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

Multi-Factor Adjustable $PtSe_2$/GaN van der Waals Heterostructure with Enhanced Photocatalytic performance

Qihao Zhang[1], Hua Zhu[1], Xiaodong Yang[2], Liang Chen[1] and Yang Shen[1, 3, *]

[1] *Institute of Optoelectronics Technology, China Jiliang University, Hangzhou 310018, China*

[2]*Key Laboratory of Ecophysics and Department of Physics, Shihezi University, Xinjiang 832003, China*

[3]*School of Materials Science and Engineering, Zhejiang University, Hangzhou 310027, China*

[*]Correspondence and requests for materials should be addressed to Y. Shen (yshen@cjlu.edu.cn)

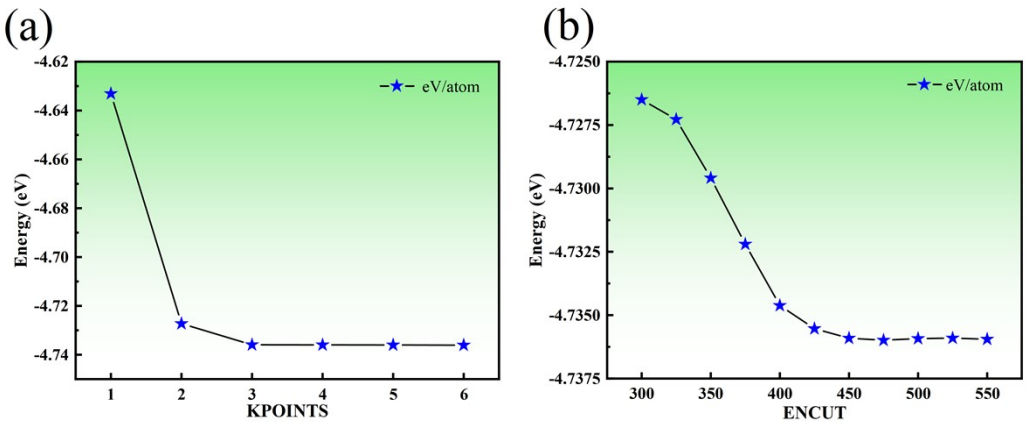

Fig. S1. The convergence test of (a) KPOINTS and (b) ENCUT. The Y-axis energy represents the average energy per atom.

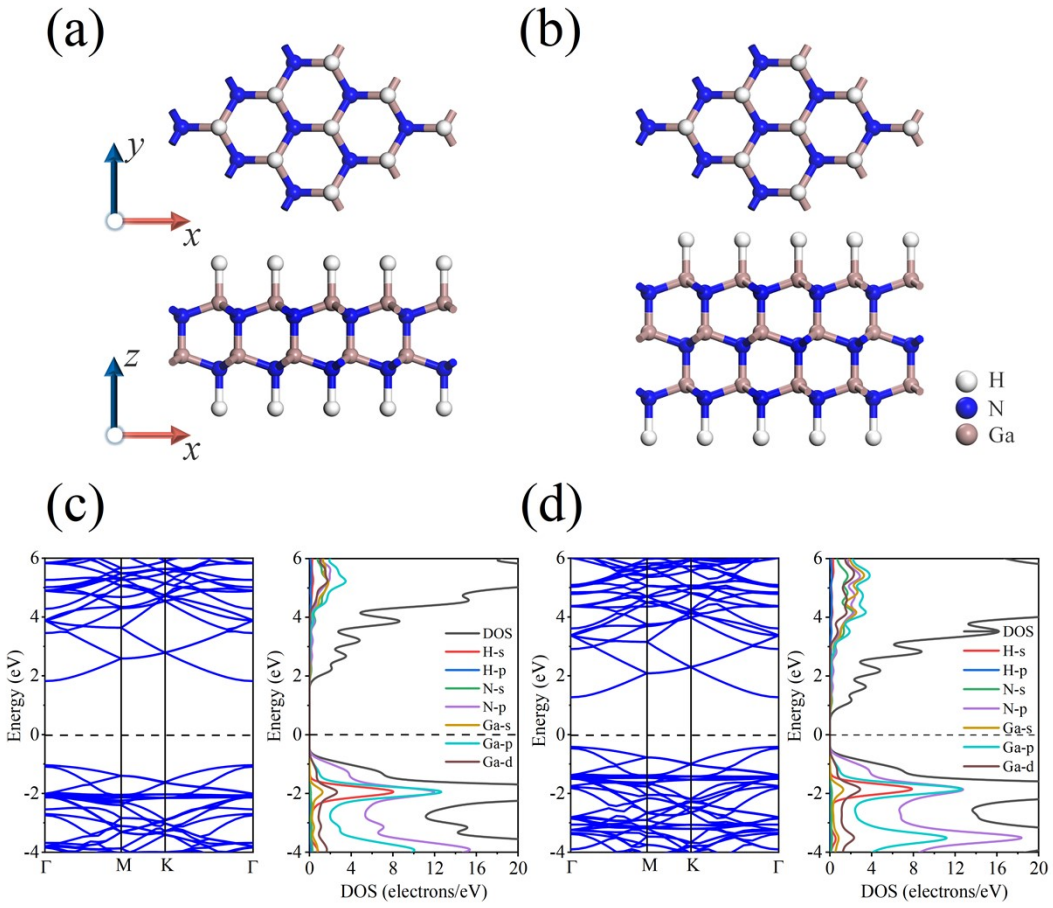

Fig. S2. The atomic structure of buckling GaN (a) BL and (b) TL. The upper part is a top view, and the lower part is a side view. Band structures and PDOS of buckling GaN (c) BL and (d) TL.

Table S1

The lattice parameter a, binding energy E_b and bandgap E_g of the $PtSe_2$ monolayer, the buckling GaN nanosheets with different thickness, and $PtSe_2$/GaN ML heterostructures with four different configurations.

	a(Å)	E_b (eV)	PBE E_g (eV)	HSE06 E_g (eV)
$PtSe_2$	3.75	-	1.4014	2.1301
Ga-1L	3.20	-	3.2838	4.3879
Ga-2L	3.22	-	1.2942	2.1695
Ga-3L	3.24	-	0.1509	1.6858
AB1	6.45	-0.08	1.1863	1.8882
AB2	-	-0.13	1.1846	1.8862
AB3	-	-0.14	1.1852	1.8861
AB4	-	-0.10	1.1869	1.8865

Table S2

The vacuum level Φ of different components of the heterostructures and bandgap value of six configurations.

	$\Phi_{GaN}\ (eV)$	$\Phi_{PtSe_2}\ (eV)$	$\Delta\Phi\ (eV)$	$E_g\ (eV)$
Ga-1L	1.611	4.894	3.283	1.866
Ga-2L	0.914	5.483	4.569	1.375
Ga-3L	1.000	5.852	4.852	0.477
N-1L	6.496	3.315	3.181	1.489
N-2L	6.576	3.340	3.236	0.356
N-3L	6.803	3.462	3.341	0.112

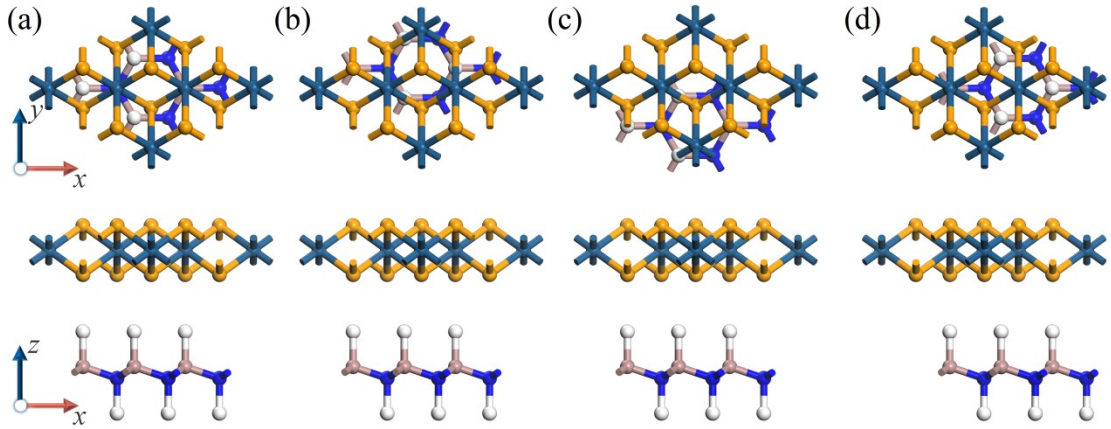

Fig. S3. The atomic structure of (a) AB1, (b) AB2, (c) AB3 and (d) AB4 configurations of the $PtSe_2$/GaN heterostructures. The upper part is a top view, and the lower part is a side view.\

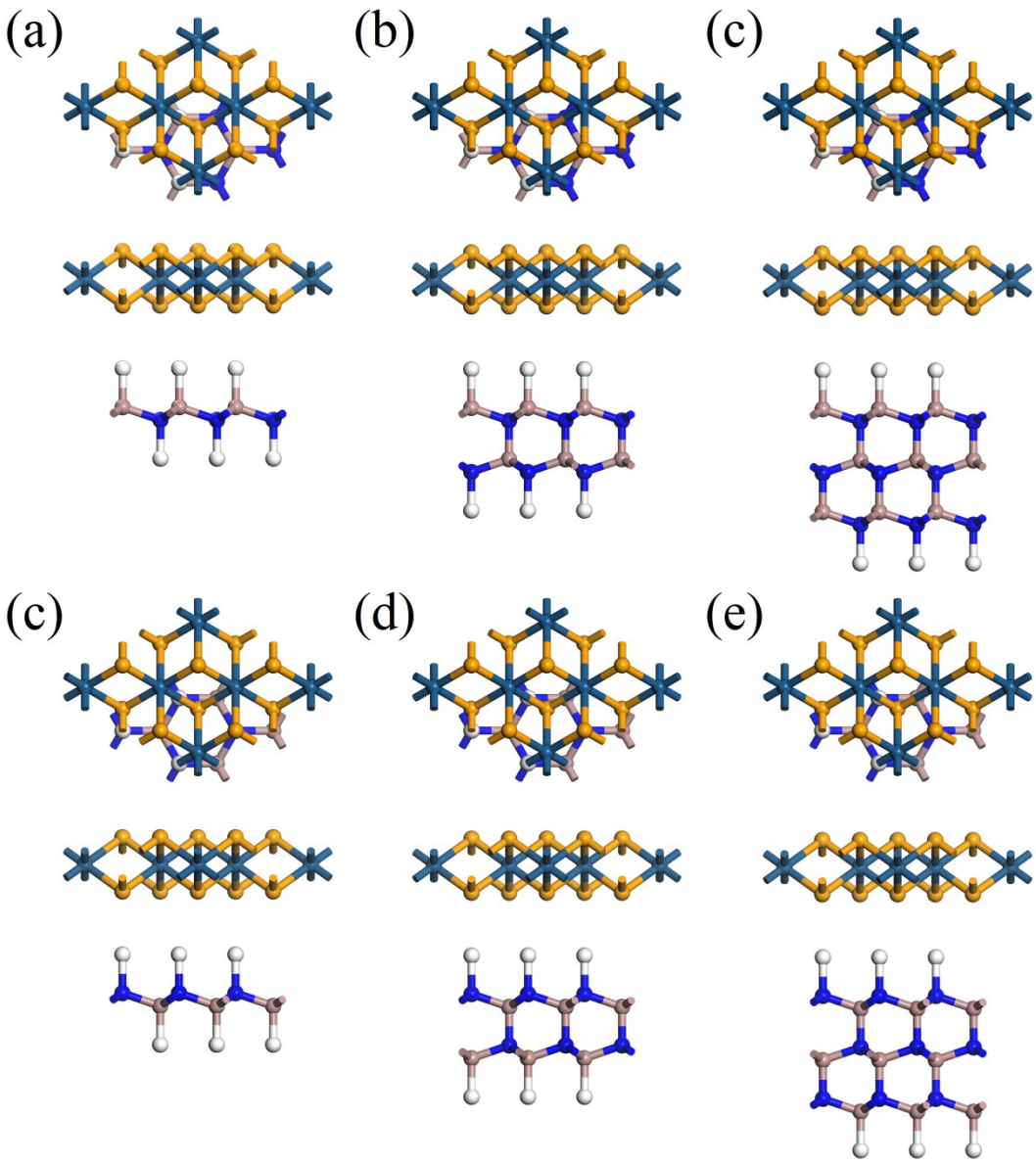

Fig. S4. The atomic structures of the six different heterostructures, respectively, (a) Ga-1L, (b) Ga-2L, (c) Ga-3L, (d) N-1L, (e) N-2L and (f) N-3L

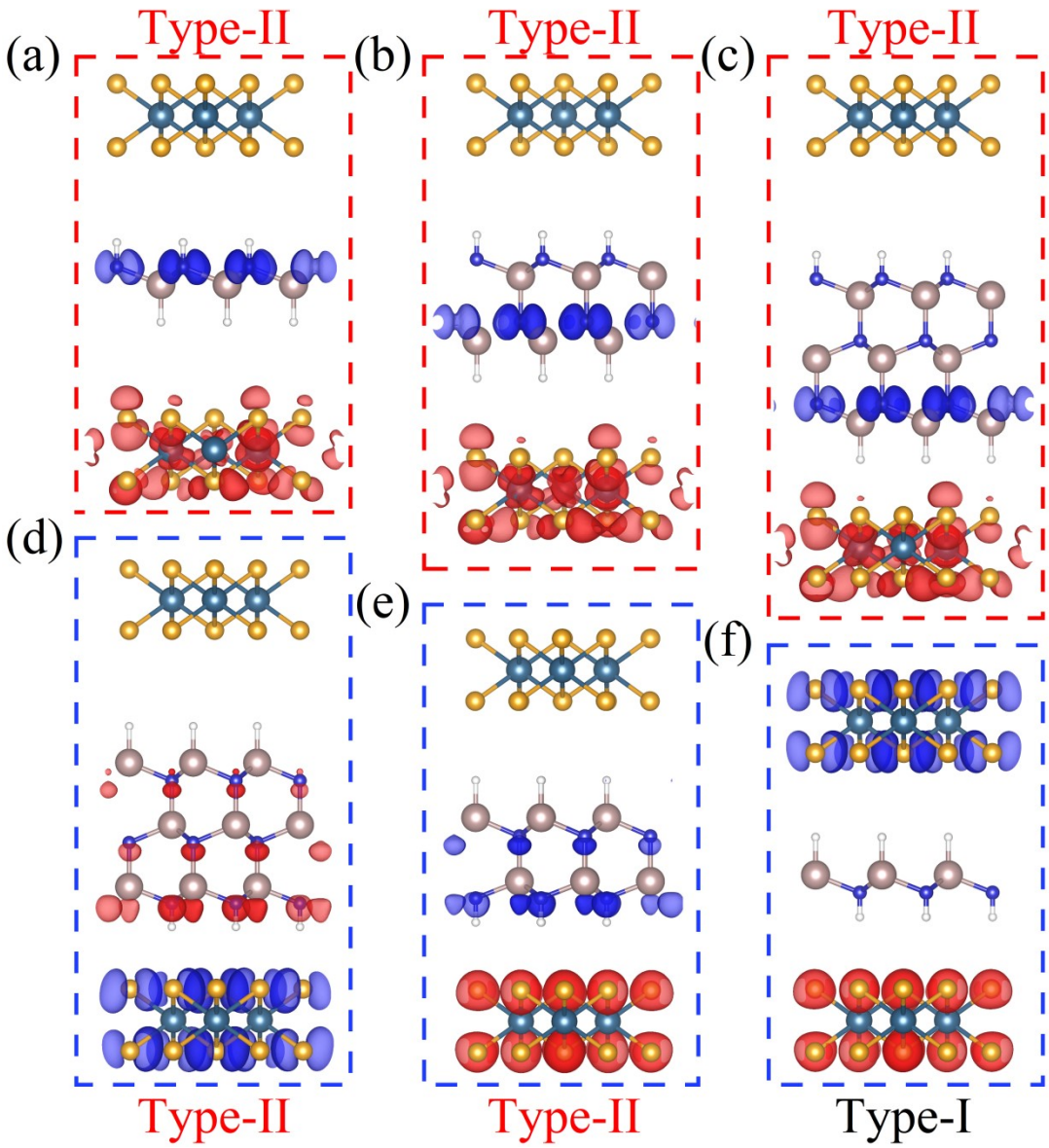

Fig. S5. The type of the band alignment and the band charge density distribution of CBM and VBM for the (a) N-1L, (b) N-2L, (c) N-3L, (d) Ga-3L, (e) Ga-2L and (f) Ga-1L.

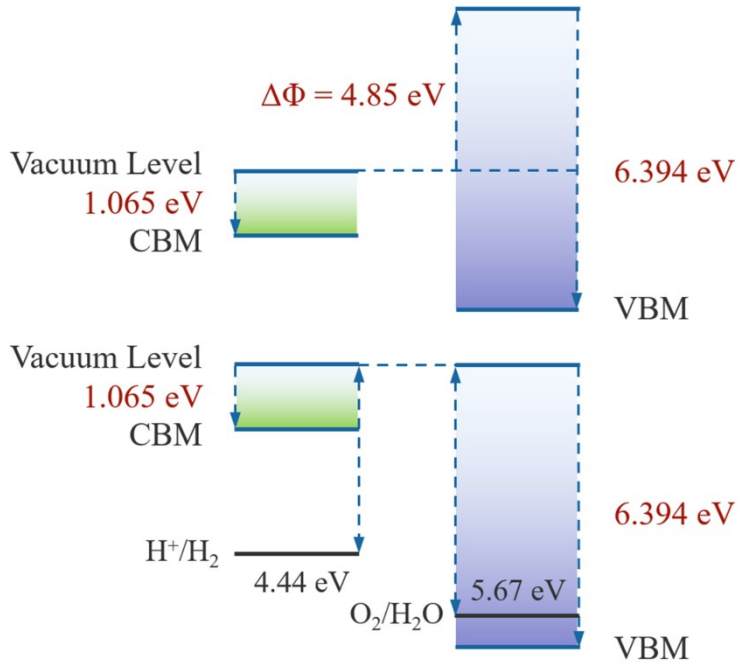

Fig. S6. The schematic diagram of the band alignment of Ga-3L with different vacuum levels (upper panel) and aligned vacuum levels (lower panel).

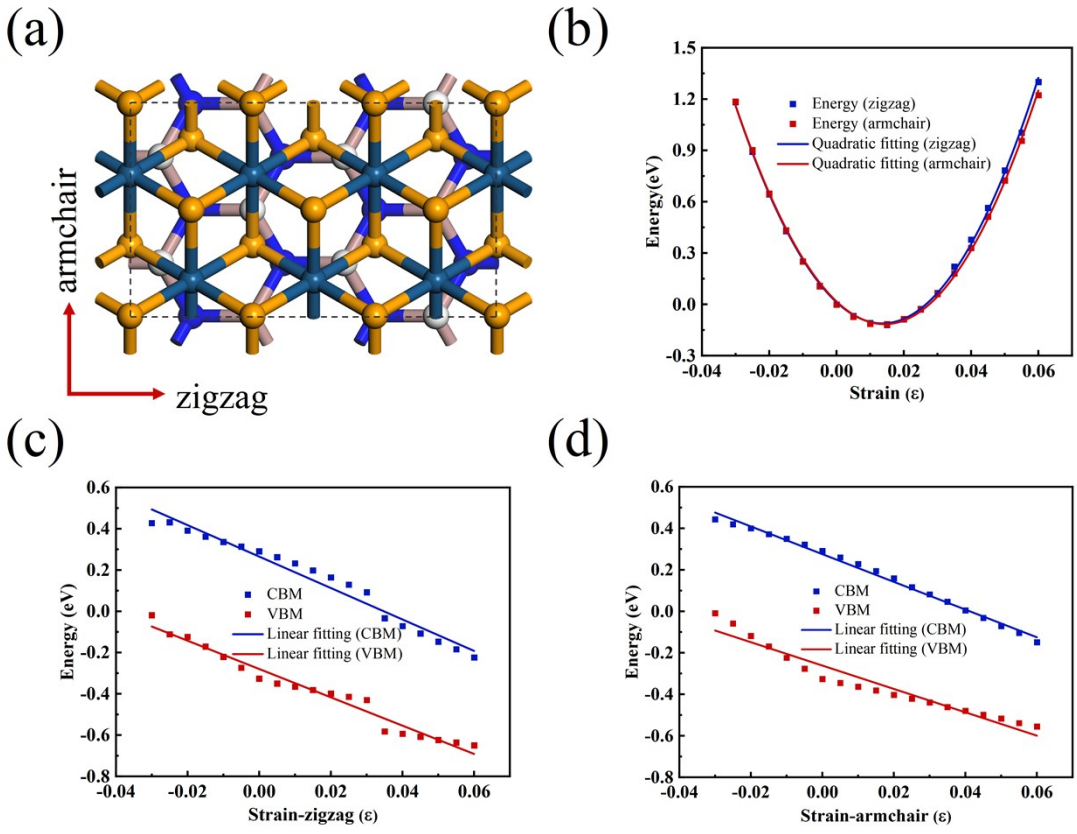

Fig. S7. (a) The carrier transport directions in the rectangular unit cell of Ga-3L. (b) The energy is a function of uniaxial strain and their quadratic fitting gives the elastic modulus. The total energy of systems without applied strain is defined as 0 eV. (c) and (d) The relationship between band edges and the uniaxial strain. Their linear fitting offers the deformational potential constant.