Supporting Information for

Ferroelectric switching driven photocatalytic overall water splitting in As/In_2Se_3 heterostructure

Rui Xiong[1,2], Lili Zhang[3,*], Cuilian Wen[1], Masakazu Anpo[4], Yee Sin Ang[2,*], Baisheng Sa[1,*]

[1]*Multiscale Multiscale Computational Materials Facility & Materials Genome Institute, School of Materials Science and Engineering, Fuzhou University, Fuzhou 350108, China*

[2]*Science, Mathematics and Technology Cluster, Singapore University of Technology and Design, 8 Somapah Road, Singapore 487372*

[3]*Key Laboratory of Material Physics, Ministry of Education, School of Physics and Microelectronics, Zhengzhou University, Zhengzhou 450001, China*

[4]*State Key Laboratory of Photocatalysis on Energy and Environment, Fuzhou University, Fuzhou 350116, China*

Corresponding Authors: *zhlili@zzu.edu.cn (L. Zhang); *yeesin_ang@sutd.edu.sg (Y. S, Ang)
*bssa@fzu.edu.cn (B. Sa)

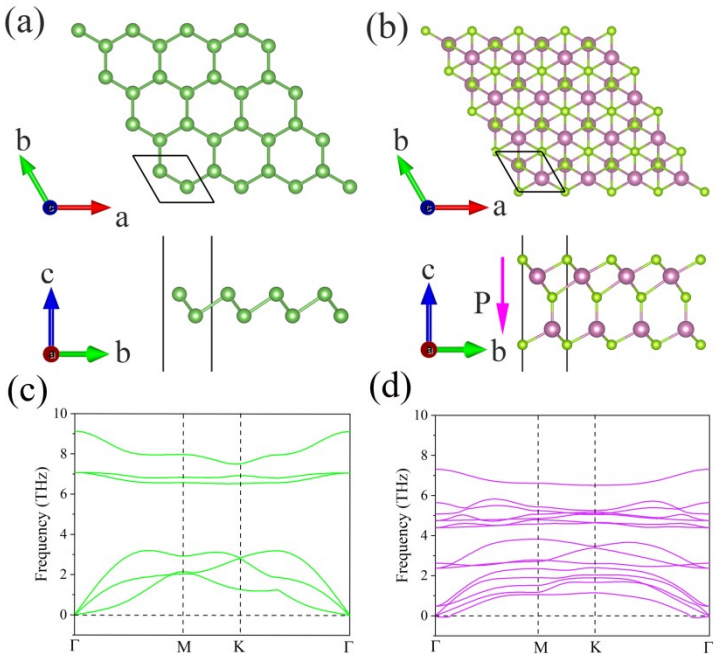

Figure S1. The top and side views of the crystal structure of (a) As and (b) In_2Se_3 monolayers. The orientation of the intrinsic electric field of In_2Se_3 points from the lower surface to the upper surface.

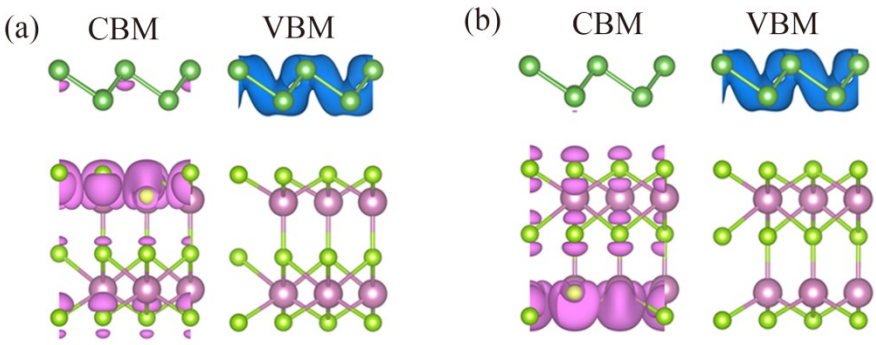

Figure S2. The band decomposed charge densities of the (a) As/In_2Se_3-P↑ and (b) As/In_2Se_3-P↓ heterostructures. The isosurface value is set 0.002 e·$bohr^{-3}$.

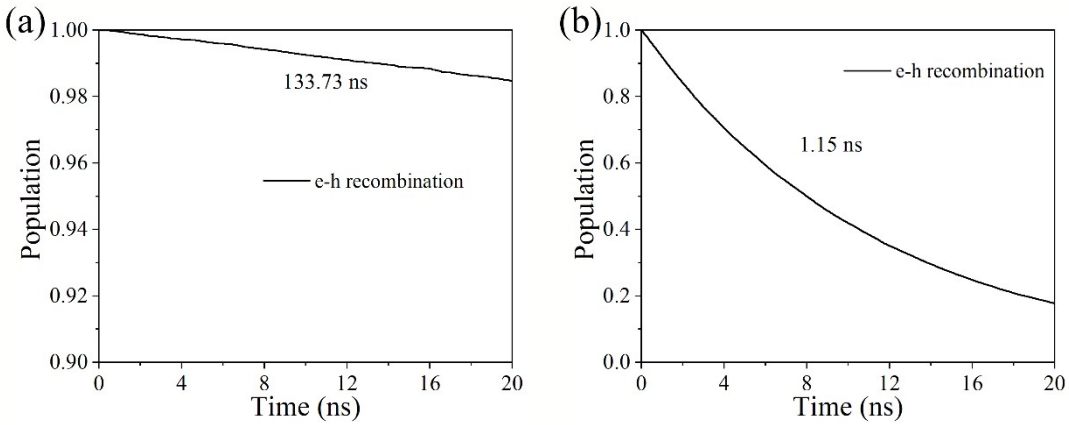

Figure S3. The electron-hole (e-h) recombination dynamic of (a) As/In_2Se_3-P↑ and (b) As/In_2Se_3-P↓ heterostructures.

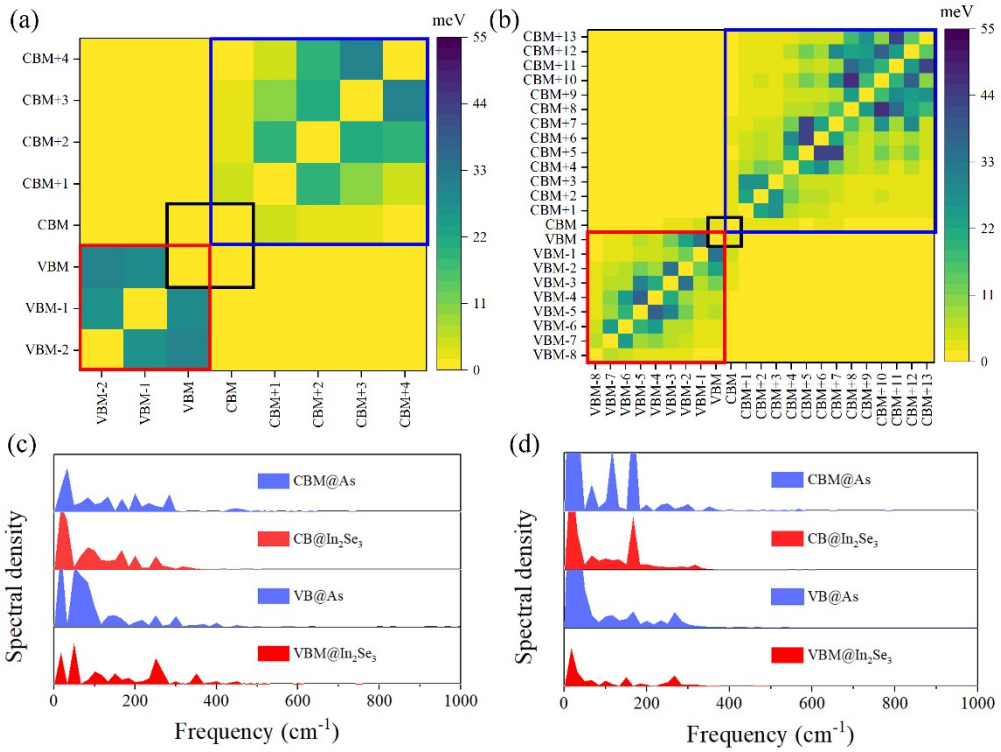

Figure S4. The averaged NAC of electronic states involved in the interface charge transfer and e-h recombination dynamics of (a) As/In_2Se_3-P↑ and (b) As/In_2Se_3-P↓. The Fourier transform (FT) spectra of some key time-dependent Kohn-Sham energy states of (c) As/In_2Se_3-P↑ and (d) As/In_2Se_3-P↓.

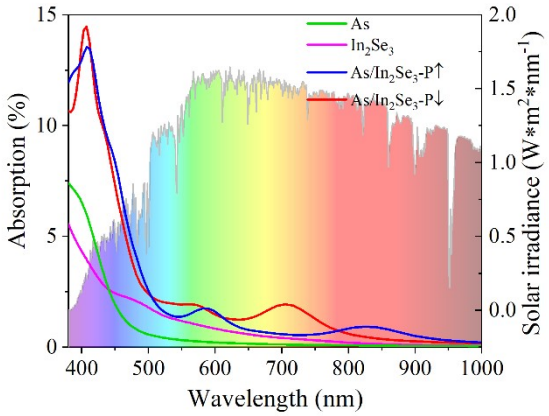

Figure S5. The calculation optical absorption coefficients of As/In_2Se_3-P↑ and As/In_2Se_3-P↓ heterostructures. The As and In_2Se_3 monolayers are also plotted for the comparison. The right axis represents the solar irradiance of the solar spectrum as shown in background.

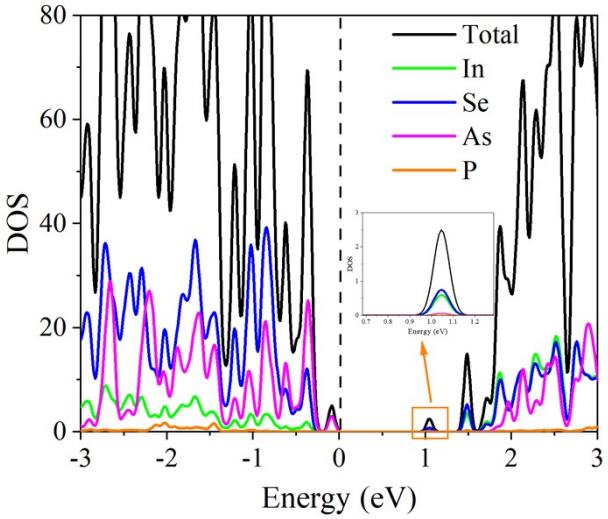

Figure S6. The atom-contributed density of states (DOS) of P-doping As/In_2Se_3-P↓ heterostructure under the HSE06 functional level.

Table S1. The calculated lattice constant a, interlayer distant d, formation energy E_f, and binding energy E_b of As/In_2Se_3-upper and As/In_2Se_3-bottom vdW heterostructure under different stacking configurations

Type	Stacking	a (Å)	d (Å)	E_f (eV)	E_b (meV/Å^2)
As/In_2Se_3↑	I	7.120	3.29	-0.6014	-14.51416
	II	7.120	3.42	-0.6062	-14.56687
	III	7.120	3.41	-0.6062	-14.56743
As/In_2Se_3↓	I	7.110	3.40	-0.6724	-13.15628
	II	7.110	3.50	-0.6750	-13.21327
	III	7.110	3.50	-0.6750	-13.21329

Table S2. The deformation potential constant (E_l), elastic modulus (C_{2D}), effective mass (m^*) and carrier mobility (μ) of As/In_2Se_3 vdW heterostructure and the corresponding As and In_2Se_3 monolayer

	Carrier type	E_l (eV)	C_{2D} (N m^{-1})	m^*/m_0	μ (cm^2 V^{-1} s^{-1})
In_2Se_3	e	6.11	209.67	0.23	1531.45
	h	5.25	209.67	2.65	15.17
As	e	1.09	80.67	0.76	1656.13
	h	6.96	80.67	0.52	87.54
As/In_2Se_3-P↑	e	5.33	327.81	0.23	3146.45
	h	2.20	327.81	0.47	4235.70
As/In_2Se_3-P↓	e	1.46	336.14	0.27	30394.75
	h	7.79	336.14	0.48	344.96