

Supplementary Information

Tunable electronic structures in MPX_3 ($M = Zn, Cd$; $X = S, Se$) monolayers by strain engineering

Hui Xiang^a, Bo Xu^{*, a}, Yidong Xia^a, Jiang Yin^{*, a, b}, Zhiguo Liu^{a, b}

^a National Laboratory of Solid State Microstructures and Department of Materials Science and Engineering, Nanjing University, Nanjing, 210093, China

^b Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing, 210093, China

*Corresponding Author: xubonju@gmail.com and jyin@nju.edu.cn.

Table S1 Fractional atomic coordinates of optimized MPX_3 ($M = Zn, Cd$; $X = S, Se$).

(a) $ZnPS_3$

| atoms | $a = b = 6.011 \text{ \AA}, c = 20 \text{ \AA}; \alpha = \beta = 0, \gamma = 120^\circ$ | | |
|-------|---|--------------------|--------------------|
| Zn | 0.0000000000000000 | 0.0000000000000000 | 0.4846545449292046 |
| Zn | 0.6666666666666667 | 0.3333333333333333 | 0.4850448430707976 |
| P | 0.3333333333333333 | 0.6666666666666667 | 0.4295803052753951 |
| P | 0.3333333333333333 | 0.6666666666666667 | 0.5401190837246037 |
| S | 0.3335097355927447 | 0.9955908975003060 | 0.4040053479529391 |
| S | 0.0044091024996939 | 0.3379188447681581 | 0.4040053479529391 |
| S | 0.6620811552318420 | 0.6664902577315357 | 0.4040053479529391 |
| S | 0.3331569243982000 | 0.3377424358330295 | 0.5656940409544120 |
| S | 0.6622575641669703 | 0.9954144885651777 | 0.5656940409544120 |
| S | 0.0045855114348224 | 0.6668430756018000 | 0.5656940409544120 |

(b) $ZnPSe_3$

| atoms | $a = b = 6.357 \text{ \AA}, c = 20 \text{ \AA}; \alpha = \beta = 0, \gamma = 120^\circ$ | | |
|-------|---|--------------------|--------------------|
| Zn | 0.0000000000000000 | 0.0000000000000000 | 0.4850713578594509 |
| Zn | 0.6666666666666667 | 0.3333333333333333 | 0.4846280301405513 |
| P | 0.3333333333333333 | 0.6666666666666667 | 0.4288297908057675 |
| P | 0.3333333333333333 | 0.6666666666666667 | 0.5408695981942314 |
| Se | 0.3328026193757395 | 0.0033116235479310 | 0.4000175915059568 |
| Se | 0.9966883764520691 | 0.3294910025035210 | 0.4000175915059568 |
| Se | 0.6705089974964790 | 0.6671973739485408 | 0.4000175915059568 |
| Se | 0.3338640406152052 | 0.3300217097854047 | 0.5696817974013944 |
| Se | 0.6699782902145952 | 0.0038423308298147 | 0.5696817974013944 |
| Se | 0.9961576691701853 | 0.6661359593847949 | 0.5696817974013944 |

(c) CdPS₃

| atoms | $a = b = 6.300 \text{ \AA}, c = 20 \text{ \AA}; \alpha = \beta = 0, \gamma = 120^\circ$ | | |
|-------|---|--------------------|--------------------|
| Cd | 0.0000000000000000 | 0.0000000000000000 | 0.4852030932124268 |
| Cd | 0.6666666666666667 | 0.3333333333333333 | 0.4844962947875754 |
| P | 0.3333333333333333 | 0.6666666666666667 | 0.4284349885625087 |
| P | 0.3333333333333333 | 0.6666666666666667 | 0.5412644004374902 |
| S | 0.3326275765813610 | 0.9771789019852107 | 0.3995776472129590 |
| S | 0.0228210980147894 | 0.3554486812718628 | 0.3995776472129590 |
| S | 0.6445513187281372 | 0.6673724167429195 | 0.3995776472129590 |
| S | 0.3340390834095838 | 0.3561544313481252 | 0.5701217416943923 |
| S | 0.6438455686518750 | 0.9778846520614729 | 0.5701217416943923 |
| S | 0.0221153479385271 | 0.6659609165904162 | 0.5701217416943923 |

(d) CdPSe₃

| atoms | $a = b = 6.618 \text{ \AA}, c = 20 \text{ \AA}; \alpha = \beta = 0, \gamma = 120^\circ$ | | |
|-------|---|--------------------|--------------------|
| Cd | 0.0000000000000000 | 0.0000000000000000 | 0.4852089136771441 |
| Cd | 0.6666666666666667 | 0.3333333333333333 | 0.4844904743228581 |

| | | | |
|----|--------------------|--------------------|--------------------|
| P | 0.3333333333333333 | 0.6666666666666667 | 0.4277471860140425 |
| P | 0.3333333333333333 | 0.6666666666666667 | 0.5419522029859563 |
| Se | 0.3325285778994166 | 0.9861077254493626 | 0.3952513264221017 |
| Se | 0.0138922745506374 | 0.3464208591257664 | 0.3952513264221017 |
| Se | 0.6535791408742335 | 0.6674714154248639 | 0.3952513264221017 |
| Se | 0.3341380820915281 | 0.3472256078839731 | 0.5744480624852494 |
| Se | 0.6527743921160269 | 0.9869124742075692 | 0.5744480624852494 |
| Se | 0.0130875257924307 | 0.6658619179084718 | 0.5744480624852494 |

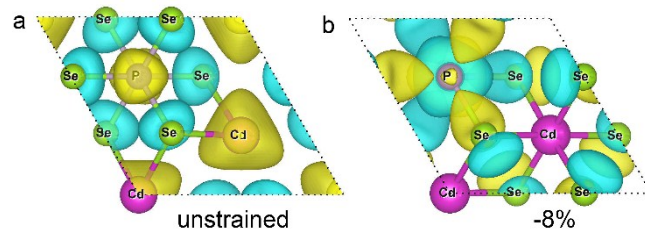


Figure S1 Spatial distributions of wave functions for the bottom of CBM of CdPSe₃. (a) unstrained state, and (b) compressive strain at 8%.

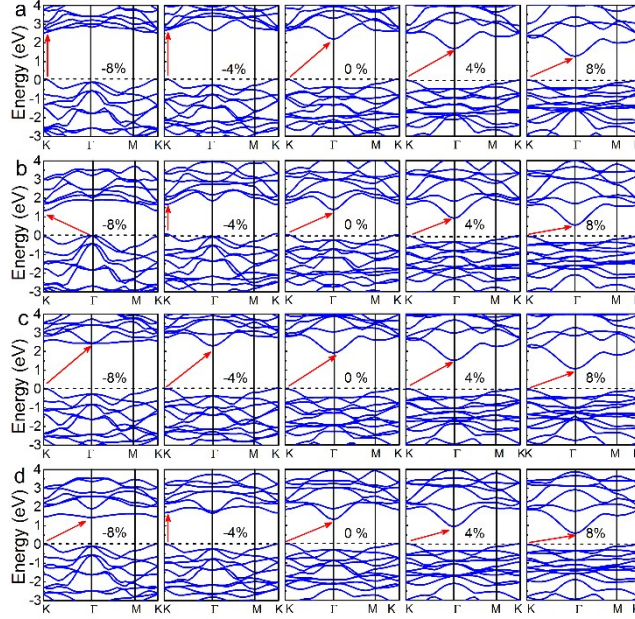


Figure S2 The evolution of band structures calculated by PBE functional under biaxial strain -8%, -4%, 0, 4%, and 8%, where the label from (a) to (d) represents ZnPS₃ (a), ZnPSe₃ (b), CdPS₃ (c), and CdPSe₃ (d), respectively. Red arrows represent the direction of band gap from the VBM to the CBM. The Fermi level is set to zero.

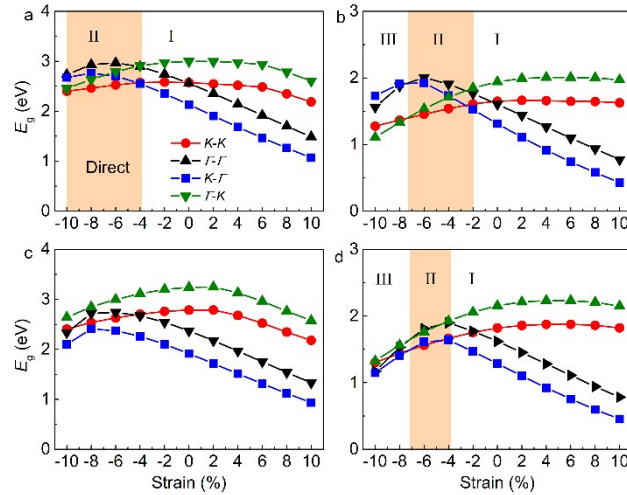


Figure S3 The change of energy gaps of (a) ZnPS₃, (b) ZnPSe₃, (c) CdPS₃ and (d) CdPSe₃ by PBE functional, including $\Gamma \rightarrow \Gamma$ (black), $K \rightarrow K$ (red), $K \rightarrow \Gamma$ (blue), and $\Gamma \rightarrow K$ (green), where the region of I, II, and III represents the indirect, direct, and indirect band gap, respectively.