Supplemental Material for "MoS_2 and Janus (MoSSe) Based 2D van der Waals Heterostructures: Emerging Direct Z-scheme Photocatalysts"

Arunima Singh*, Manjari Jain, Saswata Bhattacharya*
Department of Physics, Indian Institute of Technology Delhi, New Delhi 110016, India

I. ELECTROSTATIC POTENTIAL

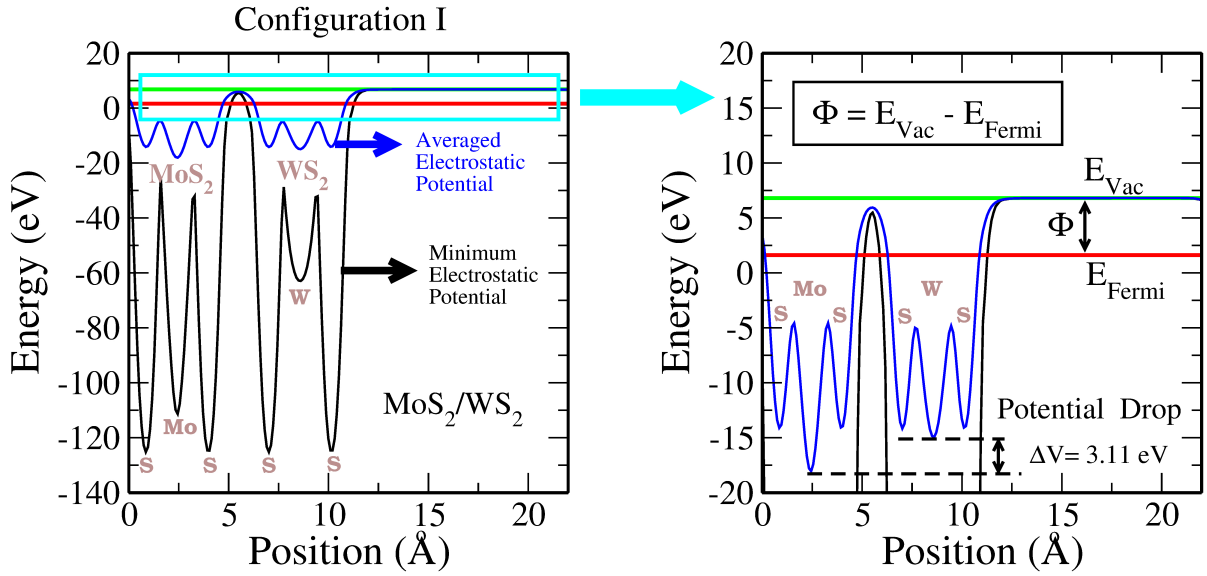

Fig.S 1: Schematic of electrostatic potential as obtained corresponding to MoS_2/WS_2 vdW HTS.

* Arunima.Singh@physics.iitd.ac.in [AS], saswata@physics.iitd.ac.in [SB]

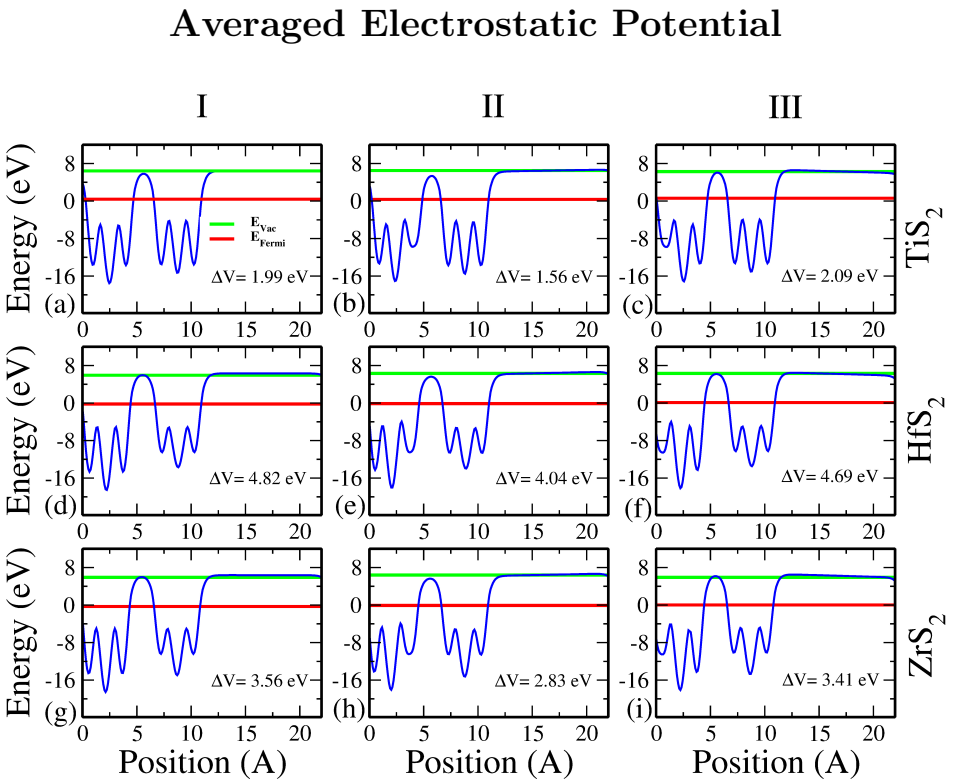

Fig.S 2: Averaged electrostatic potential corresponding to TMDs viz. TiS_2 (upper panel), HfS_2 (middle panel) and ZrS_2 (lower panel) for I, II and III configurations.

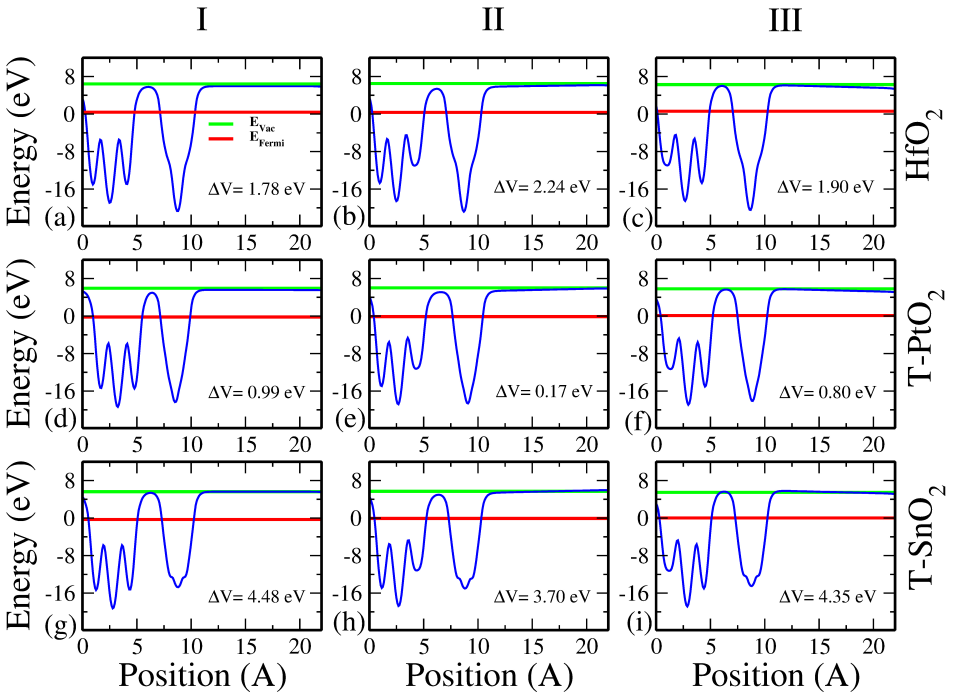

Fig.S 3: Averaged electrostatic potential corresponding to TMOs viz. HfO_2 (upper panel), T-SnO_2 (middle panel) and T-PtO_2 (lower panel) for I, II and III configurations.

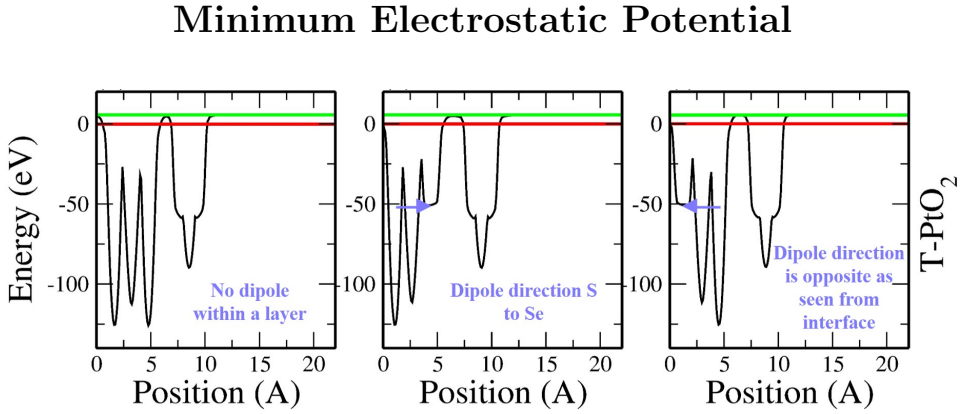

Fig.S 4: Electrostatic potential of T-PtO_2 for I, II and III configurations, showing dipole direction at the interface.

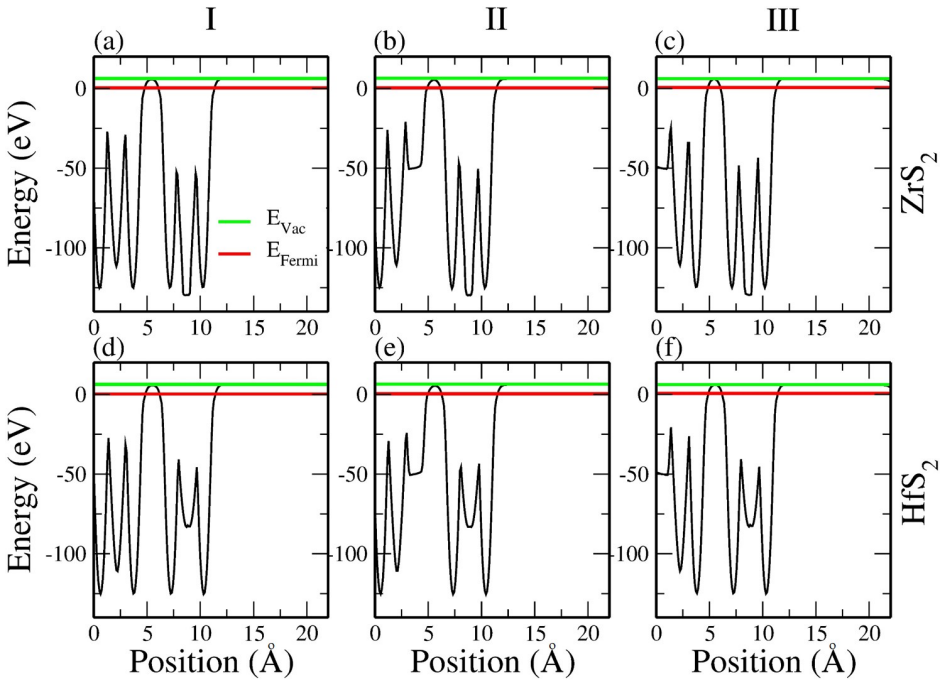

Fig.S 5: Electrostatic potential corresponding to ZrS_2 (upper panel) and HfS_2 (lower panel) for I, II and III configurations.

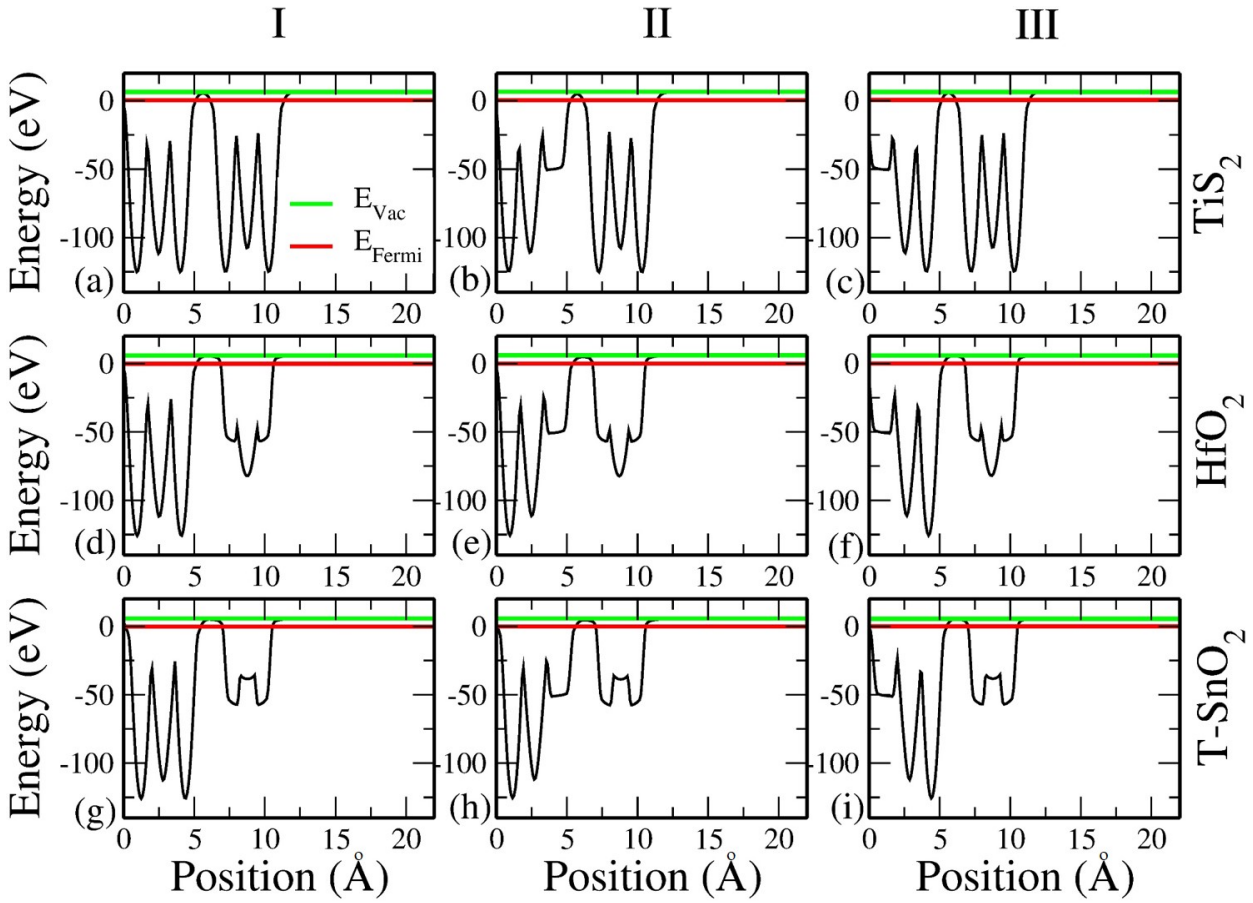

Fig.S 6: Electrostatic potential corresponding to TiS_2 (upper panel), HfO_2 (middle panel) and SnO_2 (lower panel) for I, II and III configurations.

II. LATTICE PARAMETERS

TABLE I: Lattice constants (l) of 2×2 monolayers

BX_2	MoS_2	WS_2	ZrS_2	HfS_2	TiS_2	HfO_2	T-PtO_2	T-SnO_2
l	6.321	6.321	7.120	7.019	6.660	6.243	6.296	6.450

III. BAND GAPS OF CONFIGURATIONS

TABLE II: Band gaps of the monolayers and their corresponding vdW HTSs.

BX_2	Band Gap (eV) (Indirect/Direct)			
	Monolayer	I	II	III
MoS_2	-/2.257	-	-	-
MoSSe	-/2.172	-	-	-
WS_2	-/2.447	1.636/2.038	1.879/1.885	1.589/1.928
ZrS_2	1.829/1.892	0.752/0.809	0.123/0.177	0.676/0.729
HfS_2	1.937/2.019	0.746/0.863	0.079/0.185	0.663/0.773
TiS_2	1.672/1.769	0.414/0.418	0.047/0.056	0.520/0.525
HfO_2	3.516/3.520	0.486/0.499	0.072/0.090	0.440/0.453
T-PtO_2	3.304/3.313	1.323/1.525	0.595/0.846	1.178/1.423
T-SnO_2	4.143/4.297	1.130/1.285	0.460/0.882	1.069/1.473

IV. BANDSTRUCTURES OF Z-SCHEME VDW HTSS

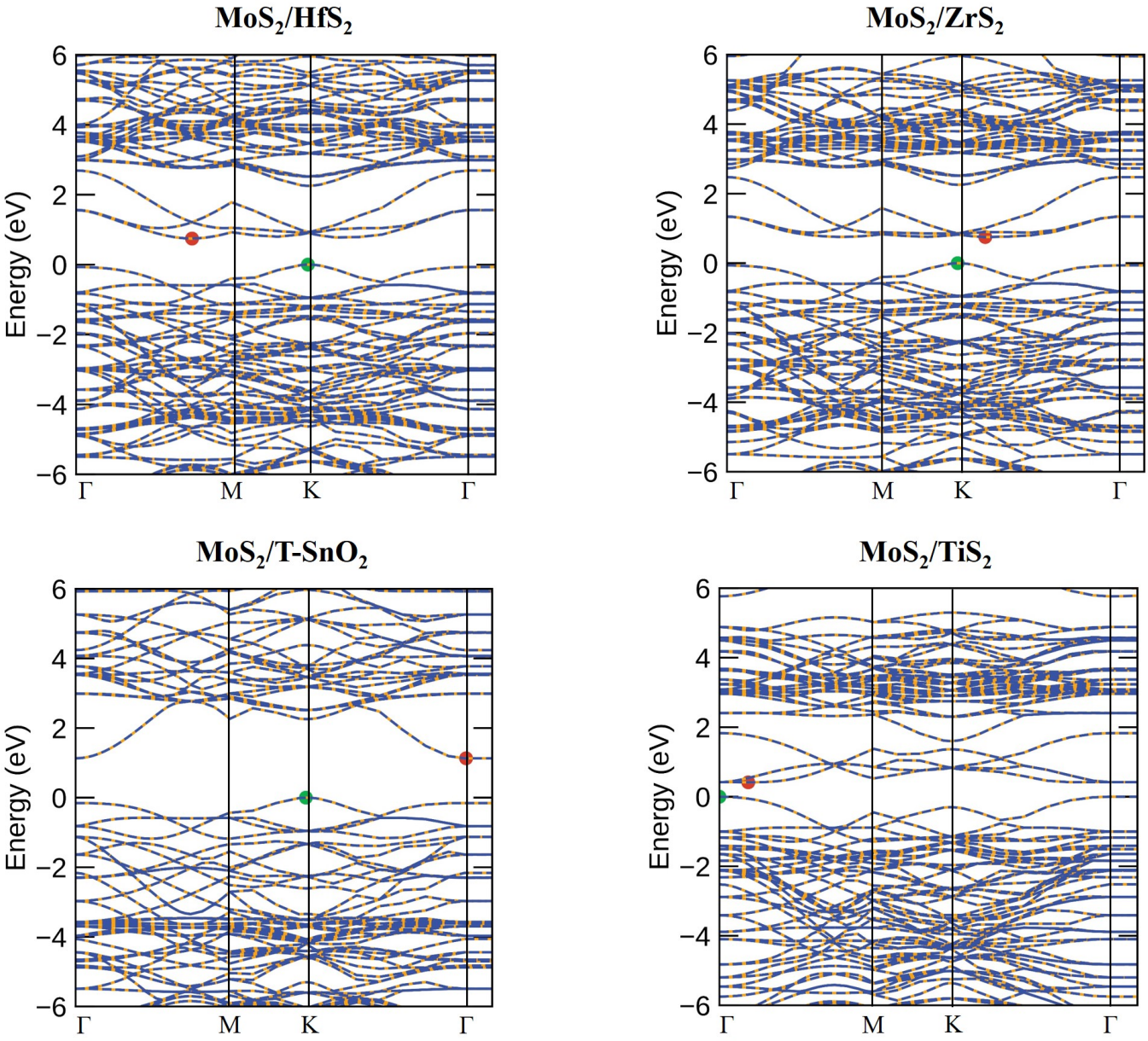

Fig.S 7: (Color online) Bandstructures corresponding to the supercell of MoS_2/BX_2 HTSs (configuration I) where MoS_2/ZrS_2 has similar bandstructure as that of MoS_2/HfS_2. The bandstructures corresponding to the MoSSe based vdW HTSs (configurations II and III) are similar with slight change in energetics. The red and green points correspond to the conduction band minimum and valence band maximum.

v. PLANAR AVERAGED CHARGED DENSITY

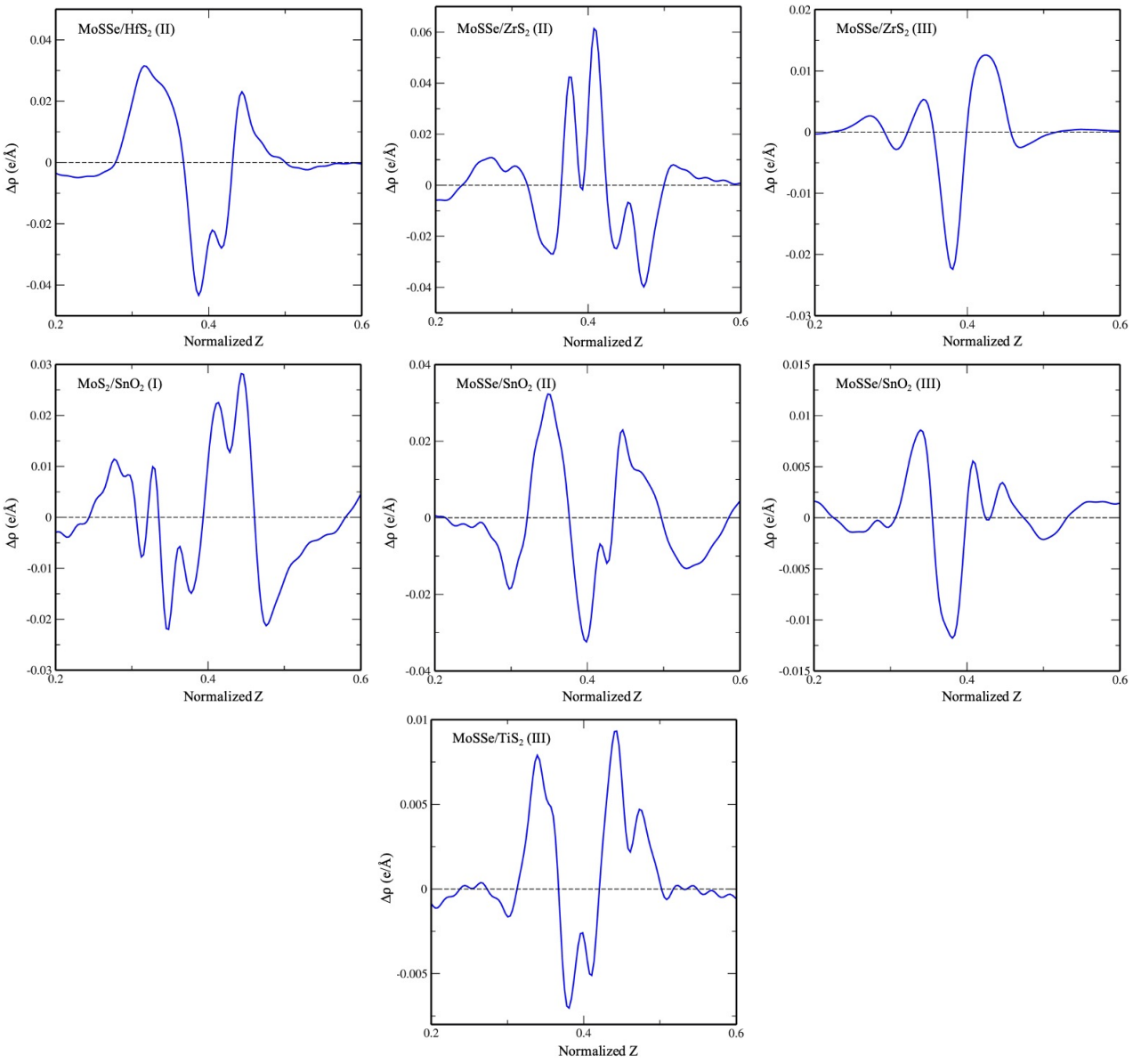

Fig.S 8: (Color online) Planar averaged charge density

VI. ABSORPTION SPECTRA OF MONOLAYERS

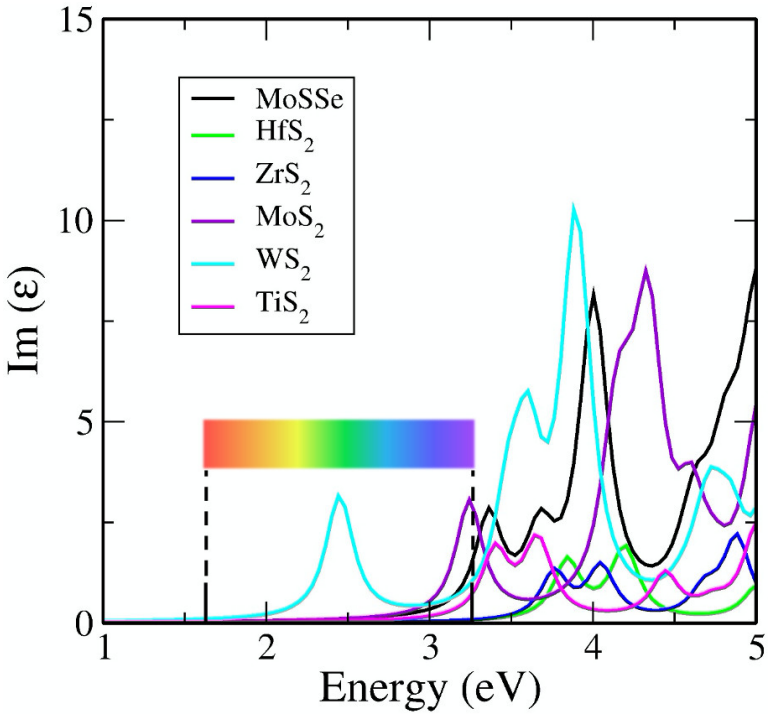

Fig.S 9: Absorption spectra of Janus (MoSSe) and monolayer TMDs i.e MoS_2, WS_2, TiS_2, HfS_2 and ZrS_2

VII. EXCITON BINDING ENERGY

We have performed mBSE calculations, with an intention to simply compare the exciton binding energy (E_B) of vdW HTSs with MoS_2 and MoSSe. We have included four valence and conduction bands for mBSE on top of hybrid functional. However, there is a huge scope in understanding the excitonic transitions in these systems with different computational approaches. Presently, considering the Z-scheme application, the smaller E_B of vdW HTSs as compared to MoS_2 (or MoSSe) would allow more e^- - h^+ recombination in vdW HTS thereby facilitating MoS_2 (or MoSSe) for HER.

TABLE III: Exciton binding energies of vdW HTSs, MoS_2 and MoSSe monolayers

	MoS_2	MoSSe	MoSSe/HfS_2 (II)	MoSSe/TiS_2 (III)	MoS_2/SnO_2 (I)	MoSSe/ZrS_2 (II)	MoSSe/ZrS_2 (III)	MoSSe/SnO_2 (III)
E_B (eV)	1.5	1.8	0.44	0.37	1.3	0.42	0.41	1.3

VIII. CARRIER MOBILITY

$$\mu = \frac{2e\hbar^3 C}{3k_B T|m^*|^2 E_1^2} \tag{1}$$

In this expression C is defined as $C = [\partial^2 E/\partial\delta^2\,]/S^0$. E refers to the total energy of the system, δ is the applied uniaxial strain, and S^0 is the area of the optimized vdW HTS. m^* is the effective mass, expressed as $m^* = \hbar^2\,(\partial E^2/\partial k^2)^{-1}$. T represents temperature, and E_1 is the deformation potential constant that is defined as $\Delta E = E_1(\Delta l/l_0\,)$. Here, ΔE is the energy shift of the band edge position with respect to the lattice strain $\Delta l/l_0$. The energies of the band edges (CBm or VBM) are obtained with vacuum level as the reference.