Electronic Supplementary Material (ESI) for RSC Advances.

Supplementary Information

Two-dimensional $Pd_3(AsSe_4)_2$ as photocatalyst for the solar-driven oxygen evolution reaction: A first principles study

Zhen Gao[a] , Xin He[a] , Wenzhong Li[a] , YaoHe[a,*], and Kai Xiong[b]

[a]*Department of Physics, Yunnan University, Kunming 650091, People's Republic of China.*

[b]*Materials Genome Institute, School of Materials and Energy, Yunnan University, Kunming 650091, People's Republic of China.*

*E-mail: yhe@ynu.edu.cn

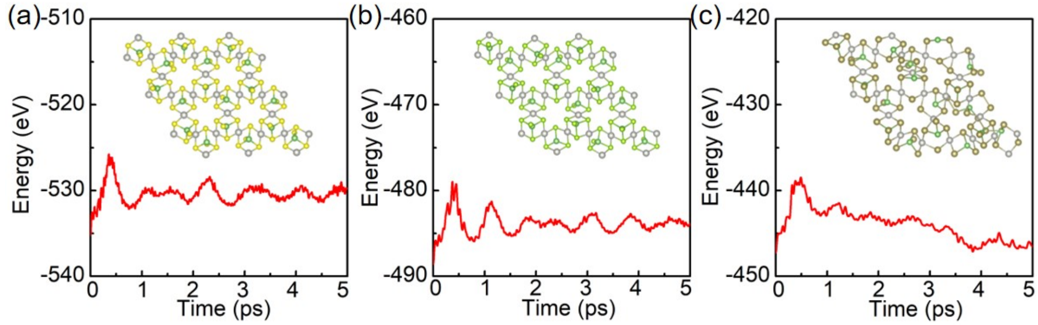

Fig. S1. Variation of the energy as a function of time for the (a) $Pd_3(AsS_4)_2$, (b) $Pd_3(AsSe_4)_2$ and (c) $Pd_3(AsTe_4)_2$ monolayers at 300K, respectively. The insets are the top view of the structure at the end of the AIMD simulation.

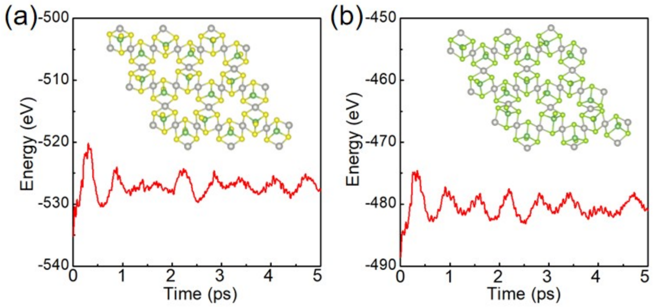

Fig. S2. Variation of the energy as a function of time for the (a) $Pd_3(AsS_4)_2$ and (b) $Pd_3(AsSe_4)_2$ monolayers at 500K, respectively. The insets are the top view of the structure at the end of the AIMD simulation.

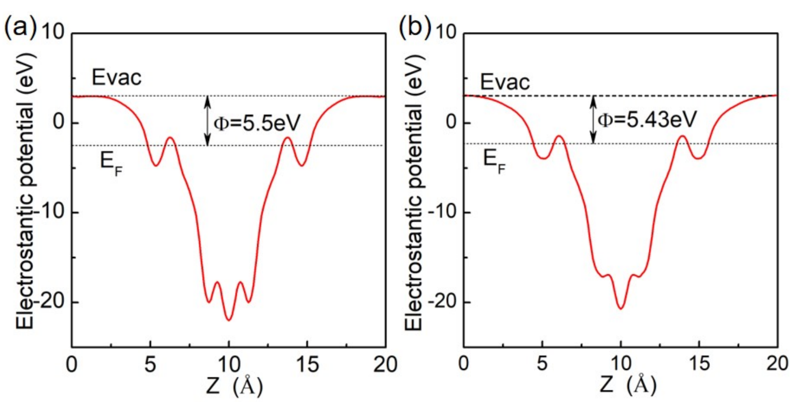

Fig. S3. The electrostatic potential diagrams of (a) $Pd_3(AsS_4)_2$ monolayer and (b) $Pd_3(AsSe_4)_2$ monolayer.

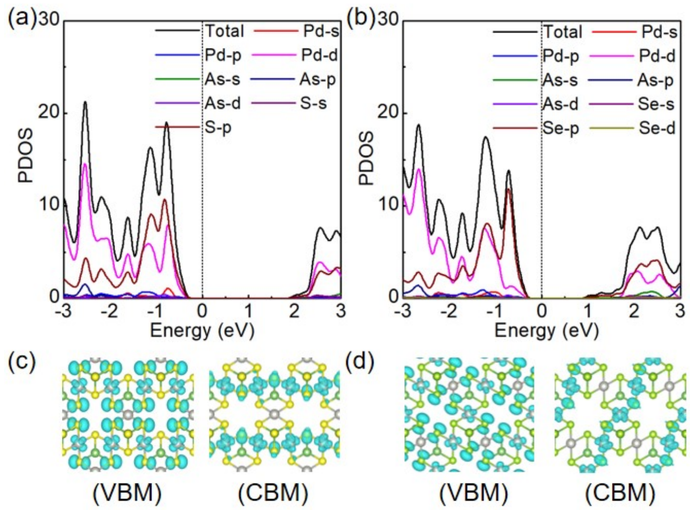

Fig. S4. Projected density of states PDOS of (a) $Pd_3(AsS_4)_2$ and (b) $Pd_3(AsSe_4)_2$ monolayers by the HSE06 method. Partial charge density distributions of the VBM and CBM for (c) $Pd_3(AsS_4)_2$ and (d) $Pd_3(AsSe_4)_2$monolayer by the HSE06 method. The isosurface is set to be 0.006eÅ^{-3}.

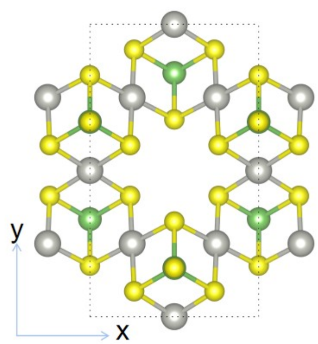

Fig. S5. The calculation of carrier migration rate is carried out in rectangular cell in the black dotted box, the x and y directions also been indicate.

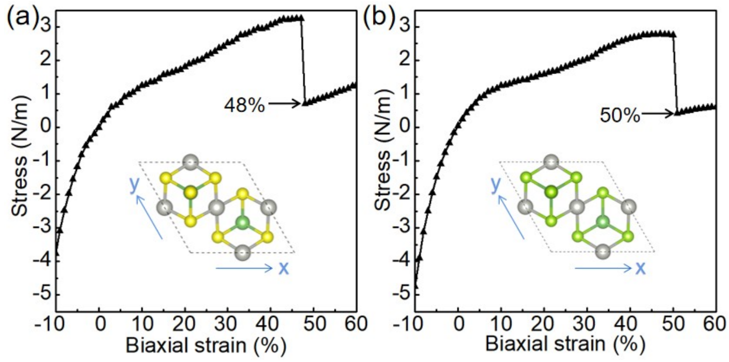

Fig. S6. Stress-strain curves for monolayer (a) $Pd_3(AsS_4)_2$ and (b) $Pd_3(AsSe_4)_2$.

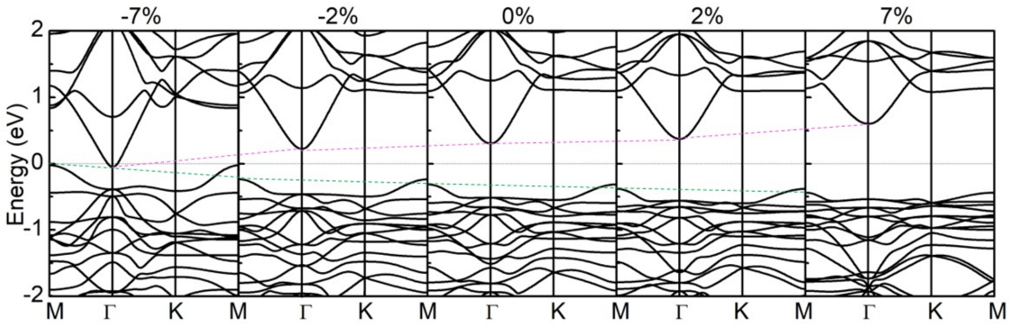

Fig. S7. Strain manipulated semiconductor-to-metal transition in a 2D Pd3(AsSe4)2 monolayer. Positive and negative strains correspond to expansion and compression, respectively. The Fermi energy was set to zero.

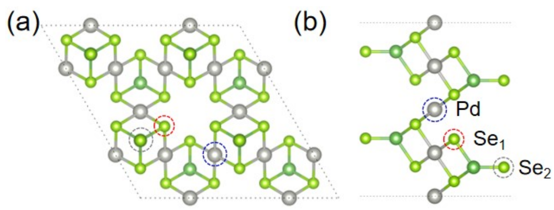

Fig. S8. Schematics for the examined surface sites of $Pd_3(AsSe_4)_2$ monolayer.

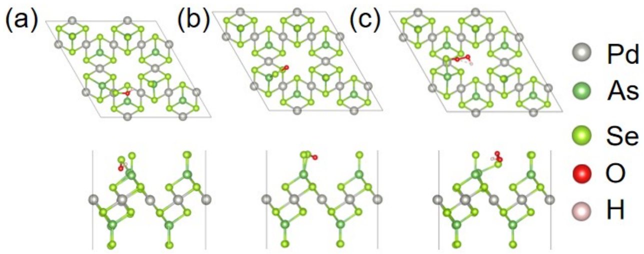

Fig. S9. (a-c)Side and top views of *OH, *O and *OOH oxygenated species on $Pd_3(AsSe_4)_2$, respectively.

Table S1 The elastic constants (in GPa·nm) of the $Pd_3(AsS_4)_2$ and $Pd_3(AsSe_4)_2$ sheets.

Phase	C_{11}	C_{22}	C_{12}	C_{66}
$Pd_3(AsS_4)_2$	22.606	23.182	6.064	7.394
$Pd_3(AsSe_4)_2$	13.074	12.59	6.182	3.232

Table S2 Total energy (E_{total}) and isolated atoms energy (E(Pd), E(As), and E(X=S,Se,Te)) for $Pd_3(AsX_4)_2$ monolayers

Phase	E_{total}	E(Pd)	E(As)	E(X)
$Pd_3(AsS_4)_2$	-61.691	-1.471	-0.178	-0.171
$Pd_3(AsSe_4)_2$	-56.723	-1.471	-0.178	-0.168
$Pd_3(AsTe_4)_2$	-52.299	-1.471	-0.178	-0.164

Table S3 Zero-point energy correction (E_{ZPE}) and entropy contribution (TS, T=300K) of molecules and adsorbates in $Pd_3(AsSe_4)_2$.

Species	E_{ZPE}(eV)	-TS(eV)
H_2	0.27	-0.40
H_2O	0.56	-0.67
O_2*	0.13	-0.15
O*	0.07	-0.06
OH*	0.34	-0.13
OOH*	0.43	-0.21

POSCAR file for $Pd_3(AsS_4)_2$, $Pd_3(AsSe_4)_2$ and $Pd_3(AsTe_4)_2$ structures

```
Pd3(AsS4)2
   1.00000000000000
     6.8818668399406446    0.0000389470571953   -0.0000231569341402
    -3.4408997051555521    5.9598833415797134    0.0000211413885933
    -0.0000666429790095    0.0000318664842659   20.0000000001029328
   As    Pd    S
     2     3     8
Direct
  0.6666662999474667  0.3333336871351923  0.3689681594826828
  0.3333336108537173  0.6666662533490844  0.6310318428069922
  0.5000000404508848  0.4999999933218414  0.5000000146102551
  0.5000001101093139 -0.0000000029229524  0.5000000042330373
  0.0000000470849879  0.5000000374120744  0.4999999808482354
  0.1745624285801977  0.8254382513302255  0.5662950314024020
  0.8254377448465952  0.1745618228567598  0.4337049243921134
  0.1745654739126793  0.3491252099826351  0.5662964650826453
  0.8254345129417553  0.6508747931111123  0.4337035184779852
  0.6508746581225910  0.8254346803666071  0.5662966722820210
  0.3491254179575750  0.1745652636473234  0.4337033841012106
  0.3333333095964030  0.6666656855232748  0.7327134838352044
  0.6666662335958379  0.3333341328868343  0.2672865184452140
```

```
Pd3(AsSe4)2
   1.00000000000000
     7.1796072722415207    0.0000443718012990    0.0000006094197159
    -3.5897652027385183    6.2176548502801197    0.0000143763468671
     0.0000041947447757    0.0000559206460244   19.9999999949340719
   As    Pd    Se
     2     3     8
Direct
  0.6666639500279478  0.3333308587052932  0.3608694336689975
  0.3333359253250735  0.6666690289436394  0.6391305918352747
  0.5000001392268388  0.5000001071719755  0.5000000015368947
  0.5000001315861209  0.0000000633286411  0.5000000320449239
  0.0000000860665329  0.5000000845987206  0.4999999502933594
  0.1712073926584062  0.8287854658342164  0.5694302966195313
  0.8287927177628914  0.1712146128704984  0.4305696808612484
  0.1712132552273379  0.3424229096522065  0.5694309275287405
  0.8287868266130912  0.6575771430622637  0.4305690219952427
  0.6575766964299972  0.8287888531853020  0.5694316375288020
  0.3424234210661334  0.1712112064740306  0.4305684369309101
  0.3333388018659977  0.6666755022874533  0.7481863684107251
  0.6666605441436363  0.3333239718857718  0.2518136207453500
```

$Pd_3(AsTe_4)_2$

```
   1.00000000000000
     7.5689797892637243    0.0000216076909168    0.0000596140111184
    -3.7844716011079242    6.5548912041316703   -0.0000377722140346
     0.0001531040591828   -0.0000129720805295   19.9999994299803880
   As    Pd    Te
     2     3     8
Direct
  0.6666642427628019  0.3333326194283239  0.3462876865428320
  0.3333138640785925  0.6666527993172540  0.6537109105764488
  0.4999894520859402  0.4999944029796883  0.5000005709346875
  0.5000223885334756  0.0000181952777669  0.5000098971631927
  0.0000377981284869  0.5000231356666678  0.4999889153708147
  0.1675986533659438  0.8324000609885931  0.5738688625735854
  0.8323976448471220  0.1675963418822187  0.4261322358277443
  0.1675977843883364  0.3351979864392782  0.5738635903078140
  0.8323988600198249  0.6648007356405918  0.4261306233054991
  0.6647835064585116  0.8323903783616097  0.5738734229386874
  0.3351974569106682  0.1675942520402371  0.4261333678006076
  0.3333340706252402  0.6666655399051337  0.7741914264493726
  0.6666641657950605  0.3333333600726492  0.2258084902087149
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