Supporting Information

Two-dimensional direct Z-scheme AlN/GaS-SiP heterojunction enhances photocatalytic hydrogen production from water: a DFT study

Zeting Gong[1], Yongsheng Yao[1], Yaozhong Liu[1], Zheng Liang[1], Junyao Li[1], Zhenkun Tang[1], Xiaolin Wei[1]**

[1] Key Laboratory of Micro-nano Energy Materials and Application Technologies, University of Hunan Province &College of Physics and Electronics Engineering, Hengyang Normal University, Hengyang 421002, China

Email: yaoys@hynu.edu.cn, xlw@xtu.edu.cn

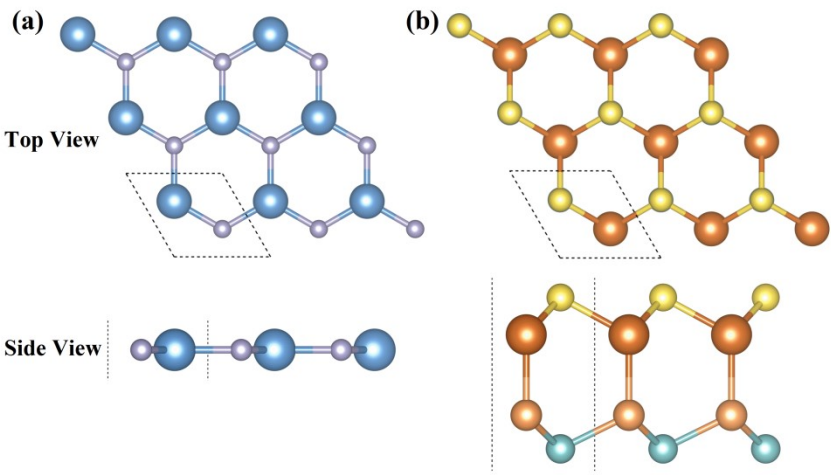

Fig. S1 Top and side views for the monolayer structure of (a) AlN and (b) SiP-GaS. Navy blue, silver, cyan, light orange, yellow, orange represent Al, N, P, Si, S and Ga atoms, respectively.

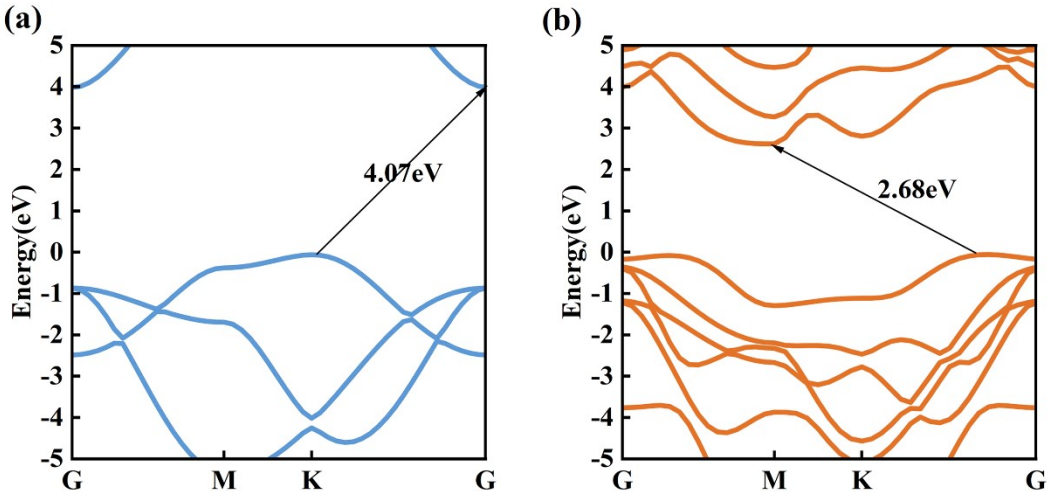

Fig. S2 Band structures of monolayer (a) AlN and (b) SiP-GaS

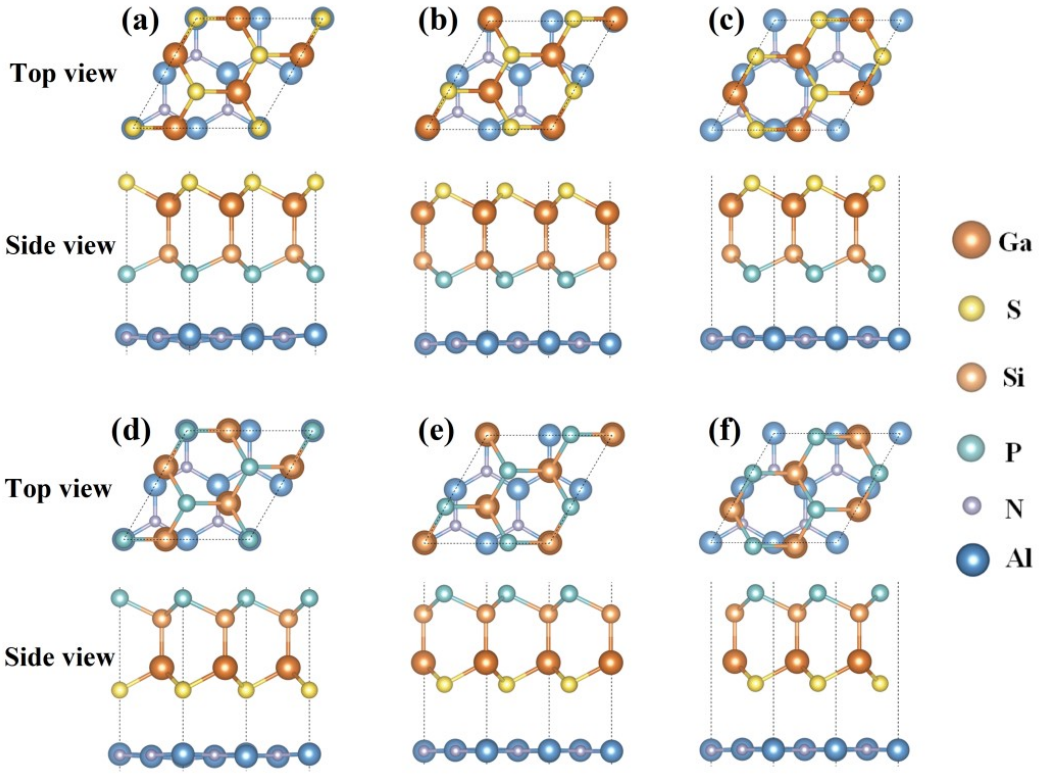

Fig. S3 Top and side views for the optimized structure of (a-c) N/P-(I-III) and (d-f) N/S-(I-III). Navy blue, silver, cyan, light orange, yellow, orange represent Al, N, P, Si, S and Ga atoms, respectively.

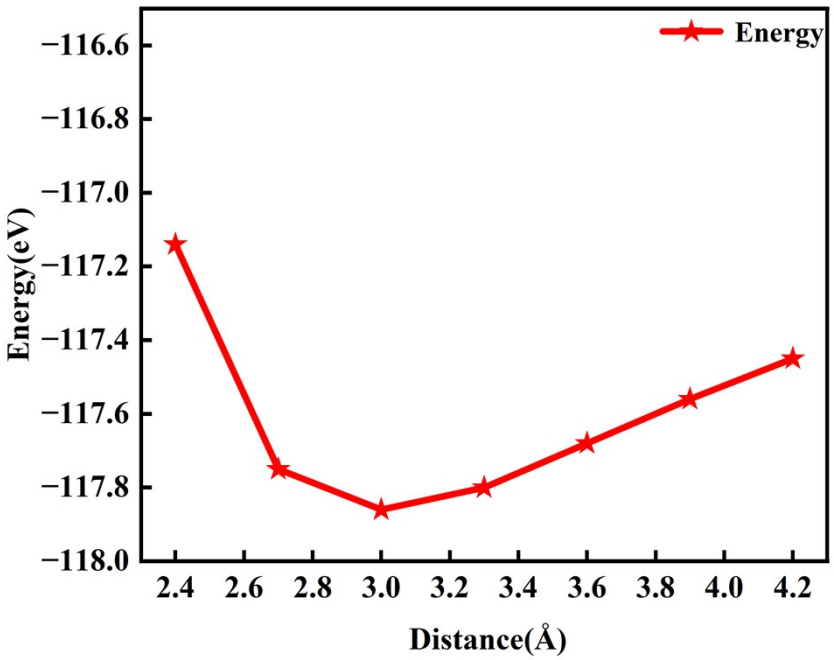

Fig. S4 The total energy change at different interlayer distances.

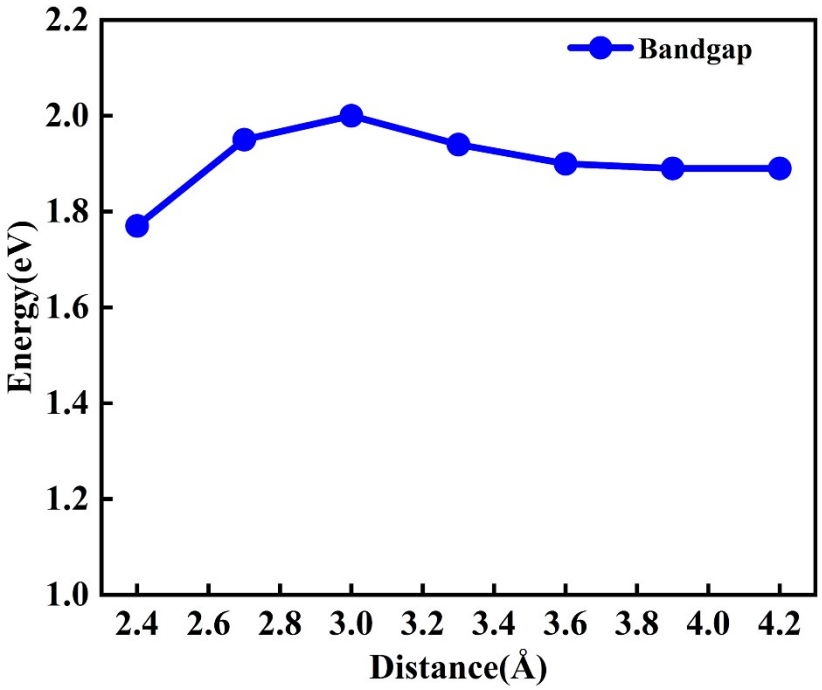

Fig. S5 The bandgap changes at different interlayer distances.

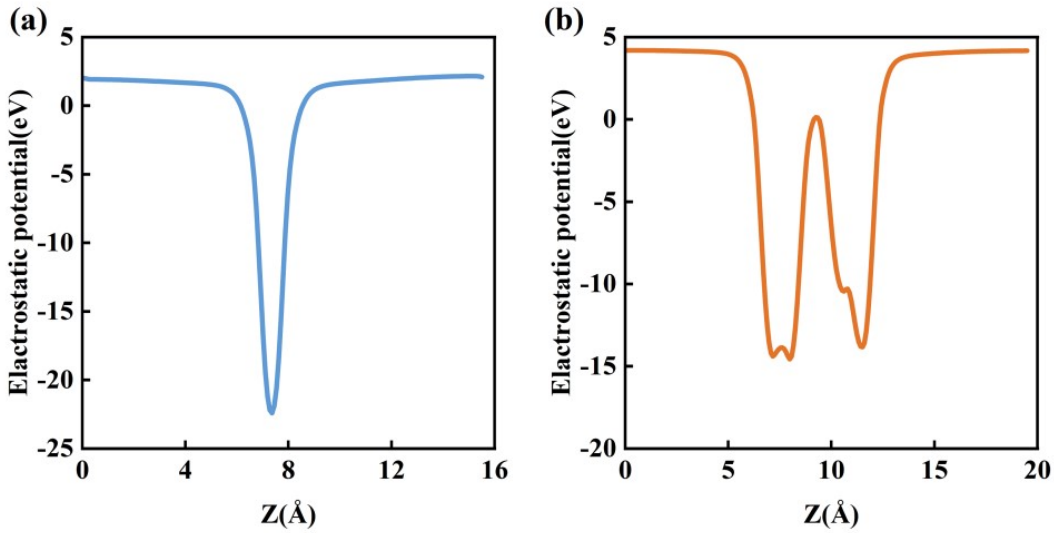

Fig. S6 The wok function of monolayer (a) AlN and (b) SiP-GaS

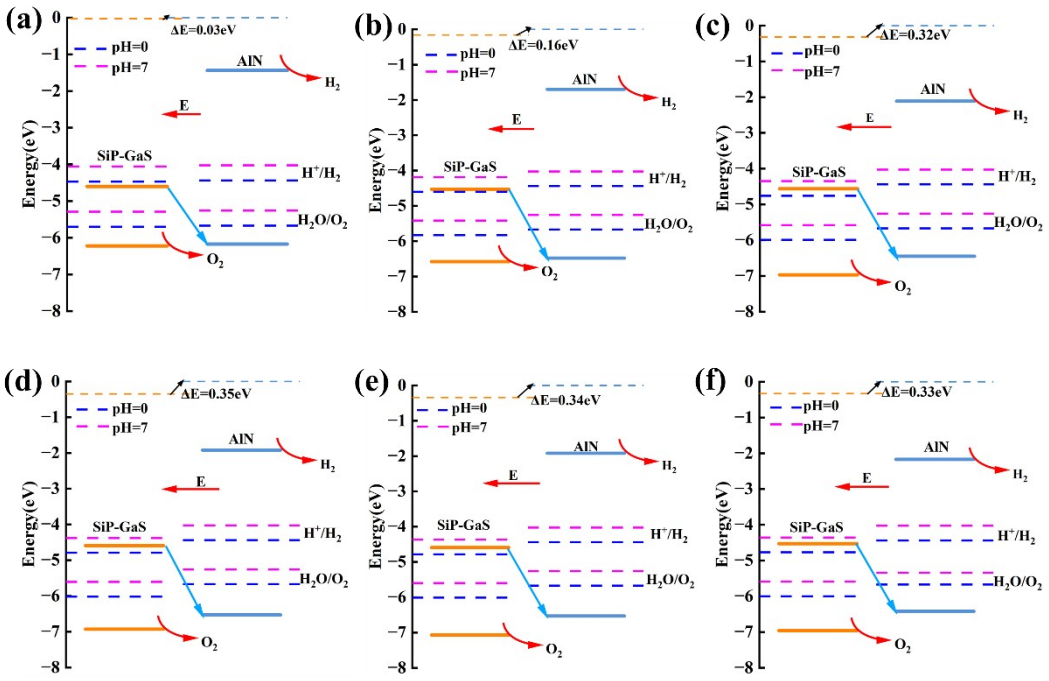

Fig. S7 The band edge position of N/P-I heterojunction at pH=0 and pH= 7 (a) d=2.4 Å, (b) d=2.7 Å，(c) d=3.3 Å，(d) d=3.6 Å，(e) d=3.9 Å and (f) d=4.2 Å.

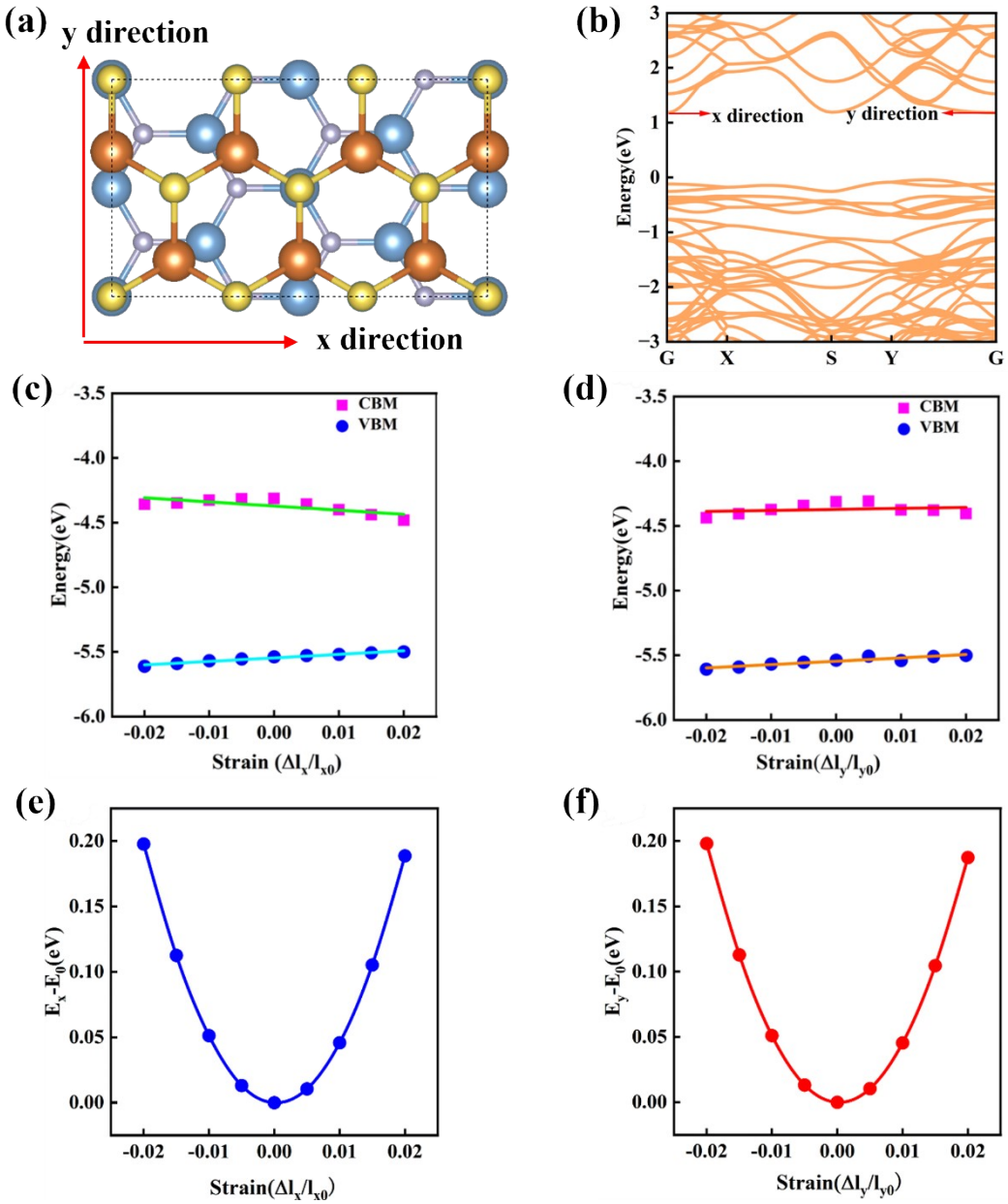

Fig. S8 (a) The geometric structure and (b) band structure of N/P-I heterojunction. The band edge positions (CBM and VBM) of N/P-I heterojunction along the (c) x and (d) y directions under uniaxial strain. The energy changes of N/P-I heterojunction along the (e) x and (f) y directions under uniaxial strain.

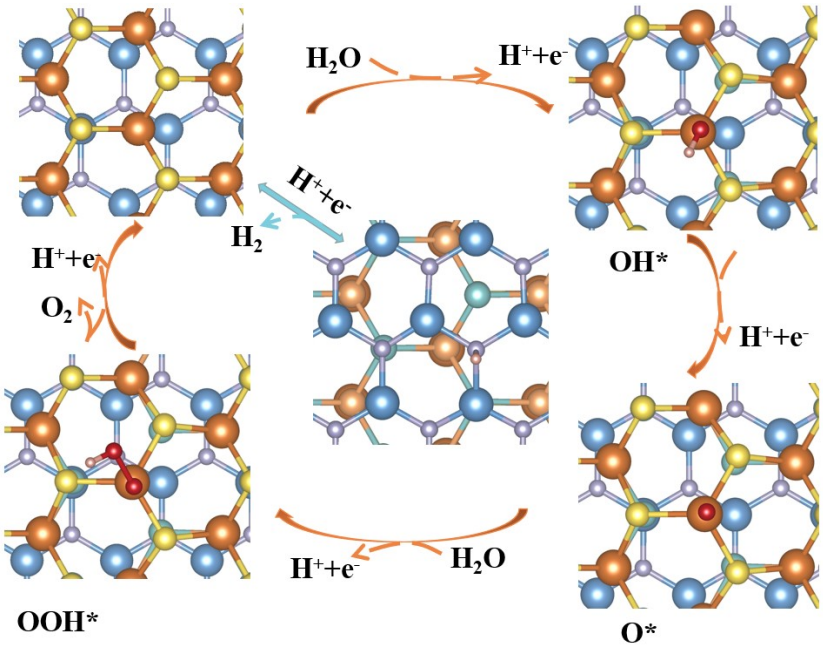

Fig. S9 The adsorption simulation process of HER and OER for N/P-I heterojunction for the most stable configurations. Navy blue, silver, cyan, light orange, yellow, orange, pink and red represent Al, N, P, Si, S, Ga, H and O atoms, respectively.

The optical driving force (U_e) for HER at a standard hydrogen electrode is defined as the energy difference between the CBM of the reduced photocatalyst and the reduction potential, while the optical driving force (U_h) for OER is the energy difference between the VBM of the oxidized photocatalyst and the reduction potential. The values of U_e and U_h will also be different under different pH. The calculation formula is as follows:

$$U_e = U_e(pH = 0) - pH \times 0.059V \quad (1)$$

$$U_h = U_h(pH = 0) + pH \times 0.059V \quad (2)$$

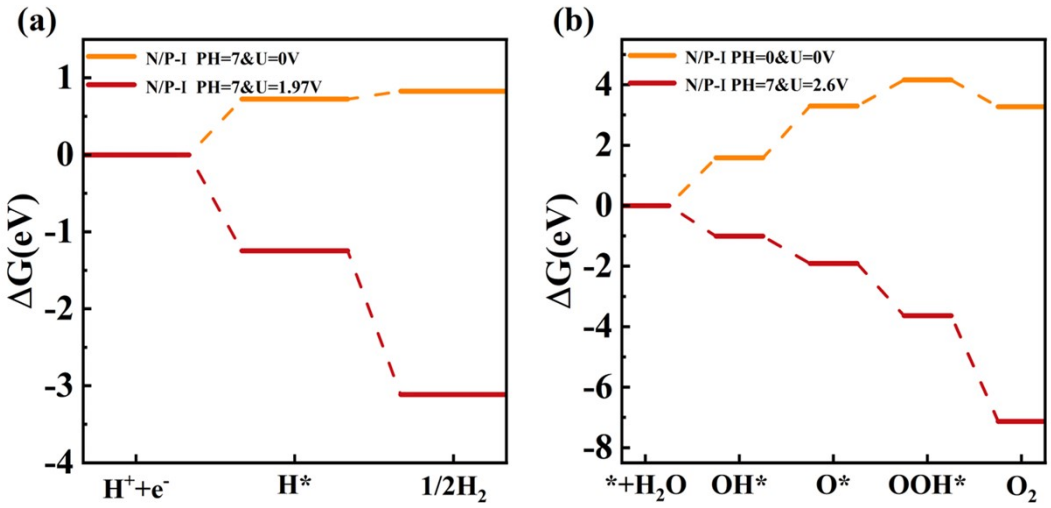

Fig. S10 The Gibbs free energy of N/P-I (a) HER and (b) OER at pH=7.

Table S1 Calculated interface binding energies (*IBE*), interlayer distances (*d*) and lattice constants (*a*) of the AlN/GaS-SiP heterojunction.

Stack	***IBE*** **(meV/Å^2)**	***d*** **(Å)**	***a*** **(Å)**
N/P-I	-20.48	3.00	6.19
N/P-II	-20.03	3.13	6.19
N/P-III	-20.22	3.12	6.19
N/S-I	-17.04	3.14	6.19
N/S-II	-16.73	3.22	6.19
N/S-III	-16.96	3.23	6.19