Supporting Information

Efficient Photocatalytic Overall Water-Splitting Characteristics Driven by Strain Engineering in Two-dimensional β-AuTe

Xiong-Fei Zhang,[a] Fei-Jie Huang,[a] Yang Huang,[b] Ju-Qi Ruan,[a] Yi-Fen Zhao,[a] Kai Xiong,[c] Yao He,[d] and Qing-Yuan Chen[a,*]

[a]*School of Physical Science and Technology, Kunming University, Kunming 650214, China*

[b]*Institute of Nanoscience and Applications, and Department of Electrical and Electronic Engineering, Southern University of Science and Technology, Shenzhen 518055, China*

[c]*Materials Genome Institute, School of Materials and Energy, Yunnan University, Kunming 650091, China*

[d]*Department of Physics, Yunnan University, No.2 Green Lake North Road, Wu Hua Qu, Kunming 650091, Yunnan Province, China*

E-mail: qingyuanchen212@163.com

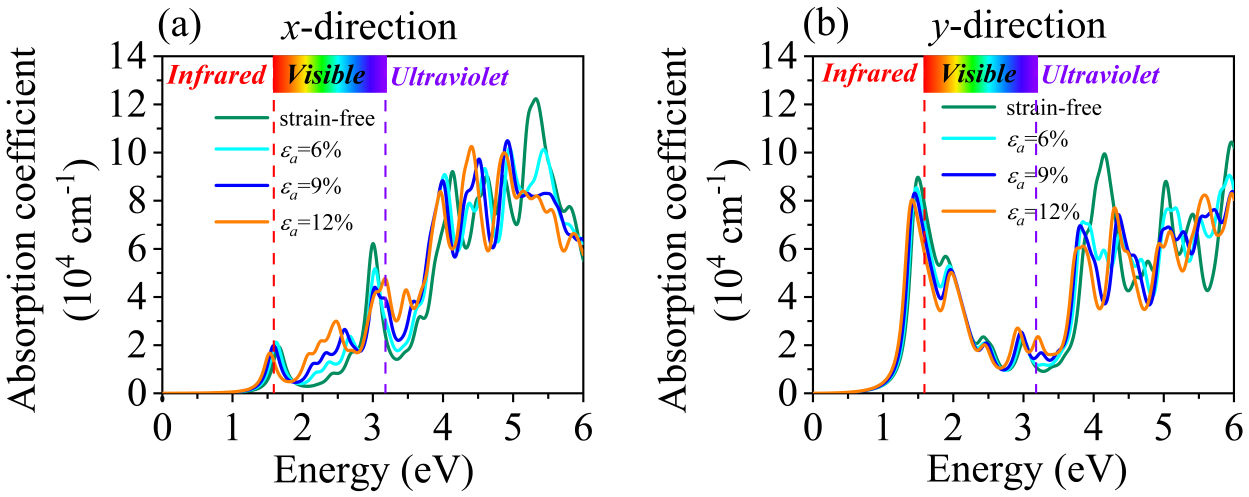

Fig. S1 The optical absorption coefficient of the monolayer β-AuTe under strain-free, uniaxial strain along the a-direction of 6%, 9%, and 12% in the x- and y-directions using the G_0W_0+BSE method.

Gibbs Free Energy

Photocatalytic water-splitting contains two half-reactions, the oxygen evolution reaction (OER) and hydrogen evolution reaction (HER), where the two half-reactions can be obtained based on the theory developed by Nørskovet al.[1] The Gibbs free energy change (ΔG) can be calculated by the following equation:[2,3]

$$\Delta G = \Delta E + \Delta E_{\mathrm{ZPE}} - T\Delta S, \tag{1}$$

where ΔE, ΔE_{ZPE}, and ΔS represent the differences in total energy, zero-point energy, and entropy of the slab with and without adsorbed intermediates. The system temperature T is set to be 298.15 K. ΔE_{ZPE} could be calculated by:

$$\Delta E_{\mathrm{ZPE}} = \frac{1}{2}\sum h\nu, \tag{2}$$

where ν represents the vibrational frequency. Then, $T\Delta S$ represents the change in entropic contributions (with T set to 298.15 K). The term TS can be calculated by:

$$TS = k_b T\left[\sum_K \ln\left(\frac{1}{1-e^{-h\nu/k_bT}}\right) + \sum_K \frac{h\nu}{k_bT}\left(\frac{1}{e^{h\nu/k_bT}-1}\right) + 1\right] \quad , \tag{3}$$

where e represents the electron charge, h represents Planck's constant, and k_b is Boltzmann's constant.

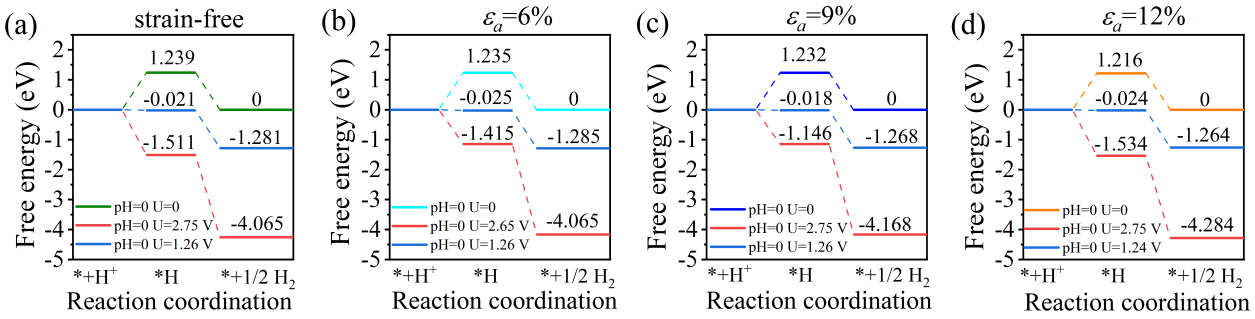

Fig. S2 The free energy for HER under different conditions of the monolayer β-AuTe under strain-free (a), uniaxial strain along the *a*-direction of 6% (b), 9% (c), and 12% (d).

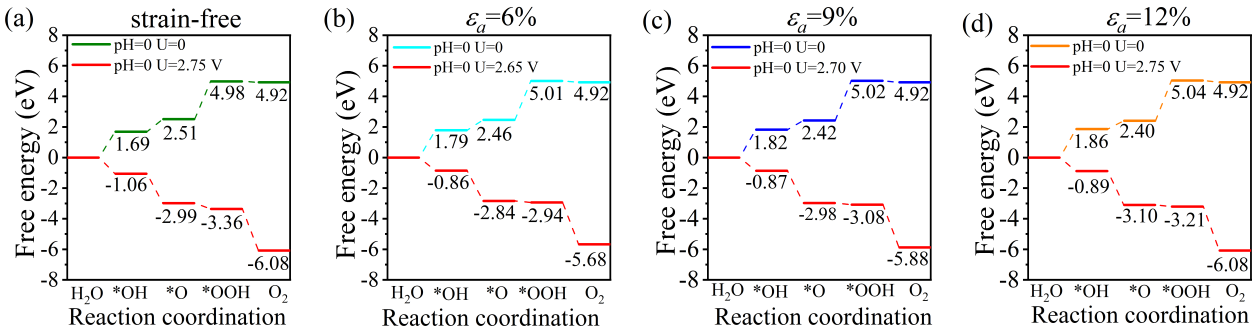

Fig. S3 The free energy for OER under different conditions of the monolayer β-AuTe under strain-free (a), uniaxial strain along the *a*-direction of 6% (b), 9% (c), and 12% (d).

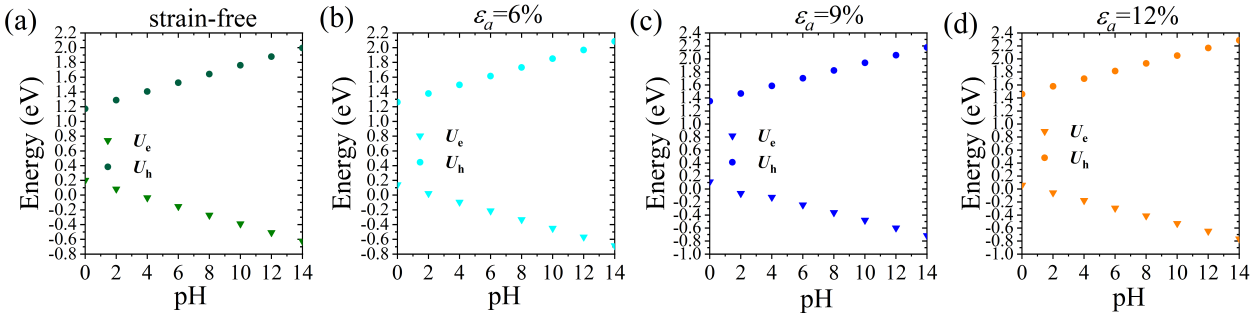

Fig. S4 The energy of U_e and the U_h changed with the pH of the monolayer β-AuTe under strain-free (a), uniaxial strain along the *a*-direction of 6% (b), 9% (c), and 12% (d).

For HER, it follows a two-electron transferred reaction pathway:

$$* + \mathrm{H^+} + \mathrm{e^-} \rightarrow \mathrm{H^*}, \tag{4}$$

$$\mathrm{H^*} + \mathrm{H^+} + \mathrm{e^-} \rightarrow \mathrm{H_2}, \tag{5}$$

For OER, it follows a four-electron transferred reaction pathway:

$$* + \mathrm{H_2O} \rightarrow \mathrm{OH^*} + \mathrm{e^-} + \mathrm{H^+}, \tag{6}$$

$$\mathrm{OH^*} \rightarrow \mathrm{O^*} + \mathrm{e^-} + \mathrm{H^+}, \tag{7}$$

$$\mathrm{O^*} + \mathrm{H_2O} \rightarrow \mathrm{OOH^*} + \mathrm{e^-} + \mathrm{H^+}, \tag{8}$$

$$\mathrm{OOH^*} \rightarrow \mathrm{O_2} + \mathrm{e^-} + \mathrm{H^+}, \tag{9}$$

where $*$ is the active site on photocatalysts, $\mathrm{O^*}$, $\mathrm{OH^*}$, $\mathrm{OOH^*}$, and $\mathrm{H^*}$ represent the adsorbed intermediates.

For HER, ΔG under the effect of electrolyte pH is given by the following equation:

$$\Delta G_{\mathrm{H^*}} = G_{\mathrm{H^*}} - \frac{1}{2}G_{\mathrm{H_2}} - G^* + 0.059 \times \mathrm{pH} - eU. \tag{10}$$

For OER, ΔG under the effect of electrolyte pH is given by the following equations:

$$\Delta G_1 = G_{\mathrm{OH^*}} + \frac{1}{2}G_{\mathrm{H_2}} - G_{\mathrm{H_2O}} - G^* - 0.059 \times \mathrm{pH} - eU, \tag{11}$$

$$\Delta G_2 = G_{\mathrm{O^*}} + \frac{1}{2}G_{\mathrm{H_2}} - G_{\mathrm{OH^*}} - 0.059 \times \mathrm{pH} - eU, \tag{12}$$

$$\Delta G_3 = G_{\mathrm{OOH^*}} + \frac{1}{2}G_{\mathrm{H_2}} - G_{\mathrm{H_2O}} - G_{\mathrm{O^*}} - 0.059 \times \mathrm{pH} - eU, \tag{13}$$

$$\Delta G_4 = G_{\mathrm{O_2}} + G^* + \frac{1}{2}G_{\mathrm{H_2}} - G_{\mathrm{OOH^*}} - 0.059 \times \mathrm{pH} - eU, \tag{14}$$

where $0.059 \times$ pH is the free energy contribution under the effect of pH, eU denotes the influence

of extra potential bias provided by the electrons or holes in the electrode, and U is the electrode potential relative to the standard hydrogen electrode (SHE).

References

(1) Novoselov, K. S.; Geim, A. K.; Morozov, S. V.; Jiang, D.-e.; Zhang, Y.; Dubonos, S. V.; Grigorieva, I. V.; Firsov, A. A. Electric field effect in atomically thin carbon films. *Science* **2004**, *306*, 666–669.

(2) Sun, R.; Yang, C.-L.; Wang, M.-S.; Ma, X.-G. High solar-to-hydrogen efficiency photocatalytic hydrogen evolution reaction with the HfSe2/InSe heterostructure. *Journal of Power Sources* **2022**, *547*, 232008.

(3) Liu, M.; Yang, C.-L.; Li, Y.; Wang, M.-S.; Ma, X.-G. Hydrogen production of overall water splitting with direct Z-scheme driven by antimonene and arsenide nanoribbon heterostructures: Insight from electronic properties and carrier nonadiabatic dynamics. *Journal of Power Sources* **2024**, *594*, 234045.