

Supplemental Information:

Mn₂P₂S₃Se₃: two-dimensional Janus room-temperature antiferromagnetic semiconductor with a large out-of-plane piezoelectricity

Peng Jiang,¹ Xiaohong Zheng,^{2, a)} Lili Kang,³ Xixi Tao,⁴ Hongmei Huang,¹ Xiaochen Dong,⁵ and Yan-Ling Li^{1, b)}

¹⁾*School of Physics and Electronic Engineering, Jiangsu Normal University, Xuzhou 221116, China*

²⁾*College of Information Science and Technology, Nanjing Forestry University, Nanjing 210037, China*

³⁾*Institute for Computational Materials Science, School of Physics and Electronics, Henan University, Kaifeng 475004, China*

⁴⁾*School of Electronic and Information Engineering, Changshu Institute of Technology, Changshu 215500, China*

⁵⁾*School of Chemistry and Materials Science, Jiangsu Normal University, Xuzhou 221116, China*

SI. SUPPLEMENTAL RESULTS

TABLE S1. The energy for different magnetic configurations with respect to the Néel-AFM configuration, the absolute magnetic moment of the Mn atom, exchange coupling constants, and magnetic critical temperatures calculated by GGA+U and HSE06 functionals. Note that critical temperatures are estimated from exchange coupling constants based on the mean-field formula. The unit cell is a square cell in the table.

	GGA + U (U=3.0 eV)	HSE06
E _{nAFM} (meV/u.c.)	0.00	0.00
E _{zAFM} (meV/u.c.)	50.11	45.85
E _{sAFM} (meV/u.c.)	64.56	54.40
E _{FM} (meV/u.c.)	131.70	111.96
M _{Mn} (μB)	4.49	4.51
J ₁ (meV)	-1.45	-1.27
J ₂ (meV)	-0.11	-0.07
J ₃ (meV)	-0.72	-0.56
T _{Néel} (K)	315	275

^{a)}Electronic mail: xhzheng@njfu.edu.cn

^{b)}Electronic mail: ylli@jsnu.edu.cn

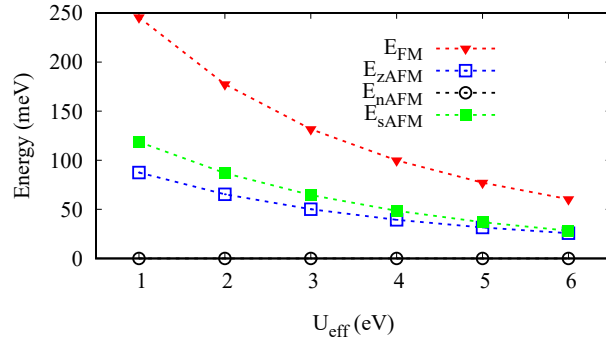


FIG. S1. The magnetic energy for nAFM, zAFM, sAFM, and FM configurations with respect to the nAFM configuration of 2D Janus $\text{Mn}_2\text{P}_2\text{S}_3\text{Se}_3$ monolayer.

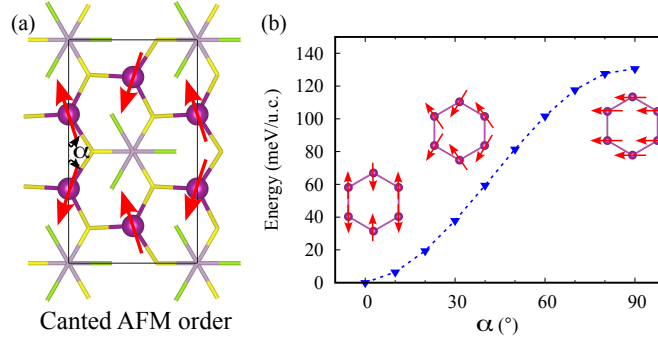


FIG. S2. (a) The canted spin configuration and (b) its magnetic energy as a function of the canted angle α with respect to the nAFM configuration of 2D Janus $\text{Mn}_2\text{P}_2\text{S}_3\text{Se}_3$ monolayer. Here, $\alpha=0/90^\circ$ denotes the nAFM/FM configuration.

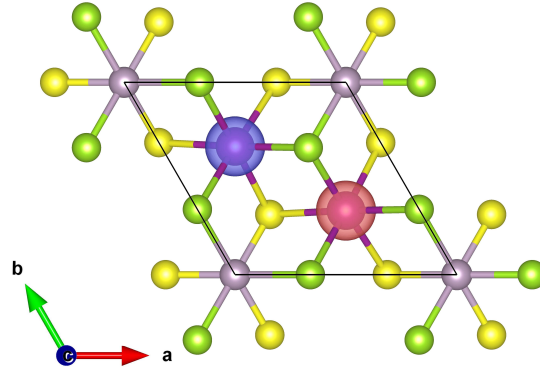


FIG. S3. 3D real-space spin charge density of 2D Janus $\text{Mn}_2\text{P}_2\text{S}_3\text{Se}_3$ monolayer. Red and blue colors denote spin-up and spin-down components, respectively.

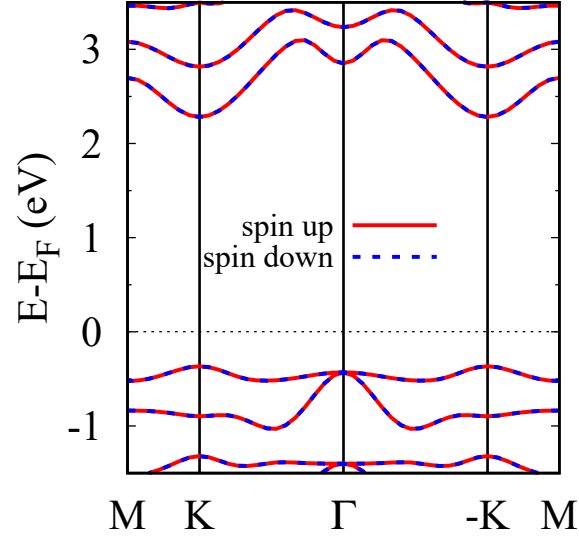


FIG. S4. The spin-polarized band structure calculated by HSE06 functional of 2D Janus $\text{Mn}_2\text{P}_2\text{S}_3\text{Se}_3$ monolayer.

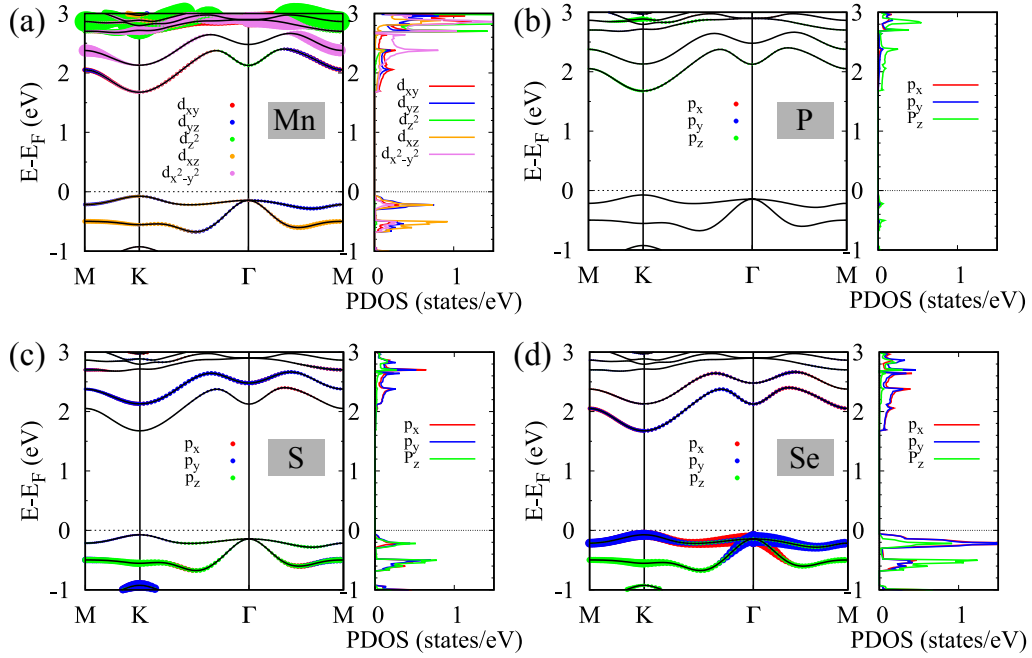


FIG. S5. The orbital-projected band structures and density of states of different elements for 2D Janus $\text{Mn}_2\text{P}_2\text{S}_3\text{Se}_3$ monolayer: (a) Mn; (b) P; (c) S; (d) Se.

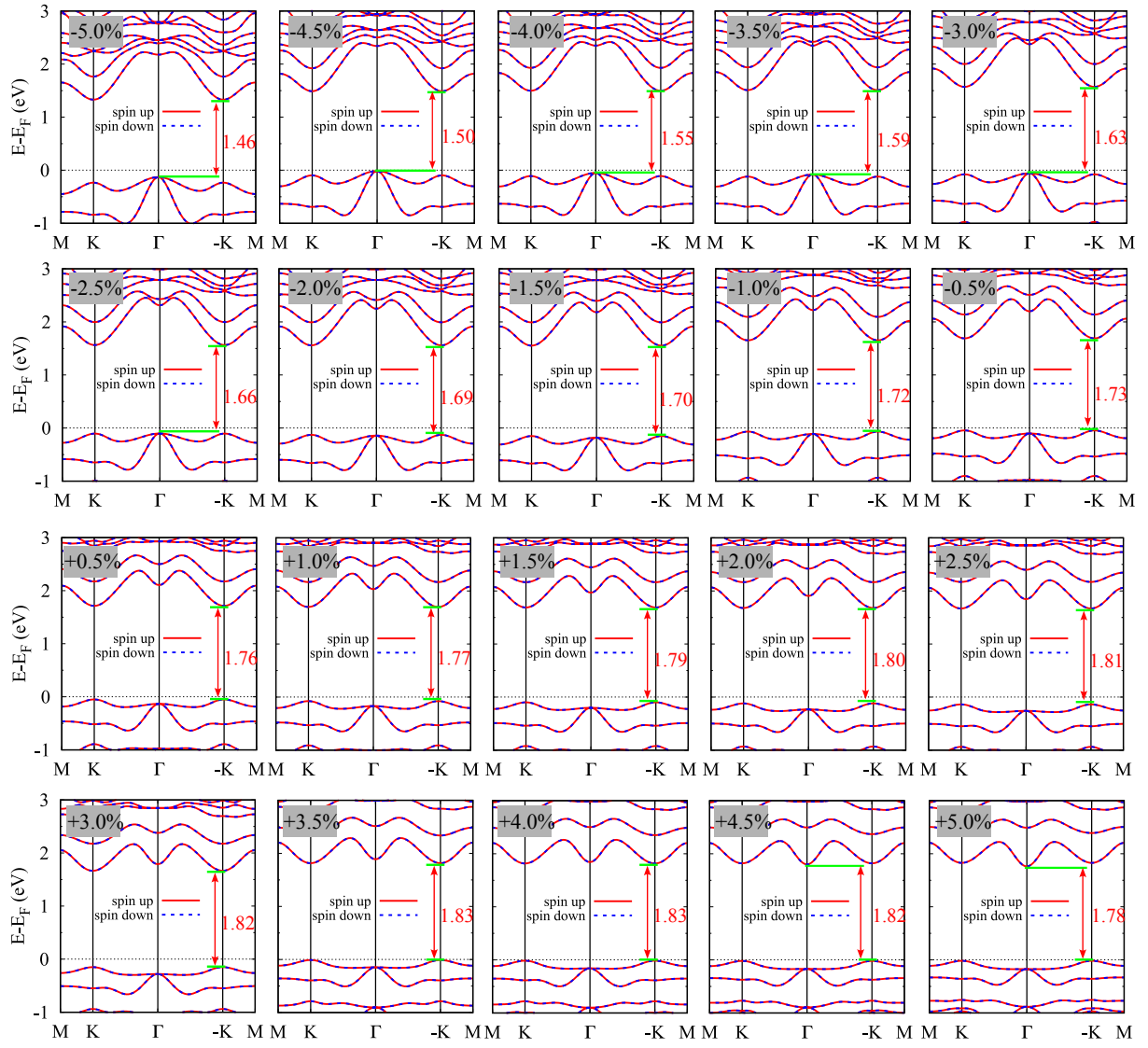


FIG. S6. (color online) The spin-polarized band structures of the Janus $\text{Mn}_2\text{P}_2\text{S}_3\text{Se}_3$ monolayer at different strains. The positive and negative values denotes biaxial tensile and compressive strains, respectively.

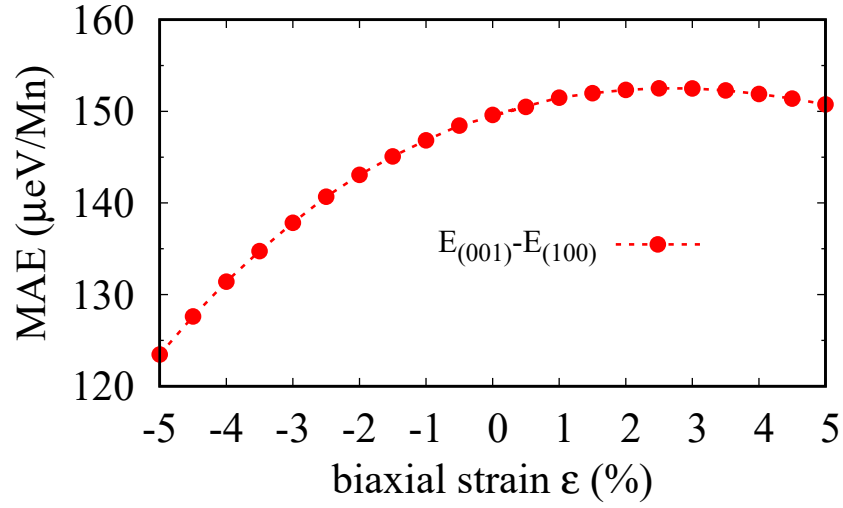


FIG. S7. (color online) The strain dependence of MAE ($E_{(001)} - E_{(100)}$) for Janus $\text{Mn}_2\text{P}_2\text{S}_3\text{Se}_3$ monolayer.

SII. THE SUPPLEMENTAL STRUCTURE FILE IN VASP FORMAT

```

Mn2P2S3Se3
1.0000000000000000
6.2865744093688845 0.0000000000000000 0.0000000000000000
-3.1432872046844422 5.4443331412946057 0.0000000000000000
-0.0000000000000000 0.0000000000000000 30.0000000000000000
Mn P S Se
2 2 3 3
Direct
0.6666666666666643 0.3333333333333357 0.4962532330799497
0.3333333333333357 0.6666666666666643 0.4962532330799497
0.0000000000000000 -0.0000000000000000 0.4638037642087067
0.0000000000000000 -0.0000000000000000 0.5380608299458799
0.6867702579917774 -0.0000000000000000 0.4448948100426130
0.0000000000000000 0.6867702579917774 0.4448948100426130
0.3132297420082227 0.3132297420082227 0.4448948100426130
0.3395913695870342 -0.0000000000000000 0.5569815025192225
-0.0000000000000000 0.3395913695870342 0.5569815025192225
0.6604086304129659 0.6604086304129659 0.5569815025192225

```