Supplementary Information (SI) for Nanoscale Advances.

Support Information

Nonvolatile metal-semiconductor transition of valley in two-dimension ferrovalley/ferroelectric van der Waals heterostructures

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Section S1. Calculation of the resistance-area product

The resistance-area (RA) product can be calculated from transmission by definition:

$$\mathrm{RA} = \frac{A}{G} = \frac{A}{T(\varepsilon_f)G_0} \tag{S1.1}$$

where A is the unit cell area, $T(\varepsilon_f)$ is the calculated transmission at the Fermi energy, and $G_0 = \frac{e^2}{h} = \frac{1}{25.8k\Omega}$ is the spin-conductance quantum. For the system we calculated, the lattice constant is $\mathrm{a} = 3.39$ Å. For example, in the zigzag direction, the area of the unit cell is given by $A = a * d = 3.39d$ Å^2, where d is the thickness of the atomic layer, which can be obtained by subtracting the coordinates of the top and bottom atoms. In the armchair direction, the area of the unit cell is $A = \sqrt{3}a * d = 5.87d$ Å^2. The RA product is then given by:

$$\mathrm{RA} = \frac{A}{G} = \frac{A}{T(\varepsilon_f)G_0} = \frac{25.8\times A}{T(\varepsilon_f)} k\Omega \cdot \text{Å}^2 = \frac{25.8\times 10^{-5}\times A}{T(\varepsilon_f)} \Omega \cdot \mu\mathrm{m}^2 \tag{S1.2}$$

Section S2. The final atomic positions and lattice vectors of TNF monolayer, SCO monolayer and four TNF/SCO vdW heterostructures.

1. TNF↑

```
   1.00000000000000
     2.8525642087844436    -1.6469288392129773     0.0000000000000000
     0.0000000000000000     3.2938576784259546     0.0000000000000000
     0.0000000000000000     0.0000000000000000    20.0000000000000000
   N    F    Ta
     1    1    1
Direct
  0.6666669999999968  0.3333330000000032  0.4566388433762541
  0.3333330000000032  0.6666669999999968  0.5538470453753345
 -0.0000000000000000 -0.0000000000000000  0.4895141112484113
```

2. SCO↑

```
   1.00000000000000
     2.9671154170146758    -1.7130647413109668     0.0000000000000000
     0.0000000000000000     3.4261294826219335     0.0000000000000000
     0.0000000000000000     0.0000000000000000    20.0000000000000000
   C    O    Sc
     1    2    2
Direct
  0.0000000000000000 -0.0000000000000000  0.5203736815113420
  0.6666669999999968  0.3333330000000032  0.4085382197626271
  0.0000000000000000 -0.0000000000000000  0.6049116164365941
  0.3333330000000032  0.6666669999999968  0.4417327216894756
  0.6666669999999968  0.3333330000000032  0.5696207605999714
```

3. TNF↑/SCO↓

```
   1.00000000000000
     2.9380690000000000    -1.6962950000000001     0.0000000000000000
     0.0000000000000000     3.3925900000000002     0.0000000000000000
     0.0000000000000000     0.0000000000000000    26.0000000000000000
   C    N    O    F    Sc   Ta
     1    1    2    1    2    1
Selective dynamics
Direct
  0.3333330000000032  0.6666669999999968  0.4018817597412522
  0.6666669999999968  0.3333330000000032  0.5789255701157606
  0.3333330000000032  0.6666669999999968  0.3361118238950393
 -0.0000000000000000  0.0000000000000000  0.4873772050549482
```

0.3333330000000032 0.6666669999999968 0.6479973578655165
0.6666669999999968 0.3333330000000032 0.4620709914703161
-0.0000000000000000 0.0000000000000000 0.3634750111655696
-0.0000000000000000 0.0000000000000000 0.6010252806916092

4. TNF↑/SCO↑

```
   1.00000000000000
     2.9291580000000002    -1.6911499999999999     0.0000000000000000
     0.0000000000000000     3.3822999999999999     0.0000000000000000
     0.0000000000000000     0.0000000000000000    26.0000000000000000
   C    N    O    F    Sc   Ta
     1    1    2    1    2    1
Direct
 -0.0000000000000000 -0.0000000000000000  0.4274993265240815
  0.6666669999999968  0.3333330000000032  0.5859287018233326
 -0.0000000000000000 -0.0000000000000000  0.4930581146429607
  0.6666669999999968  0.3333330000000032  0.3407483456099062
  0.3333330000000032  0.6666669999999968  0.6560064063094705
  0.6666669999999968  0.3333330000000032  0.4673263553548534
  0.3333330000000032  0.6666669999999968  0.3673276716081885
 -0.0000000000000000 -0.0000000000000000  0.6083980781272041
```

5. TNF↓/SCO↓

```
   1.00000000000000
     2.9354360000000002    -1.6947749999999999     0.0000000000000000
     0.0000000000000000     3.3895499999999998     0.0000000000000000
     0.0000000000000000     0.0000000000000000    26.0000000000000000
   C    N    O    F    Sc   Ta
     1    1    2    1    2    1
Direct
  0.3333040388434474  0.6666540194217256  0.4162766234140632
  0.3333665017016829  0.6666837508508368  0.6493857332843690
  0.3333730708782774  0.6666900354391370  0.3504175343218761
  0.0000659461597971  0.0000344730798948  0.5022227072513101
  0.6676579150513836  0.3338299575256928  0.5797296298634799
 -0.0000203498542664 -0.0000086749271369  0.3777691399484680
  0.6666358406658374  0.3333189203329197  0.4766117471717687
 -0.0010569634461529 -0.0005284817230764  0.6269298847446647
```

6. TNF↓/SCO↑

```
   1.00000000000000
```

```
        2.9327079999999999    -1.6932000000000000     0.0000000000000000
        0.0000000000000000     3.3864000000000001     0.0000000000000000
        0.0000000000000000     0.0000000000000000    26.0000000000000000
    C     N     O     F    Sc    Ta
      1     1     2     1     2     1
Direct
  -0.0000000000000000 -0.0000000000000000  0.4329215943263525
   0.3333330000000032  0.6666669999999968  0.6464688320544723
  -0.0000000000000000 -0.0000000000000000  0.4978971593854610
0.6666669999999968  0.3333330000000032  0.3461694160477087
0.6666669999999968  0.3333330000000032  0.5764359410339306
0.3333330000000032  0.6666669999999968  0.3726273866807242
   0.6666669999999968  0.3333330000000032  0.4724955121558322
  -0.0000000000000000 -0.0000000000000000  0.6248281583155205
```

Tables:

Table S1 Calculated cleavage energy of the TNF/SCO multiferroic vdW heterostructures. The red numbers in the table indicate the most stable configurations.

E_c (eV/ Å^2)	①	②	③
TNF↑/SCO↓	-0.007	-0.014	-0.002
TNF↑/SCO↑	-0.010	-0.014	-0.004
TNF↓/SCO↓	-0.011	-0.019	-0.006
TNF↓/SCO↑	-0.012	-0.019	-0.008

Table S2 Summary of transmission data for the TNF↓/SCO heterostructure, including the transmission for Zigzag and Armchair direction, TER, and RA.

	Zigzag		Armchair	
	TNF↓/SCO↓	TNF↓/SCO↑	TNF↓/SCO↓	TNF↓/SCO↑
T	0	0.31	0	5.49
RA (Ω·μm^2)	∞	2.20×10^{-2}	∞	2.15×10^{-3}
TER	∞		∞	

Figures:

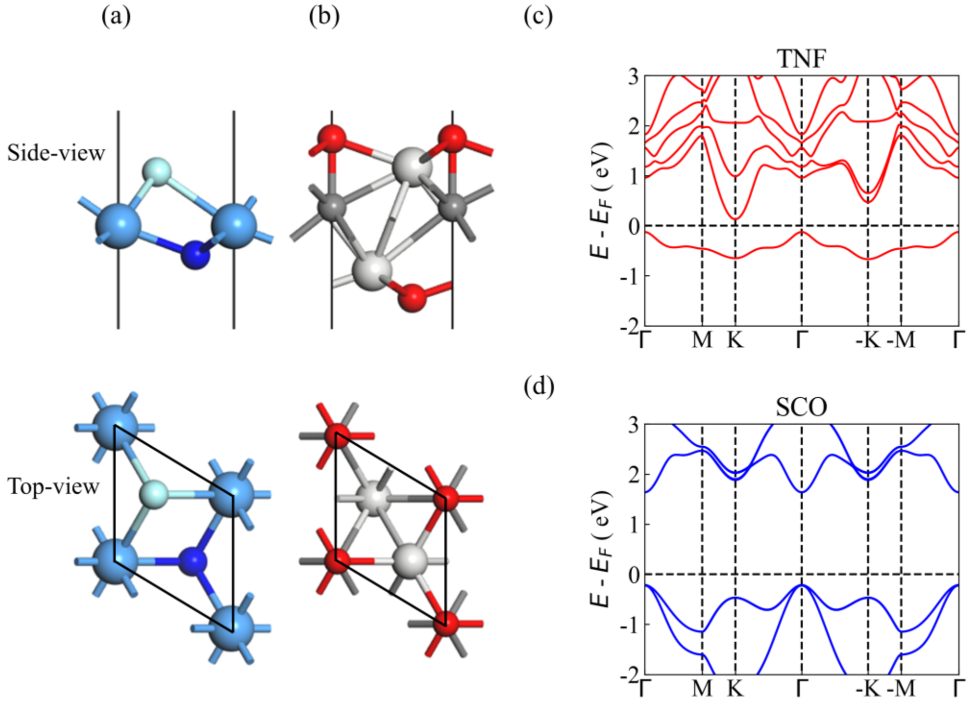

Figure S1. Crystal structures of the TNF and SCO monolayers. Side view and top view of the structures of single layers of (a) TNF and (b) SCO. The blank line represents the unit cell. Band structures of (c) TNF monolayer and (d) SCO monolayer.

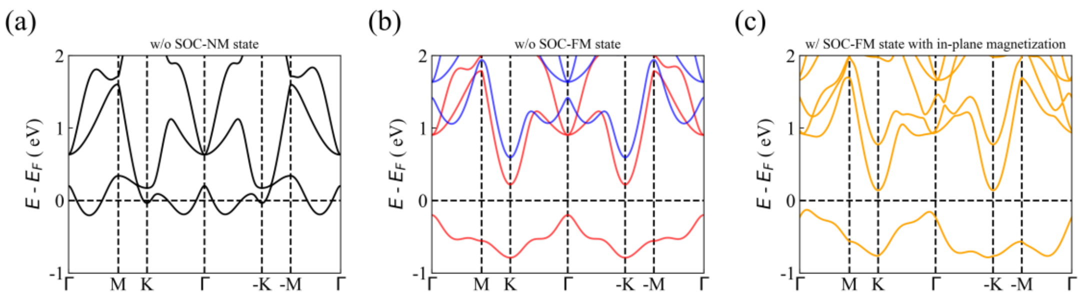

Figure S2. Band structures of TNF (a) Without SOC, NM state. (b) Without SOC, FM state. Red and blue denote the spin-up and spin-down, respectively. (c) With SOC, FM state with magnetic moment in the plane.

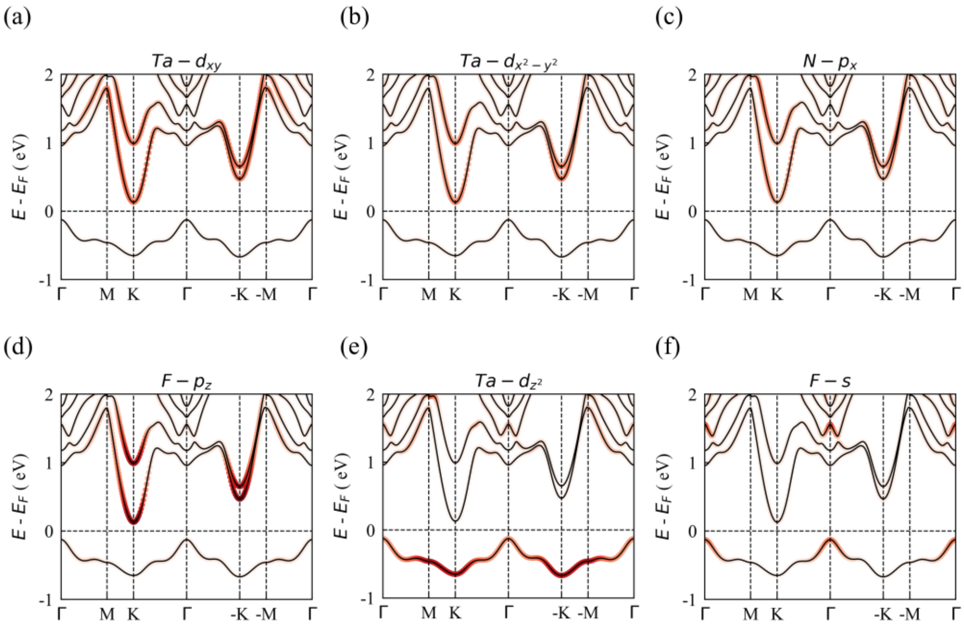

Figure S3. The projected orbital components of TNF monolayer. (a) Ta-d_{xy}, (b) Ta-$d_{x^2-y^2}$, (c) N-p_x, (d) F-p_z, (e) Ta- d_{z^2}, (f) F-s. The size and color depth of the red dot represent the degree of contribution.

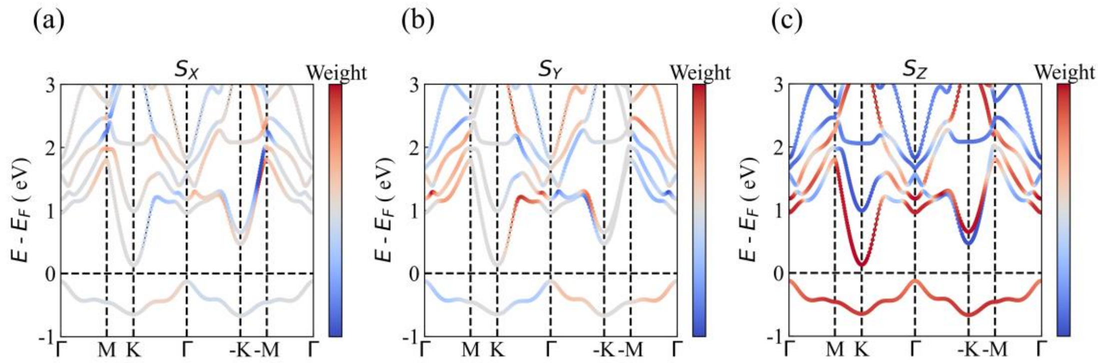

Figure S4. The spin projection bands in different directions of TNF. (a) S_x, (b) S_y, (c) S_z. Red and blue represent the positive and negative directions of spin respectively, and depth of the color represents the contribution of the spin in the corresponding direction.

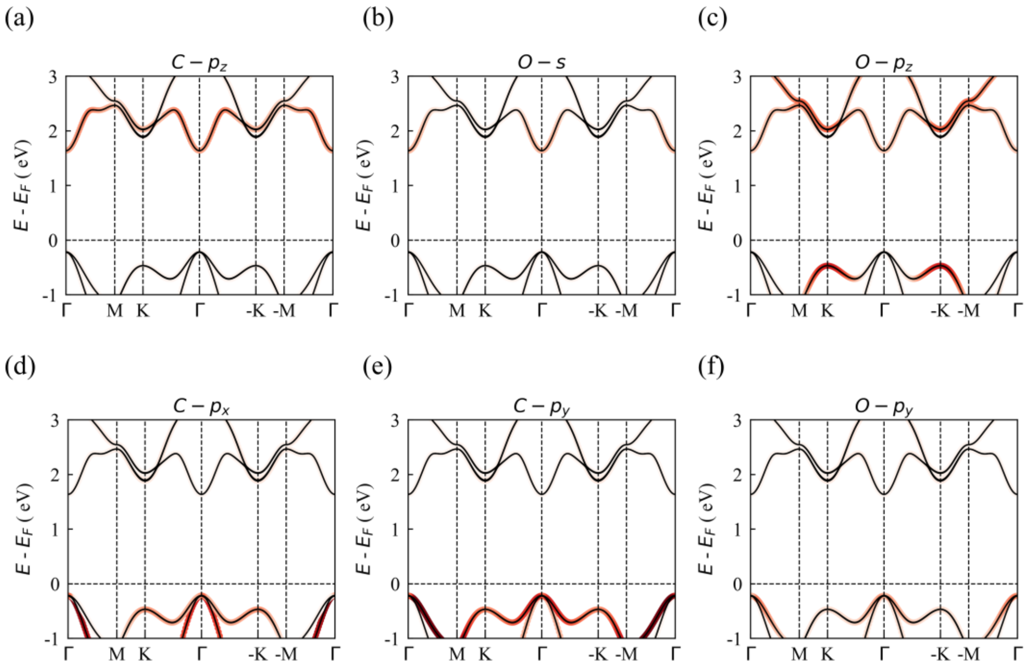

Figure S5. The projected orbital components of SCO monolayer. (a) C- p_z, (b) O-s, (c) O-p_z, (d) C-p_x, (e) C- d_y, (f) O-p_y. The size and color depth of the red dot represent the degree of contribution.

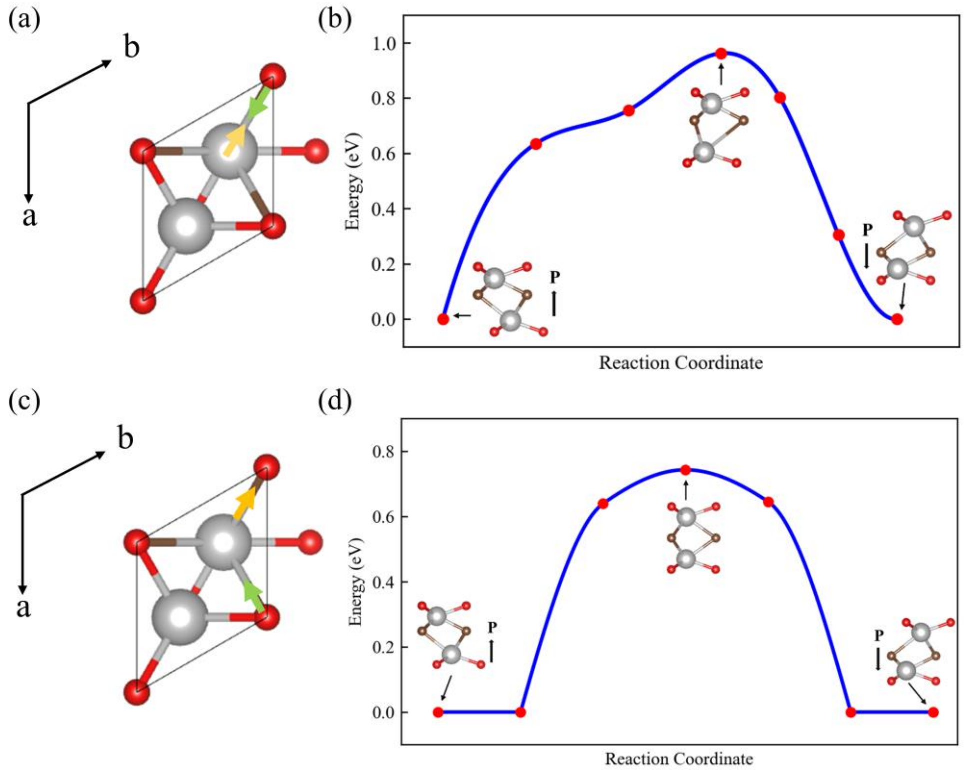

Figure S6. Kinetic pathways of polarization reversal processes. (a) and (c) show the top view of the SCO↑ monolayer. The green arrow attached to the atoms indicates the direction of the movement of C atoms during the polarization reversal process, and the yellow arrow indicates the movement directions of the lower Sc atoms and O atoms. Top views (a) and (c) correspond to the kinetic pathways in (b) and (c), respectively. (b) Path 1 of SCO from P↑ to P↓ states. (c) Path 2 of SCO from P↑ to P↓ states.

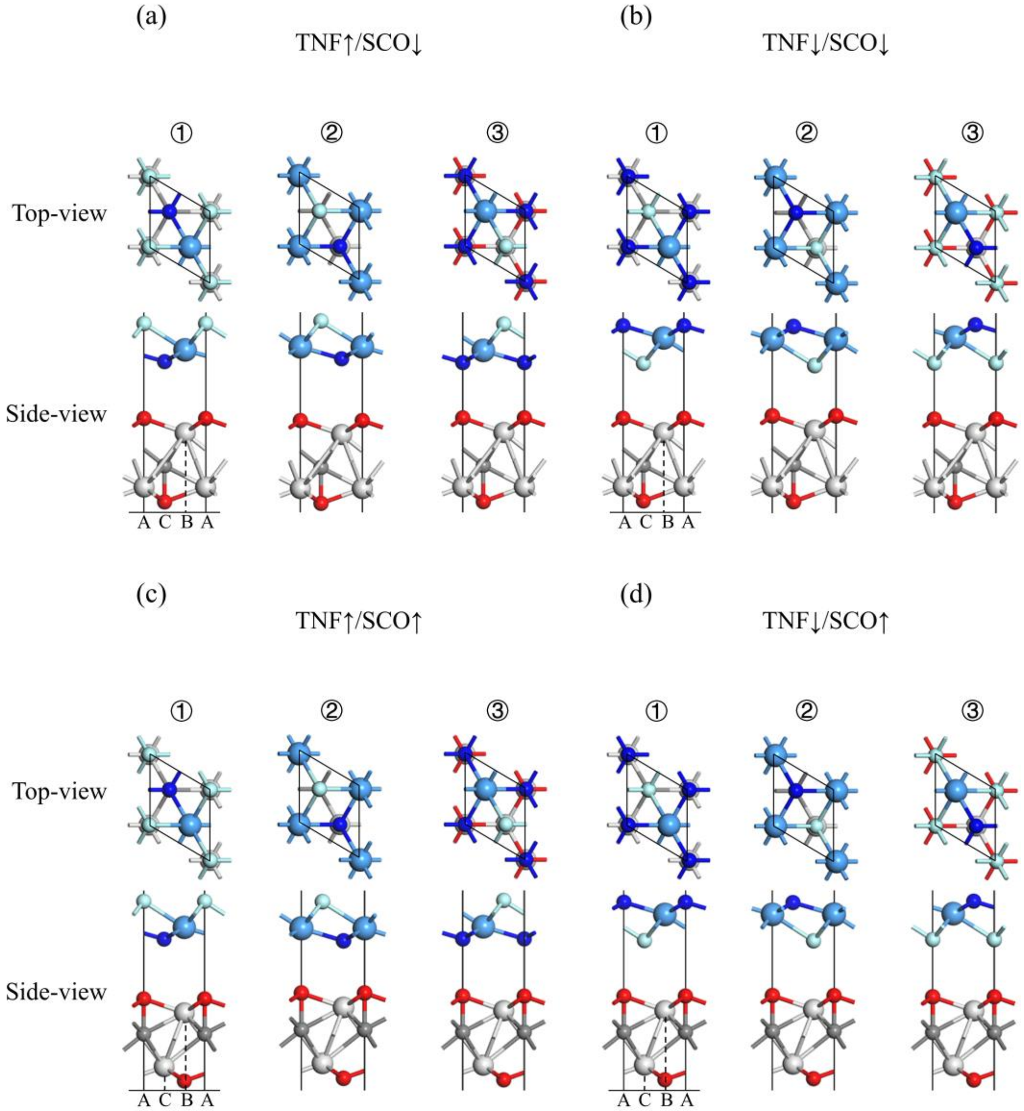

Figure S7. Top and side views of 12 stacking configurations for TNF/SCO heterostructures. (a) TNF↑/SCO↓, (b) TNF↓/SCO↓, (c) TNF↑/SCO↑ and (d) TNF↓/SCO↑.

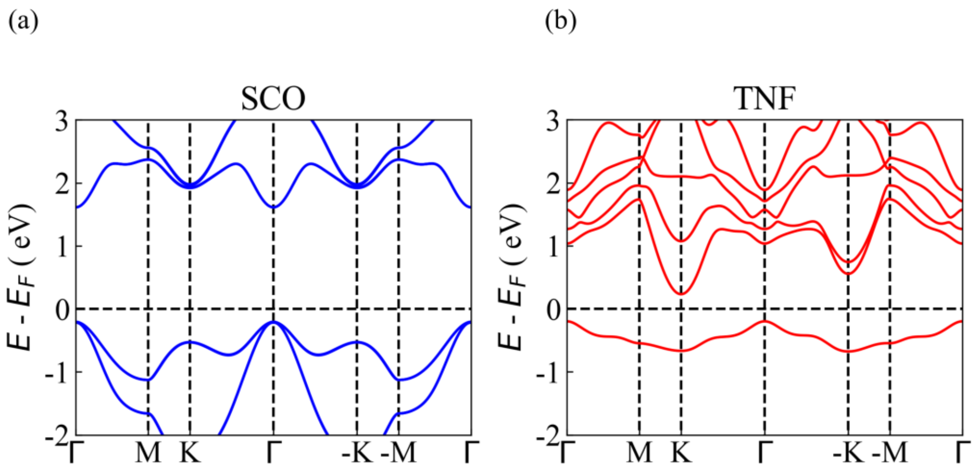

Figure S8. The band structure of the monolayer with the in-plane lattice constant of the heterostructure. (a) SCO, (b) TNF.

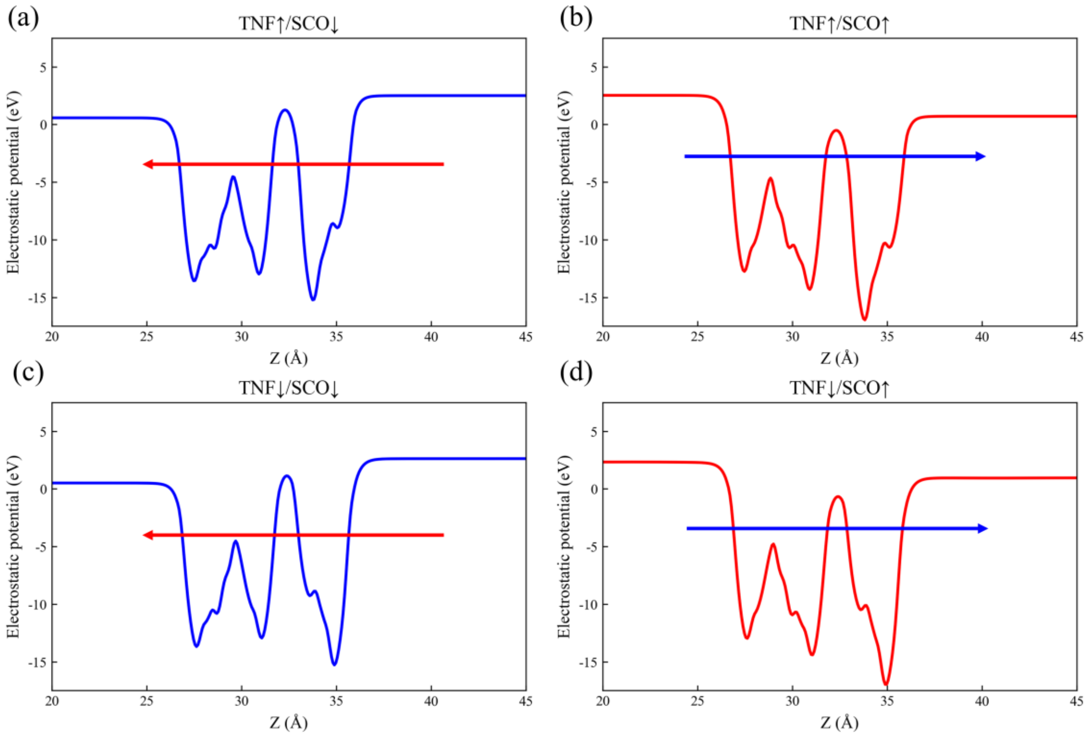

Figure S9. Electrostatic potential of (a) TNF↑/SCO↓, (b) TNF↓/SCO↓, (c) TNF↑/SCO↑ and (d) TNF↓/SCO↑.

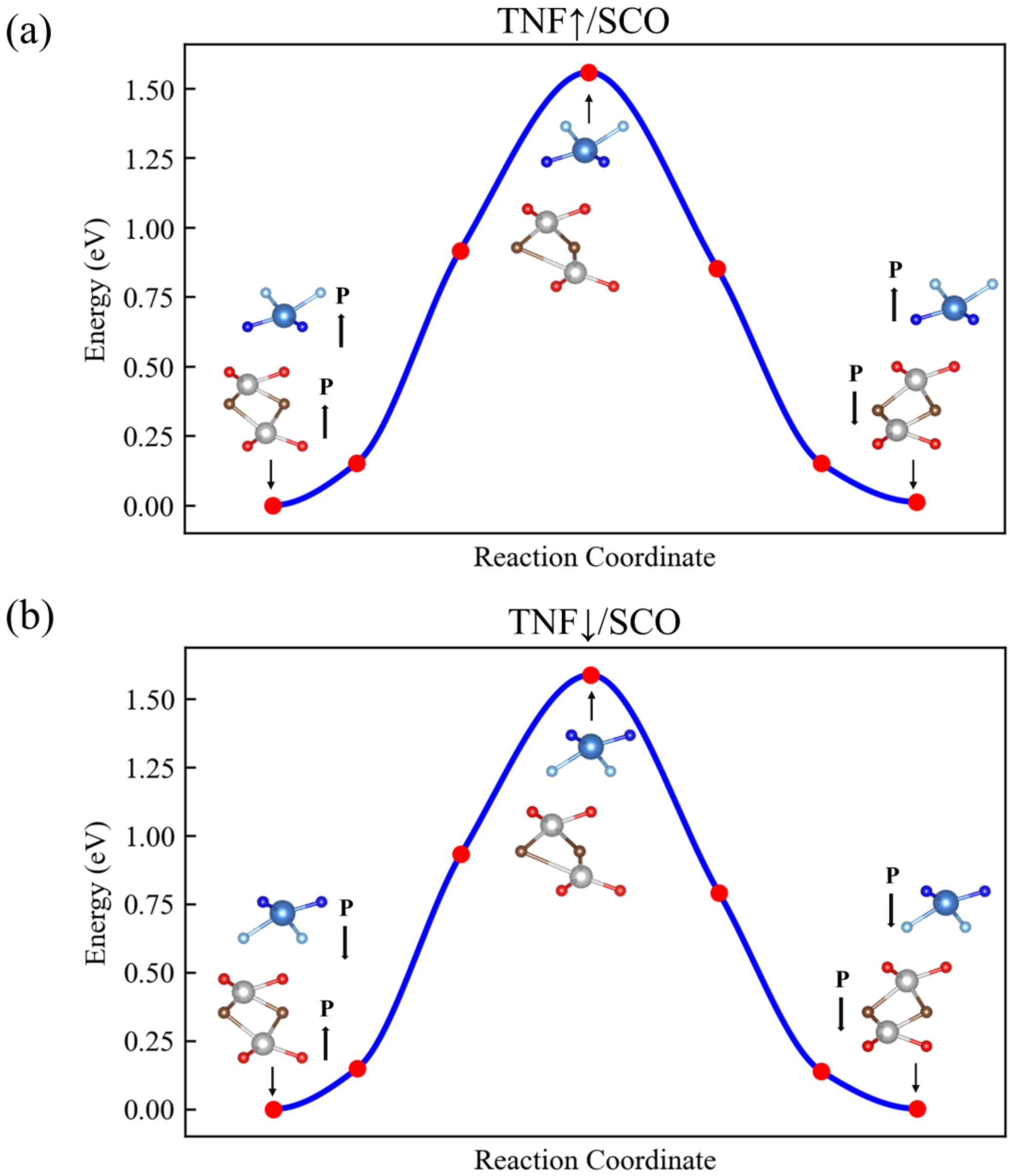

Figure S10. The kinetic pathways of polarization reversal processes. (a) TNF↑/SCO, (b) TNF↓/SCO.

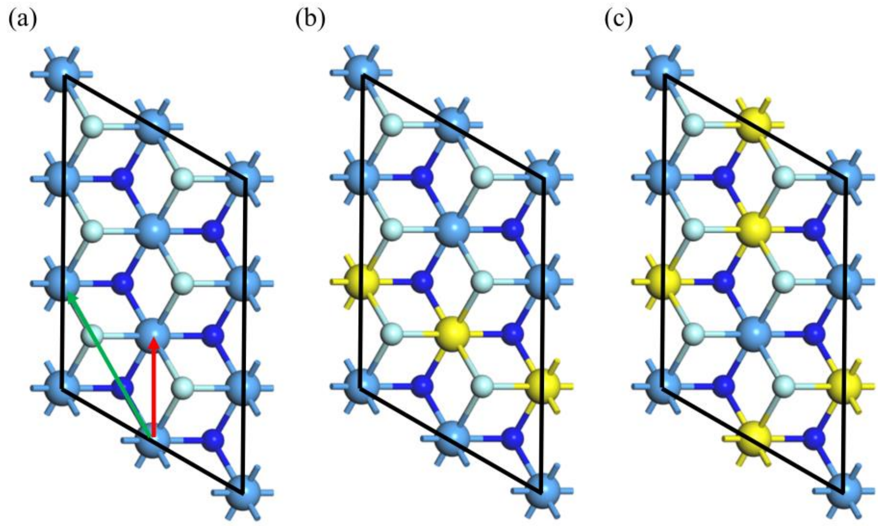

Figure S11. Magnetic configurations for (a) ferromagnetic, (b) ferrimagnetic and (c) antiferromagnetic order of TNF layer. In (a), the red arrows indicate the nearest neighbor magnetic exchange coupling, while the green arrows represent the next nearest neighbor magnetic exchange coupling. The two different colors represent the opposite directions of the magnetic moment.

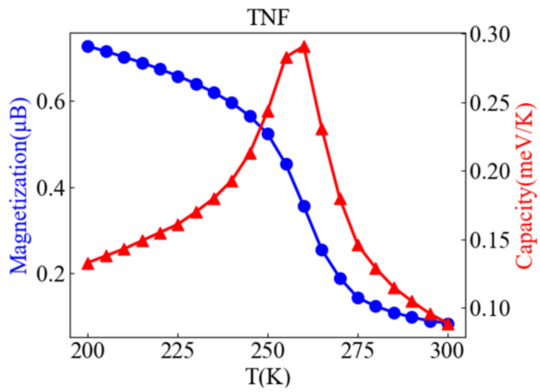

Figure S12. Temperature variation of the magnetization and capacity for the Janus TNF monolayer.

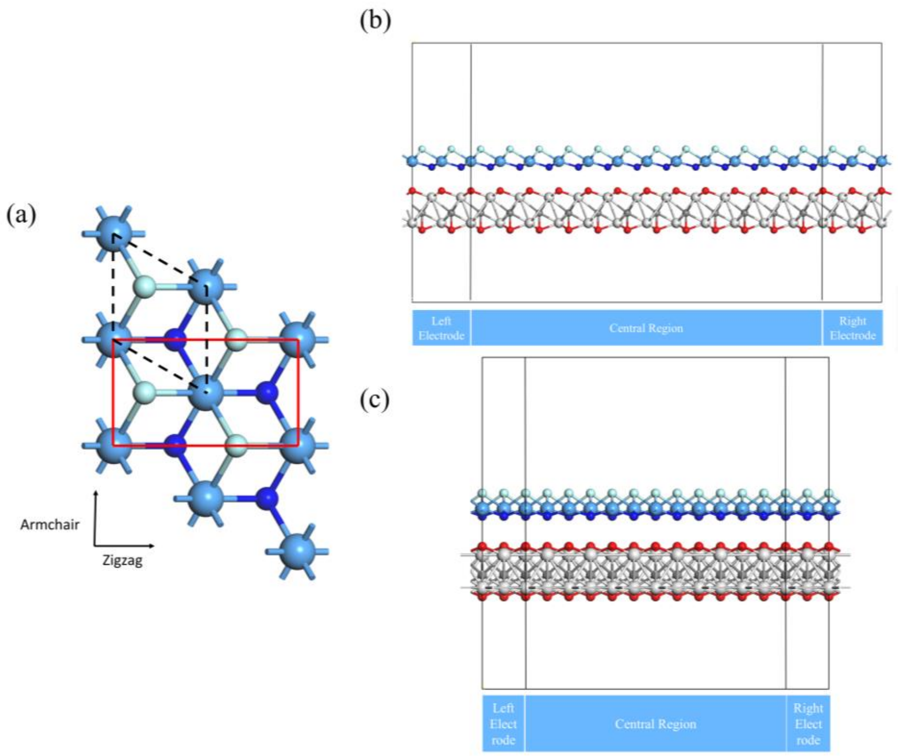

Figure S13. (a) The black dashed box represents the primitive cell, the red box represents the reconstructed $1\times\sqrt{3}$ orthogonal supercell, and the arrows represent the directions. The device configuration of TNF↑/SCO↓ heterostructure along (b) Zigzag direction and (c) Armchair direction.

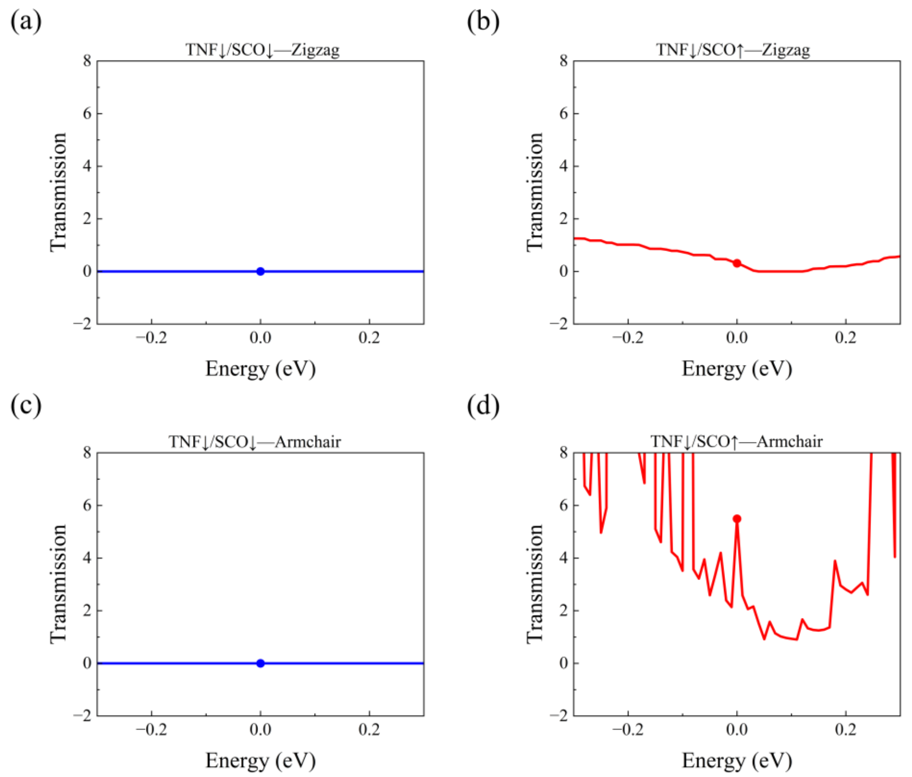

Figure S14. Transmission spectrum of the TNF↓/SCO heterostructure along the zigzag direction for (a) SCO↓ and (b) SCO↑. Transmission spectrum of the TNF↓/SCO heterostructure along the armchair direction for (c) SCO↓ and (d) SCO↑.