Supplementary Information (SI) for Nanoscale.
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A Type-I van der Waals Heterostructure formed by Monolayer WS_2 and Trilayer $PdSe_2$

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1. X-ray photoelectron spectroscopy (XPS)

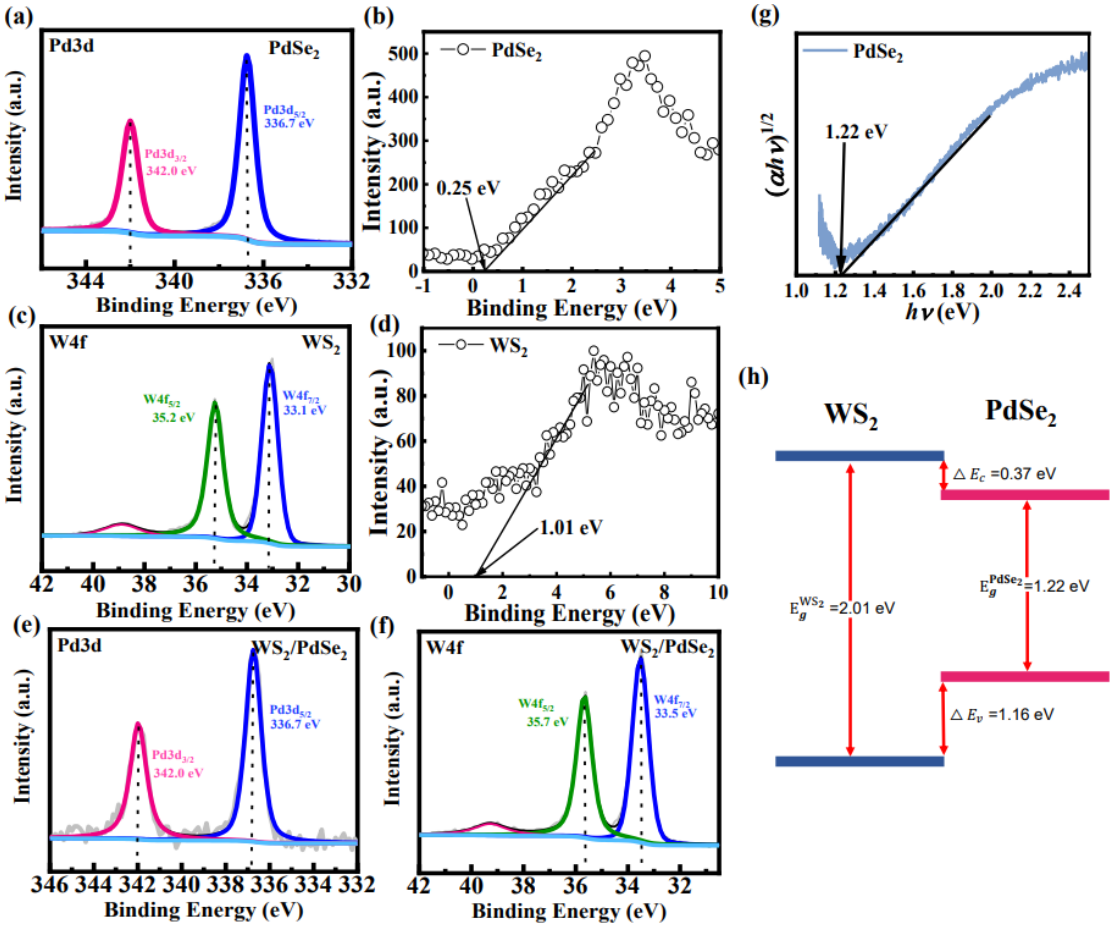

Figure S1. (a) The core-level Pd3d from trilayer $PdSe_2$. (b) The valence band spectrum obtained from trilayer $PdSe_2$. (c) The core-level W4f obtained from

monolayer WS_2. (d) The valence band spectrum obtained from monolayer WS_2. (e) The core-level Pd3d from $WS_2/PdSe_2$ heterostructure. (f) The core-level W4f obtained from $WS_2/PdSe_2$ heterostructure. (g) Indirect bandgap calculation of trilayer $PdSe_2$. (h) The schematic illustration of the band alignment of the $WS_2/PdSe_2$ heterostructure.

2. Computation Method:

All computations were carried out within the density functional theory (DFT) methods implemented in the Vienna ab initio simulation package. In the theoretical calculation process, the generalized gradient approximation PBE (GGA-PBE) of the exchange and correlation functionals were used. The cutoff energy of the selected plane wave function is 400 eV. In the calculation of geometric relaxation and single point energy, the convergence criterion for energy is 10^{-6} eV, and the convergence criterion for force is 0.015 eV/Å. Meanwhile, in order to avoid the influence of periodic effects on the two-dimensional structure, we established a 20 Å vacuum layer in the z-direction. For computing the band structure of the $PdSe_2$, the Brillouin zone of Γ-X-S-Y-Γ was selected. For monolayer WS_2, the Brillouin zone of gamma Γ-M-K-Γ was chosen. The band structure of $PdSe_2$ crystal exhibits an indirect bandgap. It can be observed that the CBM and VBM of monolayer WS_2 are located at point K, and the CBM and VBM of monolayer, bilayer, and trilayer $PdSe_2$ are all located between the CBM and VBM of monolayer WS_2. Through theoretical calculations of band alignment, it can be reasonably concluded that WS_2/1L-$PdSe_2$, WS_2/2L-PdS_2, WS_2/3L-$PdSe_2$ are both type I heterostructures.

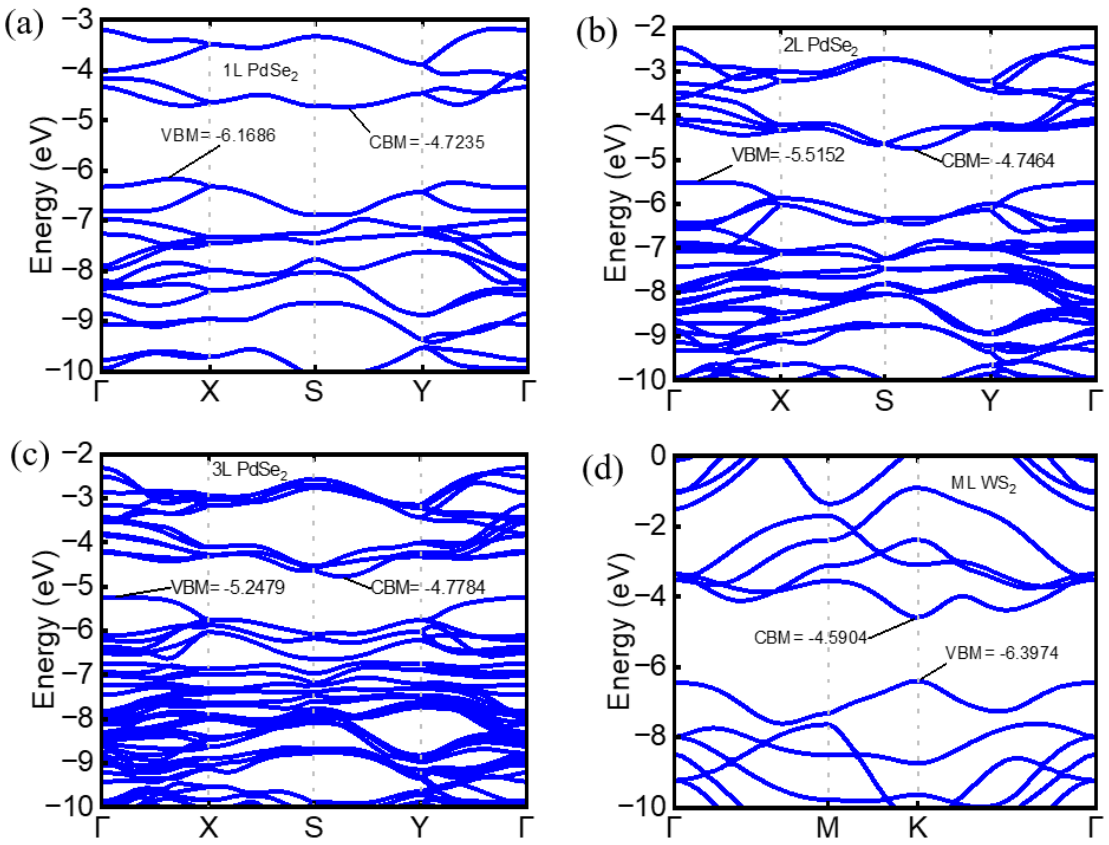

Figure S2. (a)-(c) Calculated bandstructure for the $PdSe_2$ (1-3 layers). (d) Calculated bandstructure for the monolayer WS_2.