Electronic Supplementary Information for

S-1

Tunable Valley Polarization, Magnetic Anisotropy and Dzyaloshinskii-Moriya Interaction in Two-Dimensional Intrinsic Ferromagnetic Janus 2H-VSe*X* (*X* = S, Te) Monolayers

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Table S1   Lattice constant a (Å), V-Se bond length L_{V-Se} (Å), V/Se magnetic moment $M_{V/Se}$ (μ_B), total magnetic moment of unit cell M_{total} (μ_B), magnetic anisotropic energy MAE (meV), band gap E_g (eV) and valley splitting ΔV (meV) with SOC of the unit cell 2H-VSe$_2$ monolayer with different U_{eff} (eV).

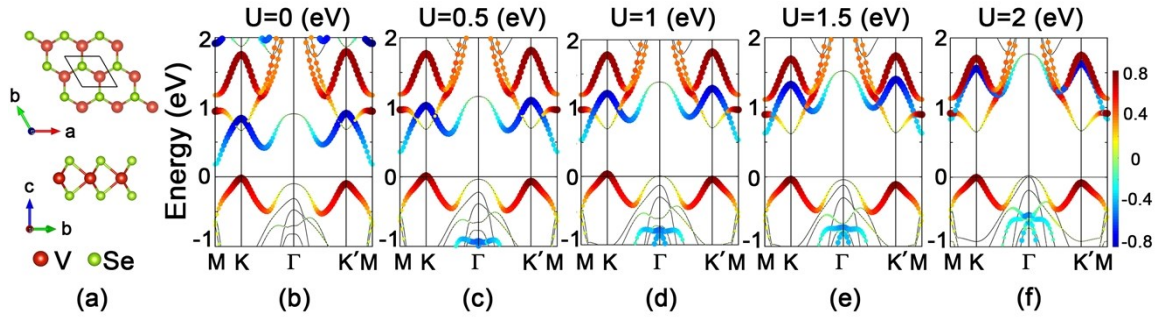| U_{ef}	a	L_{V-Se}	M_V	M_{Se}	M_{total}	MAE	E_g	ΔV
0	3.319	2.497	1.070	-0.066	0.939	0.599	0.197 (K→M)	78.4
0.5	3.328	2.500	1.123	-0.084	0.956	0.538	0.344 (K→M)	78.3
1	3.335	2.505	1.181	-0.104	0.973	0.506	0.497 (K→M)	78.1
1.5	3.342	2.509	1.244	-0.126	0.993	0.545	0.649 (K→K)	78
2	3.349	2.514	1.317	-0.150	1.016	0.513	0.627 (Γ→K)	77.6

Fig. S1 (a) Top and side views of 2H-VSe$_2$ monolayer. (b)-(f) Band structure of 2H-VSe$_2$ monolayer with SOC and different U_{eff} (eV). The circles represent V-$d_{x^2-y^2}$ and V-d_{xy} orbitals. The circle size is proportional to the contribution of the orbital components. The color scale represents the spin projection. Fermi level is set to zero.

Table S2 Calculated total energy (meV/f.u.) relative to the in-plane FM configuration among all magnetic configurations of Janus 2H-VSeX (X = S, Te) monolayers.

Magnetic configurations			VSSe	VSeTe
	FM	[001]	0.431	1.479
		[100]	0	0
		[00$\bar{1}$]	0.432	1.478
	stripy-AFM	out-of-plane	95.117	103.945
		in-plane	95.345	104.234
	zigzag-AFM	out-of-plane	98.823	100.876
		in-plane	99.309	101.146
frustrated-spin	FM	out-of-plane	...	...
	AFM1	out-of-plane	53.195	50.1
	AFM2	in-plane	129.547	140.186

Table S3 Lattice constant a (Å), V-S bond length L_{V-S} (Å), V-Se bond length L_{V-Se} (Å), S-V-Se bond angle θ (º), V/S/Se magnetic moment $M_{V/S/Se}$ (μ_B), band gap E_g (eV), valley splitting ΔV (meV) with SOC of Janus VSSe monolayer at in-plane biaxial strain (%).

| Strai | a | L_{V-S} | L_{V-Se} | θ | Magnetic moment | | | E_g | ΔV |
					M_S	M_V	M_{Se}		
-8	2.997	2.298	2.464	86.6	-0.010	0.940	-0.006	...	66.6
-6	3.062	2.311	2.465	84.3	-0.026	1.004	-0.053	...	69.3
-4	3.127	2.327	2.477	82.3	-0.038	1.050	-0.077	0.222	70.1
-2	3.192	2.345	2.492	80.5	-0.049	1.087	-0.095	0.324	69.7
0	3.257	2.363	2.508	78.7	-0.058	1.120	-0.110	0.322	68.6
2	3.322	2.383	2.526	77.0	-0.067	1.153	-0.124	0.303	66.9
4	3.387	2.404	2.546	75.4	-0.077	1.187	-0.138	0.096	64.6
6	3.453	2.424	2.567	73.7	-0.089	1.228	-0.152	...	61.6
8	3.518	2.439	2.591	72.0	-0.109	1.286	-0.171	...	56

Table S4 Lattice constant a (Å), V-Se bond length L_{V-Se} (Å), V-Te bond length L_{V-Te} (Å), Te-V-Se bond angle θ (º), V/Se/Te magnetic moment $M_{V/Se/Te}$ (μ_B), band gap E_g (eV), valley splitting ΔV (meV) with SOC of Janus VSeTe monolayer at in-plane biaxial strain (%).

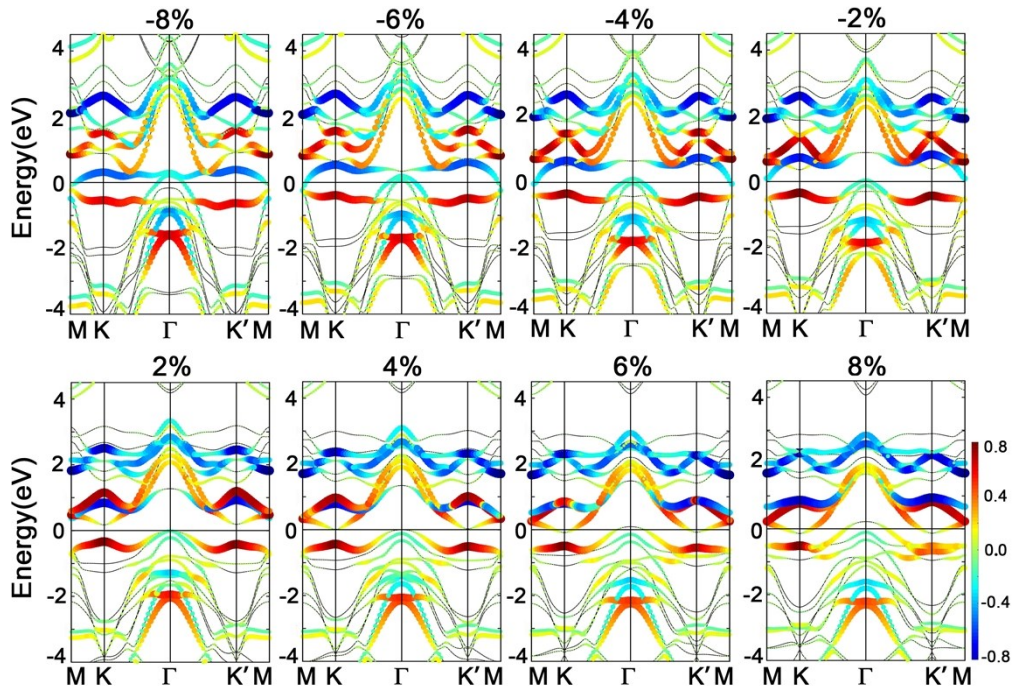| Strai | a | L_{V-Se} | L_{V-Te} | θ | Magnetic moment | | | E_g | ΔV |
					M_V	M_{Se}	M_{Te}		
-8	3.186	2.453	2.698	88.4	0.962	-0.043	0.015	...	78.1
-6	3.255	2.455	2.689	85.6	1.049	-0.048	-0.050	...	83.3
-4	3.325	2.469	2.692	83.5	1.094	-0.055	-0.082	...	92.6
-2	3.394	2.486	2.704	81.5	1.139	-0.065	-0.105	0.057	90.6
0	3.463	2.505	2.720	79.7	1.183	-0.075	-0.123	0.232	86
2	3.532	2.525	2.737	78.0	1.226	-0.086	-0.140	0.184	79.7
4	3.602	2.547	2.756	76.3	1.270	-0.096	-0.155	0.013	70.5
6	3.671	2.561	2.779	74.5	1.334	-0.116	-0.174	...	52.8
8	3.740	2.575	2.804	72.6	1.424	-0.146	-0.200	...	0.8

Fig. S2 Band structure of Janus VSeTe monolayer with SOC and M//z at different biaxial strains from -8% to 8%. The circles represent V-$d_{x^2-y^2}$ and V-d_{xy} orbitals. The circle size is proportional to the contribution of the orbital components. The color scale represents the spin projection. Fermi level is set to zero.
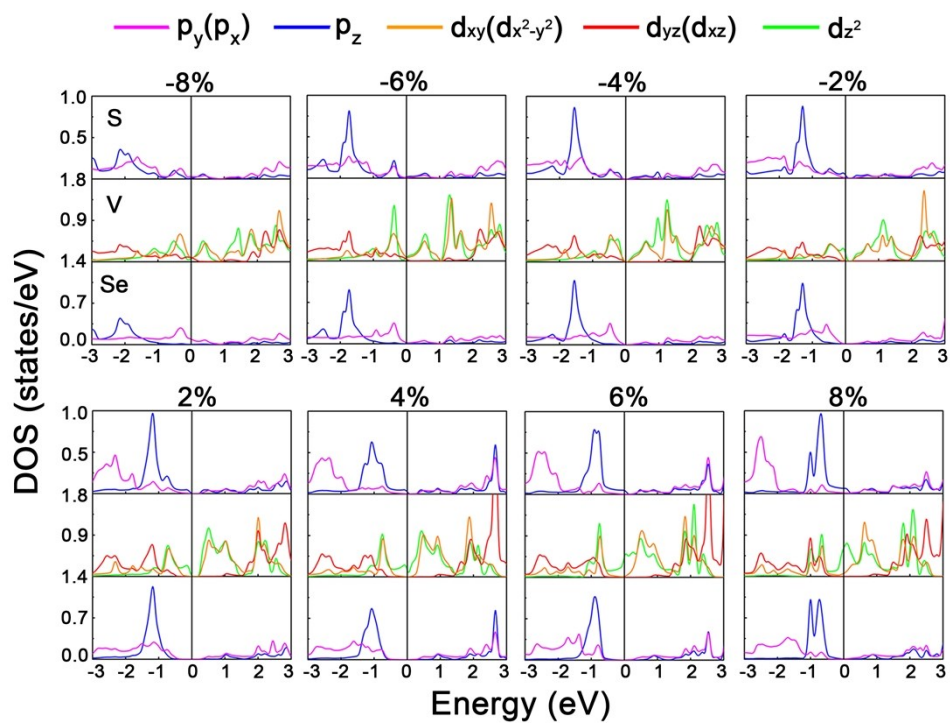
Fig. S3 Orbital-resolved DOS with SOC of Janus VSSe monolayer at different biaxial strains from -8% to 8%.
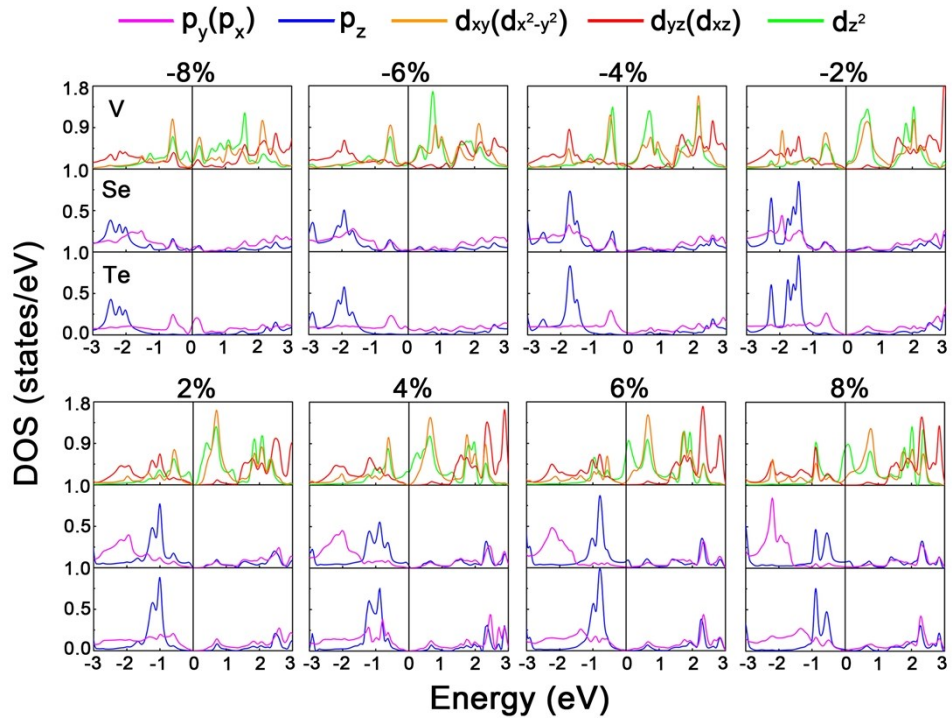
Fig. S4 Orbital-resolved DOS with SOC of Janus VSeTe monolayer at different biaxial strains from -8% to 8%.