Supplemental material for:

Large valley splitting and vacancy-induced valley polarization in two-dimensional WSeNH

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Note 1. optimized structure files of pure 2D WSeNH and 2D WSeNH with one hydrogen vacancy:

```
2D WSeNH
1.0
        3.1145000458        0.0000000000        0.0000000000
       -1.5572500229        2.6972361597        0.0000000000
        0.0000000000        0.0000000000       25.0000000000
    H    N    Se    W
    1    1    1     1
Direct
    0.333330005         0.666670024         0.607779980
    0.333330005         0.666670024         0.566820025
    0.333330005         0.666670024         0.450480014
    0.666670024         0.333330005         0.520510018
```

```
2D WSeNH with H vacancy
1.0
       12.4813003540        0.0000000000        0.0000000000
       -6.2406501770       10.8091231788        0.0000000000
        0.0000000000        0.0000000000       25.0000000000
    H    N    Se    W
   15   16   16    16
Direct
    0.083420001         0.166710004         0.546700001
    0.334809989         0.168679997         0.546190023
    0.583869994         0.168679997         0.546190023
    0.833289981         0.166710004         0.546700001
    0.083829999         0.417659998         0.546739995
    0.334809989         0.416130006         0.546190023
    0.831319988         0.416130006         0.546190023
    0.083930001         0.666970015         0.546490014
    0.333029985         0.666970015         0.546490014
    0.583869994         0.665189981         0.546190023
```

|		
0.831319988	0.665189981	0.546190023
0.083829999	0.916170001	0.546739995
0.333029985	0.916069984	0.546490014
0.582340002	0.916170001	0.546739995
0.833289981	0.916580021	0.546700001
0.083360001	0.166679993	0.505710006
0.332969993	0.165930003	0.505209982
0.582970023	0.165930003	0.505209982
0.833320022	0.166679993	0.505710006
0.083640002	0.417270005	0.505739987
0.332969993	0.417030007	0.505209982
0.583329976	0.416669995	0.501290023
0.834070027	0.417030007	0.505209982
0.083810002	0.666909993	0.505500019
0.333090007	0.666909993	0.505500019
0.582970023	0.667029977	0.505209982
0.834070027	0.667029977	0.505209982
0.083640002	0.916360021	0.505739987
0.333090007	0.916190028	0.505500019
0.582729995	0.916360021	0.505739987
0.833320022	0.916639984	0.505710006
0.083439998	0.166720003	0.389499992
0.333400011	0.167009994	0.389629990
0.583609998	0.167009994	0.389629990
0.833280027	0.166720003	0.389499992
0.083420001	0.416830003	0.389200002
0.333400011	0.416390002	0.389629990
0.583329976	0.416669995	0.386790007
0.832989991	0.416390002	0.389629990
0.083180003	0.666589975	0.389149994
0.333409995	0.666589975	0.389149994
0.583609998	0.666599989	0.389629990
0.832989991	0.666599989	0.389629990
0.083420001	0.916580021	0.389200002
0.333409995	0.916819990	0.389149994
0.583169997	0.916580021	0.389200002
0.833280027	0.916559994	0.389499992
0.165940002	0.082570001	0.459450006
0.416390002	0.082769997	0.459360003
0.666630030	0.082570001	0.459450006
0.916670024	0.083329998	0.459520012
0.165940002	0.333370000	0.459450006
0.420370013	0.335189998	0.459619999
0.664810002	0.335180014	0.459619999
0.917429984	0.333370000	0.459450006
0.166740000	0.583490014	0.459190011
0.416390002	0.583609998	0.459360003
0.664810002	0.579630017	0.459619999
0.917230010	0.583609998	0.459360003
0.166740000	0.833249986	0.459190011
0.416509986	0.833249986	0.459190011
0.666630030	0.834060013	0.459450006
0.917429984	0.834060013	0.459450006
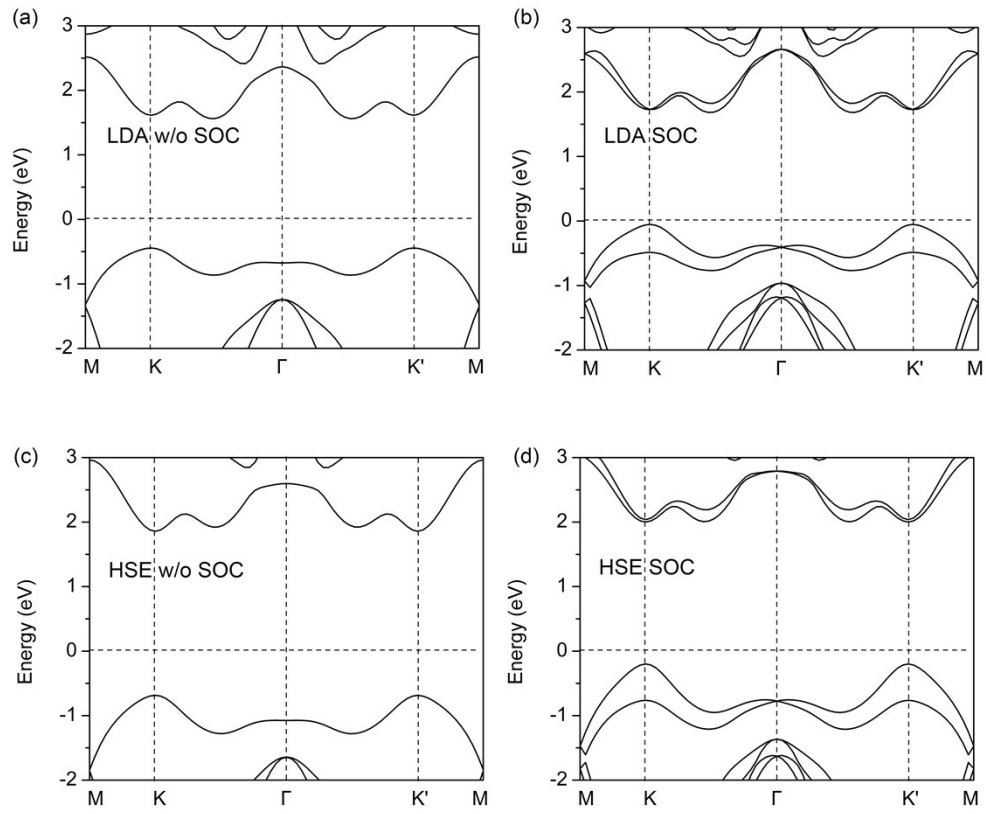
Fig. S1. Band structures of 2D WSeNH using (a-b) LDA and (c-d) HSE functionals.

Table S1. Calculated energy gaps of 2D WSeNH by using different functionals.

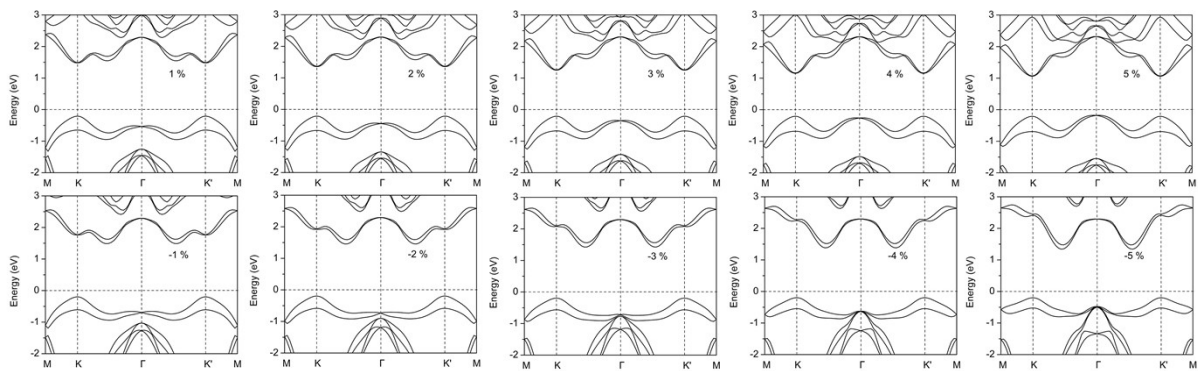|	PBE	LDA	HSE06
Eg (w/o SOC)	2.02	2.00	2.53
Eg (SOC)	1.72	1.73	2.19

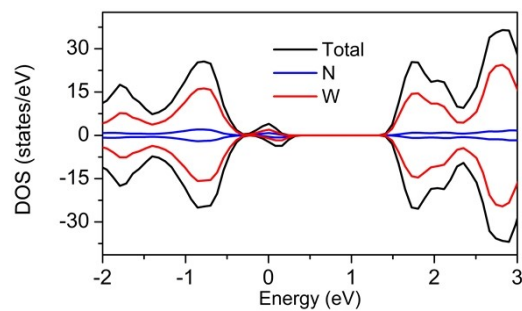Fig. S2. Band structures of 2D WSeNH under external strain in the range of -5% ~ 5%.

Fig. S3. Density of states of 4 × 4 × 1 supercell of 2D WSeNH with a hydrogen vacancy.
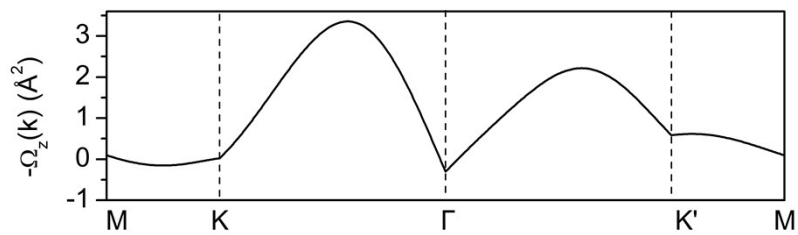

Fig. S4. The Berry curvature of 2D WSeNH with a hydrogen vacancy along high symmetry points.