Supporting Information
Tunable n-type and p-type doping of two-dimensional layered $PdSe_2$ via organic molecular adsorption

Yunzhi Gao,[1] Xiaofeng Liu,[1] Wei Hu,[1,*] and Jinlong Yang[1,†]

[1] *Hefei National Laboratory for Physical Sciences at the Microscale, Department of Chemical Physics, and Synergetic Innovation Center of Quantum Information and Quantum Physics, University of Science and Technology of China, Hefei, Anhui 230026, China*

[*] E-mail: whuustc@ustc.edu.cn
[†] E-mail: jlyang@ustc.edu.cn

TABLE S1: Total energy of momolayer $3 \times 3 \times 1$ $PdSe_2$ supercell using $3 \times 3 \times 1$, $4 \times 4 \times 1$ and $5 \times 5 \times 1$ k grids.

K grids	E_{tot} (eV)
$3 \times 3 \times 1$	-104.86824
$4 \times 4 \times 1$	-104.86825
$5 \times 5 \times 1$	-104.86828

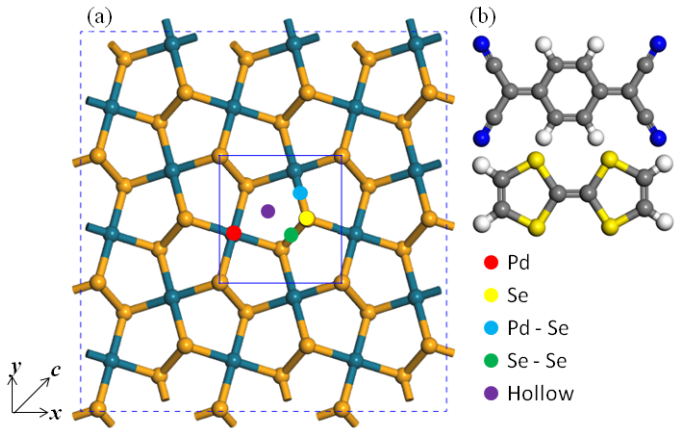

FIG. S1: (a) Adsorption sites and molecular orientations of TCNQ and TTF adsorbed on $PdSe_2$. Five possible molecular adsorption sites above Pd atom, Se atom, Pd-Se bond, Se-Se bond and the center of the pentagon are marked by red, yellow, blue, green and purple circles, respectively. Three possible molecular orientation, molecular symmetry axis are set to x axis, y axis and angle bisector between them, are represented by three arrows x, y and c, respectively. (b) The molecular configuration of TCNQ and TTF.

TABLE S2: The adsorption properties of organic TCNQ and TTF molecular adsorption on monolayer $2 \times 2 \times 1$ $PdSe_2$ supercell, including the adsorption energy E_{ads} (eV), and equilibrium distance D (Å) and charge transfer ΔQ (e).

Systems	E_{ads}	D	ΔQ
TCNQ-ML-$PdSe_2$	-1.19	3.26	0.104
TTF-ML-$PdSe_2$	-0.950	3.35	-0.0671

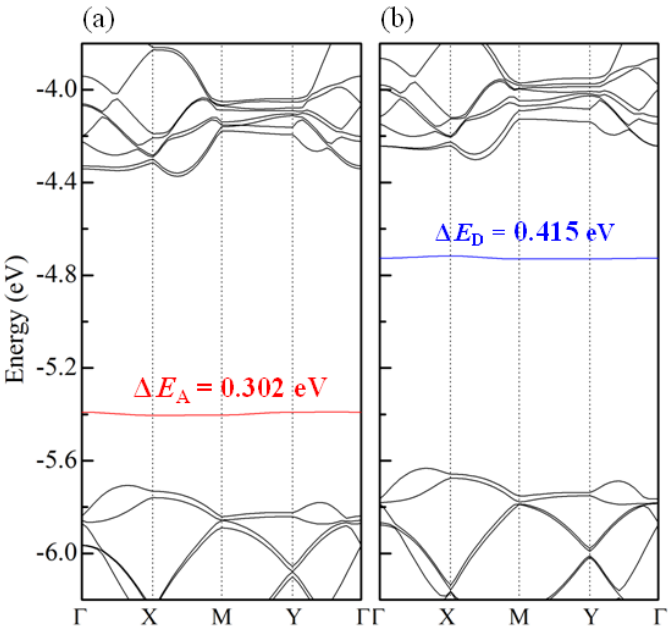

FIG. S2: Band structures of (a) TCNQ-$PdSe_2$ and (b) TTF-$PdSe_2$ by using PBE functional, respectively. The TCNQ and TTF molecules are adsorbed on monolayer $2 \times 2 \times 1$ $PdSe_2$ supercell.

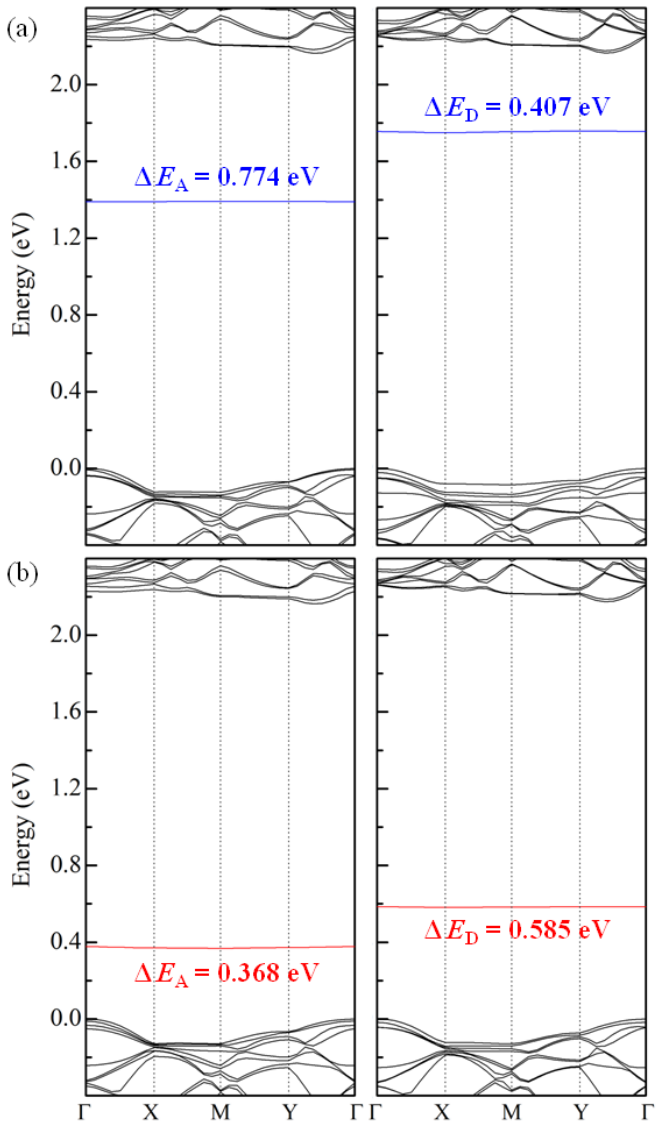

FIG. S3: Band structures of TCNQ-ML-$PdSe_2$ (the left) and TTF-ML-$PdSe_2$ (the right) by using HSE06 functional with applied electric fields of (a) +0.4 V/Å and (b) -0.4 V/Å, respectively. The VBMs of $PdSe_2$ in doping systems under external electric fields are set to zero.

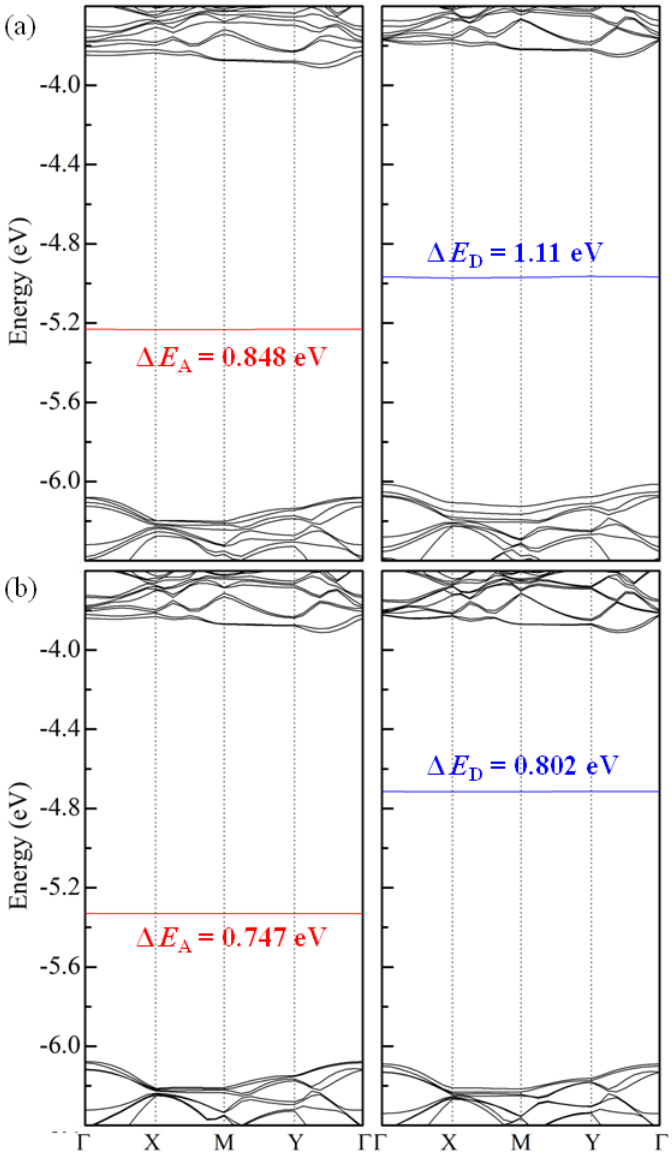

FIG. S4: Band structures of TCNQ-ML-$PdSe_2$ (the left) and TTF-ML-$PdSe_2$ (the right) by using HSE06 functional at distances of (a) -0.4 Å and (b) +0.4 Å, respectively.

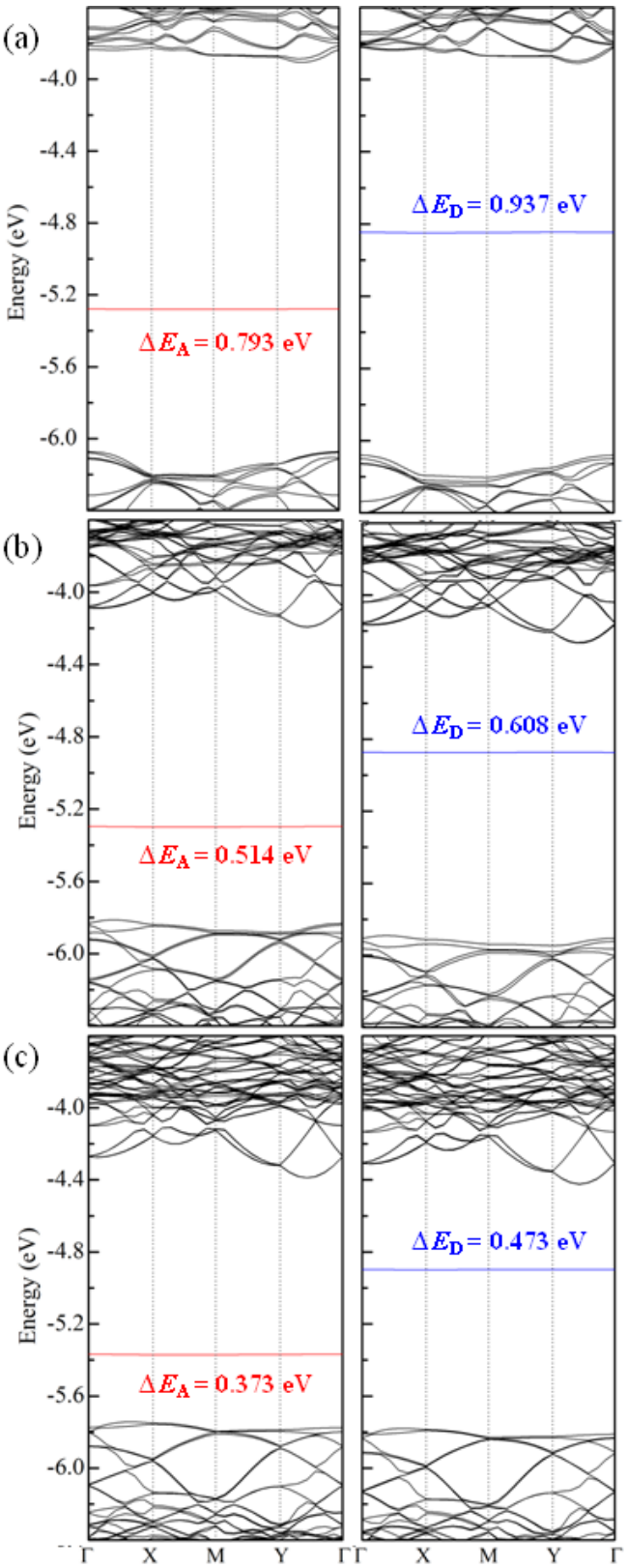

FIG. S5: Band structures of (a) TCNQ-ML-$PdSe_2$ (the left) and TTF-ML-$PdSe_2$ (the right), (b) TCNQ-BL-$PdSe_2$ (the left) and TTF-BL-$PdSe_2$ (the right), (c) TCNQ-TL-$PdSe_2$ (the left) and TTF-TL-$PdSe_2$ (the right) using HSE06 functional.