Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary Information (SI)

$In_2Si_2S_3X_3$ (X = S, Se, Te) Janus Monolayers: From Magnetic Element-Free Spin-Hall Transistor to Sustainable Energy Generation

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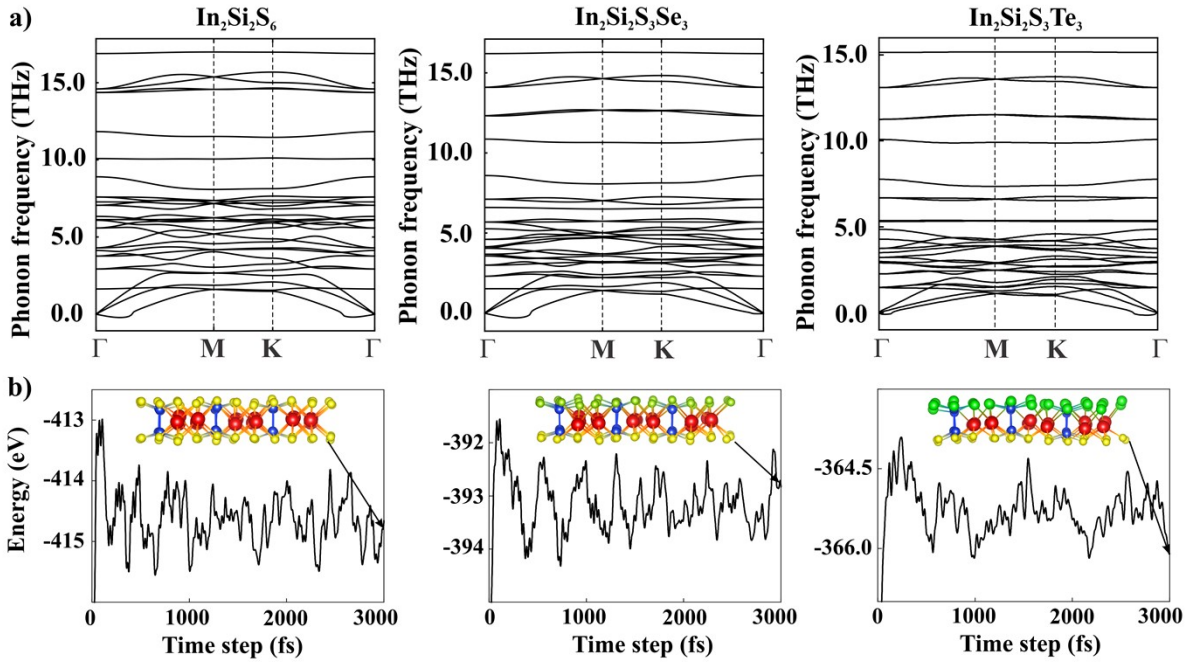

Figure S1 (a) Phonon dispersion spectra of monolayers and (b) snapshot of geometries after 3ps in ab initio molecular dynamics simulation (AIMD) at 300 K.

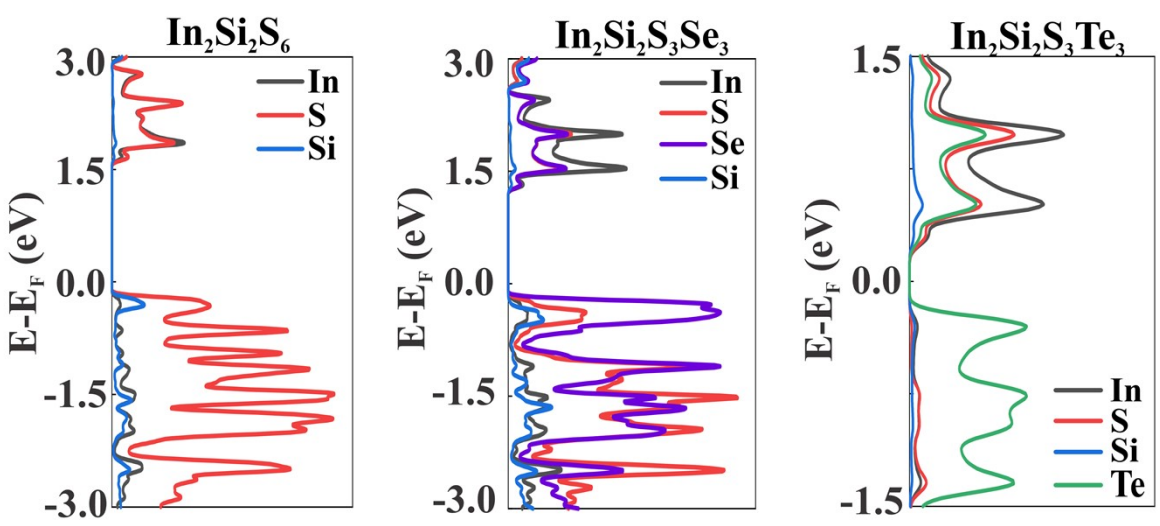

Figure S2 Projected density of states (PDOS) of monolayers near Fermi energy (set to zero).

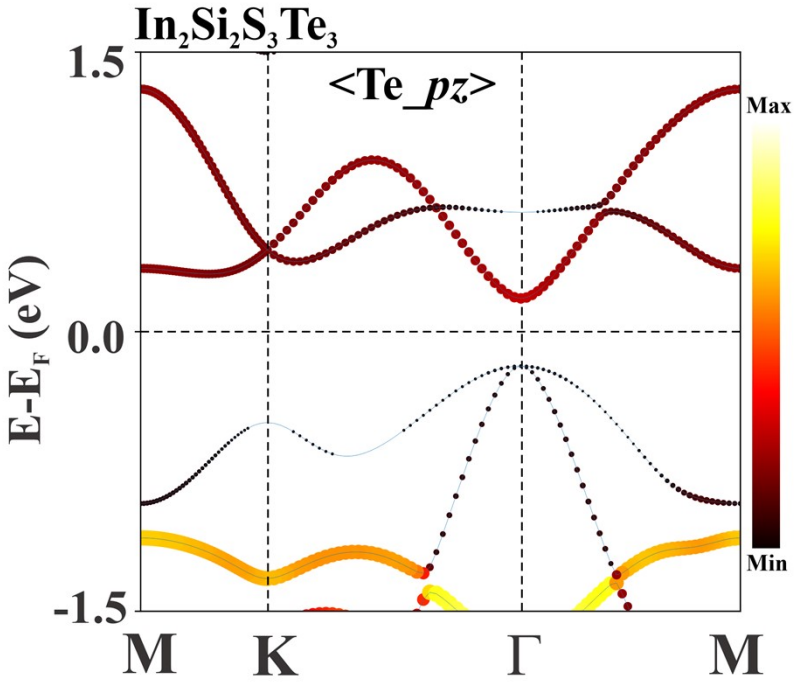

Figure S3: Te_pz orbital projected band structure of Janus $In_2Si_2S_3Te_3$ monolayer.

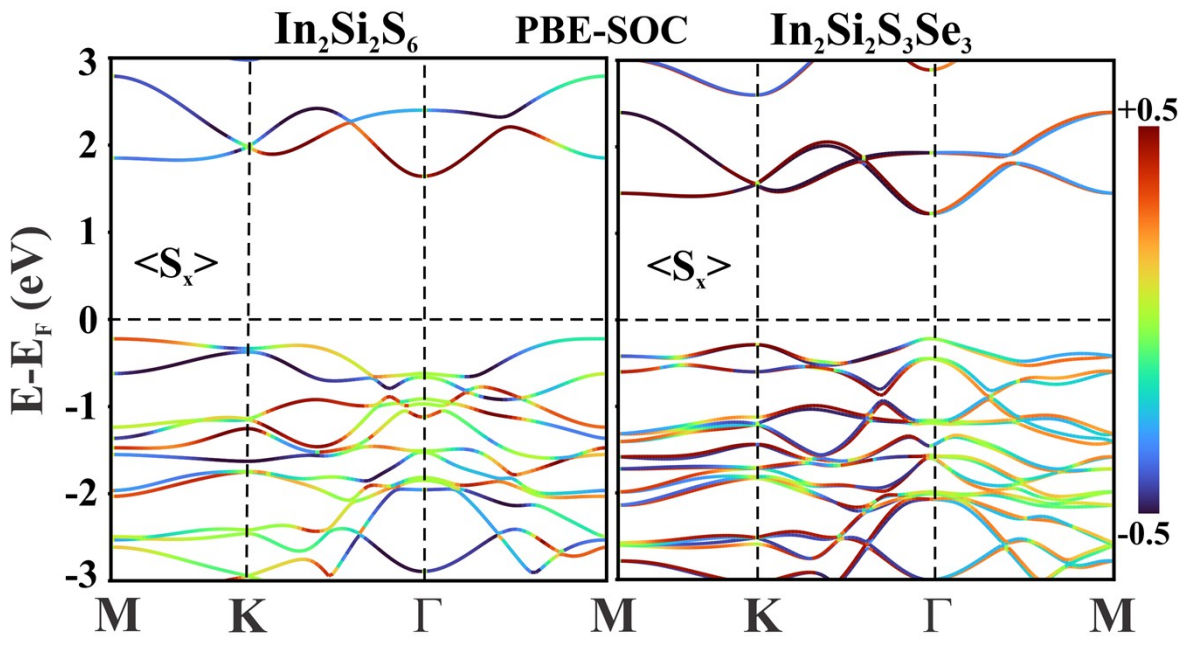

Figure S4: In-plane spin projected band structure of pristine $In_2Si_2S_6$ and $In_2Si_2S_3Se_3$ monolayer.

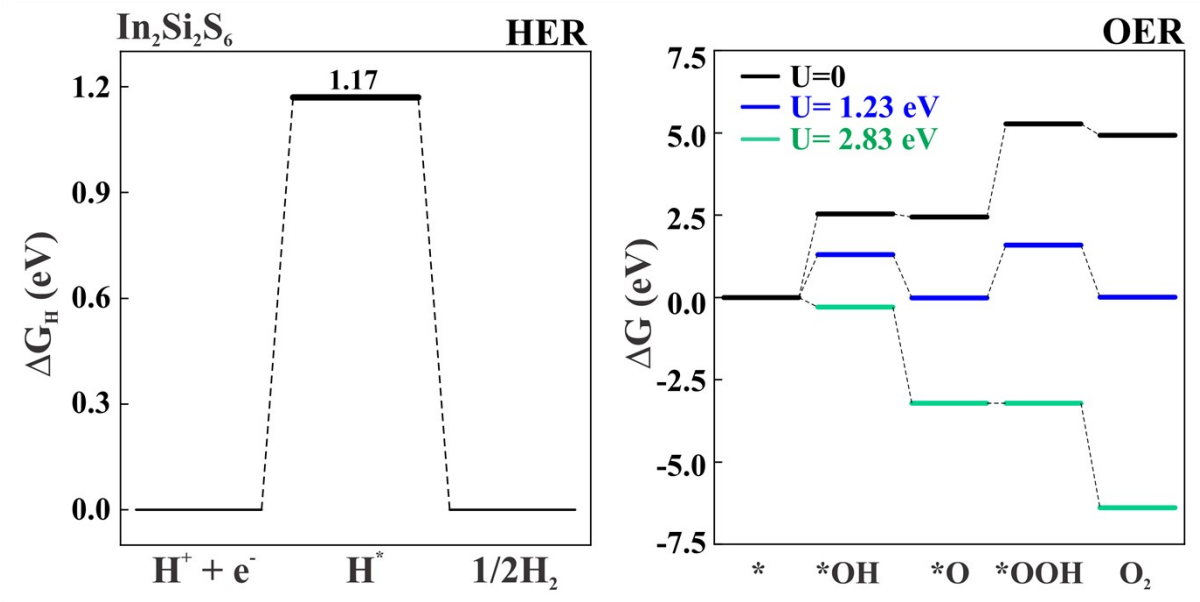

Figure S5: Gibbs free energy profile for HER and OER of pristine $In_2Si_2S_6$ monolayer

Table S1 Calculated elastic constants of monolayers			
Monolayers	C_{11}(N/m)	C_{12} (N/m)	Young's modulus Y (N/m)
$In_2Si_2S_6$	73.067	22.630	66.059
$In_2Si_2S_3Se_3$	67.916	21.041	61.397
$In_2Si_2S_3Te_3$	58.283	20.324	51.196
*$CuInP_2S_6$	62.141	18.077	56.882
*Experimentally synthesized[1–3]			

Thermodynamics for oxygen evolution reaction:

The mechanism of OER is assumed to proceed through -OH, -O and -OOH intermediates. The Gibbs free energy of each elementary step is as follows:[4]

$$\Delta G_O = E_{slab,O} - E_{slab} - (E_{H_2O} - E_{H_2})$$

$$\Delta G_{OH} = E_{slab,OH} - E_{slab} - (E_{H_2O} - \frac{1}{2}E_{H_2})$$

$$\Delta G_{OOH} = E_{slab,OOH} - E_{slab} - (2E_{H_2O} - \frac{3}{2}E_{H_2})$$

Table S2 Equations for calculating the free energies $\Delta G_1 - \Delta G_4$ and overpotentials (η) for the HER and OER		
	ΔG	η
HER	$\Delta G_H = E_{*H} - E_* - \frac{1}{2}E_{H_2}$	$\eta^{HER} = -\frac{\lvert\Delta G_{H*}\rvert}{e}$
OER	$\Delta G_1 = \Delta G_{OH}$	$\eta^{OER} = \frac{max\{\Delta G_1, \Delta G_2, \Delta G_3, \Delta G_4\}}{e} -$
	$\Delta G_2 = \Delta G_O - \Delta G_{OH}$	
	$\Delta G_3 = \Delta G_{OOH} - \Delta G_O$	
	$\Delta G_4 = 4.92eV - \Delta G_1 - \Delta G_2 - \Delta G_3$	

Table S3 Calculated DFT total energy E_{DFT} for gases and adsorbates in the HER and OER process for pristine $In_2Si_2S_6$ monolayer		
Adsorbates	E_{DFT} (eV)	ΔG (eV)
H_2	-6.77	$\Delta G_1 = 2.53$
H_2O	-14.22	$\Delta G_2 = -0.086$
*	-190.8004	$\Delta G_3 = 2.83$
*OH	-199.104	$\Delta G_4 = -0.35$
*O	-195.807	$\Delta G_H = 1.17$
*OOH	-203.81	

*H	-193.0159	

Table S4 Calculated DFT total energy E_{DFT} for gases and adsorbates in the HER and OER process for Janus $In_2Si_2S_3Se_3$ monolayer

Adsorbates	E_{DFT} (eV)	ΔG (eV)
Se-side		
H_2	-6.77	$\Delta G_1 = 2.37$
H_2O	-14.22	$\Delta G_2 = 0.673$
*	-181.72	$\Delta G_3 = 2.25$
*O	-186.12	$\Delta G_4 = -0.37$
*OH	-190.18	$\Delta G_H = 1.29$
*OOH	-194.69	
*H	-183.82	
S-side		
*	-181.72	$\Delta G_1 = 2.42$
*O	-186.76	$\Delta G_2 = -0.01$
*OH	-190.13	$\Delta G_3 = 2.95$
*OOH	-194.65	$\Delta G_4 = -0.44$
*H	-183.97	$\Delta G_H = 1.14$

References:

1 X. Jiang, X. Wang, X. Wang, X. Zhang, R. Niu, J. Deng, S. Xu, Y. Lun, Y. Liu, T. Xia, J. Lu and J. Hong, *Nature Communications*, 2022, **13**, 574.

2 J. Yao, Y. Liu, S. Ding, Y. Zhu, Z. Mao, S. V. Kalinin and Y. Liu, *arXiv preprint arXiv:2309.10045*.

3 F. Liu, L. You, K. L. Seyler, X. Li, P. Yu, J. Lin, X. Wang, J. Zhou, H. Wang, H. He, S. T. Pantelides, W. Zhou, P. Sharma, X. Xu, P. M. Ajayan, J. Wang and Z. Liu, *Nature Communications*, 2016, **7**, 12357.

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