

## Supporting Information

Electronic, Magnetic, Catalytic, and Electrochemical properties of Two-Dimensional  
Janus Transition Metal Chalcogenides

Wenzhou Chen,<sup>a</sup> Yuanju Qu<sup>b</sup>, Lingmin Yao<sup>c</sup>, Xianhua Hou<sup>d</sup>, Xingqiang Shi<sup>\*e</sup> and Hui  
Pan<sup>\*a</sup>

<sup>a</sup>Institute of Applied Physics and Materials Engineering, University of Macau, Macao  
SAR

<sup>b</sup>College of Physics and Communication Electronics, Jiangxi Normal University,  
Nanchang 330022, China

<sup>c</sup>School of Physics and Electronic Engineering, Guangzhou University, Guangzhou,  
510006, China

<sup>d</sup>School of Physics and Telecommunication Engineering, South China Normal University,  
Guangzhou 510006, P. R. China

<sup>e</sup>Department of Physics, Southern University of Science and Technology, Shenzhen  
518055, China

\* Corresponding Authors: [huipan@umac.mo](mailto:huipan@umac.mo) (H.P.); Tel: (853)88224427; Fax:  
(853)88222426; [shixq@sustc.edu.cn](mailto:shixq@sustc.edu.cn) (X. Q. S.)

Table S1 Calculated total energies of the unit cells of the MSXs (M = Ti or V; and X = C, N,  
Si or P) monolayers as a function of k-point grids.

k-grid	TiSC (eV)	TiSSi (eV)	TiSP (eV)	VSC (eV)	VSN (eV)	VSSi (eV)
10×10×1	-21.5471	-18.0983	-19.0169	-21.7394	-23.1386	-18.5501
12×12×1	-21.5471	-18.0983	-19.0198	-21.7407	-23.1386	-18.5547
14×14×1	-21.5471	-18.0983	-19.0230	-21.7398	-23.1387	-18.5582
16×16×1	-21.5471	-18.0983	-19.0248	-21.7402	-23.1387	-18.5583
18×18×1	-21.5471	-18.0983	-19.0235	-21.7399	-23.1387	-18.5564
20×20×1	-21.5471	-18.0983	-19.0225	-21.7400	-23.1387	-18.5557
22×22×1	-21.5471	-18.0983	-19.0225	-21.7401	-23.1387	-18.5562

22 Table S2 Calculated total energies of the supercell ( $3 \times 3 \times 1$  unit cells) of the MSXs (M = Ti  
 23 or V; and X = C, N, Si or P) monolayers as a function of k-point grids.

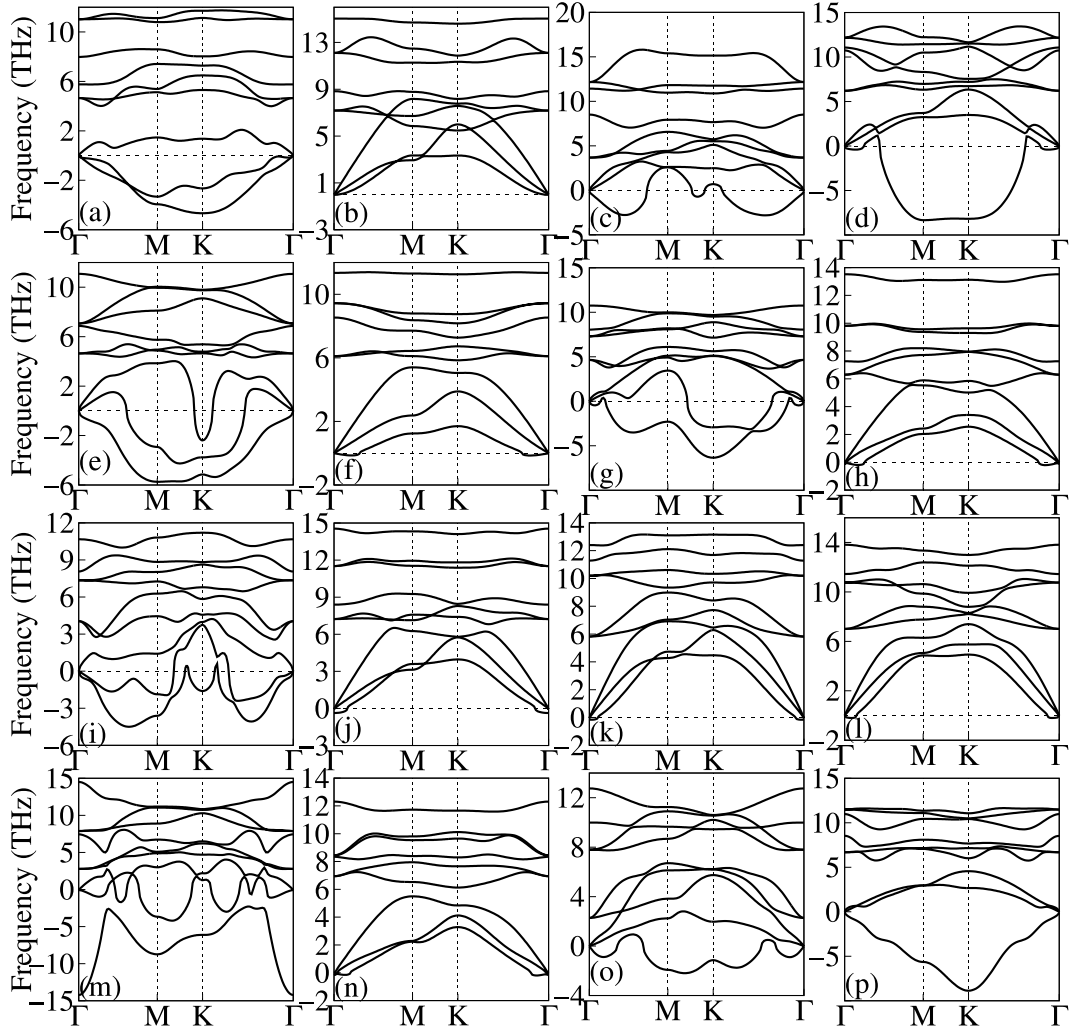
K-grid	TiSC (eV)	TiSSi (eV)	TiSP (eV)	VSC (eV)	VSN (eV)	VSSi (eV)
$1 \times 1 \times 1$	-193.8808	-162.9363	-170.4312	-195.6340	-207.8329	-167.0570
$3 \times 3 \times 1$	-193.9245	-162.8849	-171.2200	-195.6605	-208.2468	-166.9888
$5 \times 5 \times 1$	-193.9245	-162.8849	-171.1925	-195.6642	-208.2484	-167.0172

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25 Table S3 Calculated total energies of supercell ( $2 \times 2 \times 1$  unit cell) of VSSi-1H monolayer as a  
 26 function of k-point grids. The energy cutoff is fixed at 700 eV.

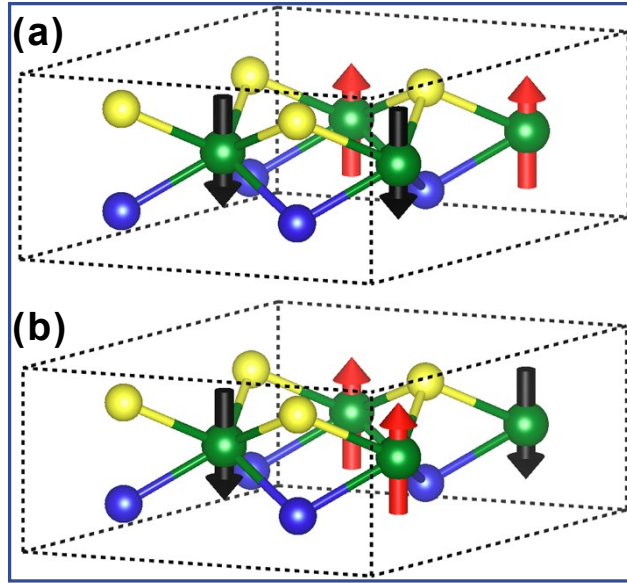
k grids	NM	FM	AFM-1	AFM-2
$5 \times 5 \times 1$	-74.2015	-74.2415	-74.2015	-74.2016
$7 \times 7 \times 1$	-74.2339	-74.2207	-74.2339	-74.2339
$9 \times 9 \times 1$	-74.2266	-74.2354	-74.2265	-74.2266
$11 \times 11 \times 1$	-74.2259	-74.2315	-74.2258	-74.2259
$13 \times 13 \times 1$	-74.2293	-74.2322	-74.2292	-74.2293

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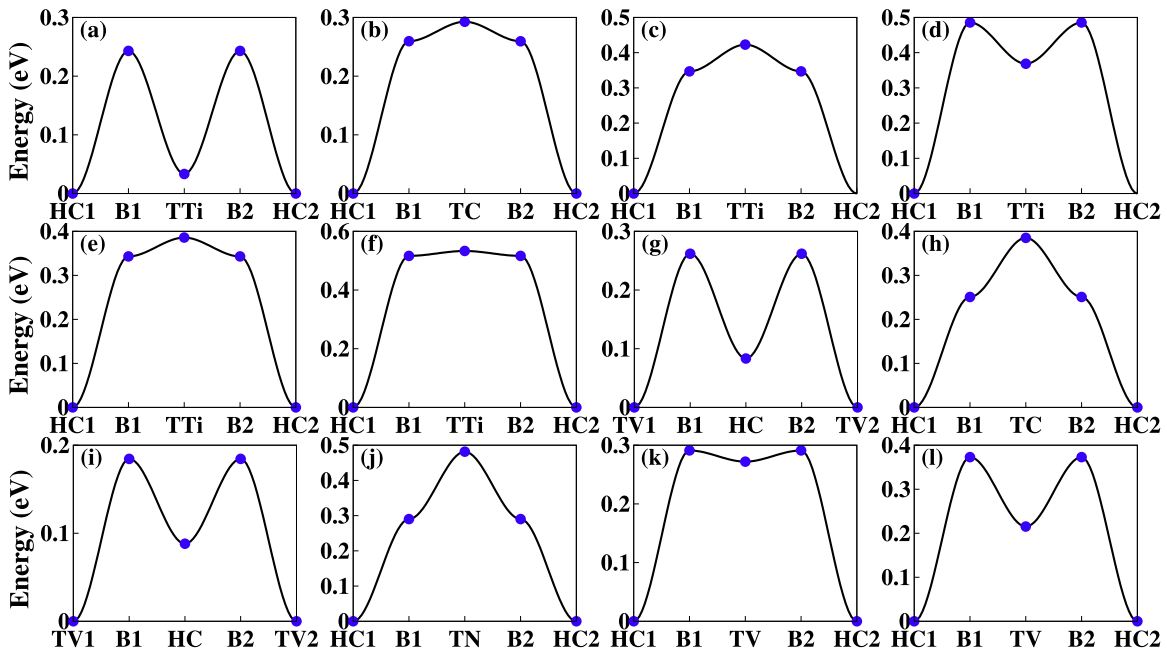
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29 Figure S1 Calculated phonon dispersions of the Janus MSXs (M = Ti or V; and X = C, N, Si or P)  
 30 monolayers: (a) TiSC-1T, (b) TiSC-1H, (c) TiSN-1T, (d) TiSN-1H, (e) TiSSi-1T, (f) TiSSi-1H, (g)  
 31 TiSP-1T, (h) TiSP-1H, (i) VSC-1T, (j) VSC-1H, (k) VSN-1T, (l) VSN-1H, (m) VSSi-1T, (n) VSSi-  
 32 1H, (o) VSP-1T, and (p) VSP-1H.



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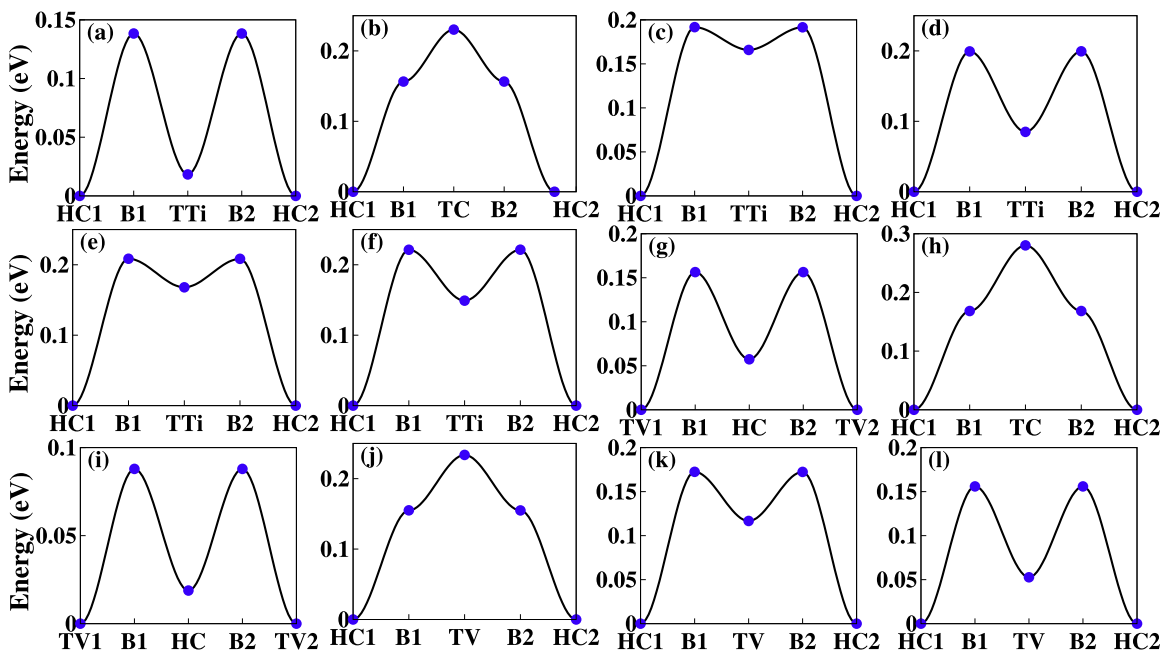
34 Figure S2 Anti-ferromagnetic (AFM) spin configurations of the VSC-1H monolayer: (a) AFM-  
 35 1 and (b) AFM-2; Yellow, green, and blue balls represent S, V, and C atoms, respectively; Black  
 36 and red arrows represent down and up spins, respectively.



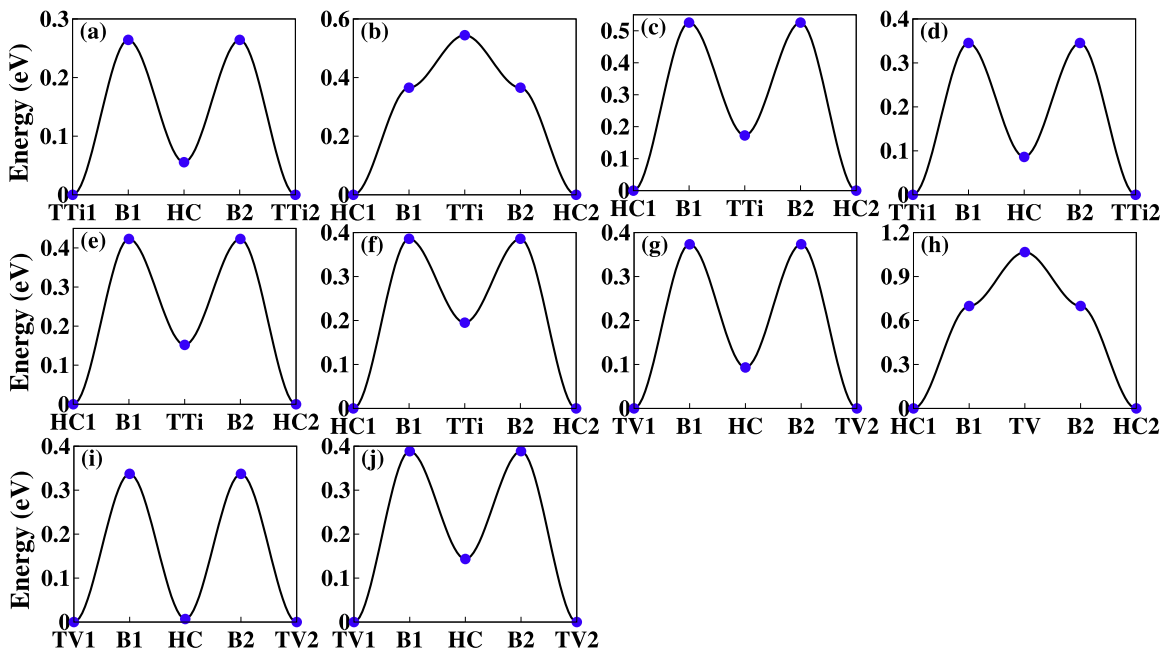
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38 Figure S3 Calculated Li diffusion energies on the surfaces of Janus MSXs ( $M = \text{Ti}$  or  $\text{V}$ ; and  $X =$   
 39  $\text{C}$ ,  $\text{N}$ ,  $\text{Si}$  or  $\text{P}$ ): (a) S-surface of  $\text{TiSC-1H}$ , (b) C-surface of  $\text{TiSC-1H}$ , (c) S-surface of  $\text{TiSSi-1H}$ , (d)  
 40 Si-surface of  $\text{TiSSi-1H}$ , (e) S-surface of the  $\text{TiSP-1H}$ , (f) P-surface of the  $\text{TiSP-1H}$ , (g) S-surface  
 41 of the  $\text{VSC-1H}$ , (h) C-surface of the  $\text{VSC-1H}$ , (i) S-surface of the  $\text{VSN-1T}$ , (j) N-surface of the  
 42  $\text{VSN-1T}$ , (k) S-surface of the  $\text{VSSi-1H}$ , and (l) Si-surface of the  $\text{VSSi-1H}$ . HC - the hexagonal

43 center, TM - on the top of M, TC - on the top of C, and B1 (B2) - the bridge site between the  
 44 corresponding stable site and intermediate position, as shown in text and figures 5&6.



46 Figure S4 Calculated Na diffusion energies on the surfaces of Janus MSXs (M = Ti or V; and X =  
 47 C, N, Si or P): (a) S-surface of TiSC-1H, (b) C-surface of TiSC-1H, (c) S-surface of TiSSi-1H, (d)  
 48 Si-surface of TiSSi-1H, (e) S-surface of the TiSP-1H, (f) P-surface of the TiSP-1H, (g) S-surface  
 49 of the VSC-1H, (h) C-surface of the VSC-1H, (i) S-surface of the VSN-1T, (j) N-surface of the  
 50 VSN-1T, (k) S-surface of the VSSi-1H, and (l) Si-surface of the VSSi-1H.



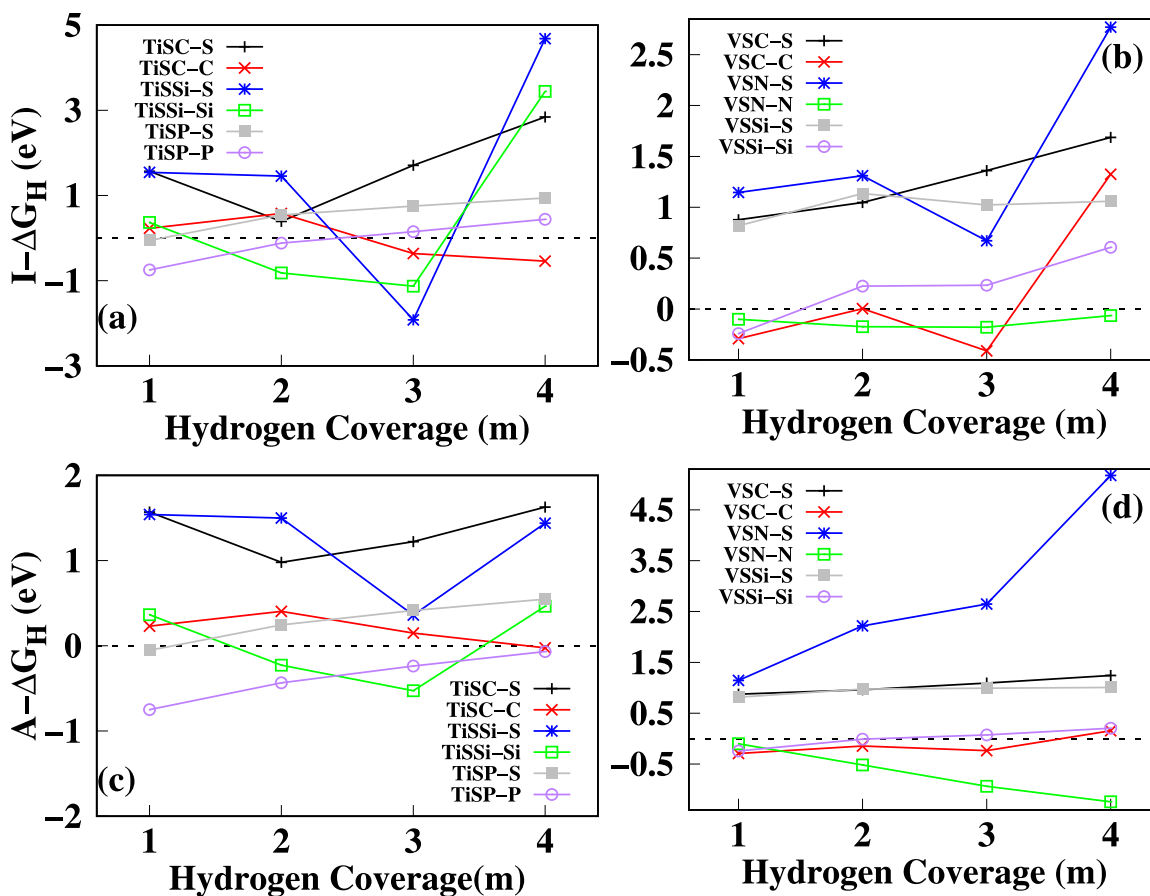
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52 Figure S5 Calculated Mg diffusion energies on the surfaces of Janus MSXs (M = Ti or V; and X =  
 53 C, N, Si or P): (a) S-surface of TiSC-1H, (b) C-surface of TiSC-1H, (c) S-surface of TiSSi-1H, (d)  
 54 Si-surface of TiSSi-1H, (e) S-surface of the TiSP-1H, (f) P-surface of the TiSP-1H, (g) S-surface  
 55 of the VSC-1H, (h) C-surface of the VSC-1H, (i) S-surface of the VSSi-1H, and (j) Si-surface of  
 56 the VSSi-1H.

57 Table S4 Calculated H-adsorption energies (eV) on MSXs (M = Ti or V; and X = C, N, Si or P)  
 58 monolayers at difference adsorption sites:  $S_{ad}$  – adsorption site; TX, HC-X, and TM-X refer to the  
 59 TX, HC, and TM sites on the X-surfaces of MSXs, respectively; TS, HC-S, and TM-S represent  
 60 the TS, HC, and TM sites on the S-surfaces of MSXs, respectively.

$S_{ad}$	TX	HC-X	TM-X	TS	HC-S	TM-S
TiSC-1H	-0.01	1.53	1.61	1.32	2.44	1.61
TiSSi-1H	0.13	0.69	0.09	1.30	2.60	1.09
TiSP-1H	-0.99	1.59	0.22	-0.29	2.25	0.50
VSC-1H	-0.56	1.00	-0.01	0.61	1.85	2.18
VSN-1T	2.04	-0.34	1.60	0.91	1.24	1.32
VSSi-1H	-0.48	0.62	-0.22	0.58	2.39	0.54

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63 Figure S6 Calculated  $I-\Delta G_H$  of (a) TiSX (X = C, Si, or P) and (b) VSX (X = C, N, or Si) monolayers  
 64 as a function of H coverage on the S- and X- surfaces; Calculated  $A-\Delta G_H$  of (c) TiSX (X = C, Si,  
 65 or P) and (d) VSX (X = C, N, or Si) monolayers as a function of H coverage on the S- and X-  
 66 surfaces;  $I-\Delta G_H$ : differential Gibbs free energy;  $A-\Delta G_H$ : average Gibbs free energy.