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## **Supporting Information**

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Living Atomically Dispersed Cu Ultrathin TiO<sub>2</sub> Nanosheet CO<sub>2</sub> Reduction Photocatalyst

Zaiyong Jiang,\* Wei Sun, Wenkang Miao, Zhimin Yuan, Guihua Yang, Fangong Kong, Tingjiang Yan, Jiachuan Chen,\* Baibiao Huang, Changhua An, and Geoffrey A. Ozin\*

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## Photocatalyst

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Figure S1. A photo of the experiment of in-situ photodeposition in air. The color of

suspension remained white.



*Figure S2.* High-resolution XPS spectra of the Cu 2p region of Cu/TiO<sub>2</sub>/Air.



Figure S3. Photocatalytic CO<sub>2</sub> reduction activities of pristine TiO<sub>2</sub> and Cu/TiO<sub>2</sub>/Air.



*Figure S4.* Photos taken (a) after completing the experiment of in-situ photodeposition in  $CO_2$  atmosphere, showing the red-pink colour of the suspension solution and (b) after the suspension was stirred in air showing the mere white colour.



*Figure S5.* Isotope (<sup>13</sup>C) tracing experiments.



*Figure S6.* An SEM image of pristine TiO<sub>2</sub> nanosheets.



Figure S7. SEM images of (a) Cu/TiO<sub>2</sub>-1, (b) Cu/TiO<sub>2</sub>-2, (c) Cu/TiO<sub>2</sub>-3 and (d) Cu/TiO<sub>2</sub>-4.



Figure S8. A zoomed-in atomic-resolution STEM image of a Cu-rich region of

Cu/TiO<sub>2</sub>-2, showing the clusters are not crystalline.



*Figure S9.* EDS nanlysis of Cu/TiO<sub>2</sub>-2.



*Figure S10.* EDS mapping images of Cu/TiO<sub>2</sub>-2.



*Figure S11.* EDS analysis of Cu/TiO<sub>2</sub>-2/24h.



Figure S12. High-resolution XPS spectra of Ti 2P region, showing the similar binding

energies of Ti<sup>4+</sup> are 458.6 and 464.4 eV, respectively, in both samples.



*Figure S13.* Photocatalytic  $CO_2 \rightarrow CO$  reduction activity of Cu/TiO<sub>2</sub>-2 for six consecutive runs.

In each cycle, the reaction solution was continuously bubbled with high purity  $CO_2$  gas for 15 min. The CO production was not deactivated but rather activated over cycling, likely due to the enrichment of dissolved  $CO_2$ .



Figure S14. The valence band XPS spectrum of Cu/TiO<sub>2</sub>-2.

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