Supporting Information

High Carrier Mobility and Pronounced Light Absorption in Methyl-Terminated Germanene: Insights from First-Principles Computations

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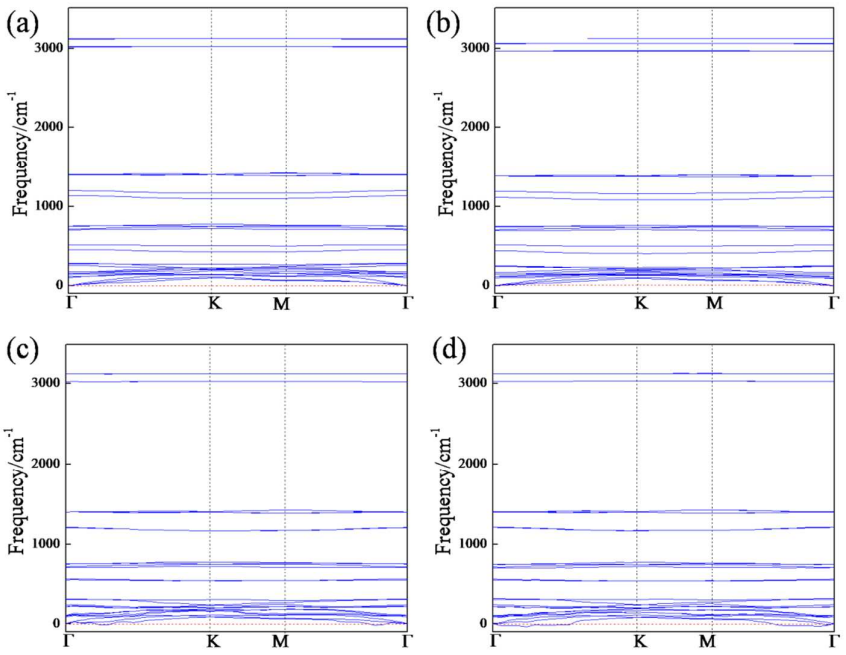

Figure S1. Phonon spectrum of $GeCH_3$ in a hexagonal primitive cell under no strain (a), 5% biaxial tensile strain (b), 2% biaxial compressive strain (c) and 3% biaxial compressive strain (d).

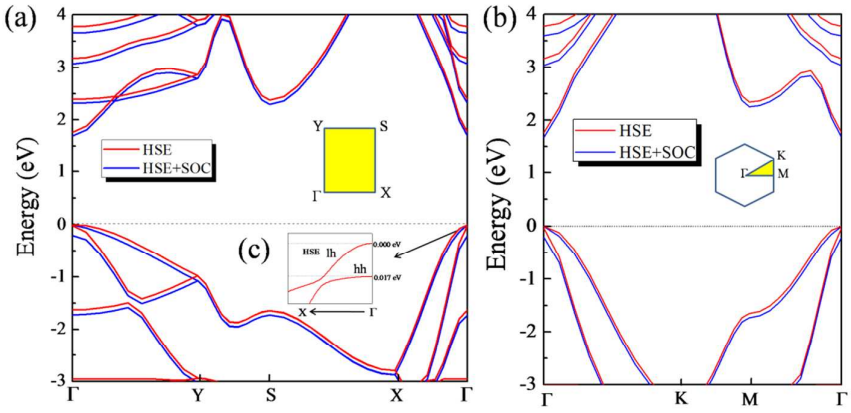

Figure S2. Band structure of $GeCH_3$ in a tetragonal cell (a) and a hexagonal primitive cell (b) at the HSE level calculated with and without SOC; Enlarged valence bands structure near the Γ point along the armchair direction (c), the heavy and light valence bands corresponding to the heavy hole (hh) and light hole (lh) are labeled inside.

Table S1. Effective masses for the heavy holes (hh), light holes (lh) and electrons (e) along the armchair and zigzag directions in the tetragonal and hexagonal cells, calculated with and without SOC effect.

Cell	m*_armchair			m*_zigzag		
	hh	lh	e	hh	lh	e
Tetra_HSE	0.14	2.35×10^{-2}	2.62×10^{-2}	1.36	0.21	0.24
Tetra_HSE+SOC	3.67×10^{-2}	3.67×10^{-2}	2.60×10^{-2}	0.30	0.30	0.19
Hexa_HSE+SOC	4.11×10^{-2}	4.25×10^{-2}	1.91×10^{-2}	0.23	0.24	0.19

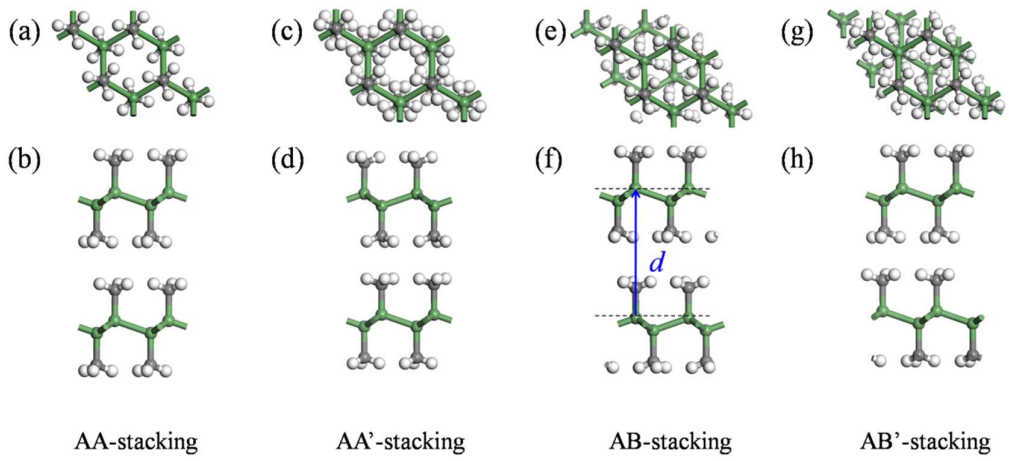

Figure S3. Atomic configurations of $GeCH_3$ bilayer (2×2 supercell of the hexagonal lattice) in different stacking ways, including the AA stacking (a and b), AA'-stacking (c and d), AB-stacking (e and f) and AB'-stacking (g and h), from the top and side views, respectively. The bilayer configurations were calculated in a hexagonal primitive cell instead of the orthogonal supercell to reduce the calculation burden. The interlayer distance (*d*) is labeled in (f).

Table S2. Interlayer distance ($d_{Ge\text{-}Ge}$) and binding energy of $GeCH_3$ bilayer (E_{bind}) in different stacking configurations.

Stacking order	AA	AA'	AB	AB'
$d_{Ge\text{-}Ge}$ (nm)	0.79	0.74	0.79	0.75
E_{bind}(J/m^2)	0.128	0.10	0.131	0.03

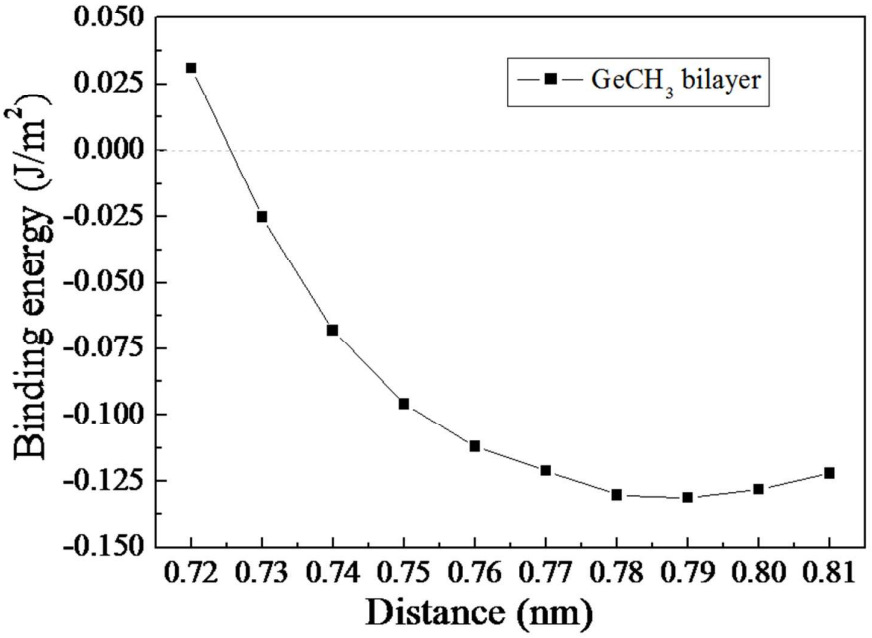

Figure S4. Binding energy as a function of the interlayer distance *d* in $GeCH_3$ bilayer in AB stacking.

As shown in Figure S4, the binding energy of $GeCH_3$ bilayer is negative when the interlayer distance (is denoted in Figure S3f) is larger than 0.72 nm. The equilibrium interlayer distance is 0.79 nm, and at this distance the $GeCH_3$ bilayer shows the lowest binding energy of 0.13 J/m^2.

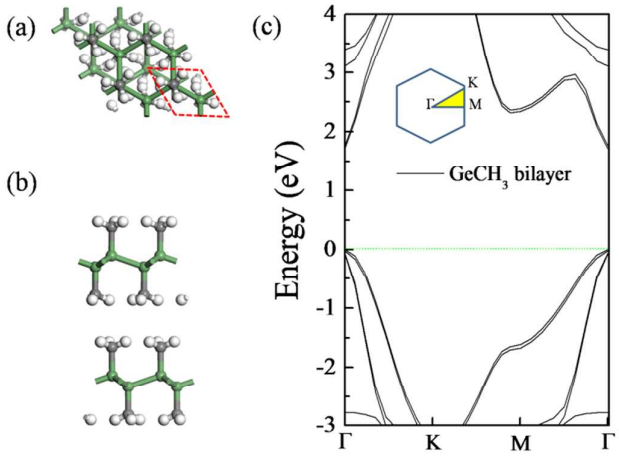

Figure S5. $GeCH_3$ bilayer (2×2×1 hexagonal supercell) in the most stable configuration (AB-stacking) from the top view (a) and side view (b) and its corresponding band structure (c).