Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Structural parameters of VX_2 monolayers

Table S1. Atomic structure, binding energies and magnetic moments of VS_2 and VSe_2 1T and 2H phases.

Structure type	Calculation method	Parameters		
		Lattice constants a, b (Å)	E_0 (eV)	Magnetic moment on unit cell (μ_B)
2H-VS_2	PBE	3.1754	-19.81	0.9570
	PBE + D3	3.1648	-20.19	0.9530
	PBE +U	3.2123	-16.91	1.0000
	HSE	3.1512	-24.36	1.0000
	PBE +U + D3	3.1979	-17.28	1.0000
1T-VS_2	PBE	3.1822	-19.76	0.4800
	PBE + D3	3.1700	-20.16	0.4614
	PBE +U	3.3321	-16.97	1.0000
	HSE	3.2656	-24.30	1.0000
	PBE +U + D3	3.3211	-17.33	1.0009
2H-VSe_2	PBE	3.3345	-18.08	0.9967
	PBE + D3	3.3204	-18.53	0.9956
	PBE +U	3.3758	-15.22	1.0000
	HSE	3.3148	-24.44	1.0000
	PBE +U + D3	3.3651	-15.66	1.0000
1T-VSe_2	PBE	3.3337	-18.03	0.6000
	PBE + D3	3.3136	-18.49	0.5737
	PBE +U	3.4786	-15.30	1.0918
	HSE	3.4083	-24.40	1.1154
	PBE +U + D3	3.4658	-15.73	1.0961

PBE equal GGA PBE

+U equal GGA+U

+D3 equal Grimme correction

E_0 – Total energy

We have calculated the lattice parameters, binding energies and magnetic moments with different combination of Grimme corrections (*D*3) and GGA+*U* method: pure PBE, PBE with only *D*3 Grimme corrections, only PBE+U (Dudarev GGA+*U*) and with both methods, i.e. PBE+*U* with *D*3. The results are provided in Table S1. In all the cases, addition of Grimme *D*3 corrections predictably results in moderate shrinking of VX_2 lattice parameter, since the *D*3 corrections are designed for simulation of long-range van der Waals attraction.

Band structure of VX_2 monolayers

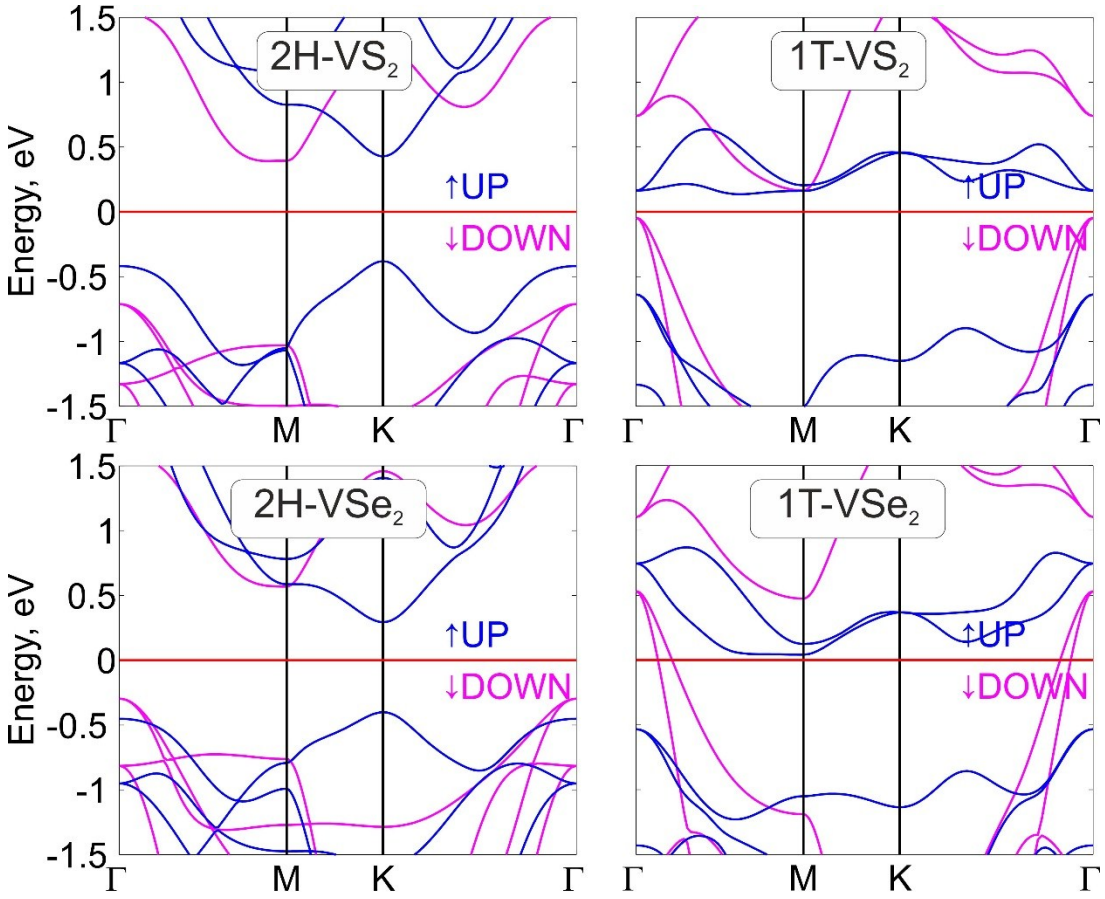

Fig. S1. Band structures of VS_2 and VSe_2 monolayers in 2H and 1T phases.