simulations

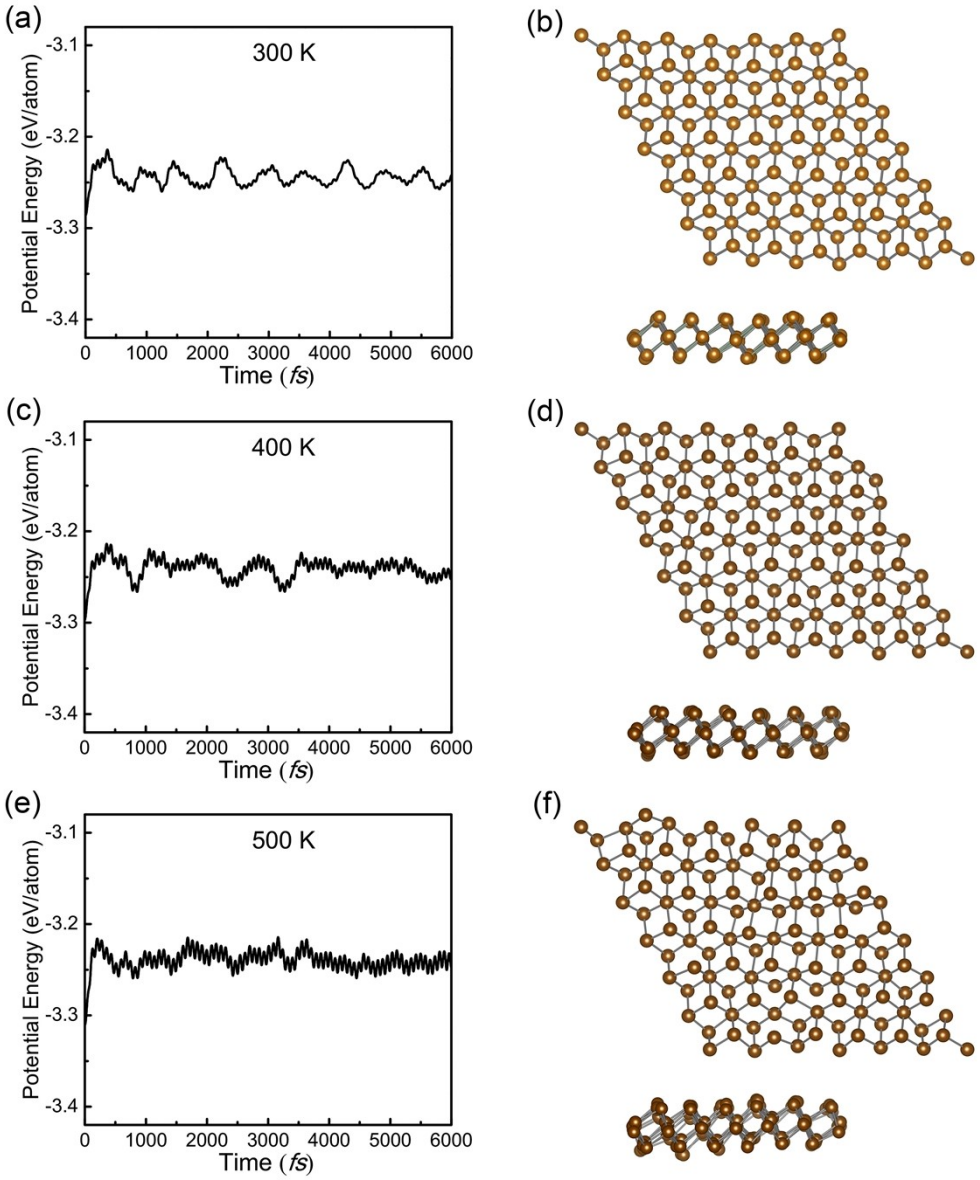

Figure S2. The potential energy fluctuations as a function of the molecular dynamic simulation step at (a)300, (c)400 and (e)500 K for polonene monolayer, respectively. The top and side views of polonene after *ab initio* MD simulations at (b)300, (d)400 and (f)500 K for 6ps in step of 2 fs, respectively. The potential energy fluctuation and structural changes of polonene are very small at both 300 and 400 K for 6 ps, while some structure disorders occur as the temperature increase to 500 K, indicating that it is thermally stable below the temperature of 400 K.

3. Lattice constants of Po films with different thicknesses

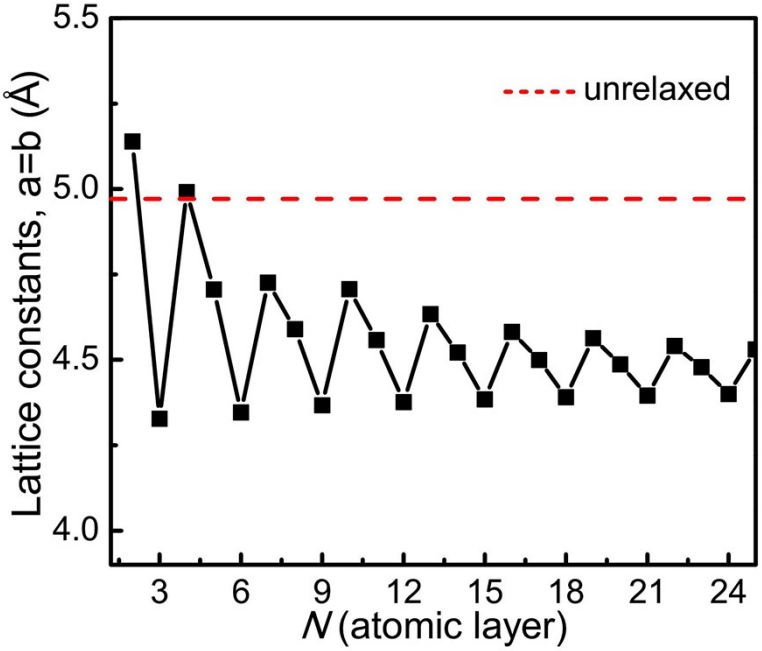

Figure S3. The calculated lattice constants in xy plane of the fully optimized bulk-truncated Po(001) films as a function of the film thickness, and the red dashed line corresponding to that of the unrelaxed β-Po(001) films.

4. Electronic density of states for different Po films

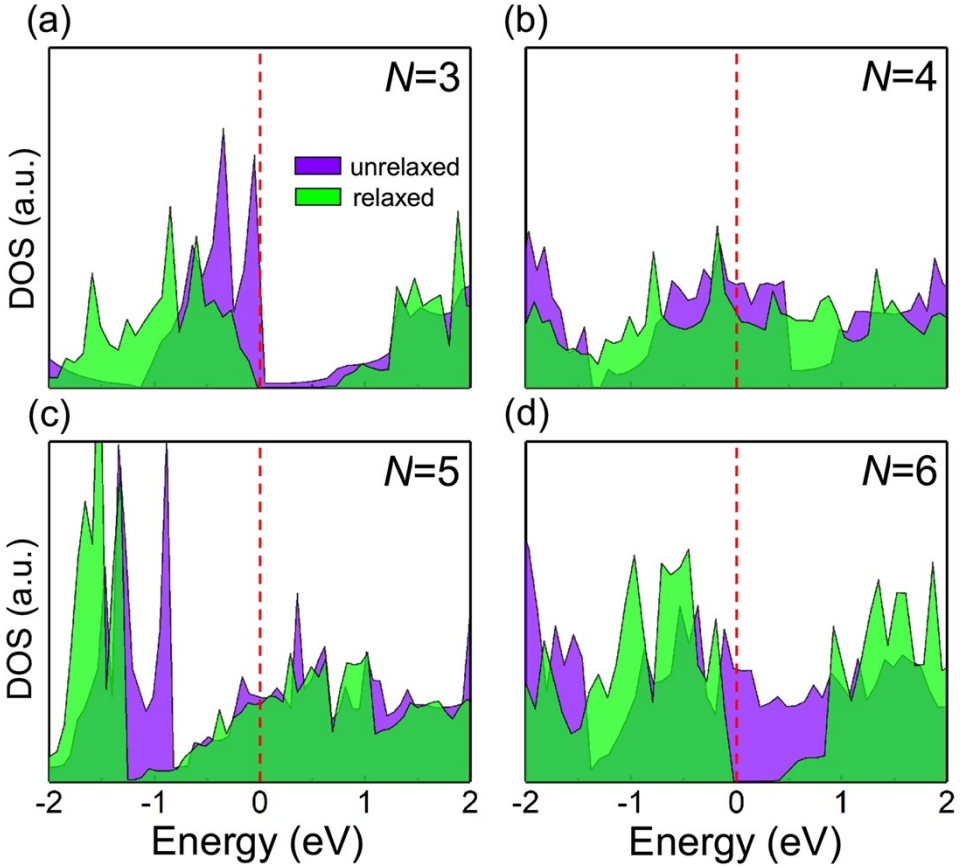

Figure S4. (a)-(d) The calculated electronic density of states for both unrelaxed (violet) and relaxed (green) Po films with thicknesses of $N = 3$, 4, 5 and 6, respectively.

5. The vibration modes

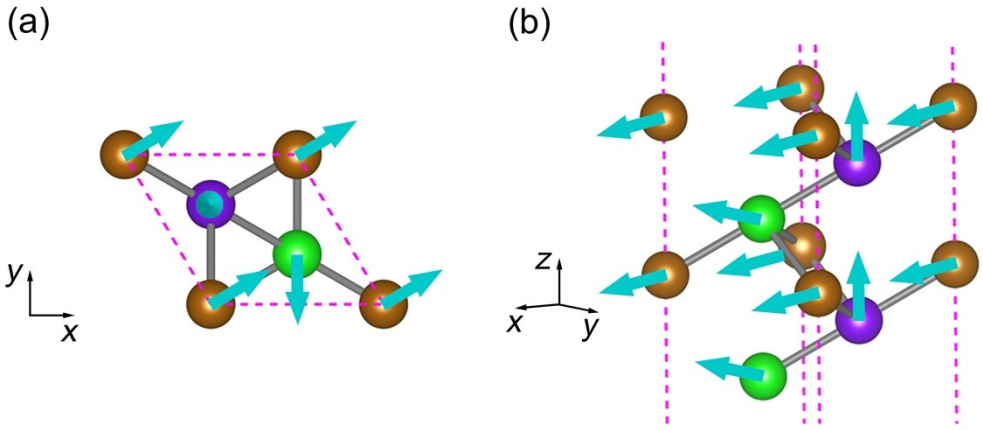

Figure S5. (a)-(b) The vibration modes of the unrelaxed β-Po(001) slab with thickness of $N = 6$ from the top and perspective view, respectively. The cyan arrows indicate the vibration directions of atoms.

6. Pb films on the CdSe(111) substrate

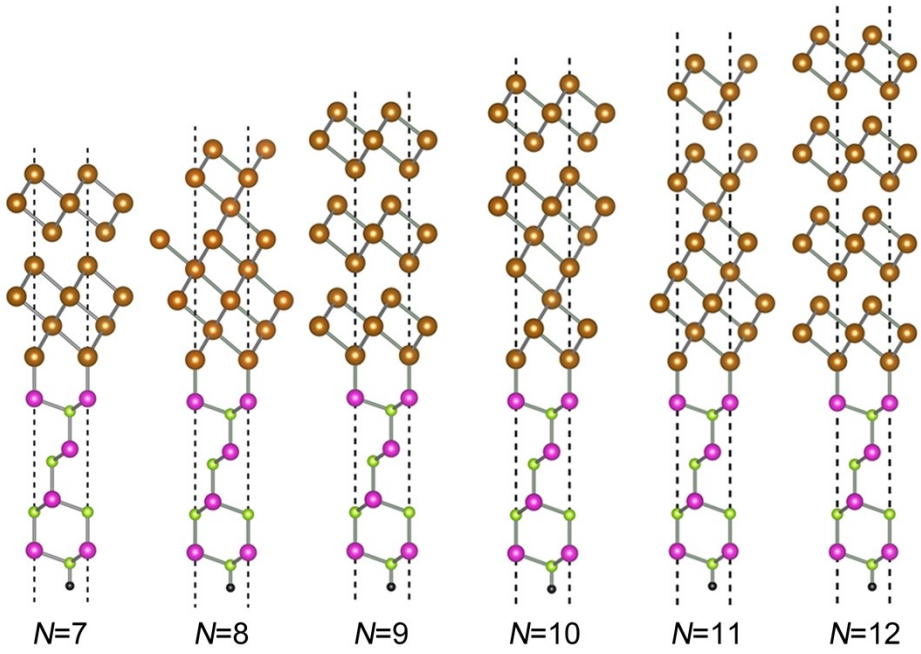

Figure S6. The relaxed geometric structures of Pb films with thicknesses from N = 7 to 12 on the CdSe(111) substrate, respectively.