# Supporting Information

## Enhanced Electron Transport and Optical Properties of Experimentally Synthesized Monolayer $Si_9C_{15}$: A Comprehensive DFT Study for Nanoelectronics and Photocatalytic Applications

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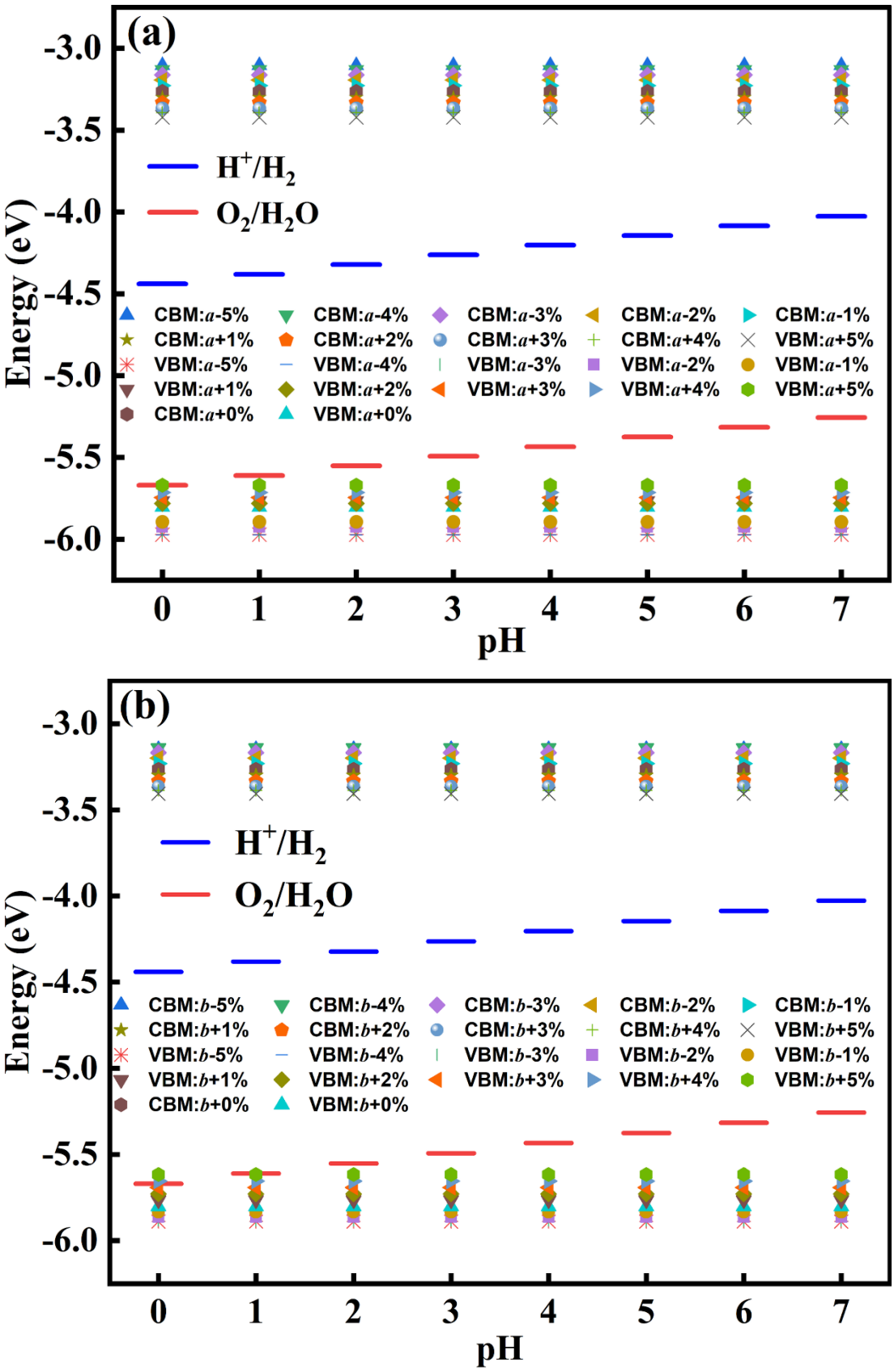


Fig.S1 Evolution of band-edge positions of $Si_9C_{15}$ as a function of applied uniaxial strain in −5%~5% in the direction a and b, respectively. The redox potentials for water oxidation and hydrogen reduction from pH = 0 to pH = 7 are marked as red and blue lines, respectively. The various symbols in the legend represent the positions of CBM and VBM at different strains, respectively.

## OER and HER free energy theory

To calculate the Gibbs free energy $\Delta G$ in the OER and HER reaction processes, we have utilized the Gibbs free energy theory proposed by Nørskov et al., as follows[1-4]:

$$\Delta G = \Delta E + \Delta E_{\mathrm{ZPE}} - T\Delta S + \Delta G_{\mathrm{pH}} + \Delta G_U$$

$\Delta E$ represents the adsorption energy, while $\Delta E_{\mathrm{ZPE}}$ and $\Delta S$ correspond to the differences in zero-point energy and entropy between the adsorbed state and the gas phase. All the $E_{\mathrm{ZPE}}$ and $S$ results can be obtained from the reference[5] and are shown in Table S1. $\Delta G_{\mathrm{pH}}$= 0.059 × pH is the free energy at different pH values, and $\Delta G_{\mathrm{U}} = -eU$ represents an extra potential bias provided by an electron or hole relative to the normal hydrogen electrode.

The equation for HER is as follows：

$$* + \mathrm{H^+} + \ + \mathrm{e^-} \rightarrow \mathrm{H^*} \quad \text{(S1)}$$

$$^*\mathrm{H} + \ \mathrm{H^+} + \ + \mathrm{e^-} \rightarrow * + \mathrm{H_2(g)} \quad \text{(S2)}$$

There are four steps involved in the conversion of $H_2O$ into $O_2$ molecules in OER reaction：

$$\mathrm{H_2O} + * \rightarrow \mathrm{OH^*} + \mathrm{H} + \ + \mathrm{e^-} \quad \text{(S3)}$$

$$\mathrm{OH^*} \rightarrow \mathrm{O^*} + \mathrm{H} + + \mathrm{e^-} \quad \text{(S4)}$$

$$\mathrm{O^*} + \ \mathrm{H_2O} \rightarrow \mathrm{OOH^*} + \mathrm{H} + \ + \mathrm{e^-} \quad \text{(S5)}$$

$$OOH^* \rightarrow * + O_2(g) + H + + e^- \quad (S6)$$

where * denotes an adsorbed species, and OH*, O*, OOH*, and H* are the adsorbed intermediates.

The change in free energy of HER as a function of electrolyte pH can be expressed as:

$$\Delta G_1 = G_{H^*} - \frac{1}{2}G_{H_2} - G^* + 0.059 \times pH - eU \quad (S7)$$

$$\Delta G_2 = G^* + \frac{1}{2}G_{H_2} - G_{H^*} + 0.059 \times pH - eU \quad (S8)$$

The potential of the photogenerated electrons for HER ($U_e$) is defined as the energy difference between the hydrogen reduction potential and the CBM of hydrogen evolution photocatalyst, thus $U_e$= eU-0.059×pH.

For OER：

$$\Delta G_3 = G_{OH^*} + \frac{1}{2}G_{H_2} - G_{H_2O} - G^* - 0.059 \times pH - eU \quad (S9)$$

$$\Delta G_4 = G_{O^*} + \frac{1}{2}G_{H_2} - G_{OH^*} - 0.059 \times pH - eU \quad (S10)$$

$$\Delta G_5 = G_{OOH^*} + \frac{1}{2}G_{H_2} - G_{H_2O} - G_{O^*} - 0.059 \times pH - eU \quad (S11)$$

$$\Delta G_6 = 2G_{H_2O} + G^* - \frac{3}{2}G_{H_2} - G_{OOH^*} + 4.92 - 0.059 \times pH - eU \quad (S12)$$

The potential of the photogenerated holes for OER ($U_h$) is defined as the energy difference between the VBM of oxygen evolution photocatalyst and the hydrogen reduction potential, thus $U_h$=e$U$+0.059×pH. The term 0.059 × pH denotes the energetic contribution resulting from changes in the

concentration of $H^+$ ions, while $eU$ accounts for the impact of potential bias on all states. This involves the transfer of energy through the movement of an electron or a hole within the electrode, with $U$ representing the electrode potential relative to the normal hydrogen electrode (NHE).

**Table S1** The entropy and zero-point energy corrections are utilized to determine the free energy of reactants, products, and intermediate species that are adsorbed on catalysts[5]. In the case of adsorbates, the ZPE values are averaged across all single atom catalyst systems as they exhibit a rather similar value.

| Species | $T \times S$ (eV) (298K) | ZPE (eV) |
|---|---|---|
| $H^*$ | 0 | 0.17 |
| $O^*$ | 0 | 0.07 |
| $OH^*$ | 0 | 0.33 |
| $OOH^*$ | 0 | 0.43 |
| $H_2(g)$ | 0.41 | 0.27 |
| $H_2O(g)$ | 0.58 | 0.57 |


1. Nørskov, J.K., et al., *Origin of the overpotential for oxygen reduction at a fuel-cell cathode.* The Journal of Physical Chemistry B, 2004. **108**(46): p. 17886-17892.
2. Greeley, J., et al., *Computational high-throughput screening of electrocatalytic materials for hydrogen evolution.* Nature materials, 2006. **5**(11): p. 909-913.
3. Rossmeisl, J., et al., *Electrolysis of water on oxide surfaces.* Journal of Electroanalytical Chemistry, 2007. **607**(1-2): p. 83-89.
4. Valdés, Á., et al., *Oxidation and photo-oxidation of water on TiO2 surface.* The

Journal of Physical Chemistry C, 2008. **112**(26): p. 9872-9879.
5. Xu, H., et al., *A universal principle for a rational design of single-atom electrocatalysts.* Nature Catalysis, 2018. **1**(5): p. 339-348.