

Supporting Information:

**PdSe₂: Flexible Two-dimensional Transition
Metal Dichalcogenides Monolayer for Water
Splitting Photocatalyst with Extremely Low
Recombination Rate**

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1. Calculation method of HER and OER activity

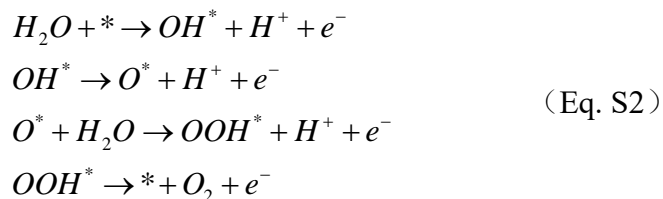
Table S1. Zero-point energy correction (E_{ZPE}), entropy contribution (TS , $T=298.15K$), total energy (E), and the Gibbs free energy (G) of molecules and adsorbates.

Species	E_{ZPE} (eV)	$-TS$	E (eV)	G (eV)
H_2	0.27	-0.41	-6.77	-6.91
H_2O	0.56	-0.67	-14.22	-14.33
OH^*	0.29	-0.21	-116.53	-116.45
O^*	0.03	-0.12	-110.75	-110.84
OOH^*	0.35	-0.25	-121.31	-121.21
H^*	0.19	-0.02	-110.12	-109.9

In the aqueous solution, the hydrogen evolution reaction can be written as:



in which the $*$ represents the adsorption site. Meanwhile, the oxygen evolution reaction processes a four-electron oxidation steps, which can be written as:



where OH^* , O^* and OOH^* denote the adsorbed intermediates. Then, the free energy change (ΔG) can be express as:

$$\begin{aligned}
 \Delta G_1 &= G_{H^*} - 1/2 G_{H_2} + G^* \\
 \Delta G_2 &= G_{OH^*} + 1/2 G_{H_2} - G_{H_2O} \\
 \Delta G_3 &= G_{O^*} + 1/2 G_{H_2} - G_{OH} \\
 \Delta G_4 &= G_{OOH^*} + 1/2 G_{H_2} - G_{H_2O} - G_{O^*} \\
 \Delta G_5 &= G^* + 1/2 G_{H_2} - G_{O_2} - G_{OOH^*}
 \end{aligned} \quad (\text{Eq. S3})$$

Table S2. The calculated ΔG of water oxidation and hydrogen reduction reactions for different sites of PdSe₂ monolayer. The most active sites for two reactions are highlight in red.

	Pd (eV)	Se _I (eV)	Se _{II} (eV)
Water Oxidation	2.155	2.75	2.685
Hydrogen Reduction	1.505	1.195	1.175

2. AIMD simulation in liquid water

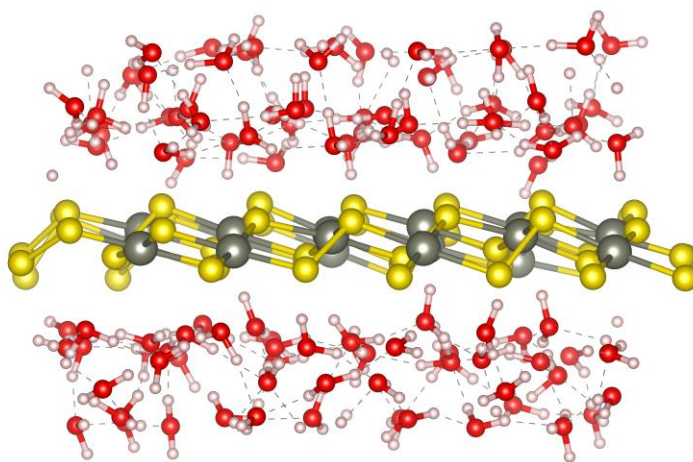


Figure S1. The structure evolution of the PdSe₂ monolayer in liquid water at 300 K for 10 ps. The yellow, grey, pink and red balls represent the Se, Pd, H and O atoms, respectively.