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Optoelectronic and solar cell applications of Janus monolayers and their Van der Waals heterostructure

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I. SUPPLEMENTARY-MATERIAL

The calculation of V_∞ is performed by using a tag LVTOT=.TRUE. in the INCAR file in the VASP calculations, which generates an output file namely LCOPOT. By using a small script on LOCPOT, we can generate a data file, which can be opened in the xmgrace or any other programme. The highest points in that data file along Y- direction gives us the value of V_∞. For further under standing, we have also plotted the electrostatic potential of models of the MXY-WXY vdW heterostructure in both model-I, and -II using PBE calculations (see FIG. S1).

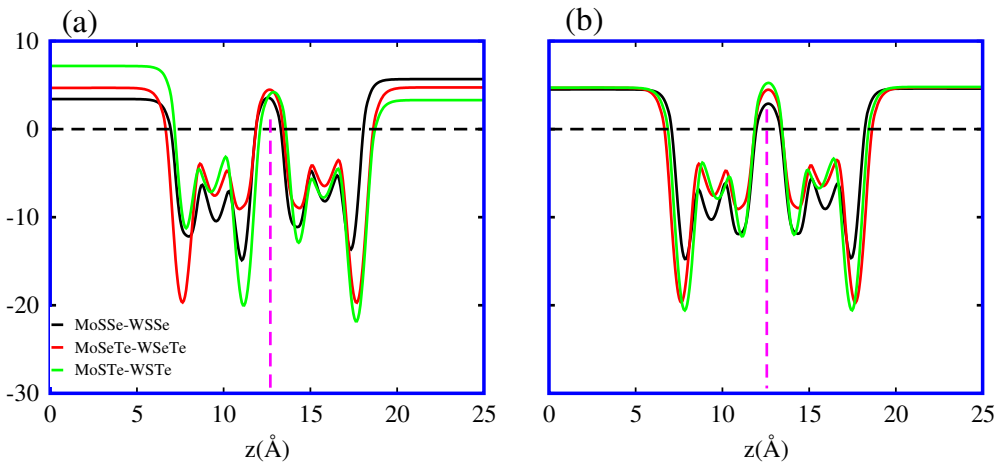

FIG. S1. Plane-averaged electrostatic potential of Janus heterostructure. The left panel is for Model-I while the right panel for Model-II.

It is well known that PBE functional systematically underestimate the band gap values of semiconductors, while a computationally expensive HSE06 (Heyd-Scuseria-Ernzerhof) functional gives the corrected value of the band gap, hence used for electronic structure calculations. Valence band maximum (VBM) and conduction band minimum (CBM) of the Janus monolayers and their corresponding heterostructure with respect to the vacuum level are also calculated. The difference of vaccum potential and VBM is called the ionization potential (IP) and the difference of vaccum potential and CBM is called to electron affinity (EA) [1]. A 20 Å vacuum gap is left between all monolayers and their heterostructure. The electrostatic potential is then calculated for each layer normal to the layers, averaged along these layers. The potential within the vacuum gap denotes the vacuum potential. The VBM and CBM can then be determined with respect to this potential. For full water-splitting, the CBM of monolayers and their corresponding heterostructure must be higher than the reduction level of hydrogen (-4.44 eV), while the VBM must be lower than the oxidation level of oxygen (-5.67 eV). As indicated in Figure 8, in the manuscript and for comparison we have also plotted the Band alignments of MoSSe monolayer (See FIG. S2) which shows that all these monolayers could well satisfy the band edge requirements for full water-splitting [2, 3].

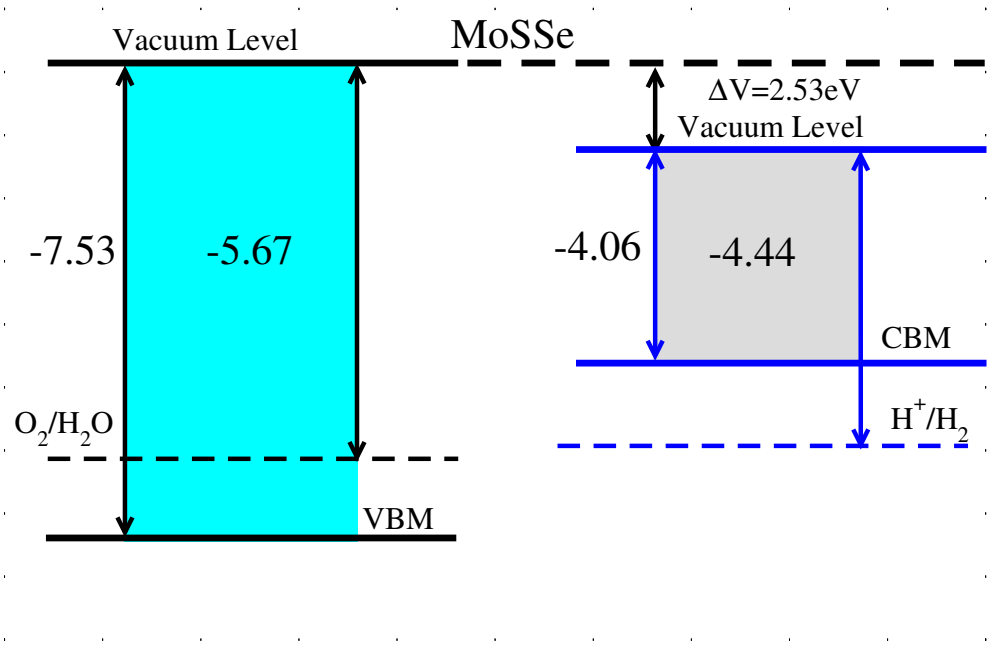

FIG. S2. Band alignments of VBM and CBM for MoSSe monolayer. The dotted lines are the standard redox potential levels of water-splitting.

The water splitting efficiency is closely related to the aqueous solution pH. Using Nernst equation the redox potentials (H^+/H_2 and H_2O/O_2) at pH=0–7 of Janus monolayers and their corresponding heterostructures are calculated and presented in the TABLE SI, -SII. The calculated values show that by increasing the value of pH, the materials tend to favorable for reduction while the value of oxidation further decreases.

pH	Band-edge	MoSSe	WSSe	MoSeTe	WSeTe	MoSTe	WSTe
0	VBM	1.86	1.99	1.45	1.54	1.38	1.54
	CBM	-0.38	-0.16	-0.39	-0.154	-0.14	0.04
1	VBM	1.80	1.94	1.39	1.49	1.32	1.48
	CBM	-0.44	-0.22	-0.45	-0.21	-0.19	-0.02
2	VBM	1.74	1.88	1.33	1.43	1.26	1.42
	CBM	-0.49	-0.28	-0.51	-0.27	-0.26	-0.08
3	VBM	1.63	1.76	1.21	1.31	1.14	1.31
	CBM	-0.61	-0.40	-0.63	-0.39	-0.37	-0.19
4	VBM	1.57	1.70	1.15	1.25	1.08	1.25
	CBM	-0.67	-0.46	-0.69	-0.45	-0.43	-0.25
5	VBM	1.51	1.64	1.09	1.19	1.03	1.19
	CBM	-0.73	-0.52	-0.75	-0.51	-0.49	-0.31
6	VBM	1.45	1.58	1.03	1.13	0.97	1.12
	CBM	-0.73	-0.52	-0.75	-0.51	-0.49	-0.31
7	VBM	1.39	1.52	0.98	1.07	0.91	1.07
	CBM	-0.79	-0.58	-0.80	-0.57	-0.55	-0.37

TABLE S I. Valence and conduction band edge potentials of Janus monolayers at pH=0–7.

[1] Y. Jao and J. Robertson, *Phys. Rev. Mat.* 2017, **1**, 044004.

[2] X. Li, Z. Li, and J. Yang, *Phys. Rev. Lett.* 2014, **112**, 018301.

[3] P. Zhao, Y. Ma, Z. Lv, M. Li, B. Huang and Y. Dai, *Nano Energy* 2018, **51**, 533.

		MoSSe-WSSe		MoSeTe-WSeTe		MoSTe-WSTe	
pH	Band-edge	Model-I	Model-II	Model-I	Model-II	Model-I	Model-II
0	VBM	1.55	1.63	0.99	1.08	0.93	0.96
	CBM	0.10	0.02	0.23	0.14	0.48	0.45
1	VBM	1.49	1.58	0.93	1.02	0.87	0.91
	CBM	0.04	-0.04	0.17	0.08	0.43	0.39
2	VBM	1.43	1.52	0.88	0.96	0.81	0.85
	CBM	-0.02	-0.10	0.11	0.02	0.37	0.33
3	VBM	1.37	1.46	0.82	0.90	0.75	0.79
	CBM	-0.07	-0.15	0.05	-0.03	0.31	0.27
4	VBM	1.31	1.40	0.76	0.85	0.69	0.73
	CBM	-0.13	-0.21	-0.01	0.09	0.25	0.21
5	VBM	1.25	1.34	0.70	0.78	0.63	0.67
	CBM	-0.19	-0.28	-0.07	-0.15	0.19	0.15
6	VBM	1.19	1.28	0.64	0.72	0.57	0.61
	CBM	-0.25	-0.33	-0.13	-0.21	0.13	0.09
7	VBM	1.14	1.22	0.58	0.66	0.51	0.55
	CBM	-0.31	-0.39	-0.19	-0.27	0.07	0.03

TABLE S II. Valence and conduction band edge potentials of model-I and model-II vdW heterostructures at pH=0–7.