

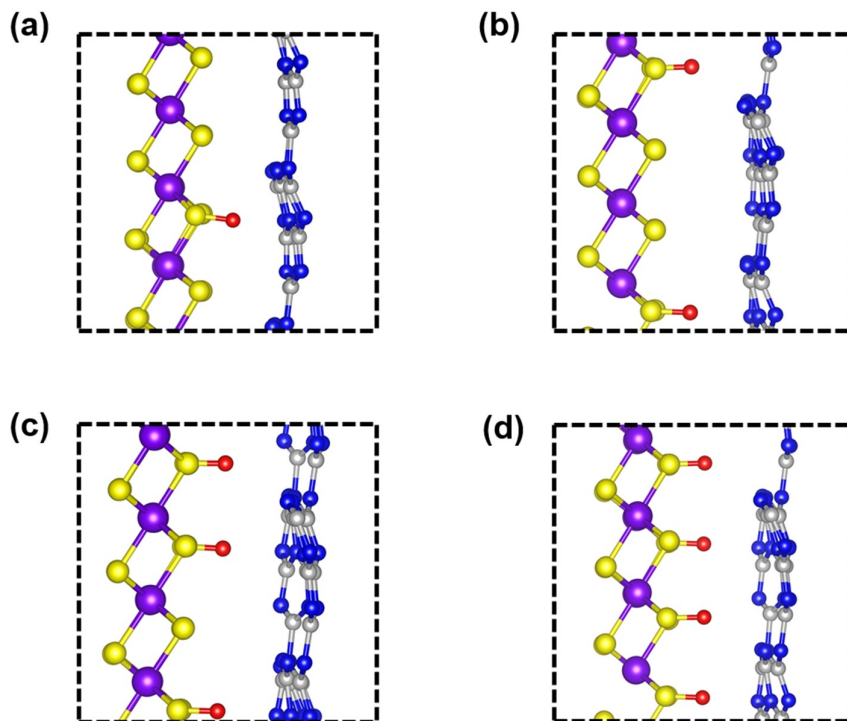
# Manipulatable Interface Electric Field and Charge Transfer in a 2D/2D Heterojunction Photocatalyst via Oxygen Intercalation

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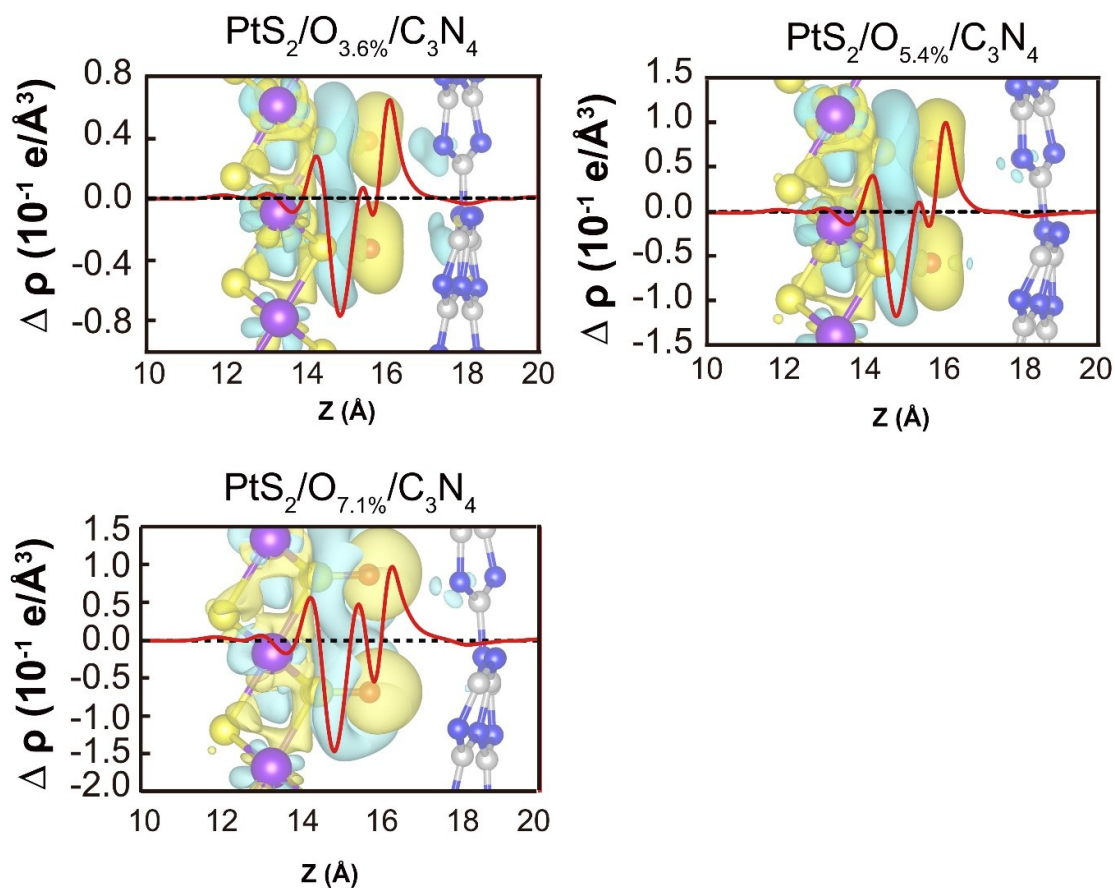
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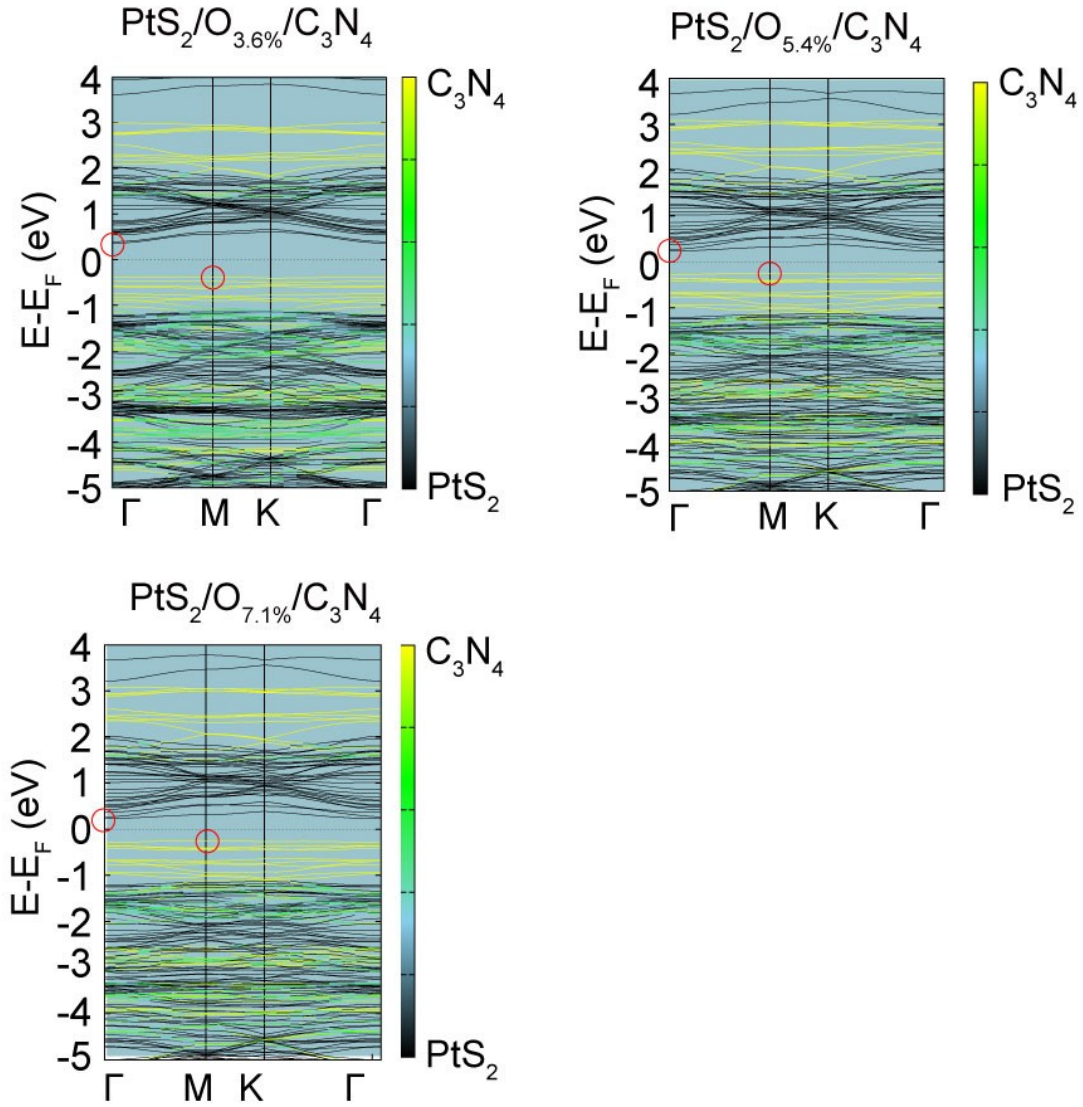
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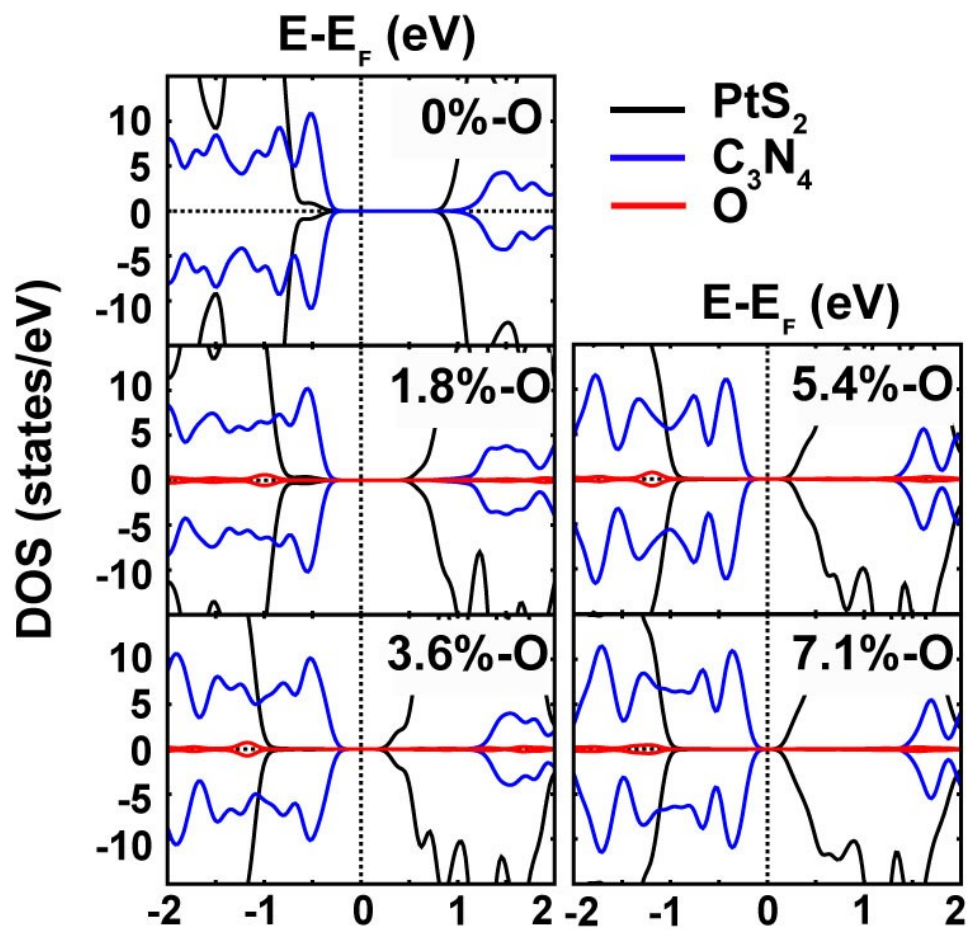
**Figure S1.** The stable configuration of the PtS<sub>2</sub>/Ox/C<sub>3</sub>N<sub>4</sub> heterostructure (x=(a)1.8, (b)3.6, (c)5.4, and (d)7.1%). The purple, yellow, blue, gray, and red colors denote the Pt, S, N, C, and O atoms, respectively.



**Figure S2.** The charge density difference and planar-averaged electron density difference for the PtS<sub>2</sub>/O<sub>3.6%</sub>/C<sub>3</sub>N<sub>4</sub>, the PtS<sub>2</sub>/O<sub>5.4%</sub>/C<sub>3</sub>N<sub>4</sub>, and the PtS<sub>2</sub>/O<sub>7.1%</sub>/C<sub>3</sub>N<sub>4</sub> heterostructure. The purple, yellow, blue, gray, and red atoms indicate Pt, S, N, C, and O, respectively. The yellow and cyan areas indicate charge accumulation and depletion, respectively. The isovalues are 0.005 eV/Å<sup>3</sup>.



**Figure S3.** The band structure of the  $\text{PtS}_2/\text{O}_{3.6\%}/\text{C}_3\text{N}_4$ , the  $\text{PtS}_2/\text{O}_{5.4\%}/\text{C}_3\text{N}_4$ , and the  $\text{PtS}_2/\text{O}_{7.1\%}/\text{C}_3\text{N}_4$  heterostructure, respectively. The contribution of each monolayer in the band structure of the  $\text{PtS}_2/\text{O}_{3.6\%}/\text{C}_3\text{N}_4$ , the  $\text{PtS}_2/\text{O}_{5.4\%}/\text{C}_3\text{N}_4$ , and the  $\text{PtS}_2/\text{O}_{7.1\%}/\text{C}_3\text{N}_4$  heterostructure. The percentages of the contributions of each layer are marked in a different color. The Fermi level was set to zero.



**Figure S4.** The projected density of states in the  $\text{PtS}_2/\text{O}_x/\text{C}_3\text{N}_4$  ( $X = 0, 1.8, 3.6, 5.4, 7.1 \%$ ) heterostructure. The black, blue, and red lines indicate the  $\text{PtS}_2$  layer, the  $\text{C}_3\text{N}_4$  layer, and the O atoms in the  $\text{PtS}_2/\text{O}_x/\text{C}_3\text{N}_4$  heterostructure. The Fermi level (vertical black dotted line) was set to zero.

**Table S1.** The intercalation energy of the  $\text{PtS}_2/\text{O}_x/\text{C}_3\text{N}_4$  heterostructure ( $x=1.8, 3.6, 5.4, 7.1\%$ ).

	$E_{\text{int}}$ (eV)
$\text{PtS}_2/\text{O}_{1.8\%}/\text{C}_3\text{N}_4$	-1.78
$\text{PtS}_2/\text{O}_{3.6\%}/\text{C}_3\text{N}_4$	8.11
$\text{PtS}_2/\text{O}_{5.4\%}/\text{C}_3\text{N}_4$	4.82
$\text{PtS}_2/\text{O}_{7.1\%}/\text{C}_3\text{N}_4$	3.25