Electronic Supplementary Information

Novel Quantum Spin Hall Insulators in Two-dimensional Hydrogenated Molybdenum and Tungsten Dinitride MN_2H_2 (M = Mo, W) with High Stability

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SURPOTING INFORMATION

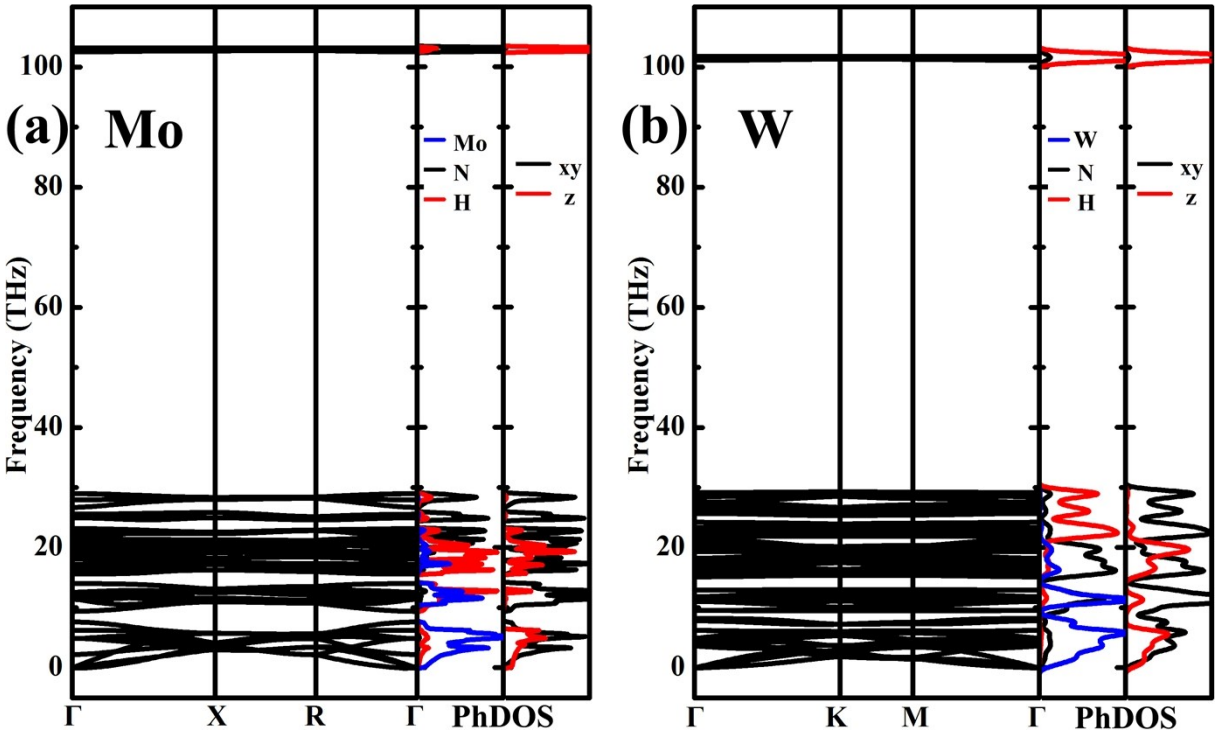

Figure S1. The phonon spectra and projected phonon density of states (PhDOS) for (a) g-MoN_2H_2 and (b) g-WN_2H_2 monolayers.

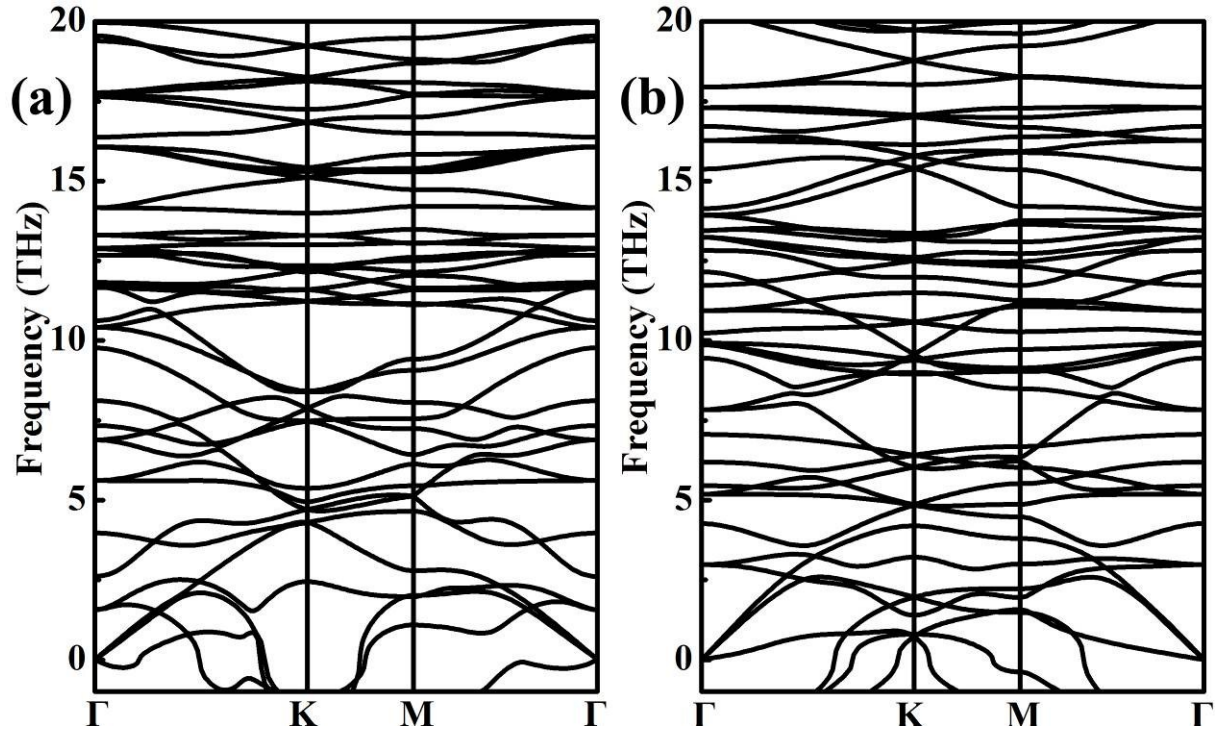

Figure S2. The phonon spectra for (a) g-MoN_2 and (b) g-WN_2 monolayers.

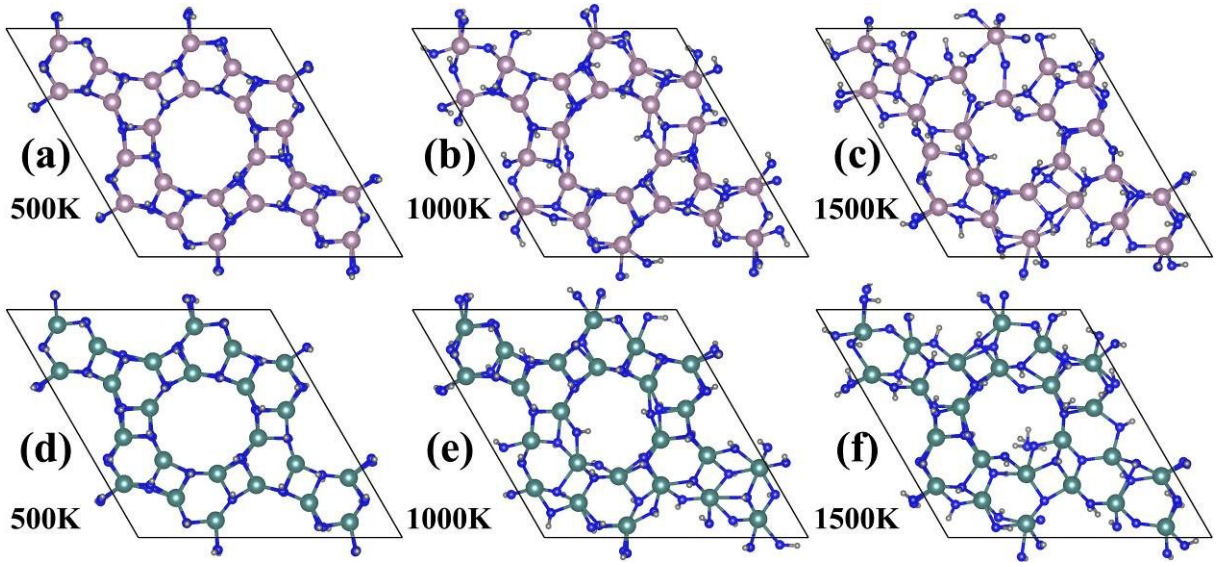

Figure S3. The top views of monolayers after the *ab-initio* molecular dynamics simulation: (a) 500 K, (b) 1000 K and (c) 1500 K for g-MoN_2H_2 monolayer; (d) 500 K, (e) 1000 K and (f) 1500 K for g-WN_2H_2 monolayer.

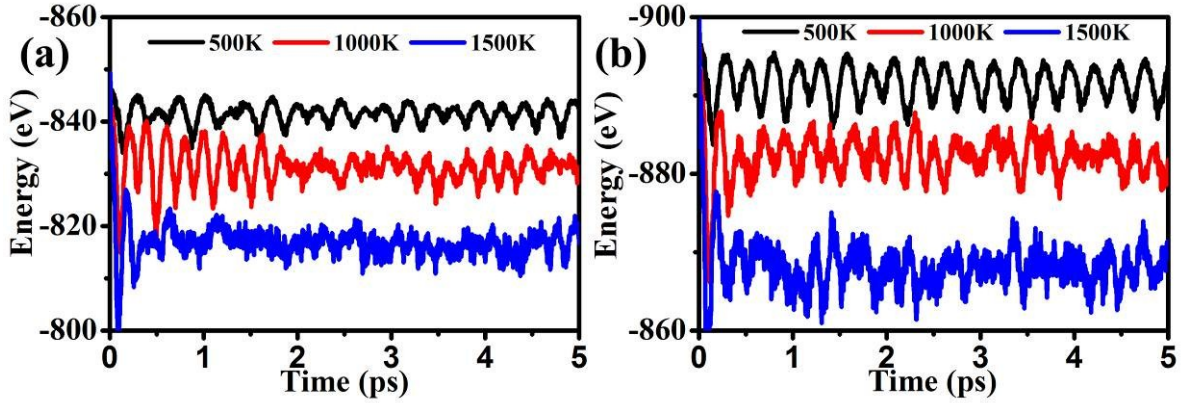

Figure S4. Energy fluctuations with respect to time in *AIMD* simulations at 500 K, 1000 K and 1500K for the (a) g-MoN_2H_2 and (b) g-WN_2H_2 monolayers, respectively.

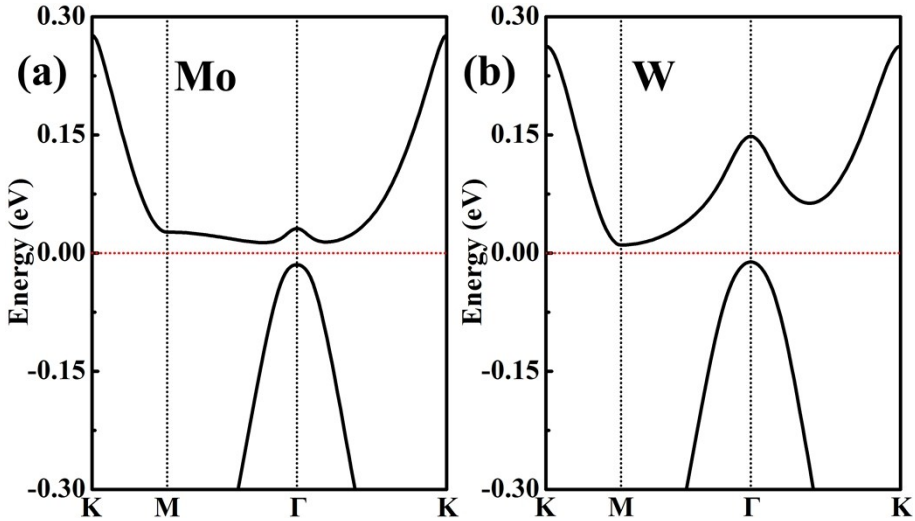

Figure S5. The zoom-in band structures for (a) g-MoN_2H_2 and (b) g-WN_2H_2 monolayers with SOC.

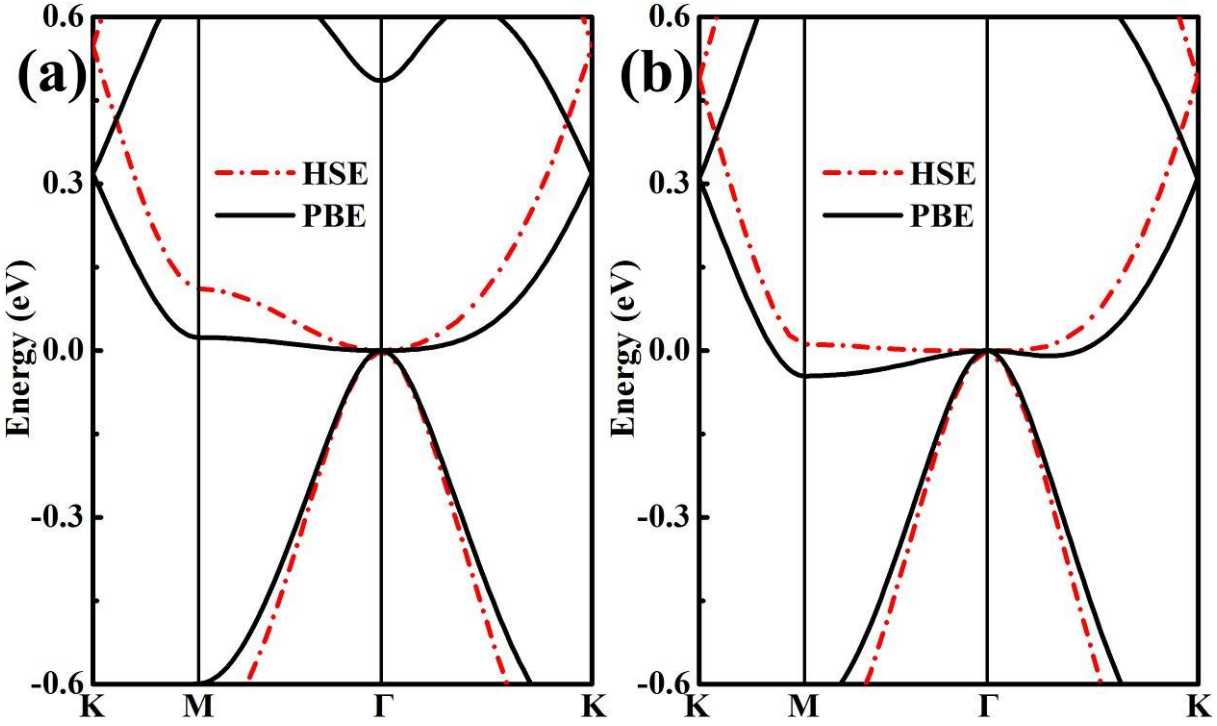

Figure S6. The band structures for (a) g-MoN_2H_2 and (b) g-WN_2H_2 monolayers based on PBE and HSE06.

Comp.	Parities of occupied spin-degenerate bands	Product
g-MoN_2H_2	+++------+++++--++------+++++----+--++++++----+-+++++-+-	Γ (+)
	-+-++-++--+--+-+-++-+-+--++-++--++--++--+--+++--+--+-+-+	M (-)
g-WN_2H_2	+++------+++++++-------+++++----+--++++++----+-+++-+++-	Γ (+)
	-+-++-++--+--+-+-++-+-+-+-++--+-++--++--+--++--++--+-+-+	M (-)

Figure S7. The parities of fifty-four occupied bands at Γ (red color) and M (black color) points for g-MoN_2H_2 and g-WN_2H_2 sheets. The total number of +1 (−1) at Γ point is twenty-eight (twenty-six), while twenty-seven (twenty-seven) for M point. So it yields that the product of Γ and M points are + 1 and – 1 for g-MoN_2H_2 and g-WN_2H_2 sheets.

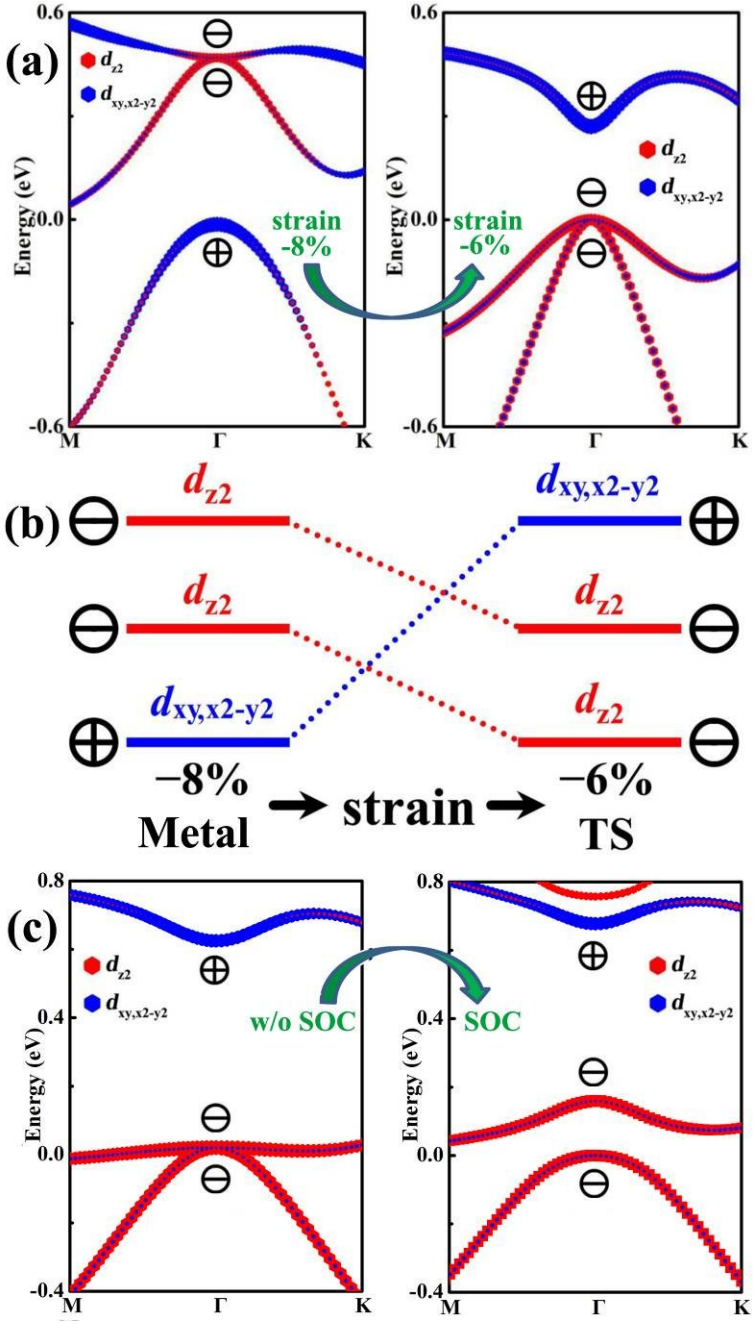

Figure S8. (a) Selected orbital-resolved band structures for g-WN_2H_2 monolayers under strain -8% and -6%. The red hexagonal dots represent the contributions from the W d_{z2} atomic orbitals; the blue hexagonal dots represent the contribution from W $d_{xy,x2-y2,}$ atomic orbitals. (b) Schematic illustration of the band evolution from the atomic orbitals d_{z2} and $d_{xy,x2-y2,}$ of the W atom around the Fermi level at the Γ point for g-WN_2H_2 monolayer. (c) Zoom-in orbital-resolved band structures for g-WN_2H_2 monolayers with and without SOC. Parity values are presented near the various orbitals.

Table S1. The contributions of the five *d* orbitals to the VBM and CBM based on PBE and HSE06 for g-MN_2H_2 sheets.

Comp.	d_{xy} (HSE06/PBE)	d_{yz}	d_{z2} (HSE06/PBE)	d_{xz}	d_{x2-y2} (HSE06/PBE)	Total (HSE06/PBE)	
g-MoN_2H_2	0.055/0.062	0	0.645/0.618	0	0.056/0.063	0.855/0.854	CBM
	0.056/0.063	0	0.645/0.618	0	0.055/0.062	0.855/0.854	VBM
g-WN_2H_2	0.055/0.058	0	0.613/0.612	0	0.055/0.060	0.835/0.829	CBM
	0.055/0.060	0	0.613/0.612	0	0.055/0.058	0.835/0.829	VBM