Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.


# Supporting Information

## Novel bonding patterns and optoelectronic properties of the two-dimensional $Si_xC_y$ monolayers

Dong Fan,[a] Shaohua Lu,[a*] Yundong Guo[b] and Xiaojun Hu[a*]

a. *College of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou 310014, China*

*b. School of Engineering and Technology, Neijiang Normal University, Neijiang, 641000, China.*

*E-mail: lsh@zjut.edu.cn; huxj@zjut.edu.cn

**Table S1** | Calculated lattice constants, bond lengths, and cohesive energy of the predicted low-energy 2D SiC monolayers at GGA (PBE) level.

| | Lattice constant Å | Bond lengths Å | Property | Cohesive energy eV/atom |
|---|---|---|---|---|
| Graphene | a=b=2.46 | C-C 1.42 | DF | 7.94 |
| L-$SiC_4$ | a= 6.25 b=2.45 buckling 0.95 | Si-Si 2.45 Si-C 1.89 C-C 1.42 | DF | 6.91 |
| *δ*-silagraphyne | a=6.26 b=4.10 | Si-C 1.77 & 1.85 C-C 1.34 & 1.26 | SM | 6.55 |
| *g*-$SiC_2$ | a=b=5.02 | Si-C 1.80 C-C 1.45 | DB | 6.46[a] |
| L-$SiC_2$ | a= 2.48 b=8.58 buckling 0.63 | Si-Si 2.48 Si-C 1.97 C-C 1.42 | IDB | 6.20 |
| *α*-silagraphyne | a=b=8.25 | Si-C 1.76 C-C 1.24 | DF | 6.09 |
| *pt*-SiC | a=2.86, b=3.88 | Si-C 1.92 C-C 1.33 | M | 6.04[b] |
| *ß*-silagraphyne | a=b=11.78 | Si-Si 2.20 Si-C1.78 C-C1.24 | DF | 5.84 |
| O-$Si_2C_3$ | a=6.79 b=3.06 | Si-C 1.75 C-C 1.33 | SM | 5.81 |
| *t*-SiC | a=3.67 b=3.21 buckling 1.42 | Si-C 1.92 & 1.87 | DB | 5.55 |
| *γ*-silagraphyne | a=b=9.27 buckling 0.48 | Si-Si 2.23 Si-C 1.79 C-C 1.23 | DS | 5.36 |
| L-silagraphyne | a=5.03 b=7.24 buckling 0.65 | Si-Si 2.28 Si-C 1.86 & 1.73 C-C 1.35 &1.25 | SM | 5.35 |
| *t*-$Si_2C$ | a=2.79 b=9.09 | Si-Si 2.44 Si-C 1.89 | SM | 4.85 |
| T-silagraphyne | a=3.17 b=10.58 | Si-C 1.82 C-C 1.40 & 1.24 | IDB | 4.50 |
| Silicene | a=b=3.87 | Si-Si 2.28 | DF | 3.93 |

* DF: Dirac Fermions. DB: Direct band gap. IDB: Indirect band gap.
* SM: Semi-metallic. M: metallic
* a: Ref.[17] b:Ref.[16]

**Table S2** | Calculated elastic constants, in-plane stiffness and Poisson's coefficients for predicted low-energy 2D SiC monolayers.[*1]

| | $C_{11}$/ GPa | $C_{22}$/ GPa | $C_{12}$/ GPa | $C_{66}$/ GPa | In-plane stiffness/ GPa·nm | Poisson's ratio |
|---|---|---|---|---|---|---|
| **Graphene** | 358 | 358 | 65 | 147 | 346.73 | 0.18 |
| **Silicene** | 69 | 69 | 22 | 23 | 61.88 | 0.32 |
| ***t*-SiC** | 152 | 123 | 22 | 95 | 105(**a**)[*2]119(**b**)[*2] | 0.18(**a**)0.14(**b**) |
| ***t*-$Si_2C$** | 153 | 173 | 24 | 60 | 150(**a**)169(**b**) | 0.14(**a**)0.16(**b**) |
| **L-$SiC_4$** | 121 | 292 | 12 | 67 | 121(**a**)291(**b**) | 0.04(**a**)0.10(**b**) |
| **L-$SiC_2$** | 254 | 139 | 7 | 49 | 253(**a**)139(**b**) | 0.05(**a**)0.03(**b**) |
| **O-$Si_2C_3$** | 219 | 125 | 46 | 26 | 202(**a**)115(**b**) | 0.37(**a**)0.21(**b**) |

[*1]: Calculated by equations in Reference: Zhang, S., et al. PNAS, 112.8(2015):2372-7. where $C_{11}$, $C_{12}$, $C_{21}$ and $C_{22}$ represent elastic contacts (GPa) calculated by VASP.

[*2]: a direction and b direction of 2D BC compounds.

**Table S3** | Calculated elastic constants, in-plane stiffness and Poisson's coefficients for predicted low-energy 2D SiC monolayers.

| | $C_{11}$/ GPa | $C_{22}$/ GPa | $C_{12}$/ GPa | $C_{66}$/ GPa | In-plane stiffness/ GPa·nm | Poisson's ratio |
|---|---|---|---|---|---|---|
| **α-graphyne** | 96 | 96 | 81 | 7 | 27.03(21.00[ref1]) | 0.85(0.88[ref1]) |
| **ß-graphyne** | 130 | 130 | 88 | 21 | 70.80(73.07[ref2]) | 0.68(0.67[ref2]) |
| **γ-graphyne** | 196 | 196 | 83 | 56 | 160.25(162.1[ref3]) | 0.43(0.429[ref3]) |
| **α-silagraphyne** | 61 | 61 | 55 | 3 | 17.00 | 0.89 |
| **ß-silagraphyne** | 68 | 68 | 53 | 8 | 26.36 | 0.78 |
| **γ-silagraphyne** | 76 | 76 | 47 | 15 | 47.55 | 0.61 |
| **δ-silagraphyne** | 253 | 105 | 32 | 7 | 243(**a**)101(**b**) | 0.30(**a**)0.13(**b**) |
| **T-silagraphyne** | 95 | 242 | 35 | 9 | 90(**a**)229(**b**) | 0.14(**a**)0.37(**b**) |
| **L-silagraphyne** | 39 | 87 | 9 | 1 | 38(**a**)84(**b**) | 0.10(**a**)0.23(**b**) |


Ref[1]: Journal of Physical Chemistry C 117.5 (2013): 2175-2182.

Ref[2]:Carbon 96 (2016): 879-887.

Ref[3]: Physical Chemistry Chemical Physics 14.38 (2012): 13385-13391.

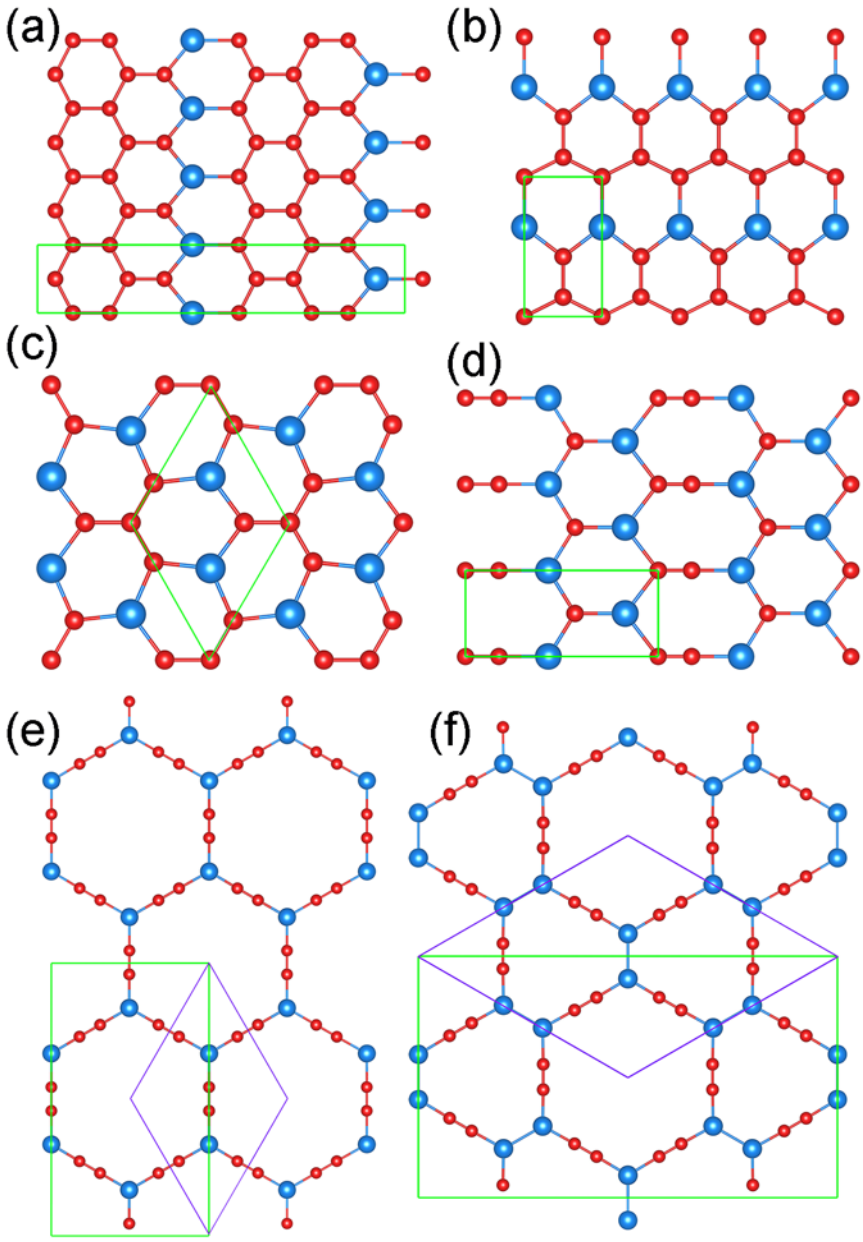


**Fig S1 |** Crystal structure of *h*-$SiC_5$, *h*-$SiC_3$, *h*-$SiC_2$, O-$Si_2C_3$, *α*-silagraphyne and *ß*-silagraphyne from top view. The blue and red balls represent Si and C atoms, respectively. The green lines denote a unit cell.

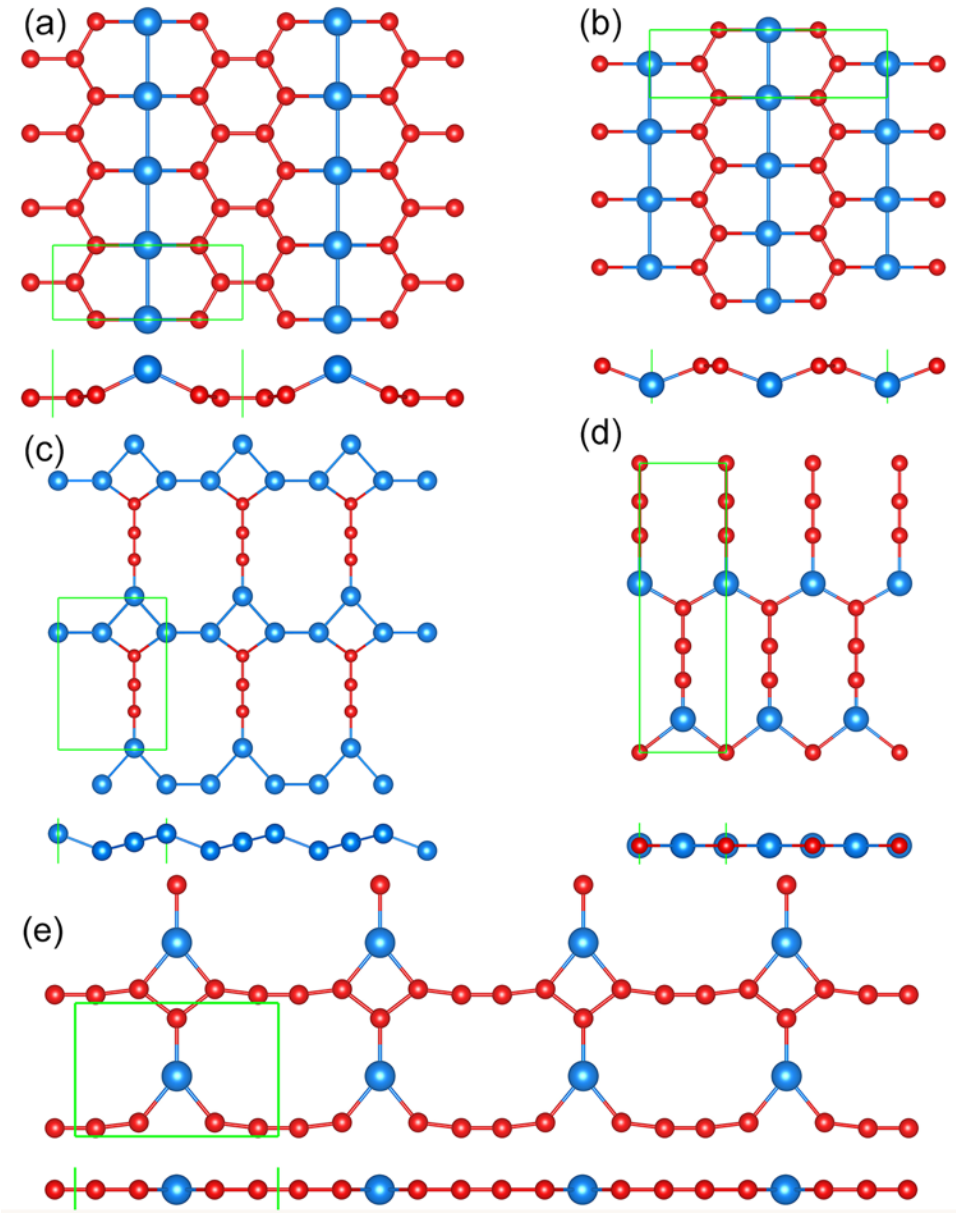


**Fig S2 |** Top (upper) and side (lower) views of geometric structure of (a) L-$SiC_4$, (b)L-$SiC_2$, (c)L-silagraphyne ,(d) T-silagraphyne and (e) $\delta$-silagraphyne. The green lines denote a unit cell. The blue and red balls represent Si and C atoms, respectively.

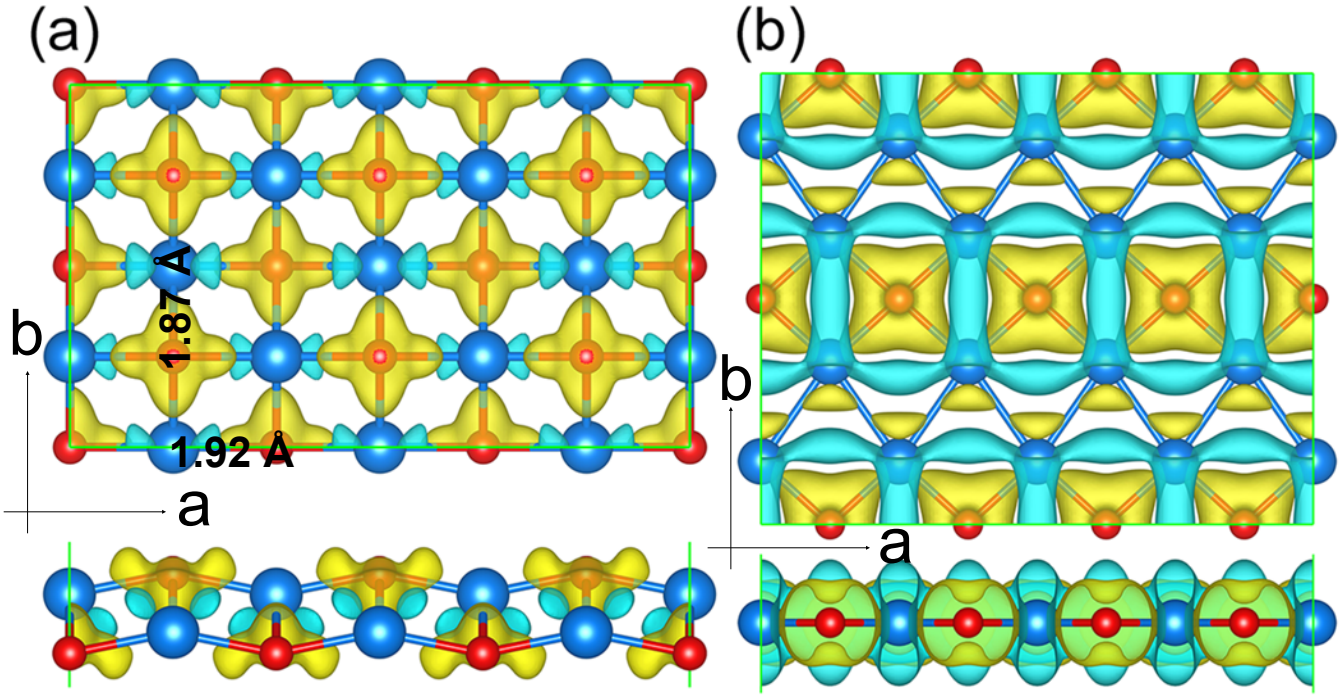


**Fig S3** | Deformation charge density of (a) *t*-SiC and (b) *t*-$Si_2C$ monolayer. Yellow and blue refer to electron accumulation and depletion regions, respectively. The blue and red balls represent Si and C atoms, respectively. The isosurface value is 0.02 and 0.005 e/au. The transferred electrons mainly originated from the Si atoms of *t*-SiC. The Si-C bond length in the buckled structure is 1.92 and 1.87 Å along **a** and **b** direction, due to the different degree of the localization of electronic states on Si-C bonds.

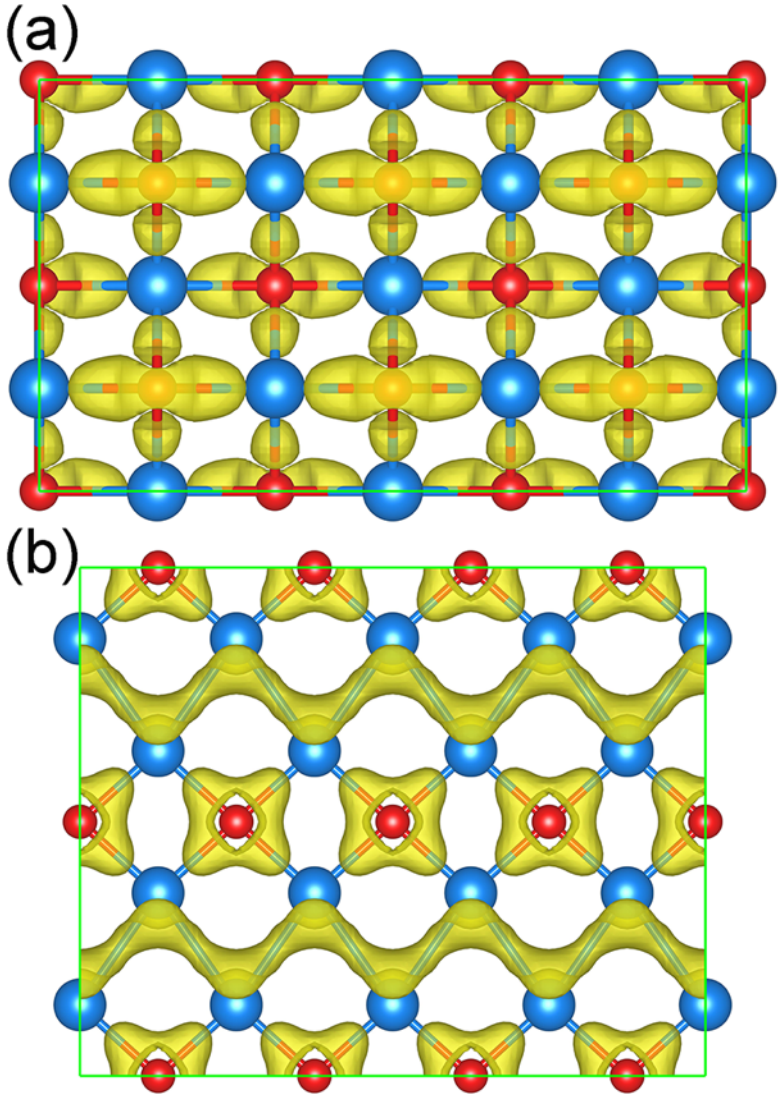


**Fig S4 |** Isosurfaces of electron location function plotted with the value of 0.80 and 0.75 au for *t*-SiC and *t*-$Si_2C$ monolayer, respectively.

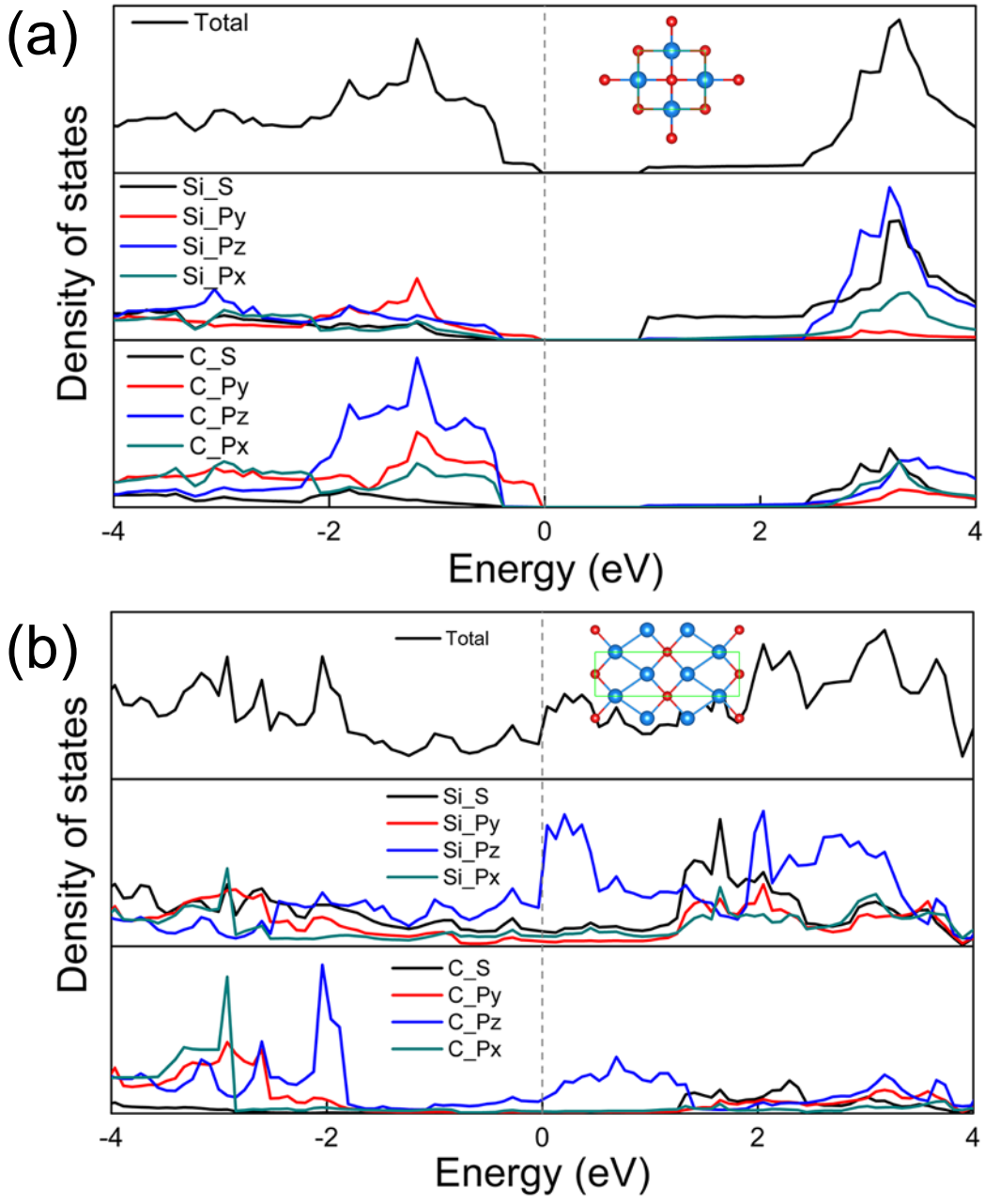


**Fig S5 |** Computed Partial density of states for (a) *t*-SiC and (b) *t*-$Si_2C$ sheets. Calculated at the PBE level of theory and the vertical dashed lines denote the Fermi level.

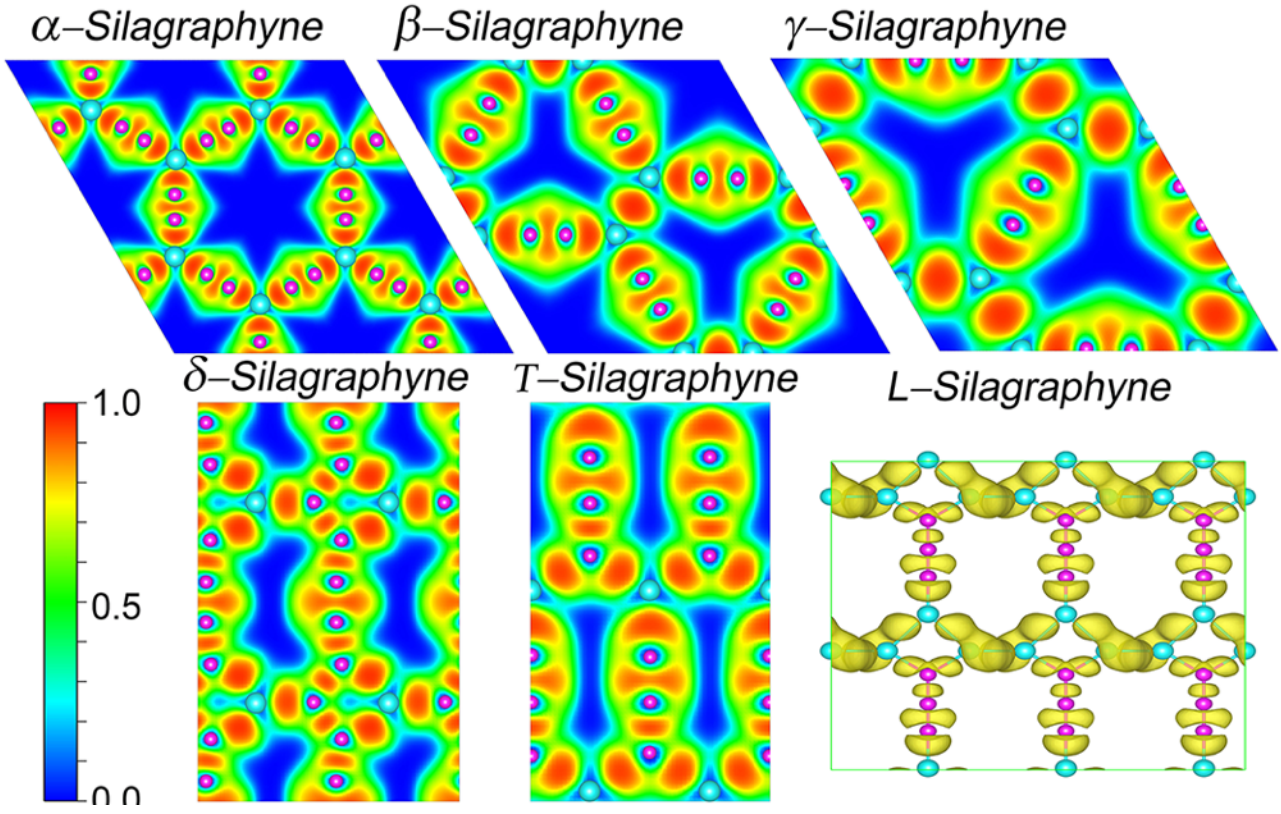


**Fig S6** | Calculated ELF of different silagraphyne. The cyan and pink balls represent Si and C atoms, respectively. For L-silagraphyne, the isosurface value is set as 0.75.

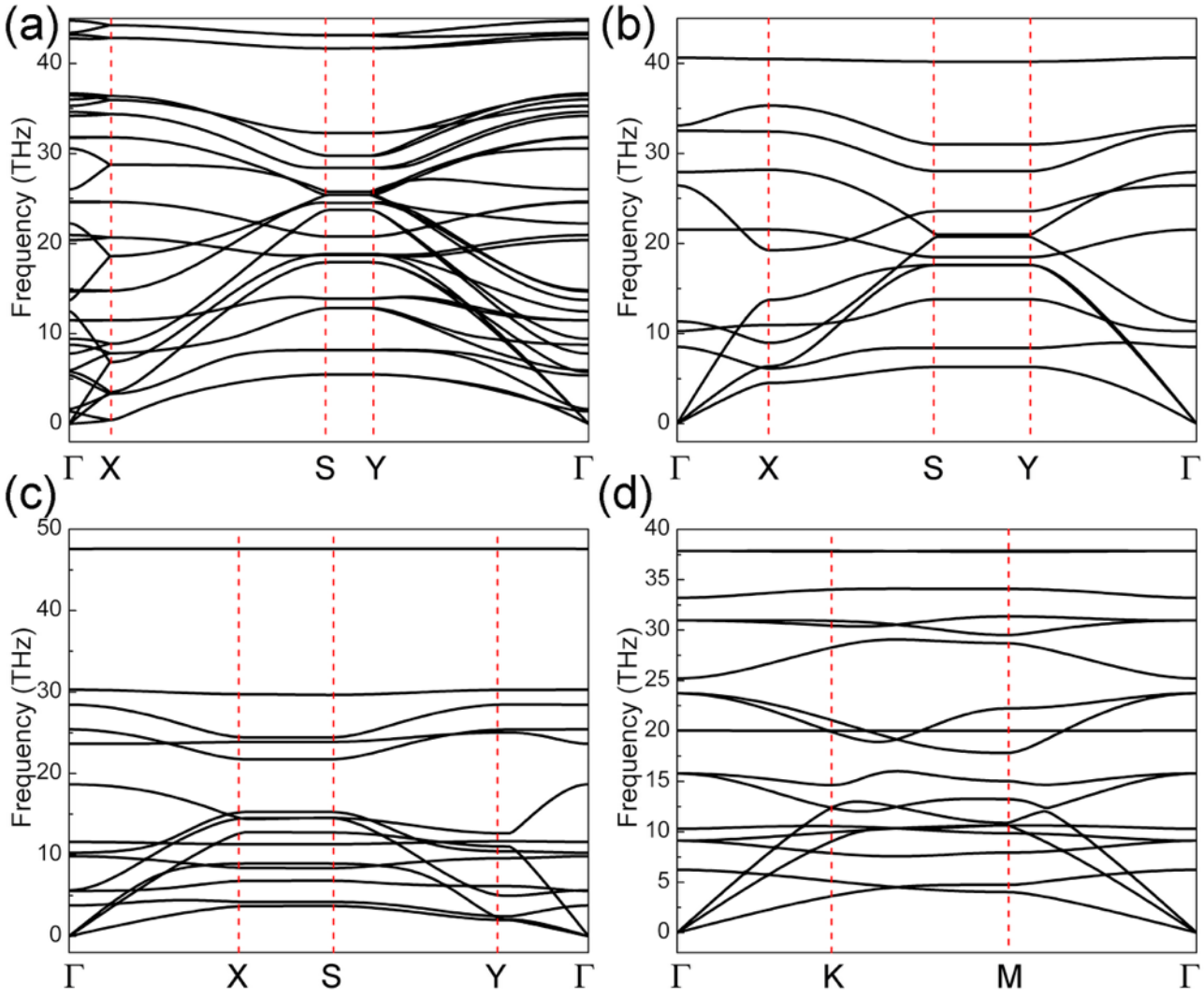


**Fig S7 |** Phonon band structures of (a) *h*-$SiC_5$ (b) *h*-$SiC_3$ (c) *h*-$SiC_2$ and (d) O-$Si_2C_3$ silagraphene calculated by linear response theory. Γ (0, 0, 0), X (1/2, 0, 0), S (1/2, 1/2, 0) , Y (0, 1/2, 0) and K(-1/3, 2/3, 0) represent the symmetry points in the first Brillouin zone in reciprocal space.

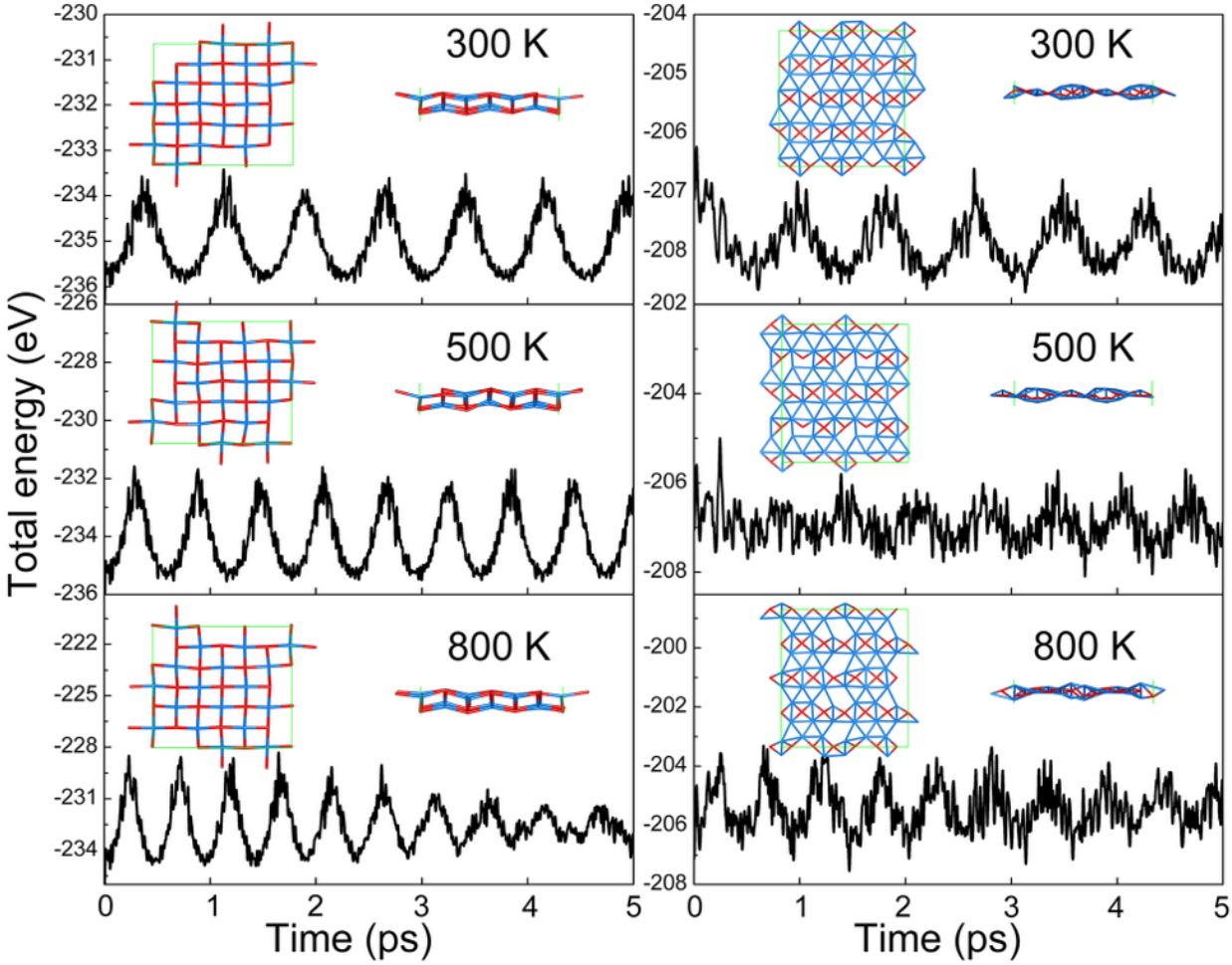


**Fig S8 |** Energy fluctuation of *t*-SiC (left) and *t*-$Si_2C$ (right) with respect to time in AIMD simulations at different temperature. Inset drawing shows the the final geometrical structure of each AIMD simulation (top and side views).

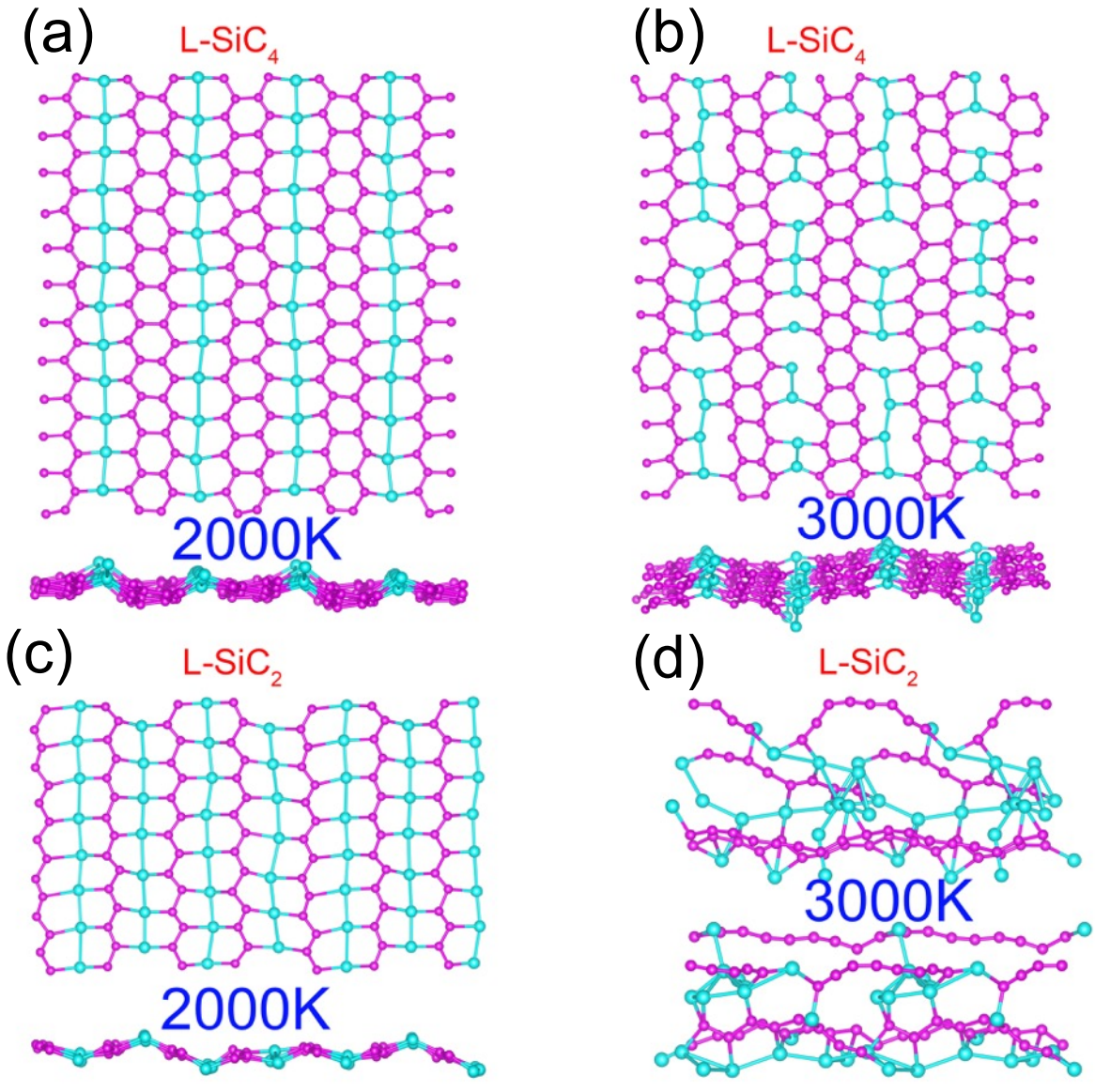


**Fig S9 |** Snapshots of the final frame of each AIMD simulation from 2000 to 3000 K (top and side views).

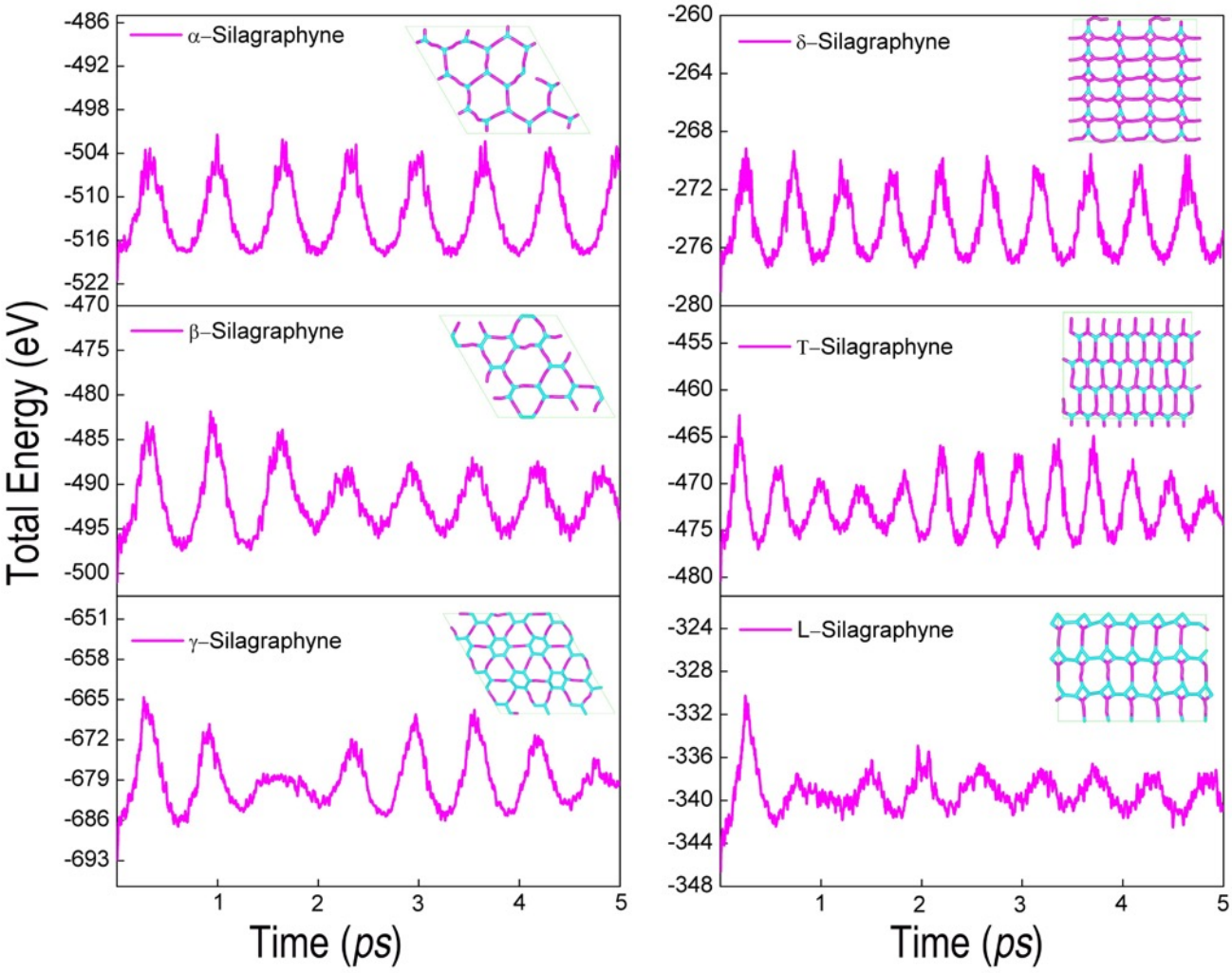


**Fig S10 |** Energy fluctuation of proposed Silagraphyne sheets with respect to time in AIMD simulations at 1000K. Inset drawing shows the the final geometrical structure of each AIMD simulation at 1000K.

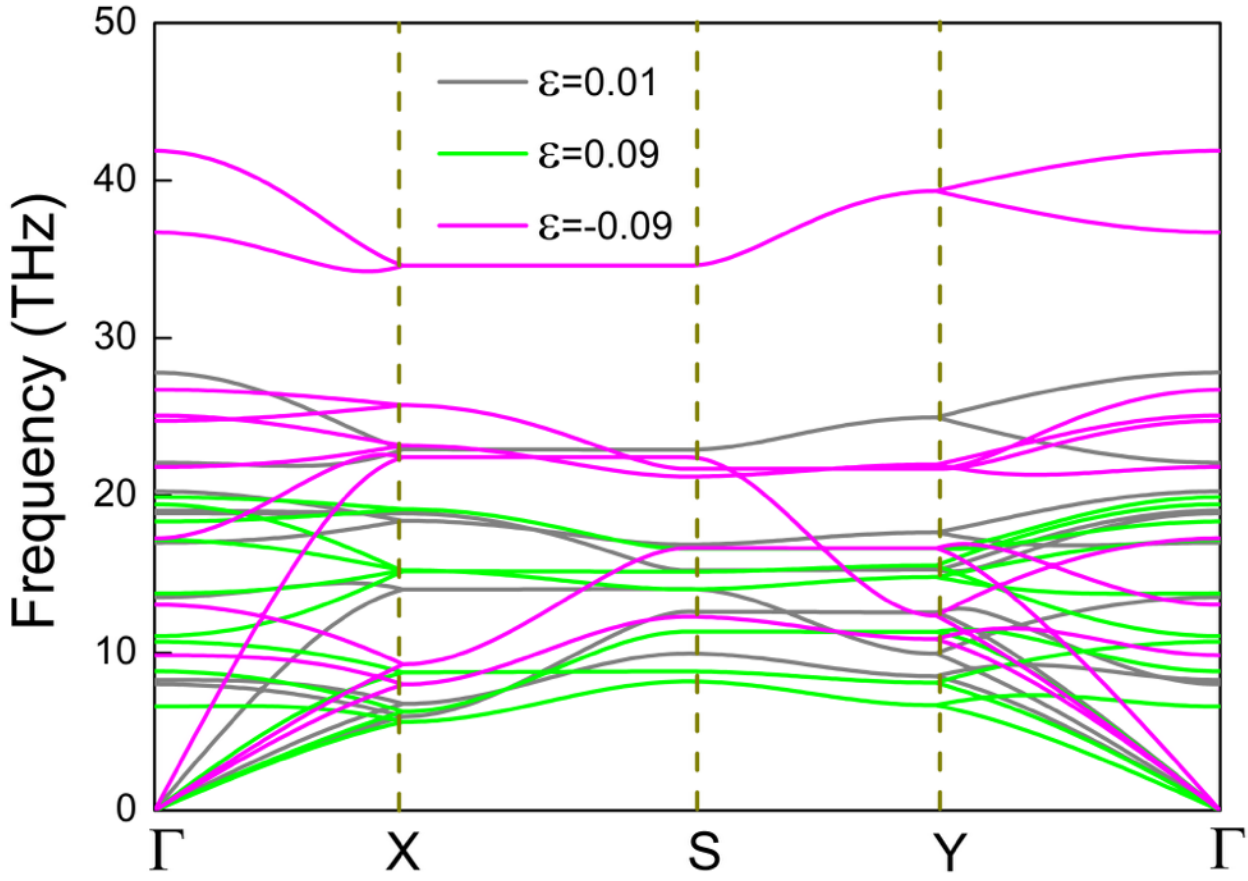


**Fig S11 |** Phonon dispersion curve for biaxial strained *t*-SiC sheet calculated by linear response theory. Γ (0, 0, 0), X (1/2, 0, 0), S (1/2, 1/2, 0) and Y (0, 1/2, 0) represent the symmetry points in the first Brillouin zone in reciprocal space.

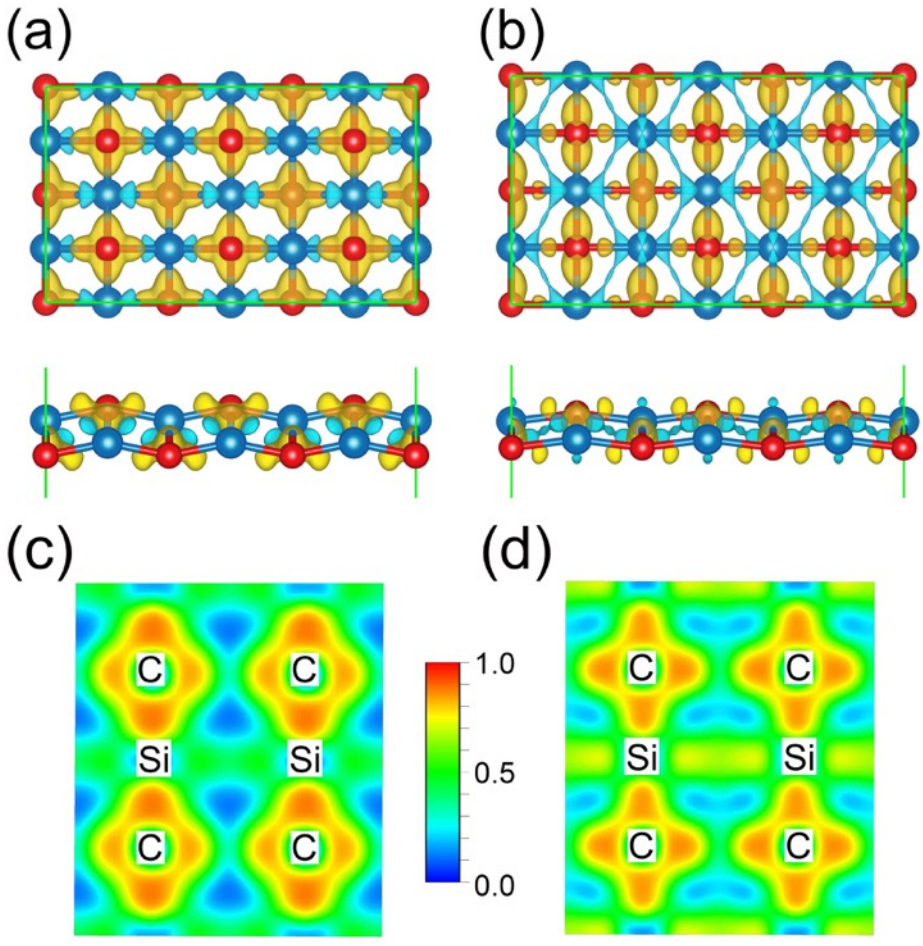


**Fig S12 |** Deformation charge density of (a) 0% strained *t*-SiC and (b) 10% strained *t*-SiC monolayer. Yellow and blue refer to electron accumulation and depletion regions, respectively. The blue and red balls represent Si and C atoms, respectively. The isosurface value is 0.02 and 0.015 e/au. (c, d) Corresponding ELF maps. The cyan and pink balls represent Si and C atoms, respectively. The isosurface value is set as 0.75.

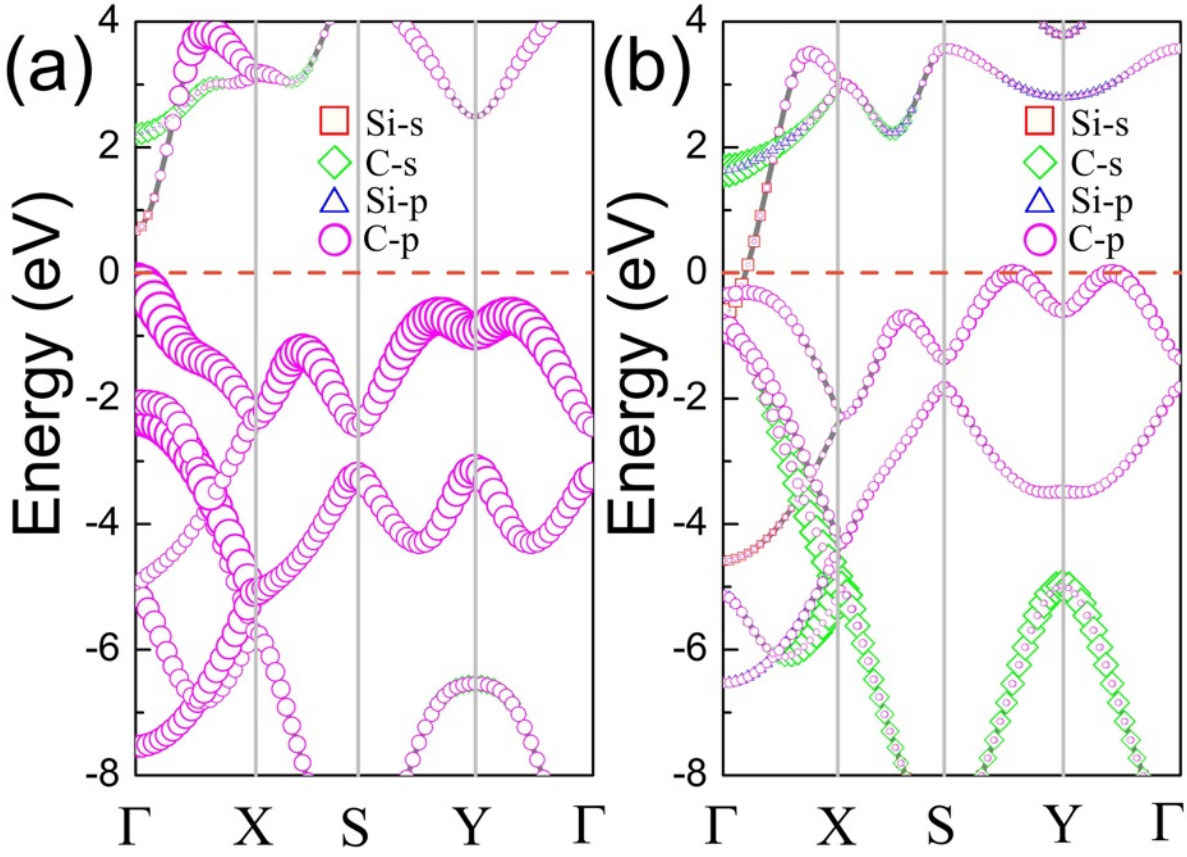


**Fig S13 |** The calculated the orbital-resolved band structures of (a) no-strained *t*-SiC and (b) 10% strained *t*-SiC at PBE level of theory. The dots are the contributions from the corresponding atomic orbital of Si or C atoms. Larger dot means higher contribution while smaller one indicates lower contribution.

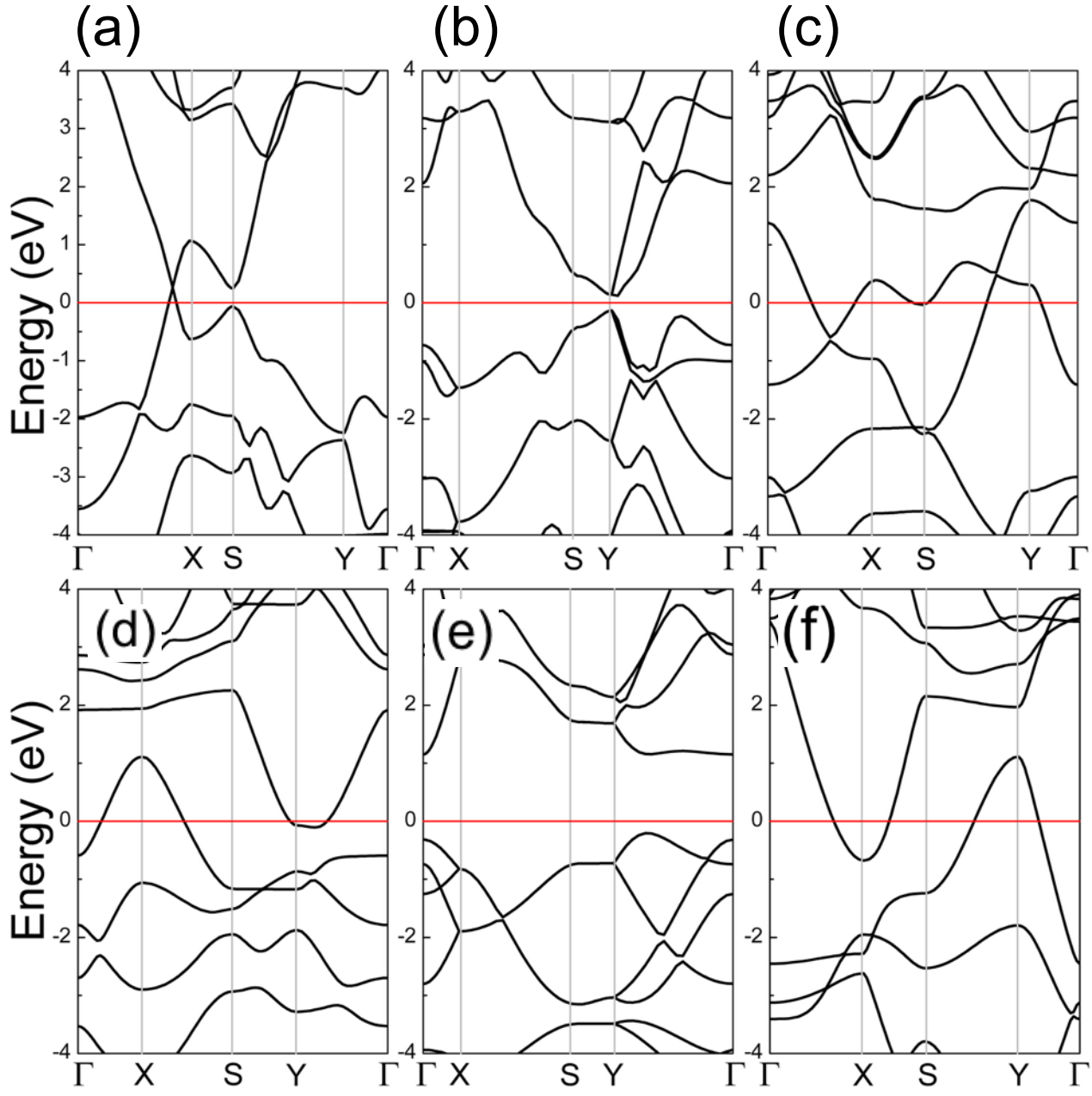


**Fig S14 |** The calculated band structure of (a) L-SiC, (b)L-$SiC_2$ (c) O-$Si_2C_3$, (d) T-silagraphyne, (e) L-silagraphyne and (f) δ-silagraphyne at GGA/PBE level. The Fermi level is set to zero and marked by red lines. The accurate band gap determination provided by the HSE06 method.