Supporting Information

Forecasting the unrevealed surface controlled photocatalytic water splitting in two-dimensional Ag_2Se with ultrafast carrier mobility: A first-principles study

Yee Hui Robin Chang [a, *], Keat Hoe Yeoh [b, *], Junke Jiang [c], Soo See Chai [d], Yusuf Zuntu Abdullahi [e, f], Heng Yen Khong [a], Thong Leng Lim [g], Moi Hua Tuh [h]

[a] Faculty of Applied Sciences, Universiti Teknologi MARA, Cawangan Sarawak, 94300 Kota Samarahan, Sarawak, Malaysia
[b] Department of Electrical and Electronic Engineering, Lee Kong Chian Faculty of Engineering and Science, Universiti Tunku Abdul Rahman, 43000 Kajang, Selangor, Malaysia
[c] Materials Simulation and Modelling, Department of Applied Physics, Eindhoven University of Technology, 5612 Eindhoven, The Netherlands
Present Address: Univ Rennes, ENSCR, CNRS, ISCR-UMR 6226, F-35000 Rennes, France
[d] Faculty of Computer Science & Information Technology (FCSIT), Universiti Malaysia Sarawak, 94300 Kota Samarahan, Sarawak, Malaysia
[e] Department of Physics, Faculty of Science, Kaduna State University, PMB 2339, Kaduna, Nigeria
[f] Department of Physics, Adnan Menderes University, Aydın 09010, Turkey
[g] Faculty of Engineering and Technology, Multimedia University, Jalan Ayer Keroh Lama, 75450 Melaka, Malaysia
[h] Faculty of Computer & Mathematical Sciences, Universiti Teknologi MARA, Cawangan Sarawak, 94300 Kota Samarahan, Sarawak, Malaysia

[*]Electronic mail: robincyh@uitm.edu.my; keathoe.yeoh@gmail.com

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S1 Unit cell POSCAR for optimized Ag_2Se monolayer

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Ag2Se
1.00000000000000
  5.8560390000000000   -0.0207510000000000   -0.0435000000000000
  0.0243210000000000    5.8809839999999998    0.0036040000000000
 -0.1850750000000000    0.0155000000000000   17.7377740000000017
  Ag   Se
   4    2
Direct
  0.6851229999999973  0.7045510000000021  0.4977590000000021
  0.1844719999999995  0.7193340000000035  0.5008369999999971
  0.6973569999999967  0.2048819999999978  0.4989769999999965
  0.1993610000000032  0.2178729999999973  0.5021509999999978
  0.9418170000000003  0.4593679999999978  0.4168280000000024
  0.4511639999999986  0.9584430000000026  0.5834479999999971
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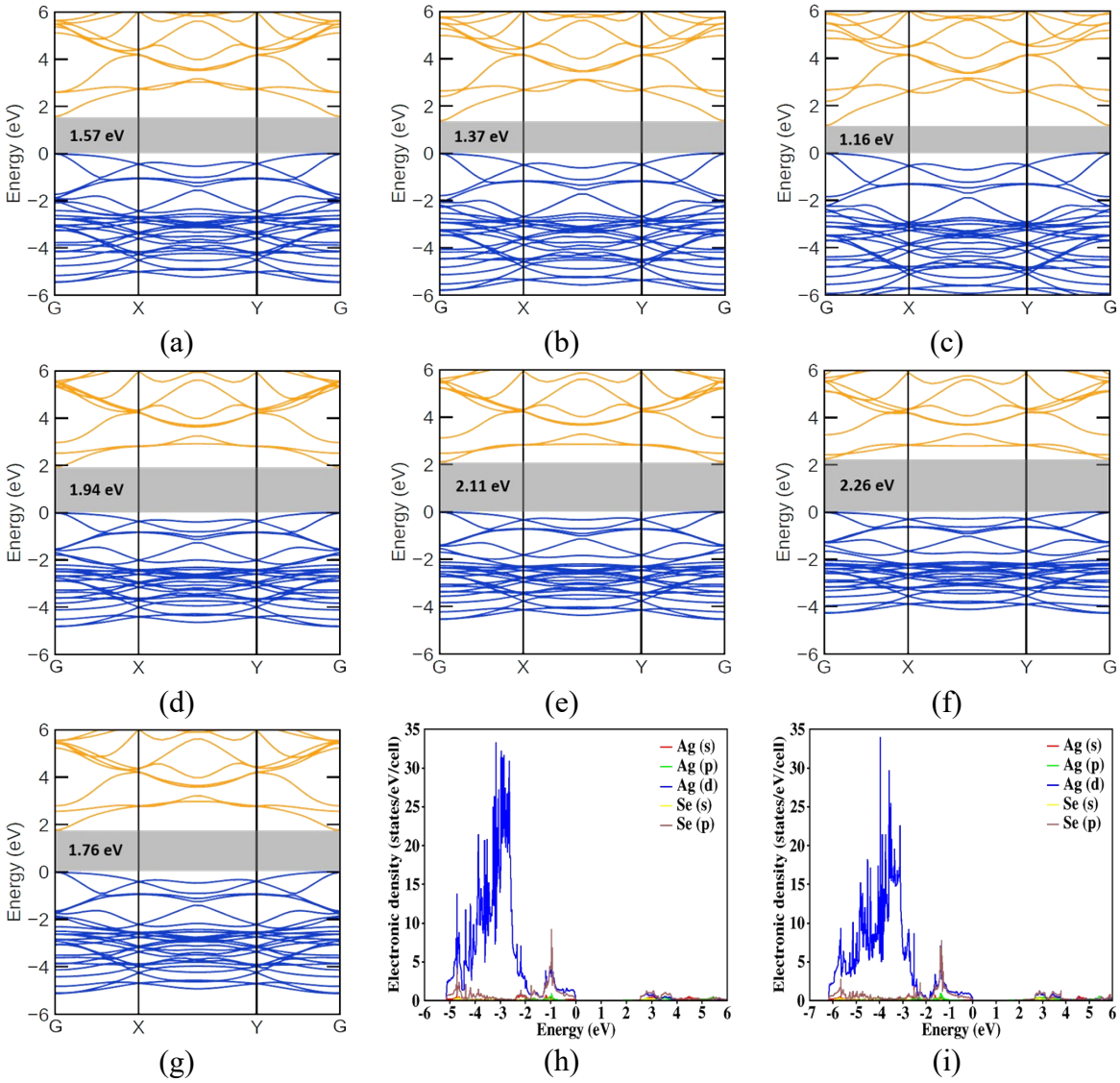
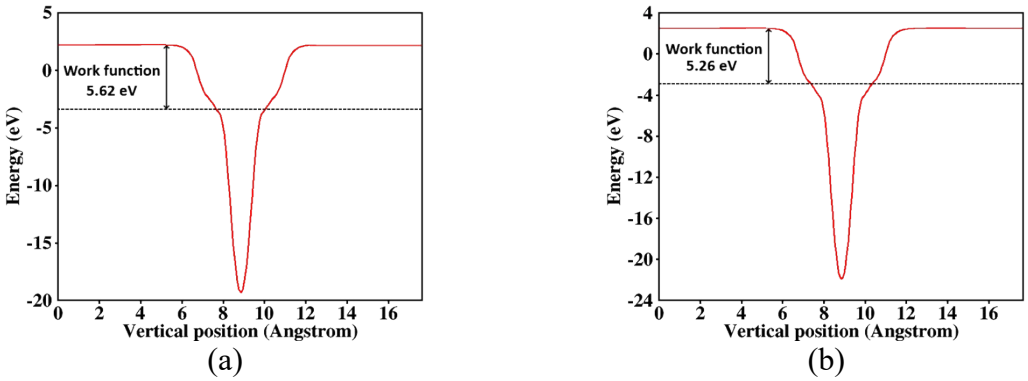


S2 Detailed electronic band structures of Ag_2Se monolayer under (a) -2%, (b) -4%, (c) -6%, (d) 2%, (e) 4%, (f) 6% and (g) 0% biaxial straining, at PBE level. Calculated PDOS of Ag_2Se monolayer under (h) 0% and (i) -6%, at PBE level.



S3 Electrostatic potential for Ag_2Se monolayer (a) 0% and (b) -6% biaxial straining.

S4 SLME computational details

Theoretical maximum solar cell efficiency can be calculated using the following formula:

$$\eta = \frac{P_m}{P_{in}}$$

where P_m is the highest achievable power density of the thin-film solar absorber material, while P_{in} is the incident power density of the entire solar spectrum. The maximum output power density P=JV of the material can be found utilizing J-V characteristics of solar cell as proposed by Yu and Zunger [1]. Here, J indicates the total current density, while V represents the potential over the absorber layer, as seen in the following relationship:

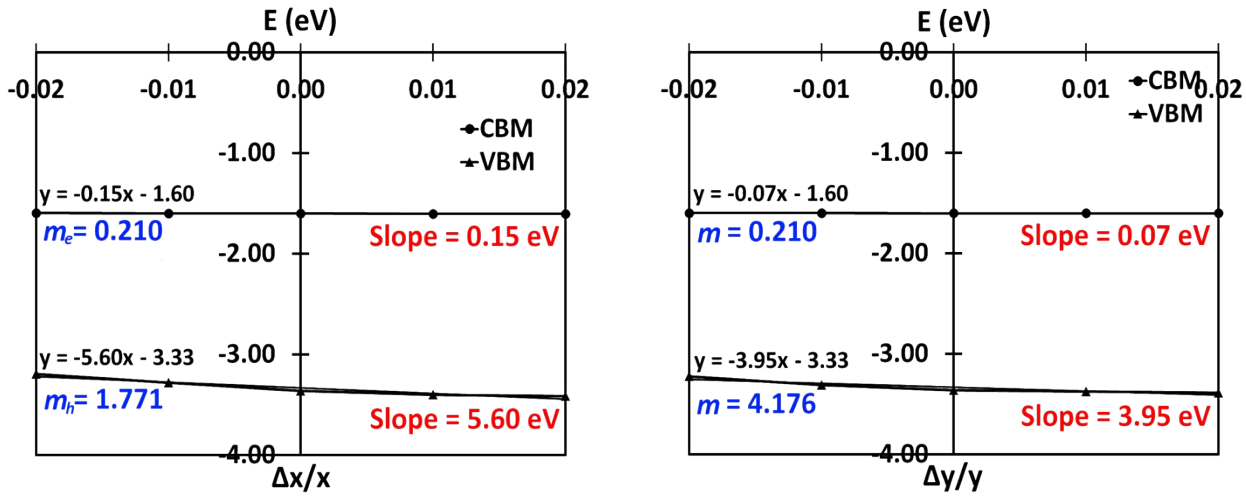
$$J = J_{sc} - J_r\left(e^{eV/k_BT} - 1\right)$$

in which J_{sc}, J_r, k_B and T denote, respectively, the short-circuit current, the total recombination current density, the Boltzmann constant, and the temperature. Absorptivity α(E), photon flux from incident solar spectrum $I_{sun}(E)$ (AM1.5G) and black-body spectrum $I_{bb}(E,T)$ can be used to calculate J_{sc} and J_r, as follows:

$$J_{sc} = e\int_0^\infty \alpha(E) I_{sun}(E) dE$$

$$J_r = e\int_0^\infty \alpha(E) I_{bb}(E,T) dE$$

where α(E) and L are the absorbance coefficient and thickness of the thin film, respectively. Total recombination current density is largely contributed by radiative recombination current density J_{rad} and non-radiative recombination current density $J_{non\text{-}rad}$.



S5 The displacement of CBM and VBM for Ag_2Se monolayer as a function of applied strain in the (x) zigzag and (y) armchair directions, respectively. The linear fit offers the deformation potentials of the monolayer under study. m_e and m_h are in electron rest mass unit.

S6 Solar-to-hydrogen (STH) efficiency

Prediction of STH for unstrained and strained Ag_2Se monolayers were estimated using modified MATLAB coding based on the following:

$$\eta_{STH} = \frac{1.23\int_{E}^{\infty}\frac{P(x)}{x}dx}{\int_{E_g}^{\infty}P(x)dx}$$

where x, P(x) and E are the photon energy hω, AM1.5G solar energy flux at hω and actual photon energy used for water splitting, computed by:

$$E = \begin{cases} E_g, [\chi(H_2) \geq 0.2, \chi(O_2) \geq 0.6] \\ E_g + 0.2 - \chi(H_2), [\chi(H_2) < 0.2, \chi(O_2) \geq 0.6] \\ E_g + 0.6 - \chi(O_2), [\chi(H_2) \geq 0.2, \chi(O_2) < 0.6] \\ E_g + 0.8 - \chi(H_2) - \chi(O_2), [\chi(H_2) < 0.2, \chi(O_2) < 0.6] \end{cases}$$

In this work, $\chi(H_2)$ and $\chi(O_2)$ represent the HER and OER overpotential, forecasted by HSE hybrid functional to be 0.74 (0.36) and 0.71 (0.31) eV for unstrained Ag_2Se (-6% strained) monolayer, respectively.

[1] L. Yu and A. Zunger (2012). Identification of Potential Photovoltaic Absorbers Based on First-Principles Spectroscopic Screening of Materials. *Phys. Rev. Lett.*, 108, 068701.