# Supplementary Information

# Deciphering the regulatory mechanism of electrocatalytic activity in Fe-anchored $CuInP_2S_6$ monolayer via ferroelectric switching and in-plane strain

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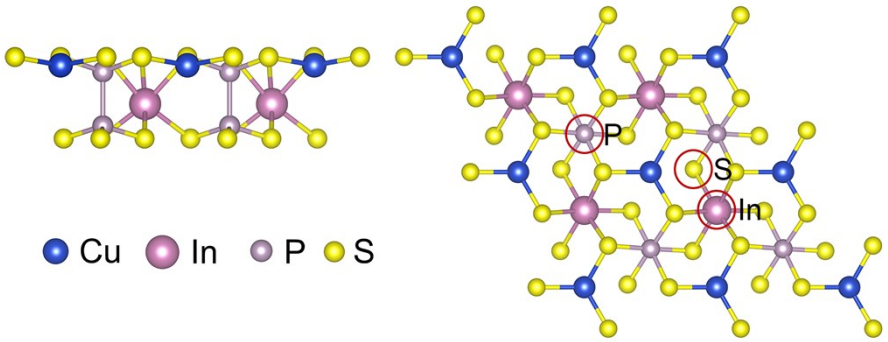


**Fig. S1** Schematic diagram of different adsorption sites on the surface of CIPS substrate.

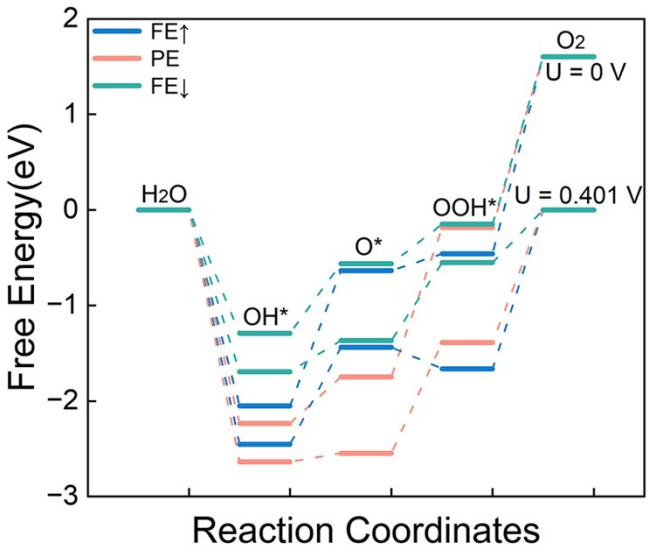


**Fig. S2** Gibbs free energy changes for individual reaction steps of the OER process in three configurations under alkaline conditions (pH = 14)

**Table. S1** Total energy ($E_{tot-H*}$), adsorption energy ($E_{ads}$ ), the Gibbs free energy ($\Delta G_{H*}$) of H*

| | $E_{total-H*}$(eV) | $\Delta G_{H*}$(eV) | $E_{ads}$(eV) |
|---|---|---|---|
| FE↑ | −195.730 | −0.508 | −3.28 |
| PE | −196.145 | −0.861 | −3.15 |
| FE↓ | −194.365 | −1.068 | −2.57 |

**Table. S2** The energy consumptions of each reaction step and overpotential in different polarization states

| | $\Delta G_1$ | $\Delta G_2$ | $\Delta G_3$ | $\Delta G_4$ | Overpotential (eV) |
|---|---|---|---|---|---|
| FE↑ | −1.223 | 2.245 | 1.007 | 2.891 | 1.661 |
| PE | −1.405 | 1.315 | 2.393 | 2.617 | 1.387 |
| FE↓ | −0.464 | 1.559 | 1.242 | 2.583 | 1.353 |

**Table. S3** Total energy ($E_{tot\text{-}H*}$), adsorption energy ($E_{ads}$ ), the Gibbs free energy ($\Delta G_{H*}$) of H* on the Cu adsorption site under −14% strain

| | $E_{total\text{-}H*}$(eV) | $\Delta G_{H*}$(eV) | $E_{ads}$(eV) |
|---|---|---|---|
| Cu | −166.835 | 0.414 | −3.27 |

**Table. S4** The energy consumption of each reaction step and the overpotential with Cu as the adsorption site under −14% strain

| | $\Delta G_1$ | $\Delta G_2$ | $\Delta G_3$ | $\Delta G_4$ | Overpotential (eV) |
|---|---|---|---|---|---|
| Cu | −1.223 | 2.245 | 1.007 | 2.891 | 1.661 |

**Table. S5** Total energy ($E_{tot-H*}$), adsorption energy ($E_{ads}$ ), the Gibbs free energy ($\Delta G_{H*}$) of H* on the Fe adsorption site under −14% strain

| | $E_{total-H*}$(eV) | $\Delta G_{H*}$(eV) | $E_{ads}$(eV) |
|---|---|---|---|
| Fe | –172.979 | –0.616 | –4.151 |

**Table. S6** The energy consumption of each reaction step and the overpotential with Fe as the adsorption site under −14% strain

| | $\Delta G_1$ | $\Delta G_2$ | $\Delta G_3$ | $\Delta G_4$ | Overpotential (eV) |
|---|---|---|---|---|---|
| Fe | –1.327 | 0.944 | 3.362 | 1.941 | 2.132 |