

**Supplementary Materials**  
**of**  
**Modulating interface performance between 2D semiconductor MoSi<sub>2</sub>N<sub>4</sub> and its native high-*k* dielectric Si<sub>3</sub>N<sub>4</sub>**

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## 1. DOS of bulk $\beta$ - $\text{Si}_3\text{N}_4$ with/without strain

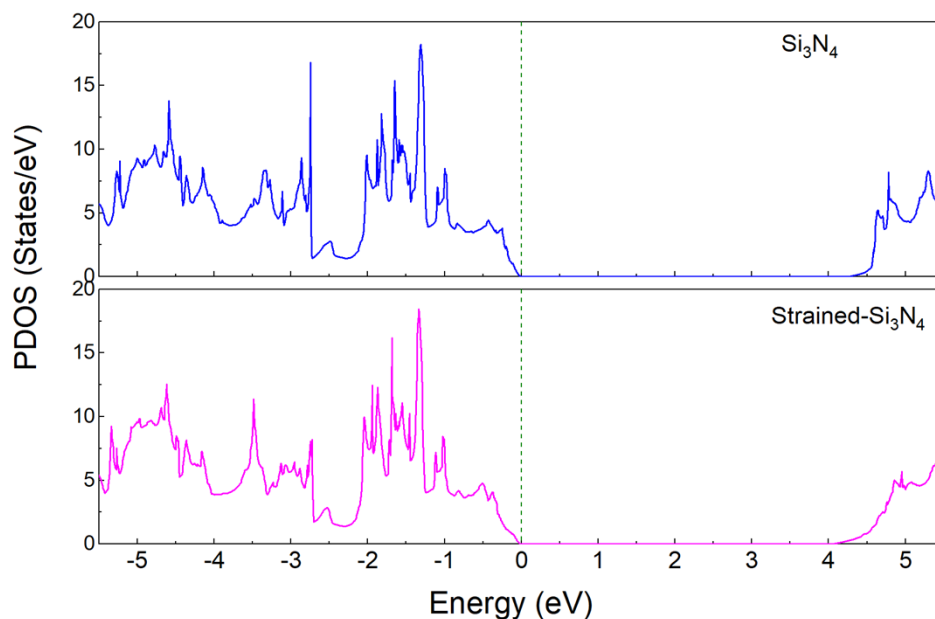


Figure S1. The Total Density of States (TDOS) of  $\text{Si}_3\text{N}_4$  bulk before and after applying the compressive strain.

## 2. PDOS projected on Mo before/after integrating with $\text{Si}_3\text{N}_4$

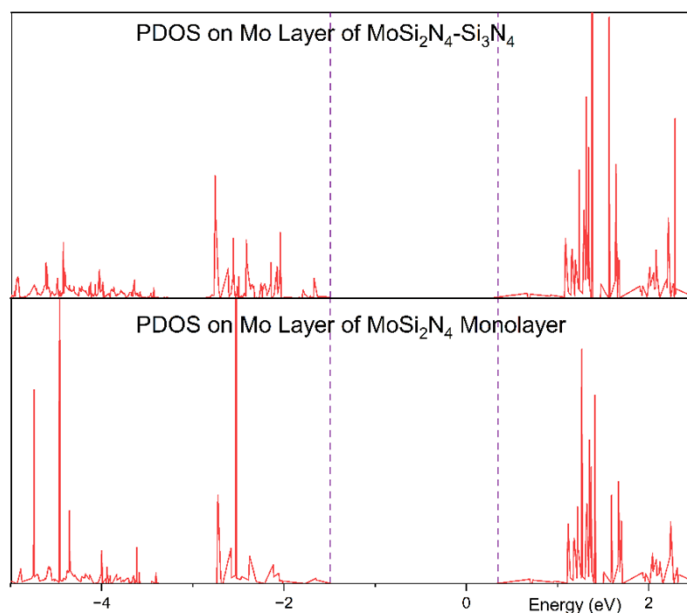


Figure S2. The Projected Density of States (PDOS) on the Mo layer of monolayer  $\text{MoSi}_2\text{N}_4$  and  $\text{MoSi}_2\text{N}_4/\text{Si}_3\text{N}_4$ .

### 3. Planar averaged potential

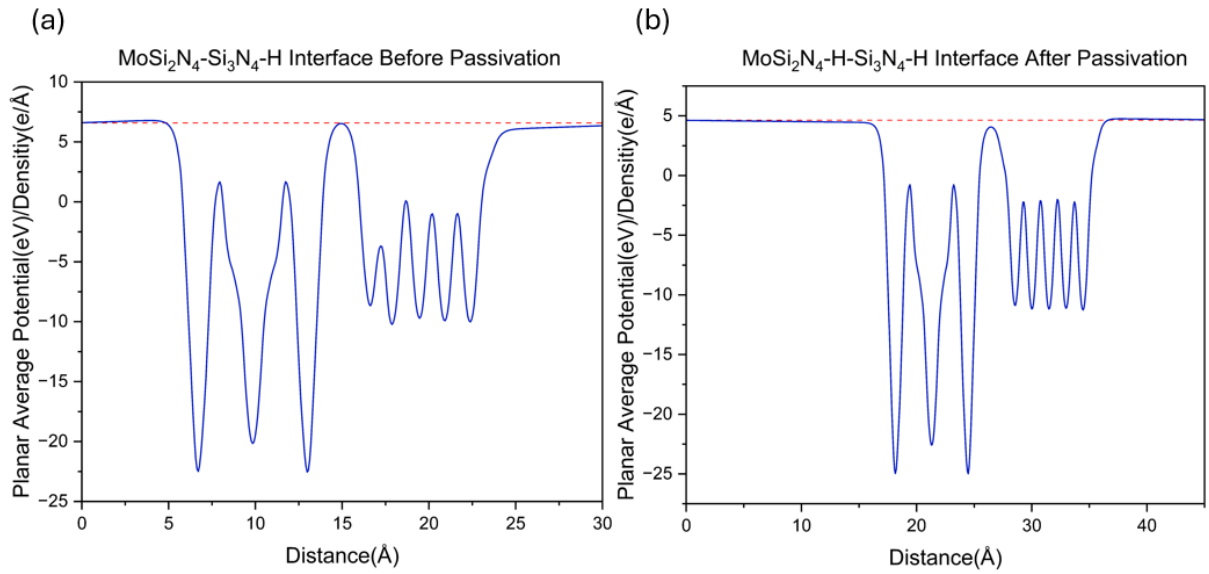


Figure S3. The planar averaged potential of MoSi<sub>2</sub>N<sub>4</sub>/Si<sub>3</sub>N<sub>4</sub> interface (a) before and (b) after hydrogen passivation.

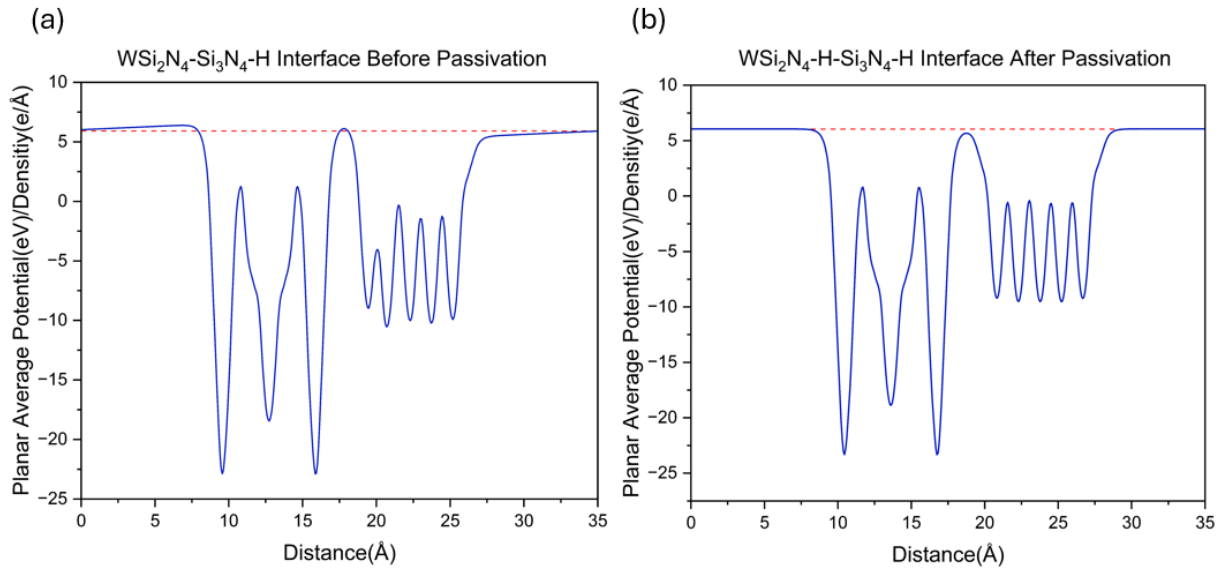


Figure S4. The planar average potential of WSi<sub>2</sub>N<sub>4</sub>- Si<sub>3</sub>N<sub>4</sub> interface (a) before and (b) after hydrogen passivation.

### 4. Charge density redistribution on W plane of 2D WSi<sub>2</sub>N<sub>4</sub>

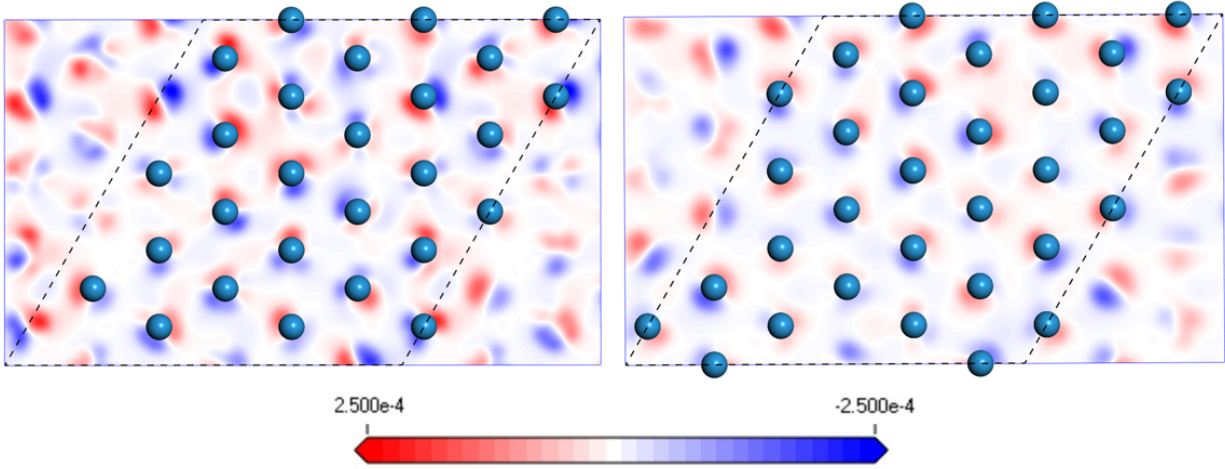


Figure S5 The charge depletion and accumulation around W atoms. The green and purple colours in (b) denote the depleted and accumulated charge density visualised by an iso-surface value of  $1.0 \times 10^{-4} \text{ e}/\text{\AA}^3$ , respectively.