Supplementary Information

Two-Dimensional Transition-Metal Dichalcogenides-Based Ferromagnetic van der Waals Heterostructures

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Table S1 Calculated lattice constants a and *b*, binding energy E_b, interlayer distance *d*, magnetic moments M, work function W, spin-up band gap $E_{g\text{-up}}$, spin-down band gap $E_{g\text{-down}}$, conduction band offset ΔEc and valence band offset ΔEv with spin-up and spin-down channels using the hybrid HSE06 functional for MoS_2, WS_2, VS_2 monolayers and MoS_2/VS_2, WS_2/VS_2 heterostructures with and without the SOC effects.

		MoS_2	WS_2	VS_2	MoS_2/VS_2	WS_2/VS_2
a=b (Å)		3.183	3.182	3.17	3.179	3.178
E_b (meV/Å^2)		—	—	—	-84.589	-119.766
d(Å)		—	—	—	3.085	3.043
M(μ_B)		0	0	1	0.905	0.898
W (eV)		5.93	5.73	6.77	5.96	5.78
Eg-up (eV)	without SOC	2.143	2.305	1.117	0.418	0.196
	with SOC	2.04	2.02	1.116	0.442	0.202
Eg-down (eV)	without SOC	2.143	2.305	1.985	0.949	0.730
	with SOC	2.04	2.02	1.116	0.442	0.202
$\Delta E_{C\text{-up}}$ (eV)	without SOC	—	—	—	1.455	1.868
	with SOC	—	—	—	1.397	1.811
$\Delta E_{V\text{-up}}$ (eV)	without SOC	—	—	—	0.638	0.832
	with SOC	—	—	—	0.659	0.868
$\Delta E_{C\text{-down}}$ (eV)	without SOC	—	—	—	1.114	1.487
	with SOC	—	—	—	1.397	1.811
$\Delta E_{V\text{-down}}$ (eV)	without SOC	—	—	—	1.063	1.243
	with SOC	—	—	—	0.659	0.868

Table S2 Calculated bond lengths $d_{Mo\text{-}S1(S2)}$, $d_{W\text{-}S1(S2)}$ and $d_{V\text{-}S1(S2)}$ in the XS_2/VS_2 heterostructures under different external E-field.

E-field (V/Å)	MoS_2/VS_2		WS_2/VS_2	
	$d_{Mo-S1(S2)}$(Å)	$d_{V-S1(S2)}$(Å)	$d_{W-S1(S2)}$(Å)	$d_{V-S1(S2)}$(Å)
-0.7	2.411(2.409)	2.359(2.360)	2.412(2.413)	2.359(2.359)
-0.6	2.410(2.409)	2.359(2.359)	2.412(2.413)	2.359(2.359)
-0.5	2.410(2.409)	2.360(2.359)	2.412(2.413)	2.360(2.359)
-0.4	2.410(2.410)	2.360(2.358)	2.411(2.413)	2.361(2.358)
-0.3	2.409(2.410)	2.361(2.358)	2.411(2.414)	2.361(2.358)
-0.2	2.409(2.409)	2.361(2.357)	2.411(2.413)	2.362(2.357)
-0.1	2.409(2.410)	2.362(2.357)	2.411(2.413)	2.362(2.357)
0	2.408(2.410)	2.362(2.356)	2.410(2.414)	2.362(2.356)
0.1	2.408(2.410)	2.362(2.356)	2.410(2.414)	2.363(2.356)
0.2	2.407(2.411)	2.363(2.355)	2.410(2.414)	2.363(2.356)
0.3	2.407(2.411)	2.363(2.355)	2.409(2.414)	2.363(2.356)
0.4	2.407(2.411)	2.364(2.355)	2.409(2.414)	2.364(2.355)
0.5	2.407(2.411)	2.363(2.355)	2.409(2.414)	2.364(2.355)
0.6	2.406(2.412)	2.364(2.355)	2.409(2.414)	2.364(2.355)
0.7	2.406(2.411)	2.365(2.354)	2.408(2.413)	2.364(2.354)

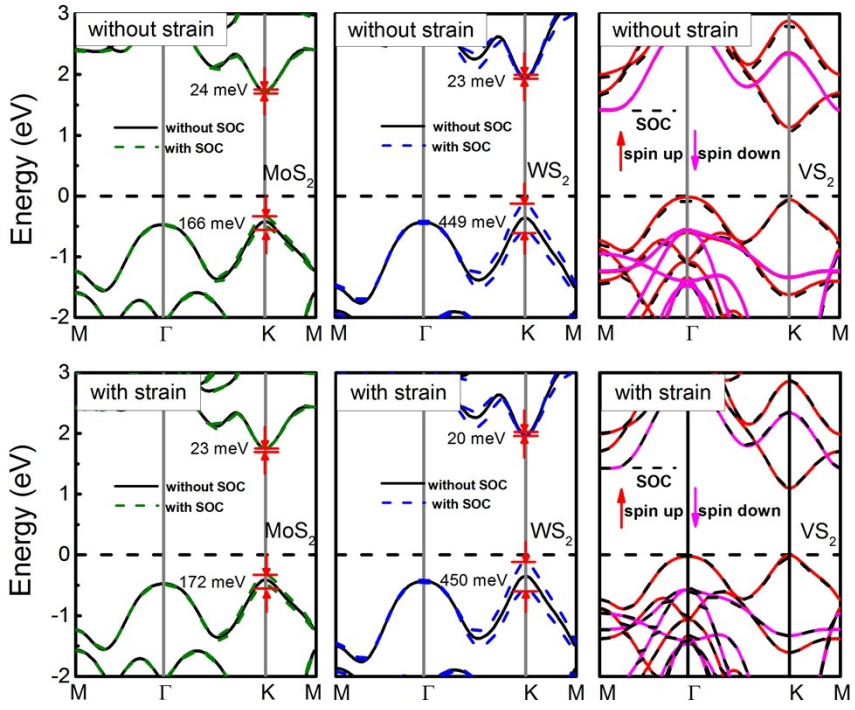

Fig. S1 The band structures of MoS_2, WS_2 and VS_2 monolayers with and without strain calculated using the HSE06 methods, considering the SOC effects.

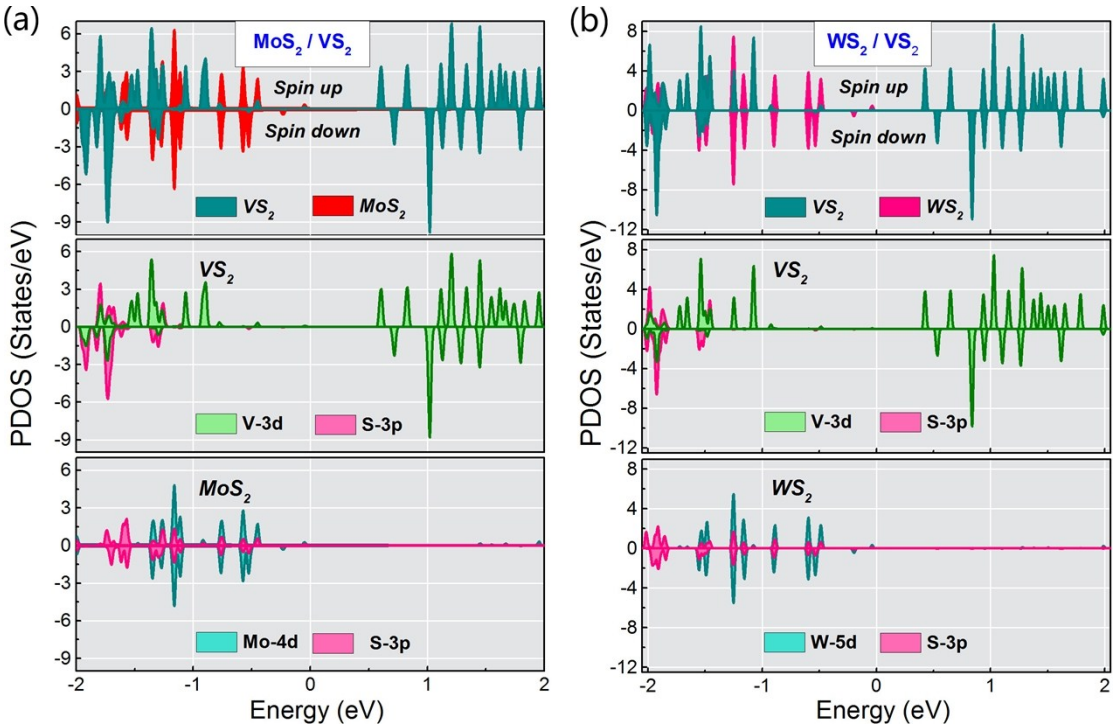

Fig. S2 The projected density of states (PDOS) of (a) MoS_2/VS_2 and (b) WS_2/VS_2 heterostructures.

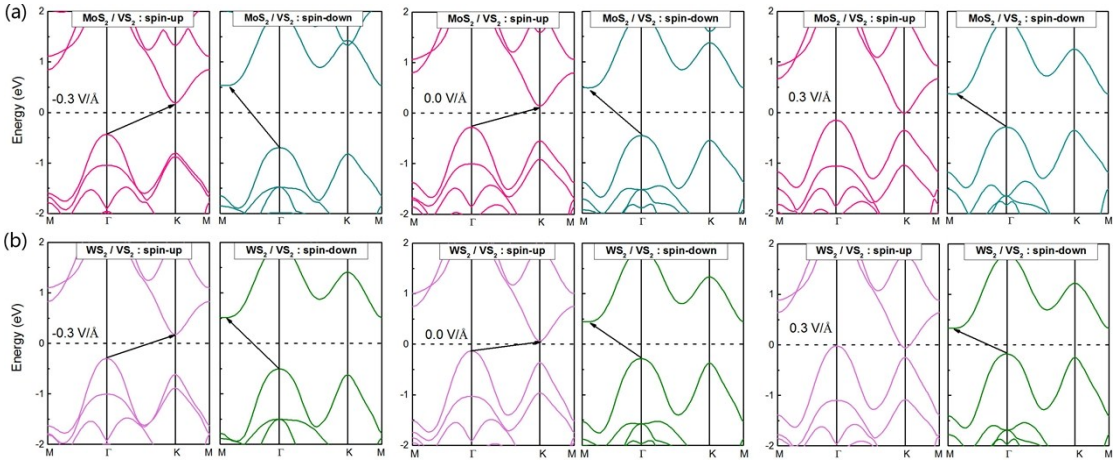

Fig. S3 The spin-up and spin-down bands obtained by HSE06 methods for (a) MoS_2/VS_2 and (b) WS_2/VS_2 heterostructure under external electric field of -0.3 V/Å, 0 V/Å, and 0.3 V/Å.

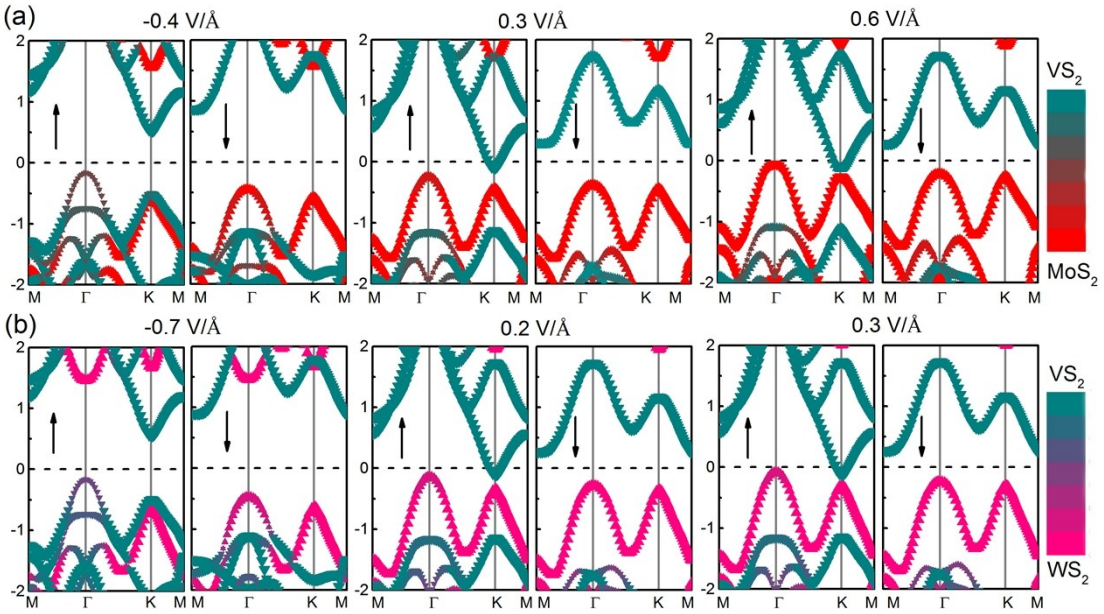

Fig. S4 The projected band structures of (a) MoS_2/VS_2 and (b) WS_2/VS_2 heterostructures under selected external electric field with majority and minority bands marked with upward arrows and downward arrows, respectively.