Supporting Information

Exfoliated Monolayer GeI_2: Theoretical Prediction of a Wide-Band Gap Semiconductor with Tunable Half-Metallic Ferromagnetism

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I. The structural and electronic properties of bulk GeI_2

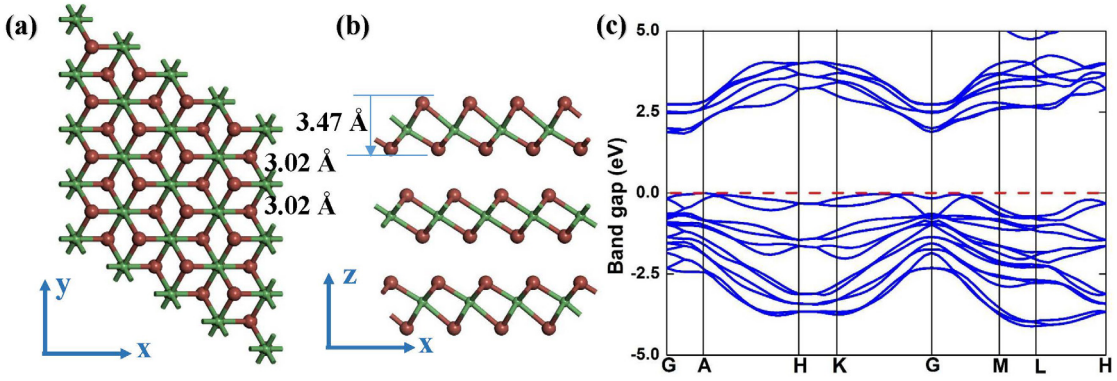

Figure S1. The optimized bulk GeI_2 structure in top a) and side b) views. c) The band structures of bulk GeI_2 with HSE+SOC functional.

II. Molecular dynamics simulations of monolayer GeI_2

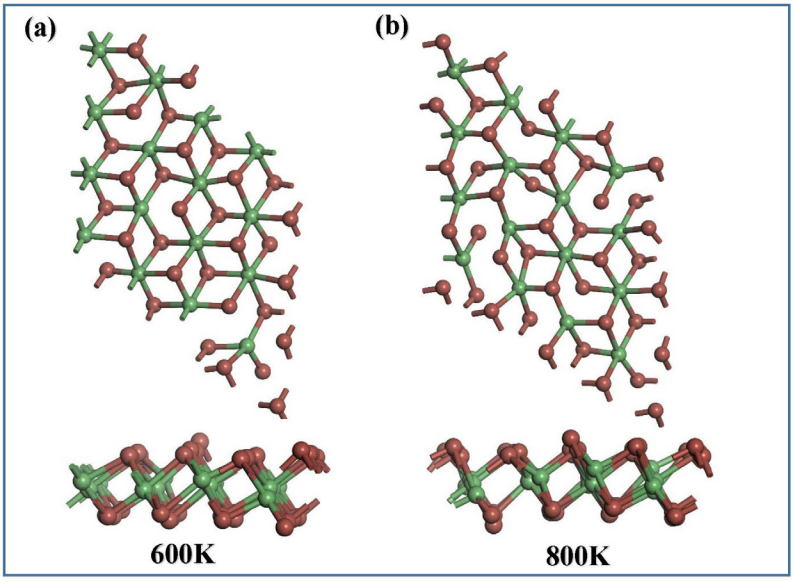

Figure S2. The snapshots of top and side views of the final atomic configurations at a) 600 and b) 800 K.

III. In-plane stiffness (C_{2D})

C_{2D} is defined as $(E - E_0)/S_0 = C_{2D}(\Delta l/l_0)^2/2$, where E_0 is the total energy of cell without dilation and S_0 is the equilibrium lattice area. The strain step of this data calculation is 0.5%. The relationship between $\Delta E = E - E_0$ and $\Delta l/l_0$ is shown in Figure S3. According to our computation, the in-plane stiffness C_{2D} of monolayer GeI_2 is 30.79 (30.80) N/m along the *a* (*b*) direction.

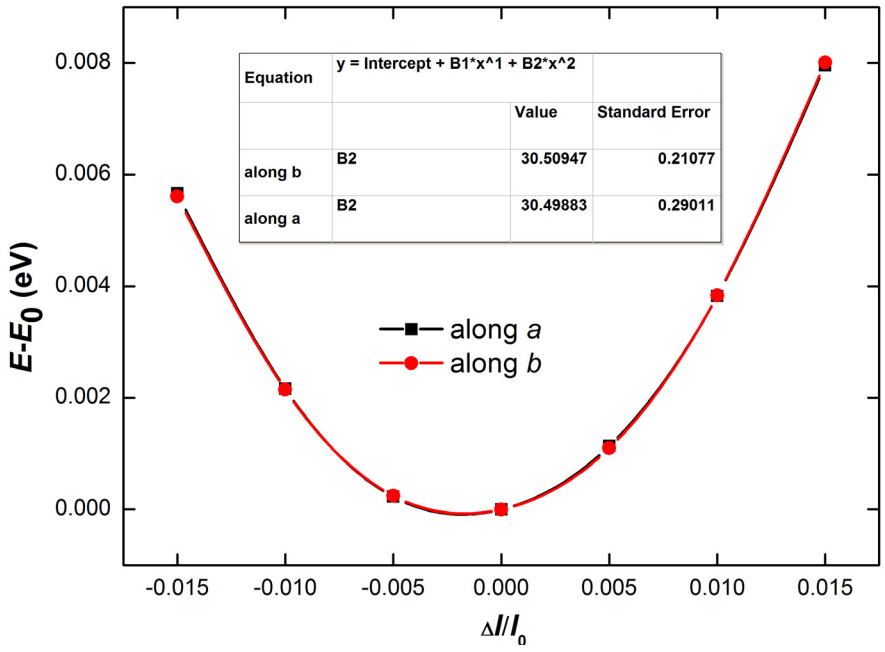

Figure S3. The total energy shift E-E_0 as a function of lattice deformation $\Delta l / l_0$ along a and b directions of monolayer GeI_2.

IV. The electronic properties of multi-layered GeI_2 systems

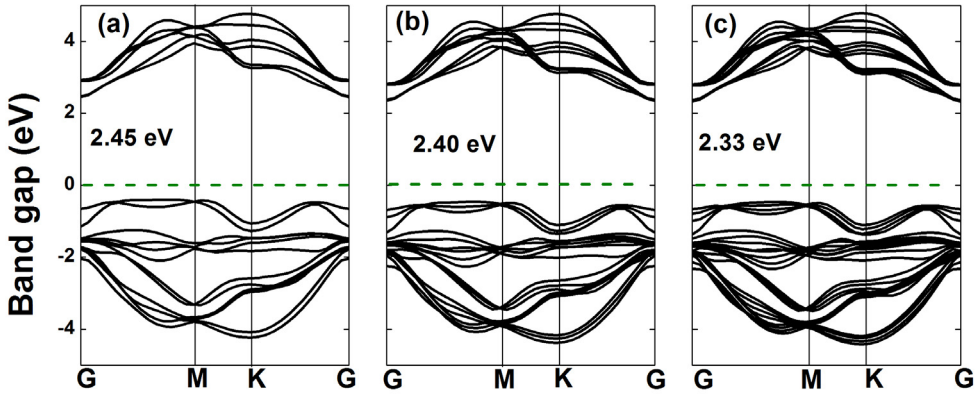

Figure S4. The band structures of (a) bi-, (b) three-, and (c) four-layered GeI_2 systems from HSE06+SOC functional.

V. Carrier mobility

According to the deformation potential#(DP) theory, the carrier mobility of 2D materials can be estimated by the following expression:

$$\mu = \frac{e\hbar^3 C_{2D}}{K_B T m_i^* m_d (E_1^i)^2}$$

where $\hbar$, K_B, T, and m^* are the reduced Planck constant, Boltzmann constant,

temperature (300 K), and effective mass, respectively. m_i^*(i = *h* for holes, i = *e* for electrons) is based on the effective mass approximation $m^* = \hbar^2(\partial^2 E/\partial K^2)^{-1}$. The effective masses along the *a* (*b*) direction are 0.25 (0.28) m_0 for electron and 0.72 (0.44) m_0 for hole. The average effective mass m_d is determined by $m_d = \sqrt{m_x^* m_y^*}$. The deformation potential constant E_1 is defined by $\frac{\Delta V}{\Delta l / l_0}$, which represents the energy of CBM for electrons and the VBM for holes along the transport (*a* and *b*) direction. ΔV is the energy change (conduction band or valence band) under certain strain. The l_0 is the lattice constant along the transport direction, and Δl is the deformation of l_0. The relationship between E_1 and dilation $\Delta l/l_0$ is shown in Figure S5. The deformation potentials (E_1) are -6.31 (-6.37) eV for electron and -3.60 (-5.49) eV for hole along the *a* (*b*) direction, respectively.

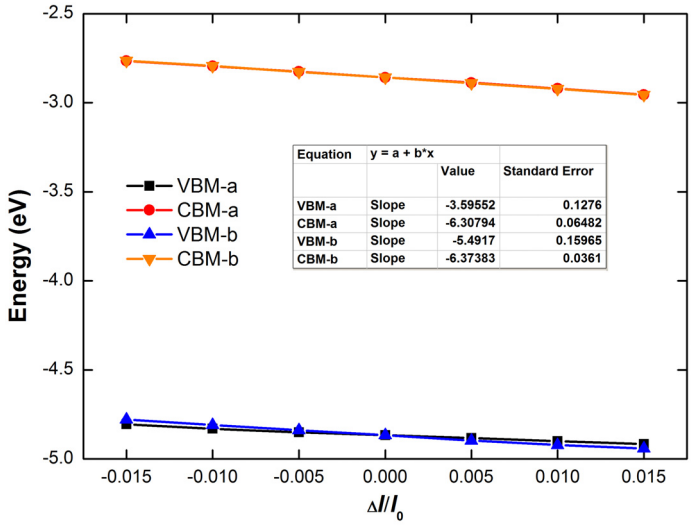

Figure S5. The relationship between the energy shift of the band edge position with the dilation $\Delta l / l_0$.

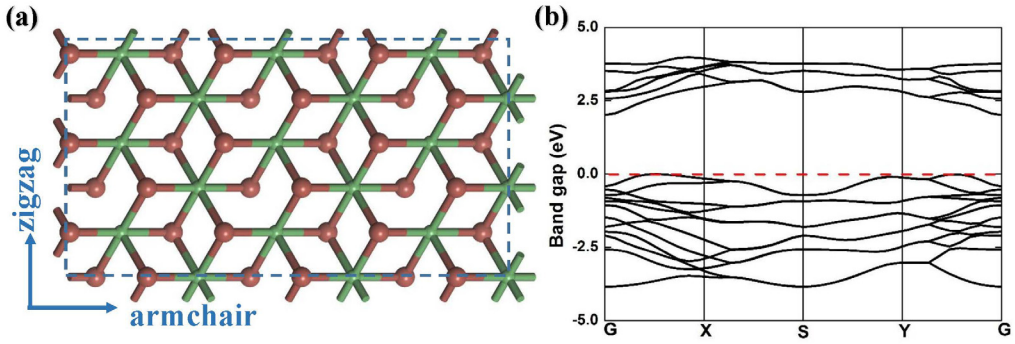

Figure S6. (a) The structure of monolayer GeI_2 with the rectangular supercell. (b) The band structure of its rectangular supercell.

VI. The deformation charge density, phonon spectrum, magnetism orders, and band structure of GeI_2H

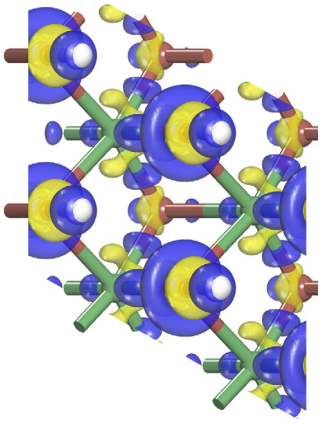

Figure S7. The deformation charge density plots of GeI_2H. The iso-surface value is 0.015 eÅ^3. The yellow and blue areas represent the electron depletion and accumulation regions, respectively.

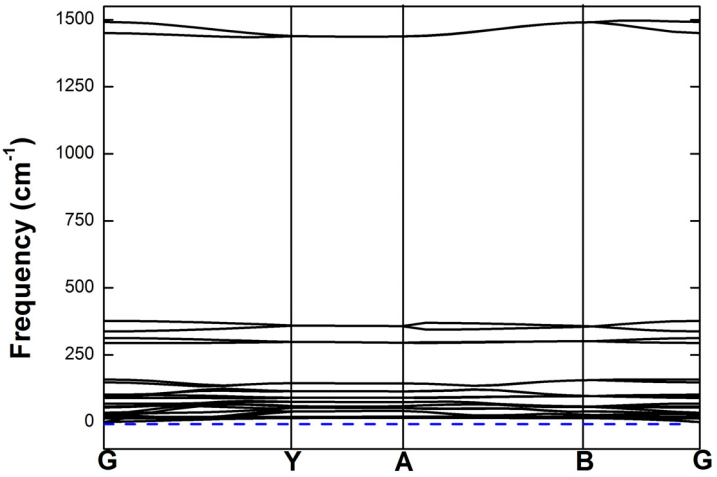

Figure S8. The phonon spectrum of hydrogenated GeI_2.

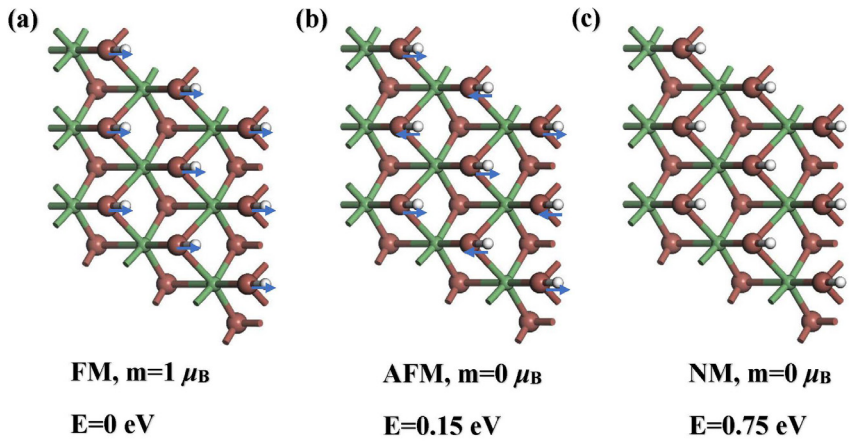

Figure S9. Three magnetic configurations of hydrogenated GeI_2, including the magnetic moments and their relative energies with respect to the FM state for a) FM, b) AFM, and c) NM states.

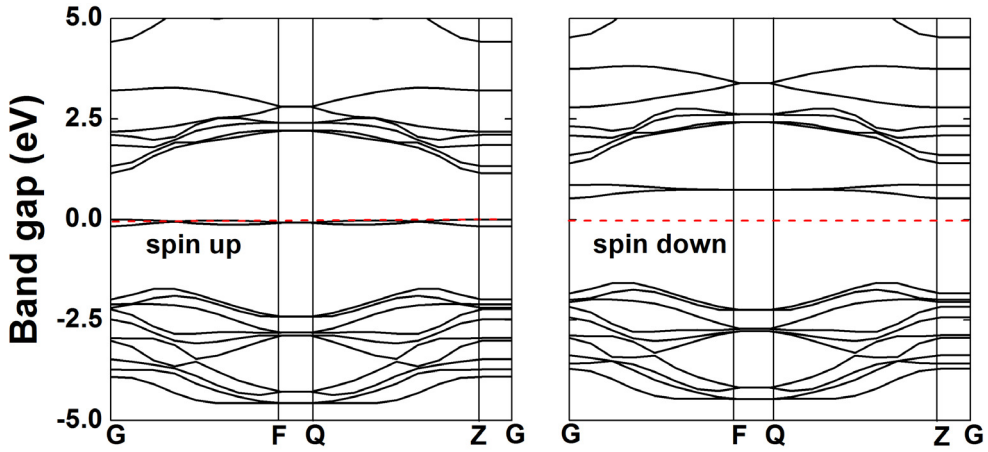

Figure S10. The spin-up and the spin-down band structures of GeI_2H.