## **Supporting information**

## Tunable Heteroassembly of 2D CoNi LDH and Ti<sub>3</sub>C<sub>2</sub> Nanosheets with Enhanced Electrocatalytic Activity for Oxygen Evolution

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Figure S1. SEM images of CoNi LDH nanosheets after freeze drying.



Figure S2. TEM image and elemental mappings of CoNi LDH nanosheets after exfoliation.



Figure S3. XRD pattern of CoNi LDH nanosheets after freeze drying.



Figure S4. XRD pattern of the multilayered  $Ti_3AlC_2$  MAX.



Figure S5. Zeta potential profiles of CoNi LDH and  $Ti_3C_2$  nanosheet dispersion.



Figure S6. Crystal structure of monolayer CoNi LDH and  $Ti_3C_2$  nanosheets and the area match model for design of the heterostructures.



Figure S7. LSV curves for CoNi LDH, Ti<sub>3</sub>C<sub>2</sub> nanosheets and CoNi LDH/Ti<sub>3</sub>C<sub>2</sub> materials.

![](_page_8_Figure_0.jpeg)

Figure S8. CV profiles of CoNi LDH/Ti $_3C_2$  at various scan rates and the capacitive currents at 1.1 V plotted as a function of scan rate.

![](_page_9_Figure_0.jpeg)

Figure S9. EIS spectra of CoNi LDH,  $Ti_3C_2$  and CoNi LDH/ $Ti_3C_2$  superlattice heterostructures.

![](_page_10_Picture_0.jpeg)

Figure S10. Different active site positions at the CoNi LDH/Ti $_3C_2$  interface.

![](_page_11_Figure_0.jpeg)

Figure S11. Adsorption energies of different active sites for the intermediates (OH\* and OOH) of the OER reaction.

![](_page_12_Figure_0.jpeg)

Figure S12. Electron localization function (a-c) and charge density difference (d-i) diagrams for the adsorption of  $OH^*$  (a, d, f),  $OOH^*$  (b, e, h), and  $O^*$  (c, f, i) on the H site.

![](_page_13_Figure_0.jpeg)

Figure S13. Electron localization function (a-c) and charge density difference (d-i) diagrams for the adsorption of OH\* (a, d, f), OOH\*(b, e, h), and O\*(c, f, i) on the H1 site.

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Figure S14. Electron localization function (a-c) and charge density difference (d-i) diagrams for the adsorption of  $OH^*$  (a, d, f),  $OOH^*$  (b, e, h), and  $O^*$  (c, f, i) on the H2 site.

![](_page_15_Figure_0.jpeg)

Figure S15. Electron localization function (a-c) and charge density difference (d-i) diagrams for the adsorption of  $OH^*$  (a, d, f),  $OOH^*$  (b, e, h), and  $O^*$  (c, f, i) on the O site.