Supporting Information

Transition Metal Dihydride Monolayers: A New Family of Two-Dimensional Ferromagnetic Materials with Intrinsic Room-Temperature Half-Metallicity

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Table S1. Relative energies (*meV/MH*$_2$) between ferromagnetic (*FM*), three antiferromagnetic (*AFM1*, *AFM2* and *AFM3*) states, and nonmagnetic (*NM*) states for MH_2 (M=Sc, Ti, V, Cr, Fe, Co, Ni) monolayers. Ground states are highlighted with yellow.

System	*FM*	*AFM1*	*AFM2*	*AFM3*	*NM*
ScH_2	0	40.5	58.5	31.6	104.1
TiH_2	0	-112.9	-97.3	-19.5	104.1
VH_2	0	-365.1	-357.8	-198.1	718.6
CrH_2	0	-17.0	-67.7	31.0	1360.3
FeH_2	0	-220.0	-184.8	-99.5	445.0
CoH_2	0	151.0	342.5	57.3	764.0
NiH_2	0	-138.2	-215.5	16.9	1.5

Table S2. Ground states (GS), magnetic moment (M, μ_B) on metal atoms, average metal-hydrogen bond lengths (d_{M-H}), metal-metal bond length (d_{M-M}), charge (C, e), formation energies (E_{form}), lattice parameters (*Lattice*) and atomic fractional coordinates (*Coordinates*) for MH_2 (M=Sc, Ti, V, Cr, Fe, Co, Ni) monolayers. FM, AFM1 and AFM2 denote a ferromagnetic and two antiferromagnetic states, respectively. SC, M, HM and QHM indicate semiconductor, metal, half-metal and quasi-half-metal, respectively.

System	*GS*	*M* (μ_B)	d_{M-H} (Å)	d_{M-M} (Å)	*C* (*e*)	E_{form} (eV/MH_2)	*Lattice*	*Coordinates*
ScH_2	FM QHM	0.59	2.05	3.19	1.58	-5.30	a = 3.19 Å b = 3.19 Å γ = 60°	Sc (0.333, 0.333, 0.392) H (0.000, 0.000, 0.445) H (0.667, 0.667, 0.339)
TiH_2	AFM1 SC	1.24	1.95	2.96	1.38	-4.97	a = 5.10 Å b = 2.96 Å γ = 90°	Ti (0.748, 0.269, 0.470) Ti (0.248, 0.769, 0.470) H (0.416, 0.272, 0.415) H (0.916, 0.772, 0.415) H (0.080, 0.266, 0.525) H (0.580, 0.766, 0.525)
VH_2	AFM1 SC	2.40	1.89	2.82	1.20	-4.65	a = 4.77 Å b = 2.89 Å γ = 90°	V (0.745, 0.253, 0.533) V (0.245, 0.753, 0.533) H (0.409, 0.253, 0.472) H (0.909, 0.753, 0.472) H (0.082, 0.253, 0.593) H (0.582, 0.753, 0.593)
CrH_2	AFM2 SC	3.52	1.90	2.87	1.08	-2.71	a = 4.94 Å b = 5.76 Å γ = 90°	Cr (0.757, 0.134, 0.562) Cr (0.257, 0.863, 0.562) Cr (0.257, 0.363, 0.562) Cr (0.757, 0.634, 0.562) H (0.072, 0.149, 0.623) H (0.572, 0.348, 0.623) H (0.436, 0.084, 0.500) H (0.936, 0.413, 0.500) H (0.072, 0.649, 0.623) H (0.572, 0.848, 0.623) H (0.436, 0.584, 0.500) H (0.936, 0.913, 0.500)
FeH_2	AFM1 SC	3.02	1.80	2.69	0.85	-3.03	a = 4.85 Å b = 2.57 Å γ = 90°	Fe (0.751, 0.273, 0.623) Fe (0.251, 0.773, 0.623) H (0.093, 0.273, 0.677) H (0.593, 0.773, 0.677) H (0.409, 0.273, 0.569) H (0.909, 0.773, 0.569)
CoH_2	FM HM	1.19	1.71	2.53	0.55	-3.49	a = 2.53 Å b = 2.53 Å γ = 60°	Co (0.333, 0.333, 0.625) H (0.000, 0.000, 0.677) H (0.667, 0.667, 0.572)
NiH_2	AFM2 SC	1.17	1.73	2.57	0.62	-3.41	a = 4.43 Å b = 5.14 Å γ = 90°	Ni (0.745, 0.115, 0.508) Ni (0.245, 0.873, 0.508) Ni (0.245, 0.373, 0.508) Ni (0.745, 0.615, 0.508) H (0.083, 0.134, 0.568) H (0.583, 0.354, 0.568) H (0.407, 0.134, 0.449) H (0.906, 0.354, 0.449) H (0.083, 0.634, 0.567) H (0.583, 0.854, 0.567) H (0.407, 0.634, 0.449) H (0.907, 0.854, 0.449)

Table S3. Relative energies between T and H phases for 2D MH_2 (M=Sc, Ti, V, Cr, Fe, Co, Ni) monolayers with ferromagnetic ground states. E_T and E_H indicate the total energy for the T and H phases of MH_2 monolayers, respectively.

System	E_H-E_T (eV/MH_2)
ScH_2	0.025
TiH_2	0.239
VH_2	0.734
CrH_2	0.595
FeH_2	0.520
CoH_2	0.745
NiH_2	1.238

Table S4. Magnetic anisotropy energies in $\mu eV/MH_2$ of different directions compared with (001) direction for CoH_2 and ScH_2 monolayers, respectively.

System	E(100)-E(001)	E(010)-E(001)	E(110)-E(001)	E(111)-E(001)
CoH_2	-442.6	-442.1	-442.8	-300.3
ScH_2	20.4	20.4	20.4	13.7

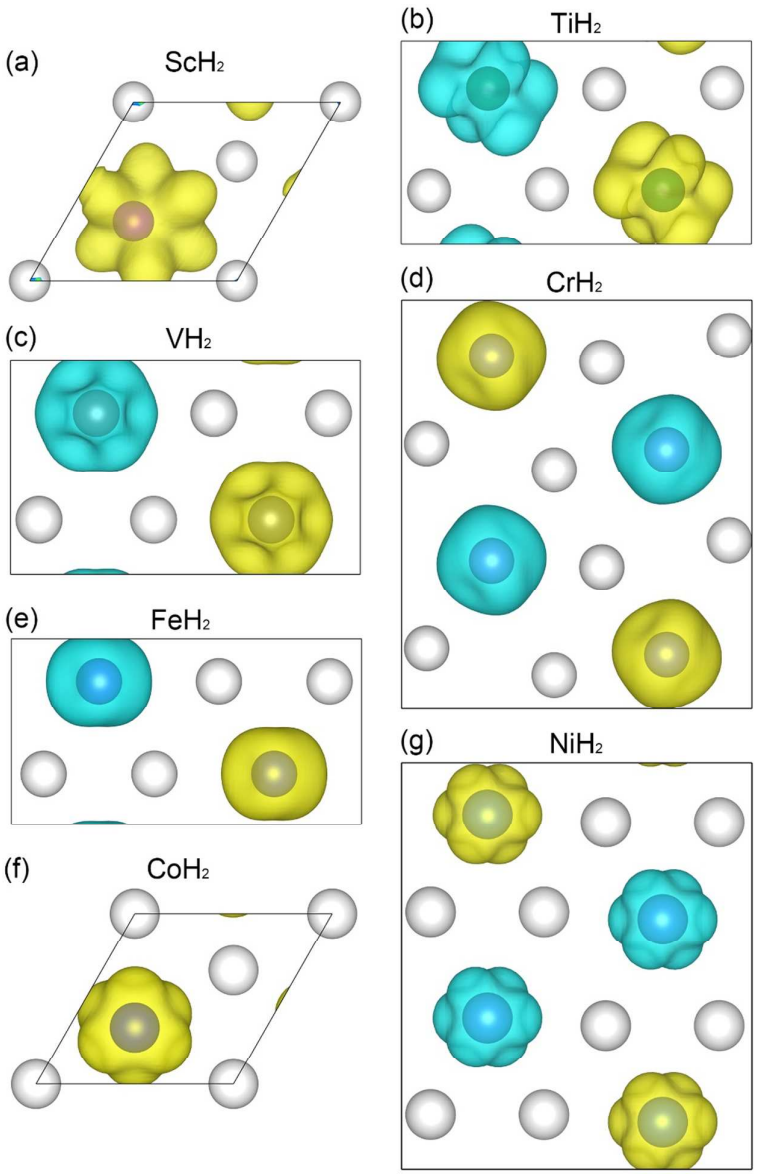

Figure S1. Spin densities for ScH_2 (a), TiH_2 (b), VH_2 (c), CrH_2 (d), FeH_2 (e), CoH_2 (f), and NiH_2 (g) monolayers. Colored and white spheres denote transition-metal and hydrogen atoms, respectively. Yellow and blue areas indicate spin up and spin down, respectively. Black lines indicate primitive cells.

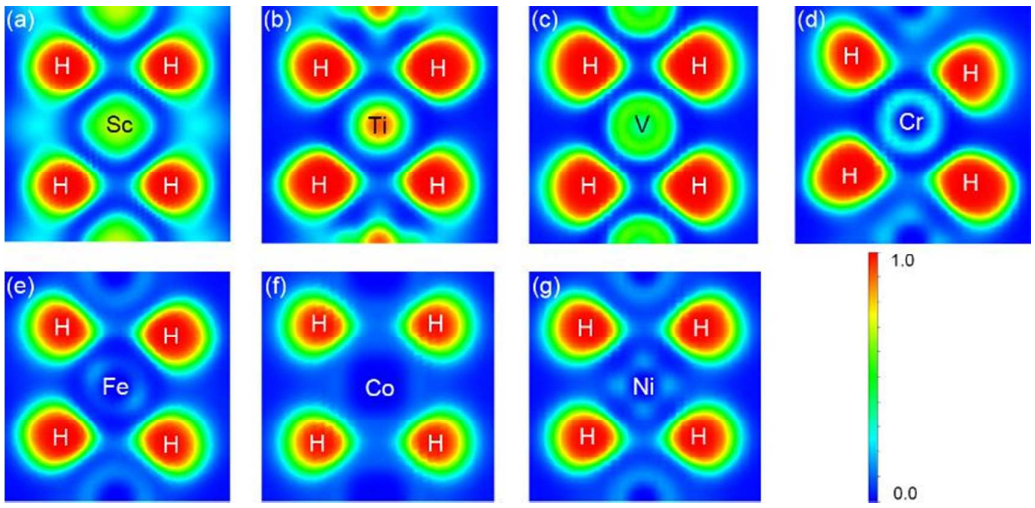

Figure S2. Electron localization functions (ELF) for ScH_2 (a), TiH_2 (b), VH_2 (c), CrH_2 (d), FeH_2 (e), CoH_2 (f), and NiH_2 (g) monolayers. Transition metal and hydrogen atoms are annotated accordingly.

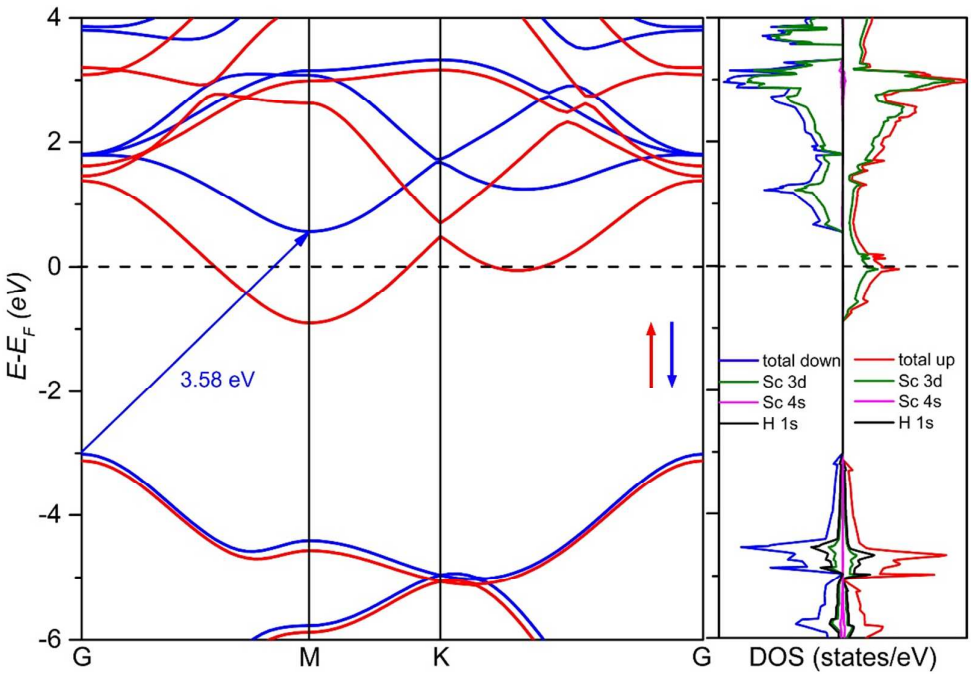

Figure S3. Spin-polarized band structures and projected density of states (DOS) for ScH_2 monolayer doping with -0.5*e*. Red and blue lines represent spin-up and spin-down parts, respectively. Dashed line denotes the Fermi level set to zero.

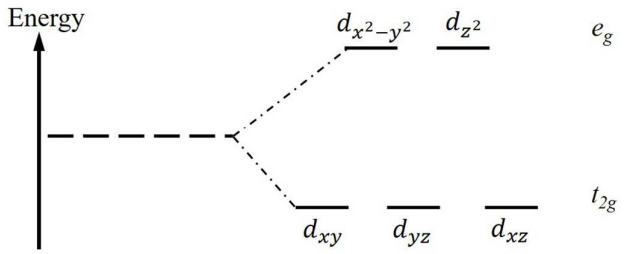

Figure S4. Energy diagram for an octahedral crystal field.

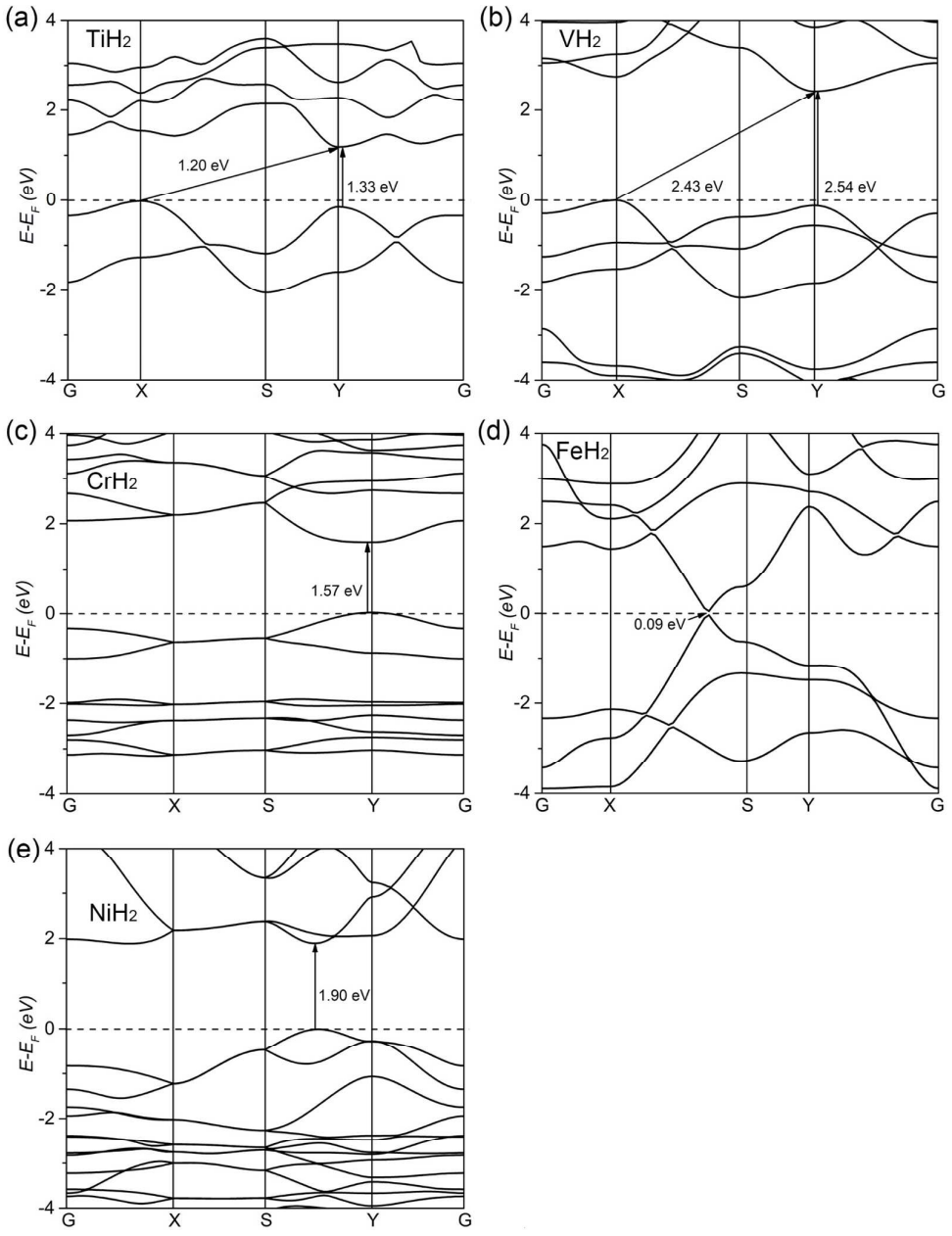

Figure S5. Computed band structures based on the HSE06 functional for TiH_2 (a), VH_2 (b), CrH_2 (c), FeH_2 (d), and NiH_2 (e) monolayers.

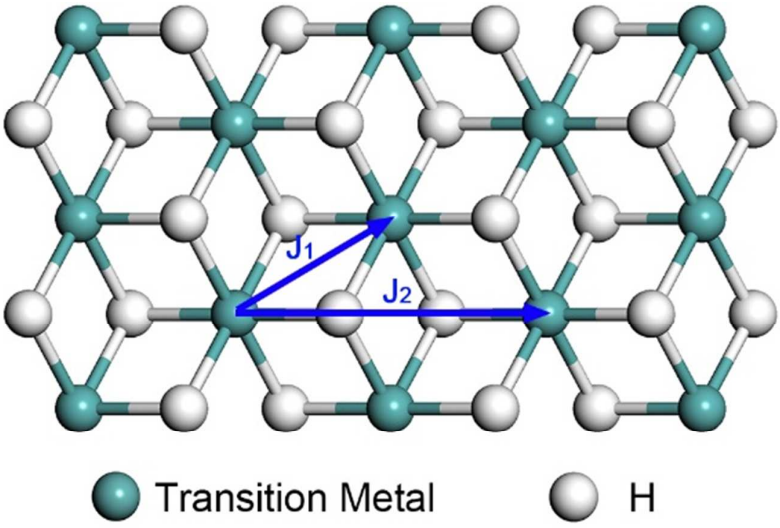

Figure S6. Spin configuration for estimating the exchange-interaction constants. J_1 and J_2 are the nearest and next nearest magnetic exchange interaction parameters, respectively.

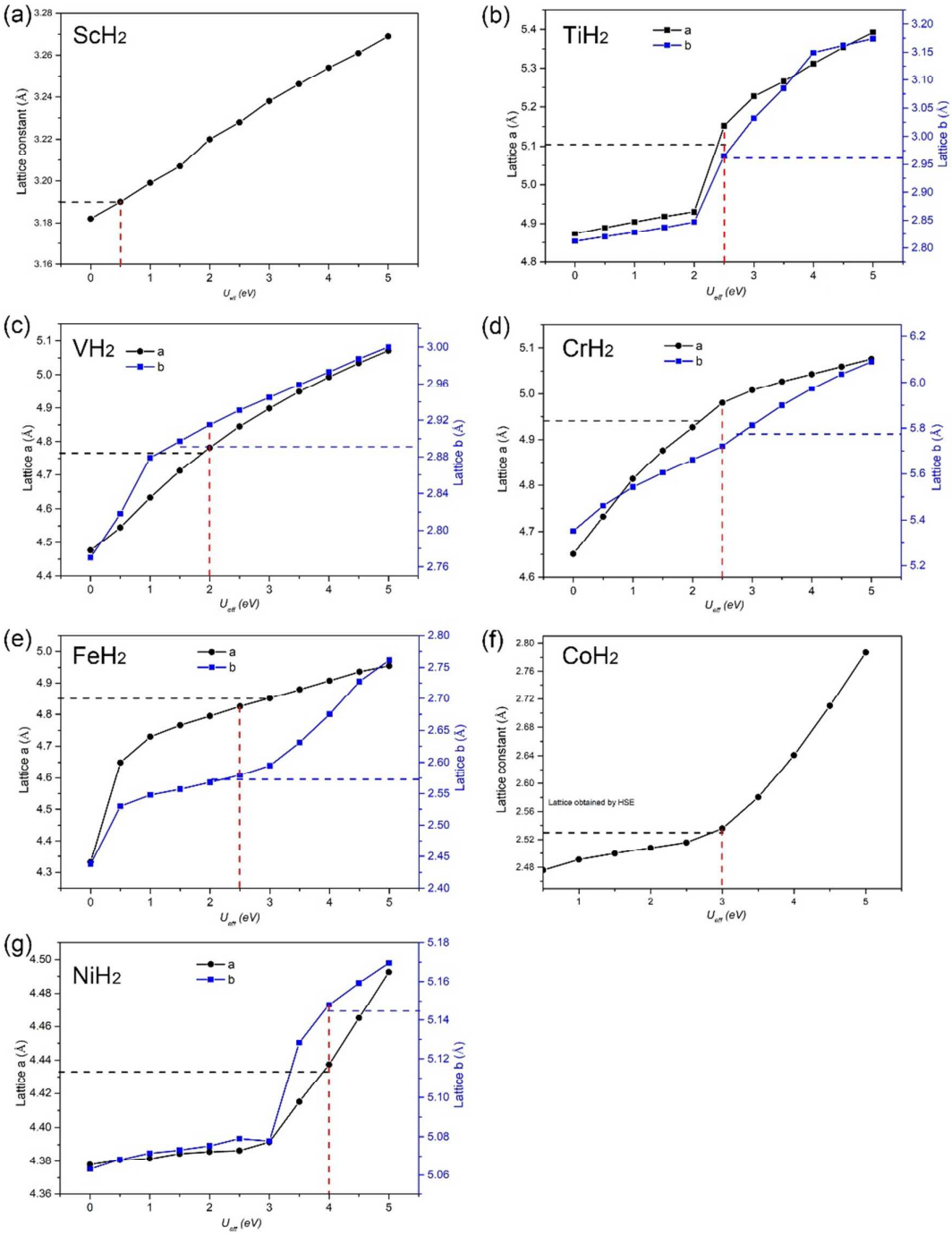

Figure S7. Test on various *U* values, compared with the results at HSE06 level for ScH_2 (a), TiH_2 (b), VH_2 (c), CrH_2 (d), FeH_2 (e), CoH_2 (f), and NiH_2 (g) monolayers. Solid lines denote lattice constants calculated using *GGA+U* method. Black and blue dashed lines indicate constants calculated at HSE06 level, respectively. Red dashed lines mark the *U* values used in phonon dispersion calculations and *ab initio* molecular dynamics simulations.

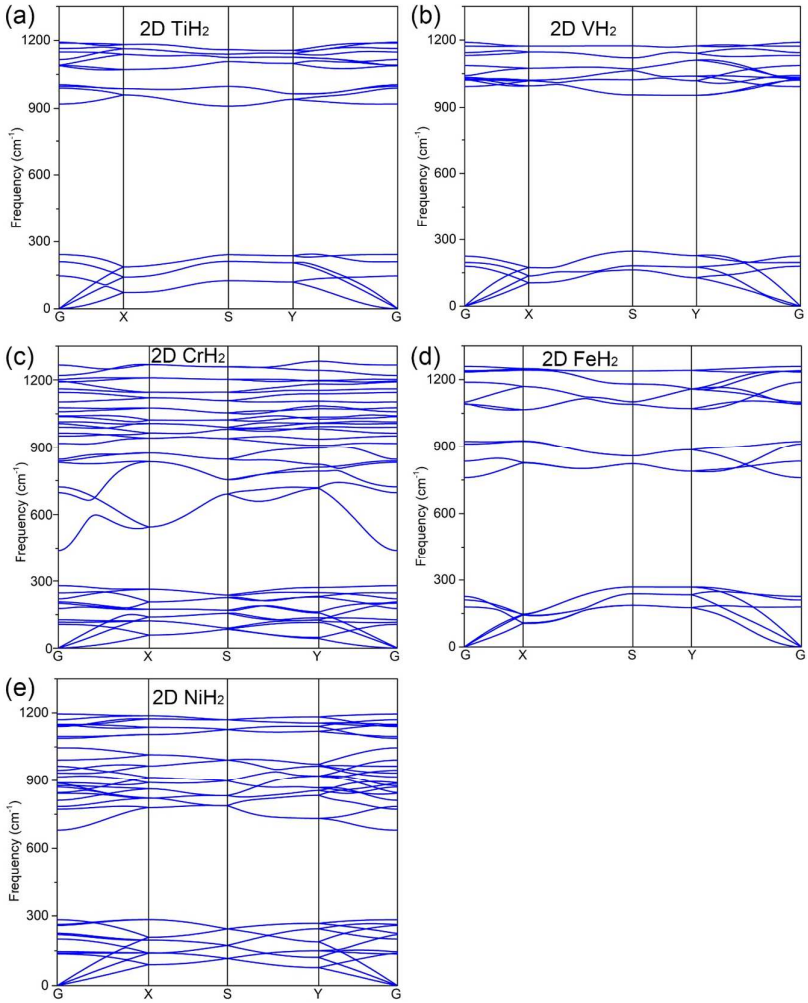

Figure S8. Phonon dispersion spectra for TiH_2 (a), VH_2 (b), CrH_2 (c), FeH_2 (d) and NiH_2 (e) monolayers.

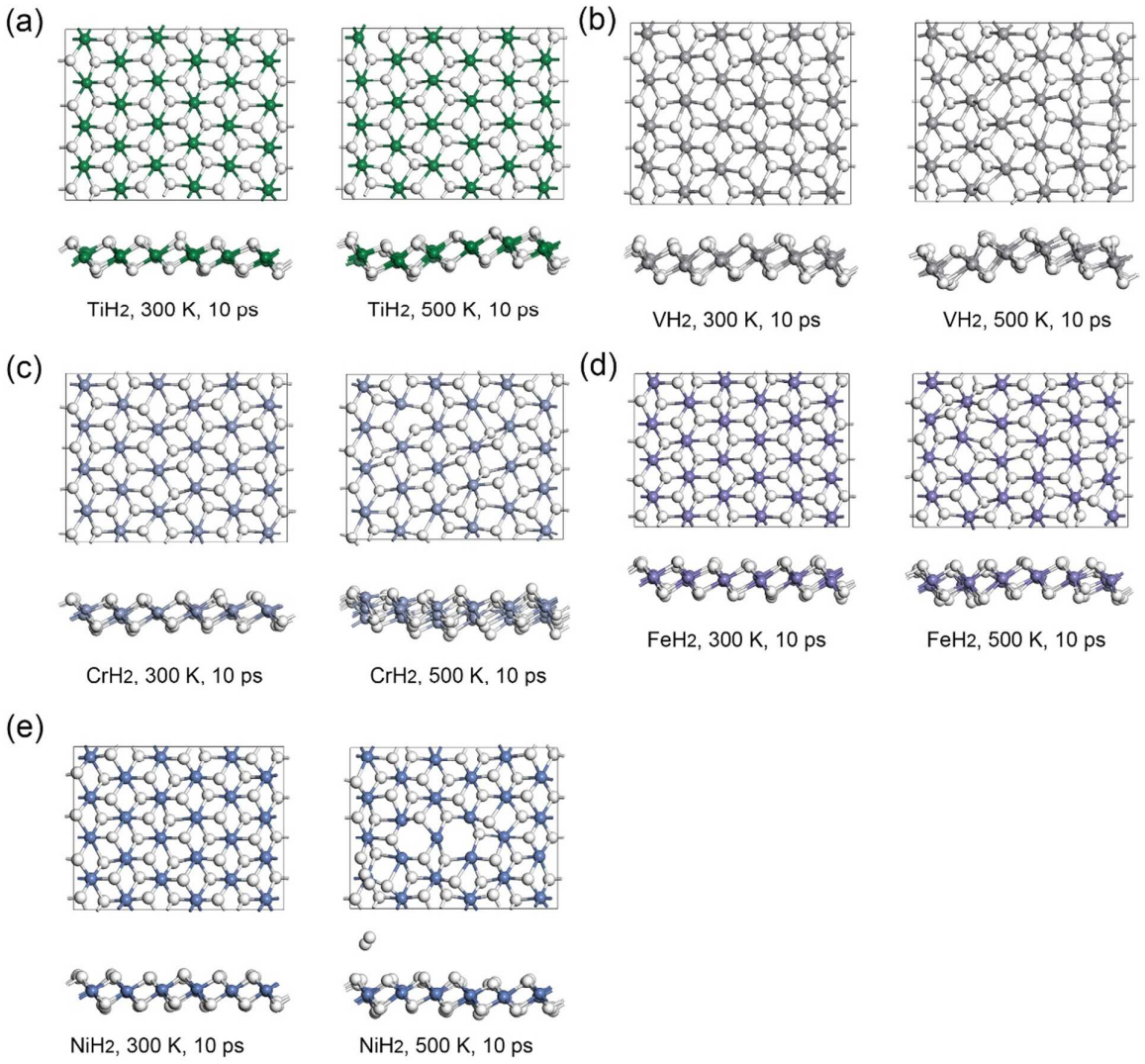

Figure S9. *Ab initio* molecular dynamics (AIMD) snapshots (top and side views) of TiH_2 (a), VH_2 (b), CrH_2 (c), FeH_2 (d) and NiH_2 (e) monolayers, after 10 ps run at 300 K and 500 K, respectively.

(a)

Magnetism	Relative energy (meV/MnH_2)
FM	0.0
AFM1	-258.0
AFM2	-253.5
AFM3	-198.3
NM	1620.0

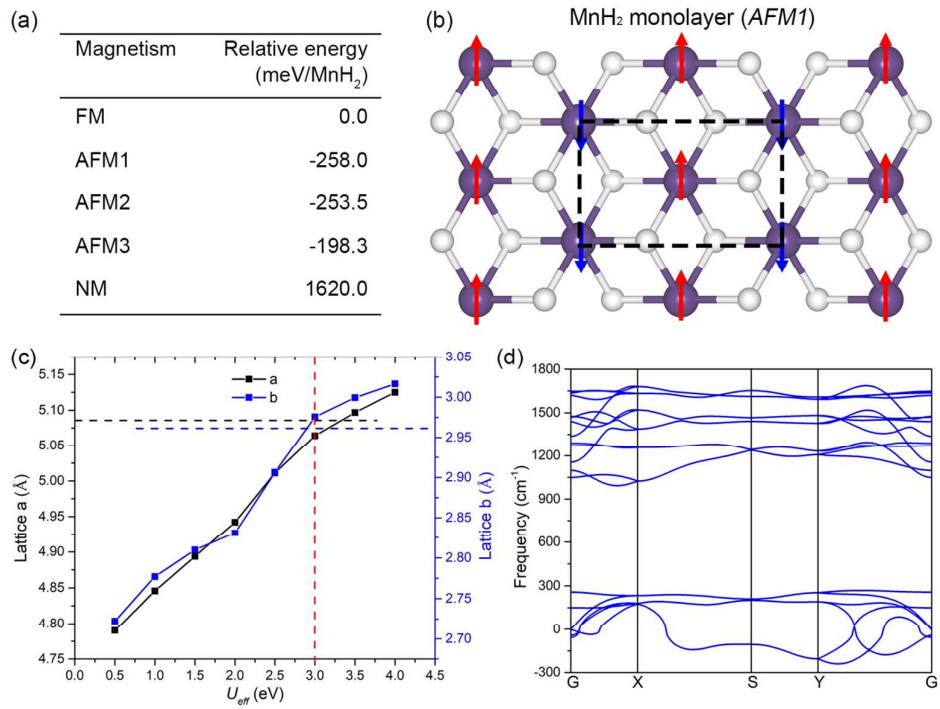

Figure S10. (a) Relative energies between different magnetic states for MnH_2 monolayer. AFM1 is the ground state for MnH_2 monolayer. (b) The geometric structure for MnH_2 monolayer. Red and blue arrows denote spin up and down, respectively. Black dashed lines indicate unit cell. (c) Test on various U values, compared with the results at HSE06 level for MnH_2 monolayer. Solid lines denote lattice constants calculated using GGA+U method. Black and blue dashed lines indicate constants calculated at HSE06 level, respectively. Red dashed lines mark the U value used in phonon dispersion calculations. (d) Phonon dispersions for MnH_2 monolayer. The existence of large imaginary frequencies suggest that MnH_2 monolayer is unstable.