

Supporting Information

**Versatile Electronic Properties of VSe₂ Bulk, Few-layer, Monolayer,
Nanoribbons and Nanotubes: A Computational Exploration**

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Table S1. The optimized lattice parameters (in Å) of *T*-, *H1*-, *H2*- and *H3*-VSe₂ bulk from PBE-D2 computations.

	<i>T</i> ^a	<i>T</i>	<i>H1</i>	<i>H2</i>	<i>H3</i>
<i>a/b</i>	3.35	3.33	3.33	3.32	3.32
<i>c</i>	6.12	6.17	18.91	12.78	19.30

^a Experimental measurements from Ref [Adv. Phys. **1969**, 18, 193].

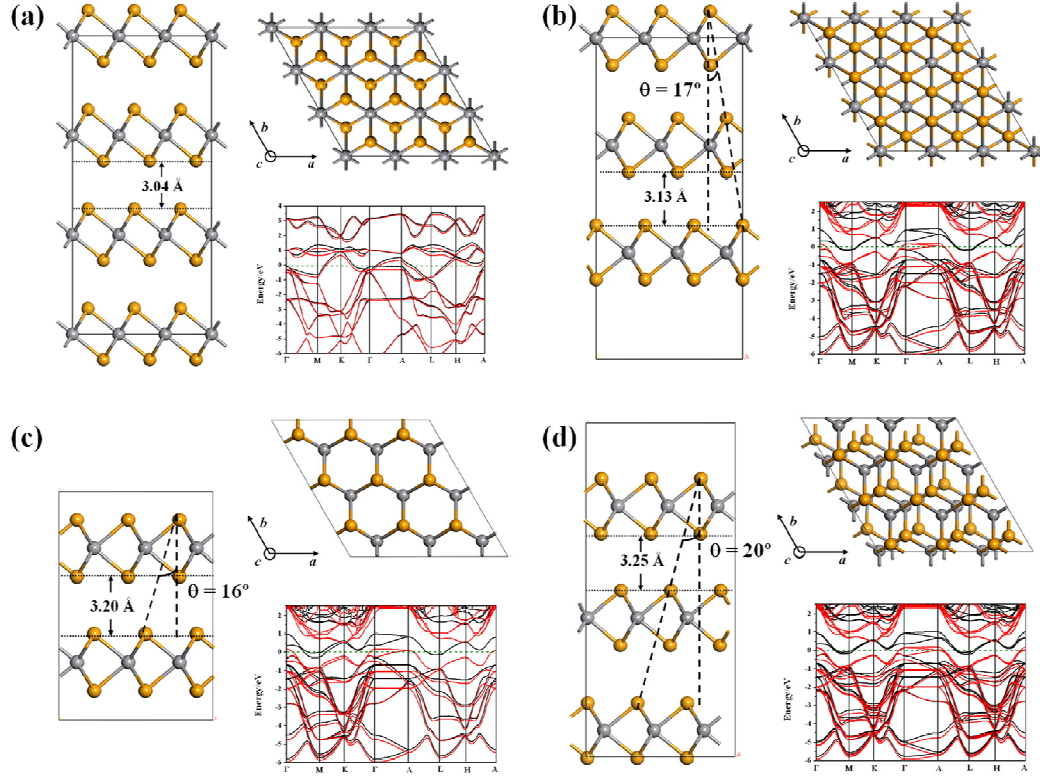


Figure S1. Side and top views of PBE-D2 optimized *T*- (a), *H1*- (b), *H2*- (c), and *H3*-VSe₂ (d) bulk (a 3×3×3 supercell), and their band structures of spin-up (black) and spin-down (red) channels. The grey and khaki atoms represent V and Se atoms, respectively. The Fermi energy is denoted by a green dashed line.

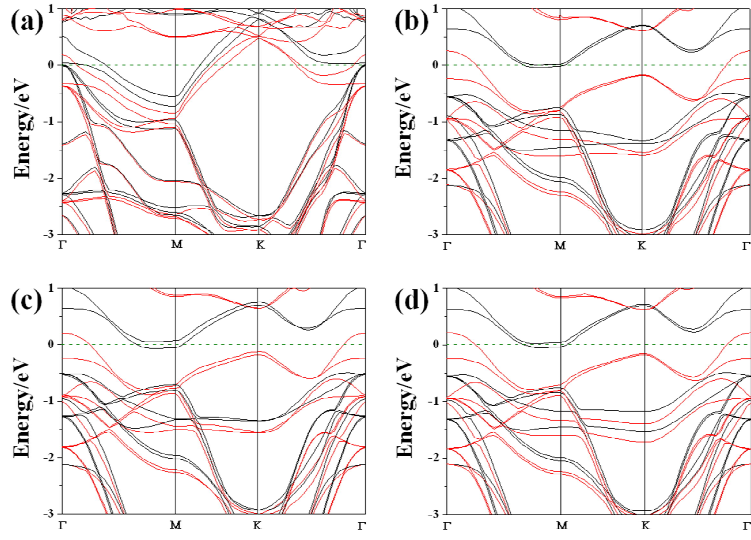


Figure S2. Band structures of spin-up (black) and spin-down (red) channels for 2-layer sheet in T (a), $H1$ (b), $H2$ (c) and $H3$ (d) phases.

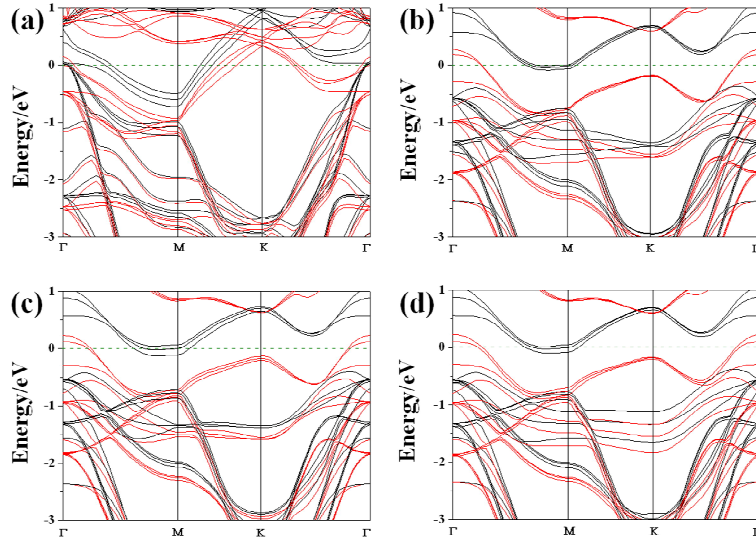


Figure S3. Band structures of spin-up (black) and spin-down (red) channels for 3-layer sheet in T (a), $H1$ (b), $H2$ (c) and $H3$ (d) phases.

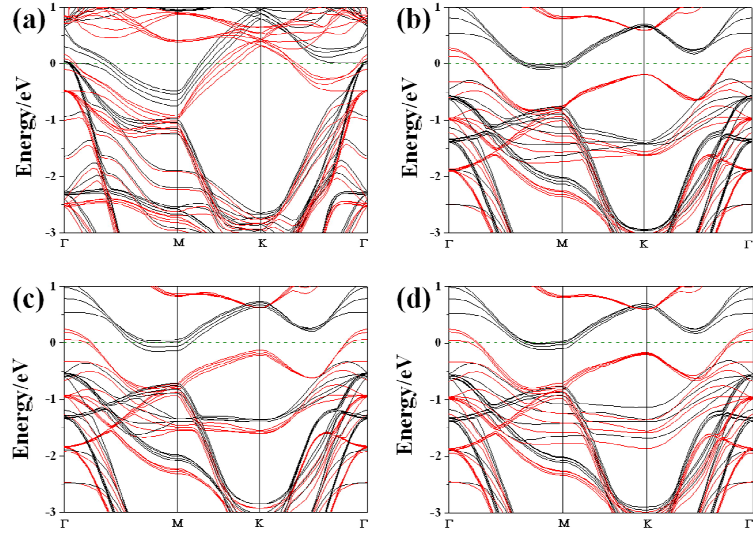


Figure S4. Band structures of spin-up (black) and spin-down (red) channels for 4-layer sheet in T (a), $H1$ (b), $H2$ (c) and $H3$ (d) phases.

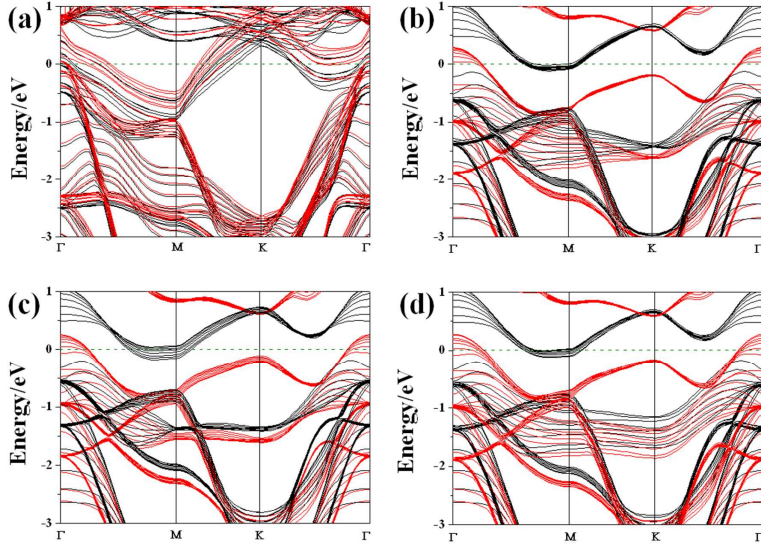


Figure S5. Band structures of spin-up (black) and spin-down (red) channels for 8-layer sheet in T (a), $H1$ (b), $H2$ (c) and $H3$ (d) phases.

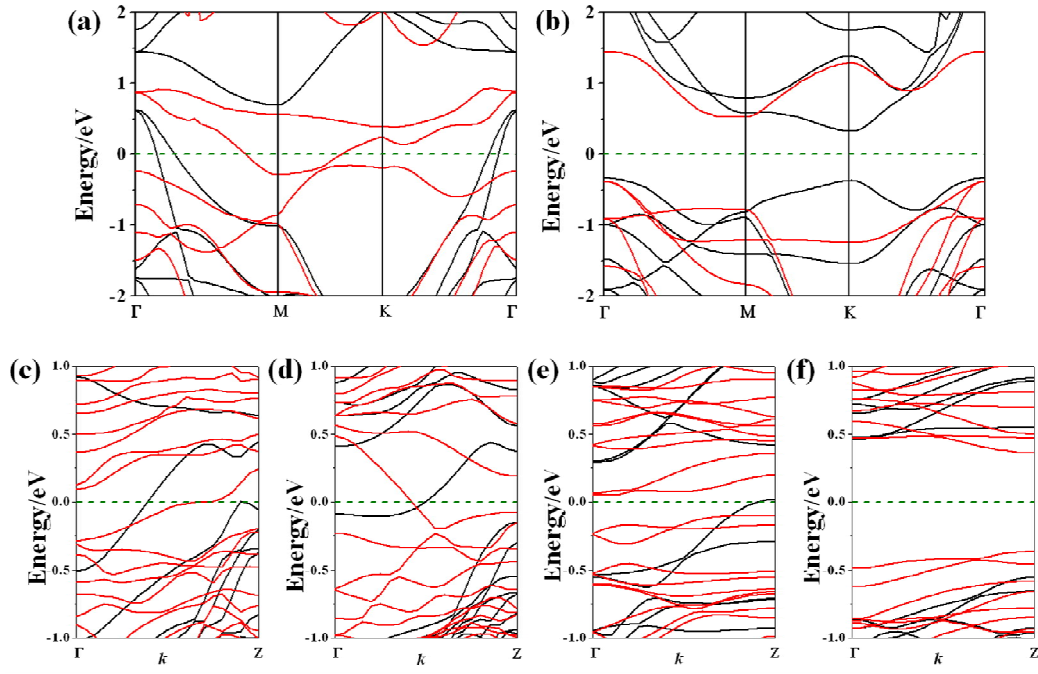


Figure S6. Band structures of spin-up (black) and spin-down (red) channels for VSe_2 monolayer in T (a) and H (b) phases, bare z -5 T -NR (c) and z -6 H -NR (d), hydrogenated a -8 T -NR (e) and a -7 H -NR (f).

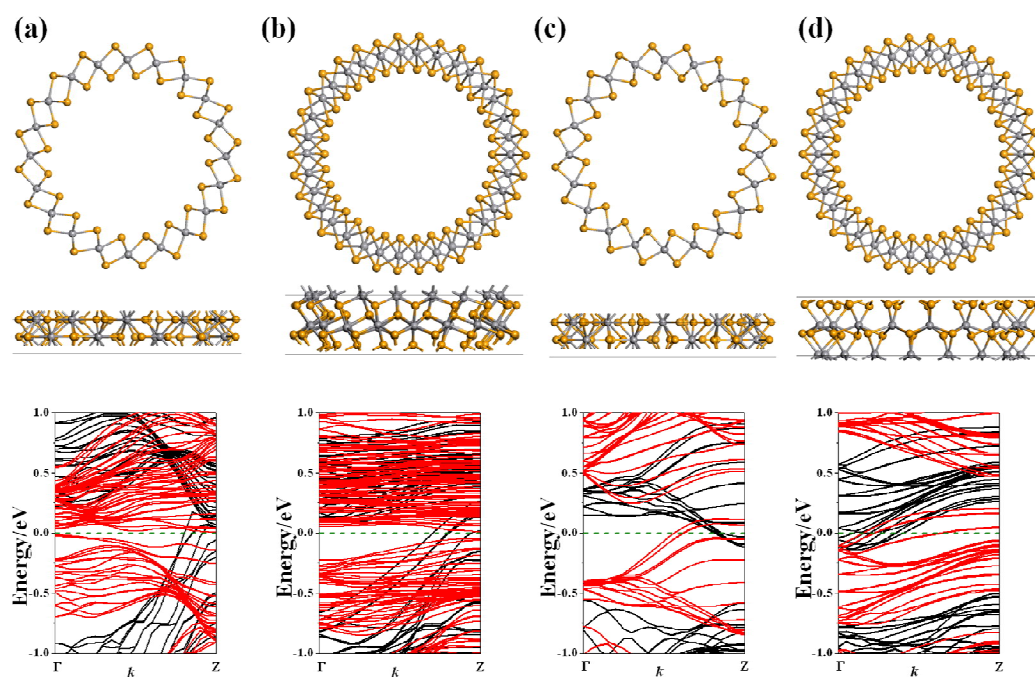


Figure S7. Top and side views of (10,10) and (17,0) nanotubes in the *T* (a, b) and *H* phases (c, d), and the corresponding band structures. The grey and khaki atoms represent V and Se atoms, respectively. The spin-polarized band structures with both spin-up and spin-down channel are presented in black and red, respectively. The Fermi energy is denoted by a green dashed line.