Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.


# Supplementary information

# Role of Defects Presented in Graphitic SiC Layer and their Consequences in Exfoliation of Layers – First Principles Approach

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Table S1: FE, various bond distances, and total magnetic moment presented in various defective systems and results are deduced by employing DFT-D2 method.

| System | FE(eV) | Si-C ( Å) | C-C ( Å) | Si-Si ( Å) | M ($\mu_B$) |
|---|---|---|---|---|---|
| $V_{Si}$ | 9.29 | 1.76-1.79 | - | - | 4.1 |
| $V_C$ | 4.40 | 1.79-1.81 | - | - | 2.0 |
| $V_{Si}$+$V_C$ | 6.00 | 1.75-1.86 | 1.62 | 2.44 | 0.0 |
| $Si_C$+$C_{Si}$ | 3.09 | 1.79-1.87 | 1.47 | 2.14 | 0.0 |

Table S2: Doping of Al and P atoms on different sites (refer Fig. S5) of divacancy SiC layer. Binding energy ($\Delta E$) and the spin moment of both cases are provided.

| | Al-doped | | P-doped | |
|---|---|---|---|---|
| Sites | ($\Delta$ E) (eV) | M($\mu_B$) | ($\Delta$ E) (eV) | M($\mu_B$) |
| 1 | -2.66 | 0.98 | -2.41 | 1.04 |
| 2 | -2.24 | 1.00 | -2.65 | 1.03 |
| 3 | -3.21 | 1.01 | -3.03 | 1.02 |
| 4 | -4.30 | 1.01 | -4.76 | 1.06 |

## Binding Energy ($\Delta E$)

$$\Delta E = E_{d+DV} - (E_{DV} + E_d) \tag{S1}$$

where, $E_{d+DV}$ and $E_{DV}$ are the total energies of doped divacancy SiC layer and without doped SiC layer, respectively. $E_d$ represents the chemical potential of Al or P atom.

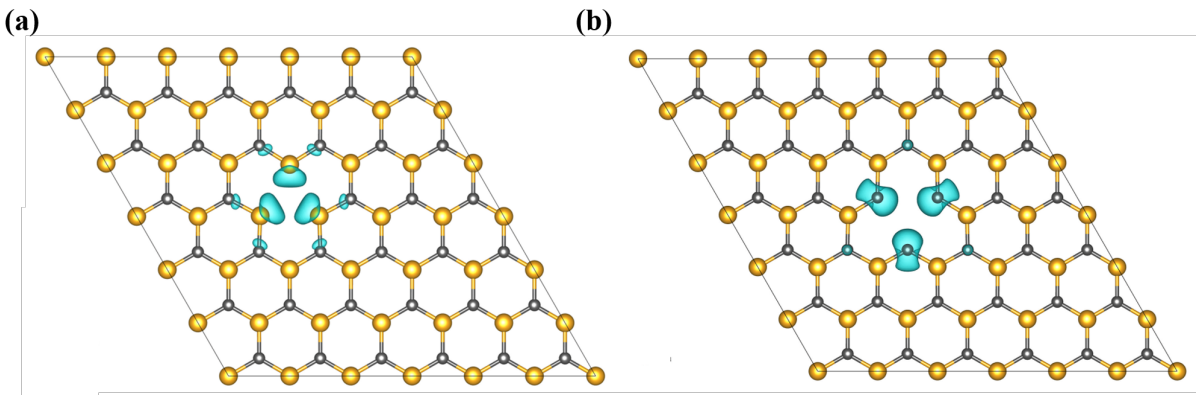


Figure S1: Spin polarized charge density distribution in a) $V_C$ and (b) $V_{Si}$ cases. The isovalue of 0.03 eV/Å$^3$ is used.

## Stacking energy

$$E_S = \frac{E_{total} - \Sigma(E_{Ind})}{n} \tag{S2}$$

where $E_{total}$ and $E_{Ind}$ are the total energies of the entire system and individual layers, respectively. $n$ is the total number of atoms presented in the entire system.

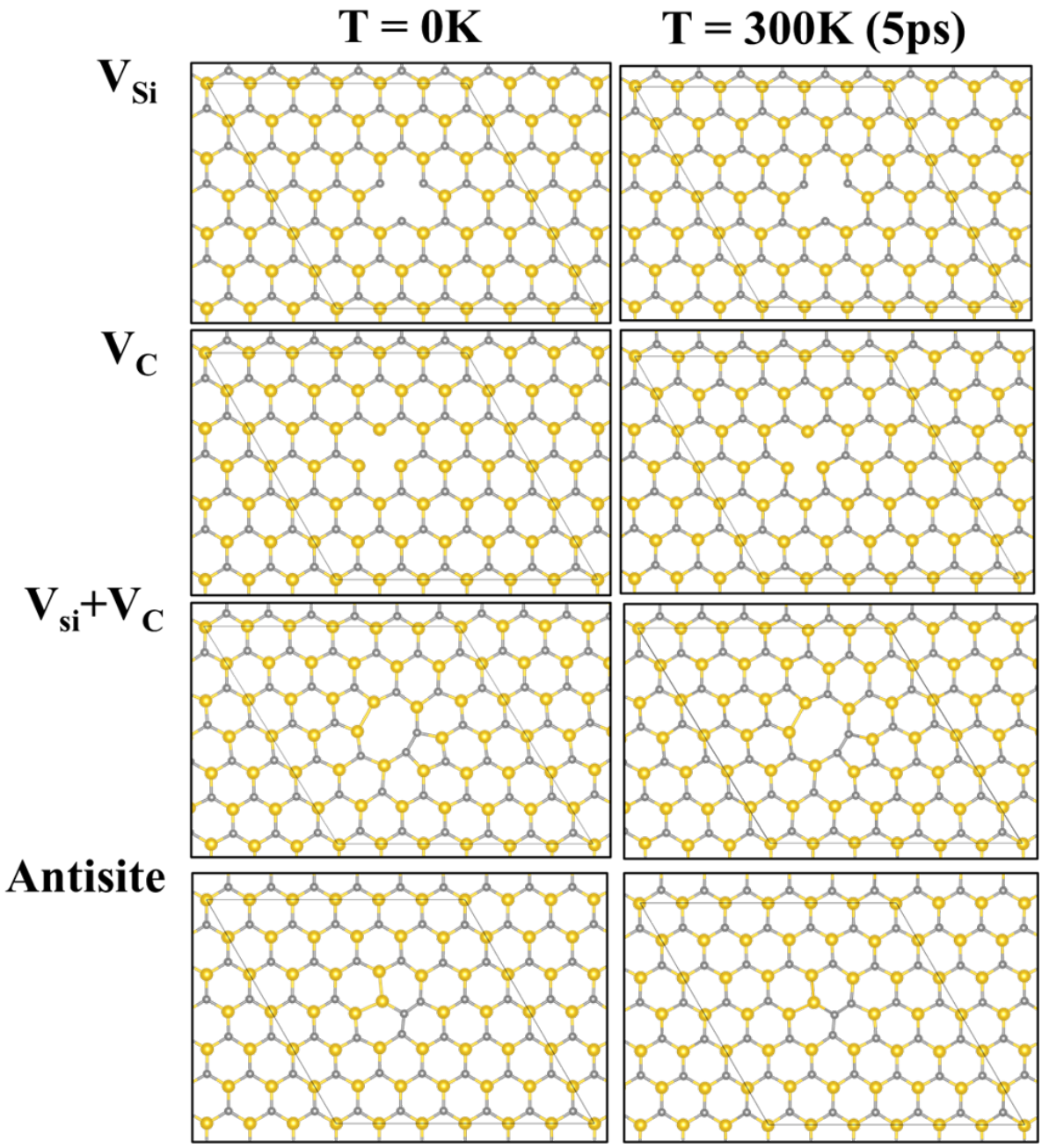


Figure S2: Thermally stable structures of the various defects and antisite cases presented in SiC layer during AIMD simulations at 300 K.

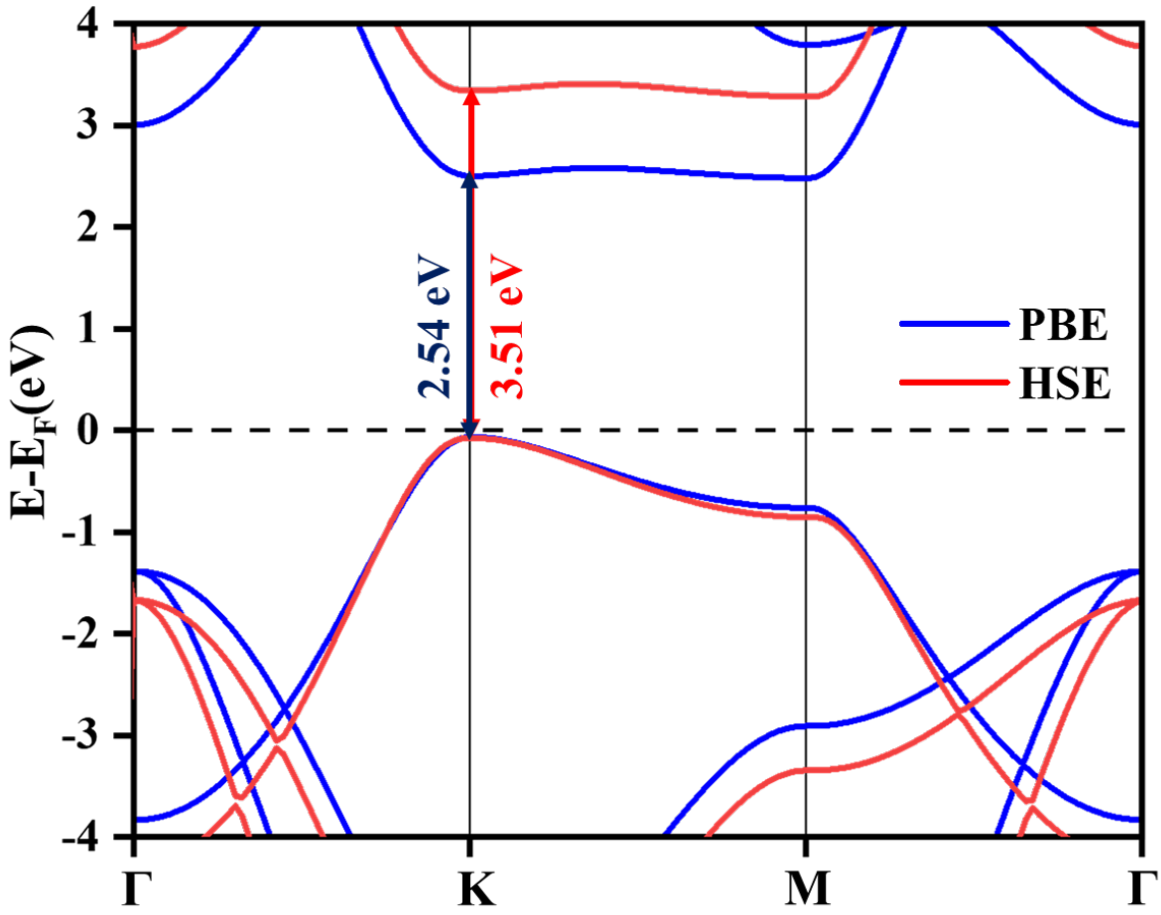


Figure S3: The comparision of BSs of graphitic SiC monolayer that are obtained using PBE and HSE calculations. The curvature of both cases is quite similar, whereas PBE band gap is underestimated by ≈1.0 eV as compared to HSE gap.

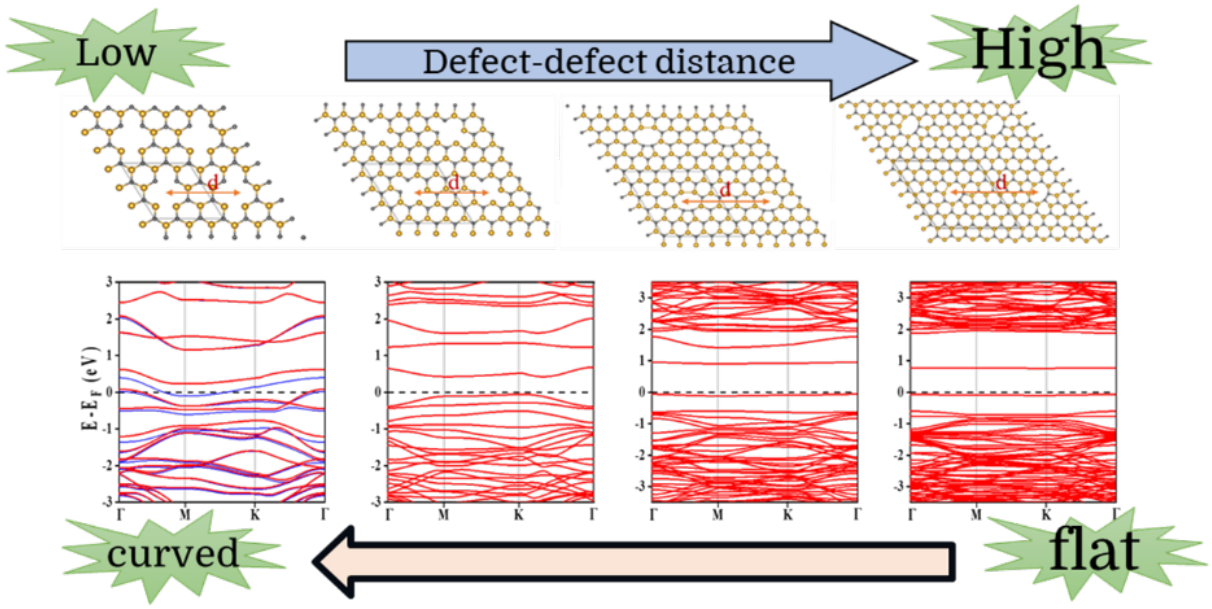


Figure S4: The optimized structure and BS of various n×n supercells (n=3-6) to infer the interaction between defects.

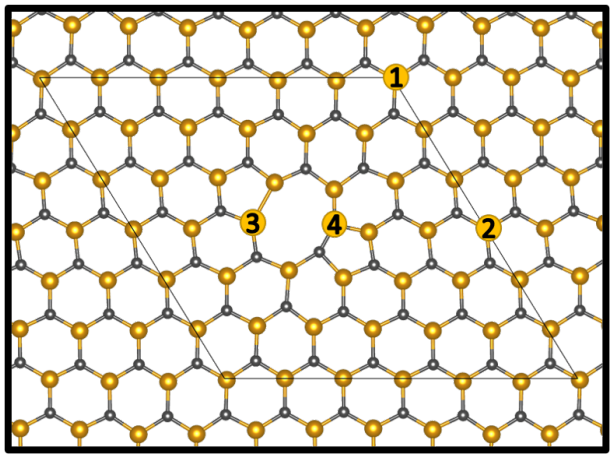


Figure S5: Four different sites are chosen for doping of Al or P atom and respective energtics are given in Table S1.

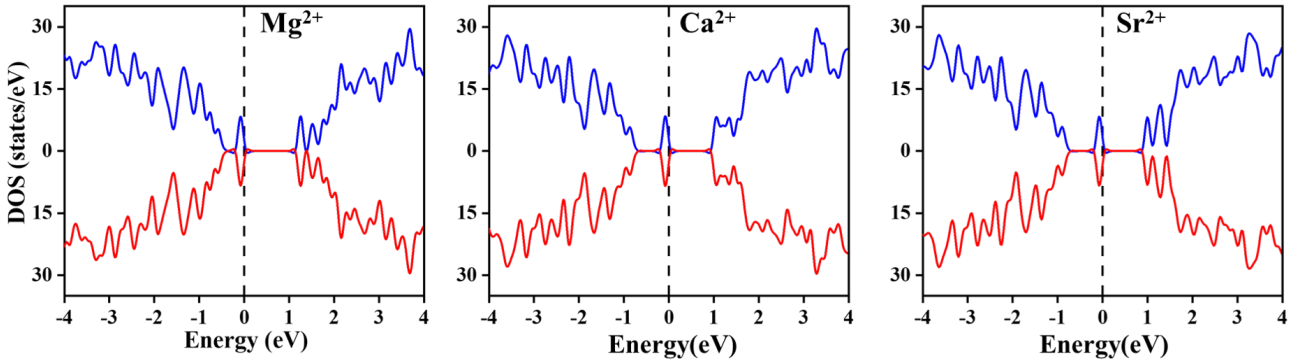


Figure S6: DOS of X atom (X = Mg, Ca, Sr) absorbed on the top of eight membered ring in divacancy SiC layer.

Table S3: Various C-C, Si-Si, and Si-C bond distances ( Å) for divalent ion adsorbed on the top of eight membered ring of divacancy case. For comparison purpose pure divacancy case is also given

| Deposited ions | C-C | Si-Si | Si-C (Octa-ring) | Si-C (Pentagon near C-C) | Si-C (Pentagon near Si-Si) | Si-C (outer) |
|---|---|---|---|---|---|---|
| No ions | 1.622 | 2.444 | 1.783-1.864 | 1.740-1.845 | 1.759-1.768 | 1.764-1.835 |
| $Mg^{2+}$ | 1.783 | 2.461 | 1.797-1.929 | 1.749-1.866 | 1.767-1.838 | 1.776-1.825 |
| $Ca^{2+}$ | 1.706 | 2.441 | 1.803-1.938 | 1.753-1.860 | 1.756-1.811 | 1.777-1.833 |
| $Sr^{2+}$ | 1.693 | 2.415 | 1.803-1.935 | 1.754-1.859 | 1.756-1.817 | 1.788-1.829 |

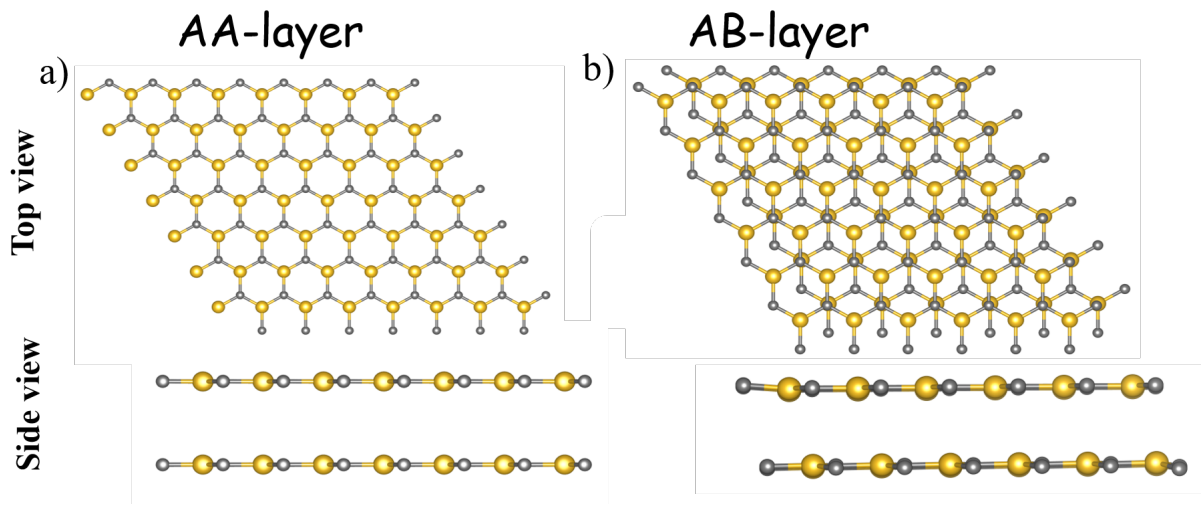


Figure S7: Optimized structure for (a) AA and (b) AB stacking of pristine SiC layers

Table S4: The stacking energy ($E_S$) and spin moment (M) of various bilayered and trilayered systems.Values are deduced by employing DFT-D2 method

| | AA-layer | | AB-layer | |
|---|---|---|---|---|
| System (Bi-layer) | $E_S$ (meV/atom) | M ($\mu_B$) | $E_S$ (meV/atom) | M ($\mu_B$) |
| graphene | -20.75 | 0.0 | -26.58 | 0.0 |
| silicene | -02.02 | 0.0 | -190.17 | 0.0 |
| SiC | -21.11 | 0.0 | -56.42 | 0.0 |
| pristine SiC with VSi | -22.68 | 4.0 | -74.88 | 4.0 |
| pristine SiC with VC | -24.41 | 2.0 | -59.68 | 2.0 |
| pristine SiC with VSi+VC | -50.27 | 0.0 | -63.28 | 0.0 |
| pristine SiC with antisite | -57.52 | 0.0 | -59.77 | 0.0 |
| | A-B($V_{Si}$)-A | | A-B($V_C$)-A | |
| System ( Tri-layer) | -87 | 3.8 | -78 | 2.0 |

## Absorption in divacancy bilayer case

We also extended this work by depositing Mg, Ca and Sr (di-valent) ions onto divacancy layer of bilayer case. Since, divacancy layer layer is strongly interacted with pristine layer, we could not observe any noticeable change in the curvature of DV layer as observed in monolayer system Fig. S8.

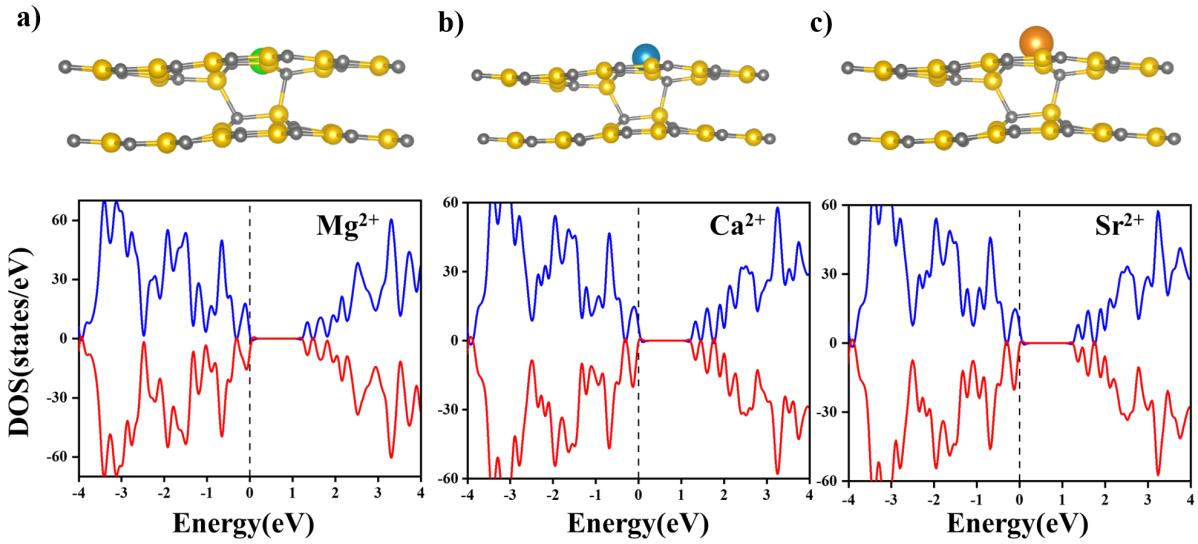


Figure S8: Optimized structures and DOS of X atom (X = Mg, Ca, Sr) absorbed on divacancy layer of bilayer case.