Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

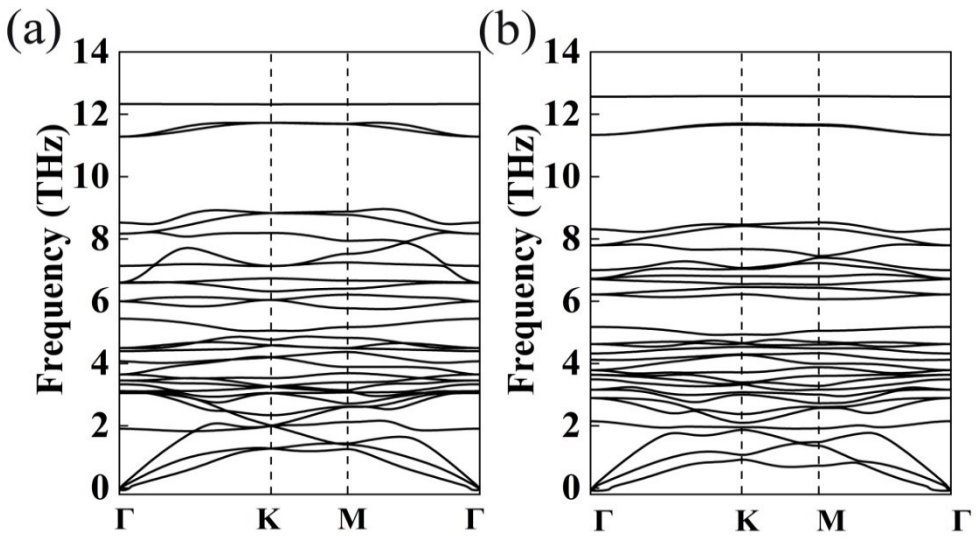

Fig. S1 Phonon spectra of the (a) $Sc_2P_2Se_6$ and (b) $ScCrP_2Se_6$ monolayer

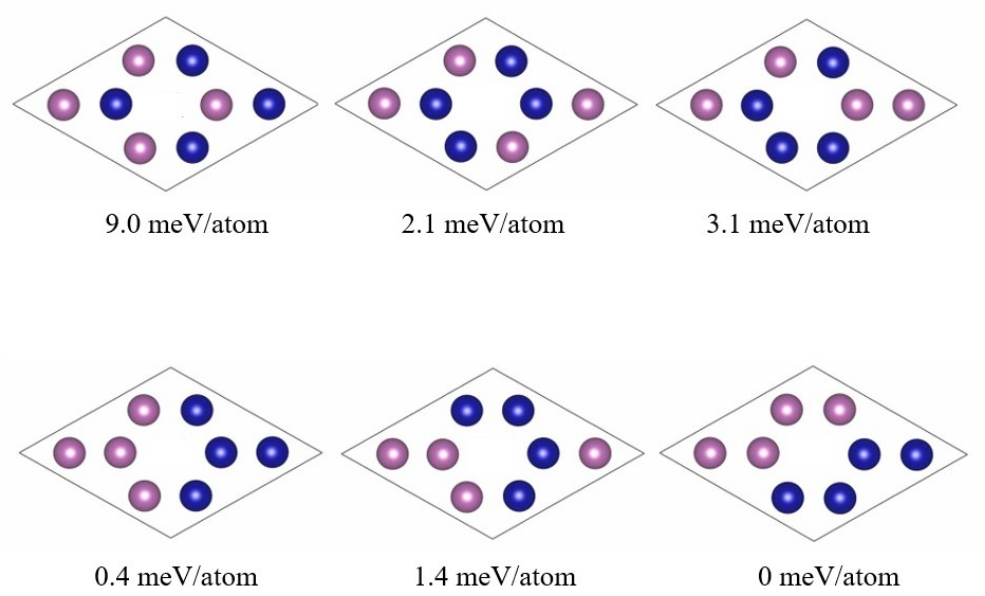

Fig. S2 Different patterns of $ScCrP_2Se_6$ monolayers. The number indicates the relative formation energy.

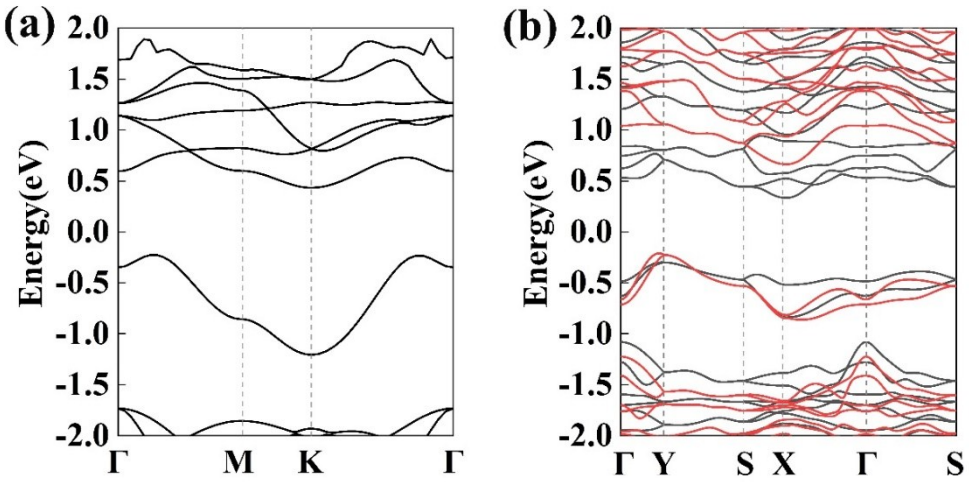

Fig. S3 Band structures of $Sc_2P_2Se_6$ (a) FE and $ScCrP_2Se_6$ (b) AFE phase

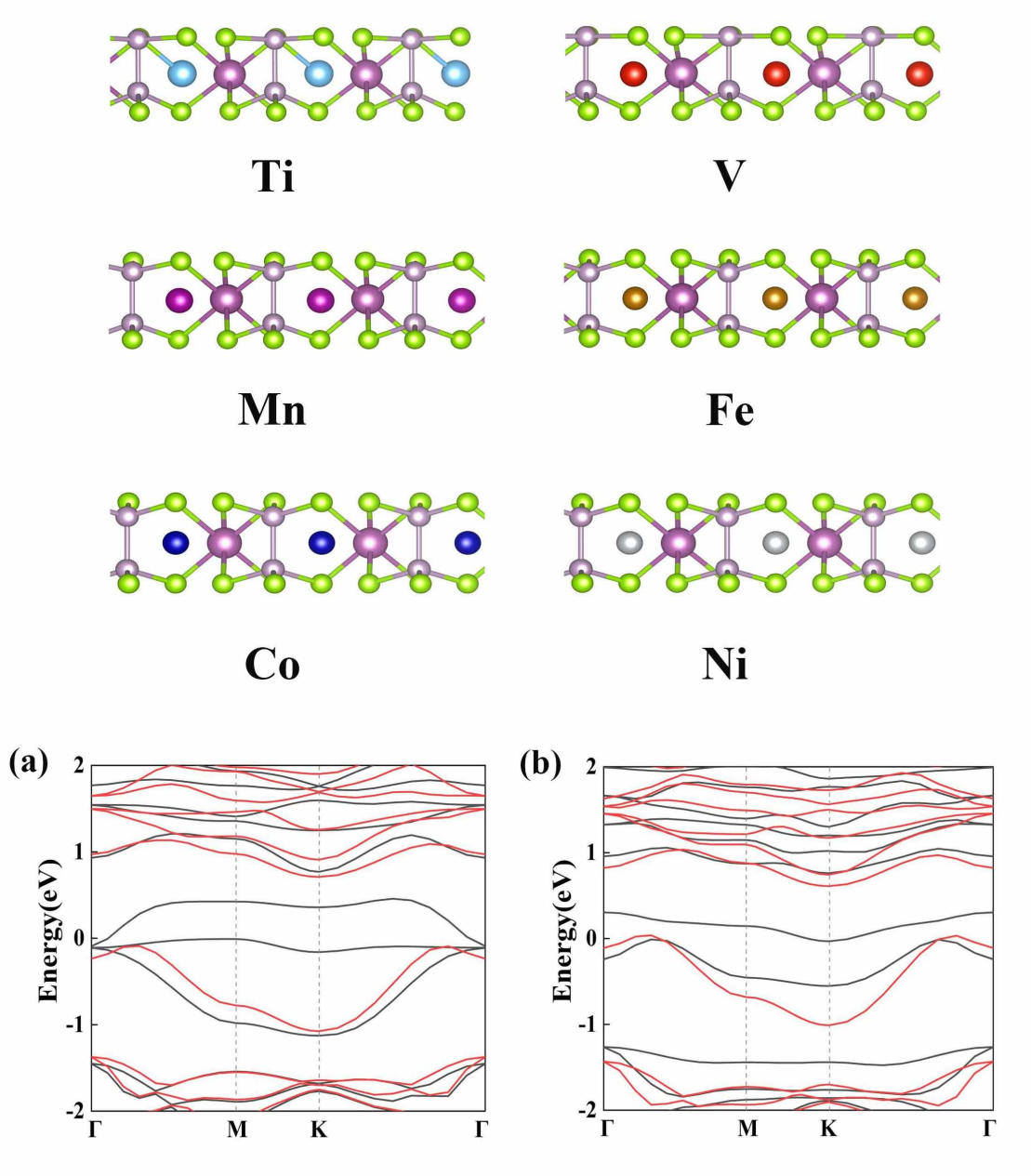

Fig. S4 Optimized structures of the $ScXP_2Se_6$ monolayer with X = Ti, V, Mn, Fe, Co and Ni. The band structures of $ScTiP_2Se_6$ and $ScVP_2Se_6$. Ferromagnetic ordering was found to be energetically most favorable.

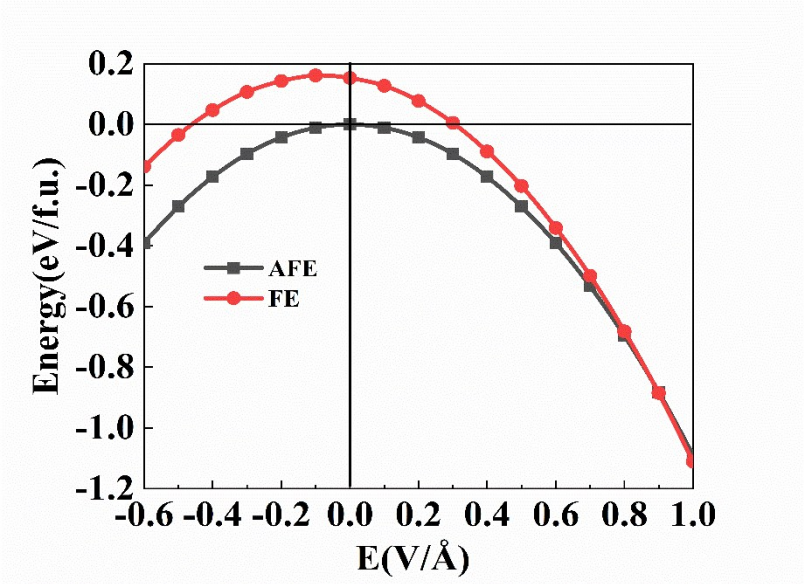

Fig. S5 Energy variation of $ScCrP_2Se_6$ monolayer under external electric field. The energy of AFE phase is set to be zero. Both curves represent FM state.

```
Sc2P2Se6 FE phase (space group: P31m)
1.0
       6.6838002205        0.0000000000        0.0000000000
      -3.3419001102        5.7883407848        0.0000000000
       0.0000000000        0.0000000000       23.2509994507
   Sc    P   Se
    2    2    6
Direct
     0.333333343        0.666666687        0.601080000
     0.666666687        0.333333343        0.601080000
     0.000000000        0.000000000        0.568709970
     0.000000000        0.000000000        0.667680025
     0.688449979        0.688449979        0.530079961
     0.311550021        0.000000000        0.530079961
     0.000000000        0.311550021        0.530079961
     0.364730000        0.364730000        0.669910014
     0.635270000        0.000000000        0.669910014
     0.000000000        0.635270000        0.669910014


Sc2P2Se6 PE phase (space group: P-31m)
1.0
       6.6838002205        0.0000000000        0.0000000000
      -3.3419001102        5.7883407848        0.0000000000
       0.0000000000        0.0000000000       23.2525997162
   Sc     P    Se
    2     2     6
Direct
     0.333333343        0.666666687        0.600179970
     0.666666687        0.333333343        0.600179970
     0.000000000        0.000000000        0.550700009
     0.000000000        0.000000000        0.649659991
     0.661840022        0.661840022        0.530189991
     0.338159978        0.000000000        0.530189991
     0.000000000        0.338159978        0.530189991
     0.338129997        0.338129997        0.670080006
     0.661870003        0.000000000        0.670080006
     0.000000000        0.661870003        0.670080006
```

```
Sc2P2Se6 AFE phase (space group: P1)
1.0
       6.4703001976        0.0000000000        0.0000000000
       0.0000000000       11.8629999161        0.0000000000
       0.0000000000        0.0000000000       23.7810001373
   Sc    P   Se
    4    4   12
Direct
     0.014440000        0.999239981        0.499359995
     0.014410000        0.334100008        0.499300003
     0.485590011        0.499220014        0.518260002
     0.485619992        0.834110022        0.518289983
     0.506160021        0.166600004        0.436430007
     0.507070005        0.166679993        0.532840014
     0.992770016        0.666689992        0.484770000
     0.993879974        0.666649997        0.581149995
     0.351509988        0.971580029        0.436520010
     0.869130015        0.166600004        0.429309994
     0.351440012        0.361779988        0.436500013
     0.676339984        0.314740002        0.571190000
     0.186210006        0.166710004        0.567799985
     0.148530006        0.471599996        0.581040025
     0.823520005        0.518660009        0.446390003
     0.313730001        0.666700006        0.449880004
     0.823549986        0.814740002        0.446410000
     0.148629993        0.861790001        0.581070006
     0.631049991        0.666639984        0.588299990
     0.676329970        0.018610001        0.571210027
```

ScCrP2Se6 FE phase (space group: P3)

```
1.0
       6.5349001884        0.0000000000        0.0000000000
      -3.2674500942        5.6593895744        0.0000000000
       0.0000000000        0.0000000000       23.1893005371
   Sc   Cr    P    Se
    1    1    2     6
Direct
     0.000000000        0.000000000        0.581849992
     0.333333343        0.666666687        0.580139995
     0.666666687        0.333333343        0.550549984
     0.666666687        0.333333343        0.648939967
     0.357250005        0.006640000        0.512290001
     0.993359983        0.350610018        0.512290001
     0.649389982        0.642750025        0.512290001
     0.048310000        0.697780013        0.649809957
     0.302219987        0.350529999        0.649809957
     0.649470031        0.951690018        0.649809957
```

ScCrP2Se6 PE phase (space group: P312)

```
1.0
       6.5349001884        0.0000000000        0.0000000000
      -3.2674500942        5.6593895744        0.0000000000
       0.0000000000        0.0000000000       23.1891002654
   Sc   Cr    P    Se
    1    1    2     6
Direct
     0.000000000        0.000000000        0.580894976
     0.333333343        0.666666687        0.580900013
     0.666666687        0.333333343        0.531684994
     0.666666687        0.333333343        0.630079984
     0.329730004        0.979140005        0.512140006
     0.020859986        0.350590005        0.512140006
     0.649410009        0.670270026        0.512140006
     0.020834991        0.670240014        0.649654984
     0.329759985        0.350594982        0.649654984
     0.649405032        0.979165017        0.649654984
```

```
ScCrP2Se6 AFE phase (space group: P1)
1.0
       6.3017001152        0.0000000000        0.0000000000
       0.0000000000       11.6492996216        0.0000000000
       0.0000000000        0.0000000000       23.7647991180
   Sc   Cr    P   Se
    2    2    4   12
Direct
    0.989820004        0.000980000        0.533680022
    0.510200024        0.500919998        0.502449989
    0.982900023        0.334120005        0.521950006
    0.517159998        0.834129989        0.514249980
    0.493640006        0.165079996        0.496300012
    0.496060014        0.176609993        0.591449976
    0.003980000        0.676720023        0.444689989
    0.006260000        0.665199995        0.539849997
    0.335640013        0.007680000        0.460590005
    0.824580014        0.170049995        0.462139994
    0.308499992        0.310020000        0.455240011
    0.660080016        0.366939992        0.585820019
    0.126139998        0.188869998        0.597299993
    0.164399996        0.507799983        0.575550020
    0.859960020        0.467130005        0.443610013
    0.373879999        0.688929975        0.438879997
    0.839959979        0.867009997        0.450309992
    0.191400006        0.810119987        0.580919981
    0.675320029        0.669990003        0.574020028
    0.640240014        0.967079997        0.592589974
```