Electronic Supplementary Material (ESI) for Nanoscale.
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Supplemental Material

Sliding ferroelectricity and moiré effect in Janus bilayer MoSSe

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Table S1. The relaxed lattice parameters, and structural parameters such as bond lengths and bond angles of the Janus bilayer MoSSe structure with AA stacking. The parameter D represents the interlayer distance between the nearest neighboring atomic layers of the upper and lower layers.

	Lattice parameter (Å)	Bond length (Å)		Angle (°)			D (Å)
configuration	a=b	Mo-S	Mo-Se	∠SMoSe	∠SMoS	∠SeMoSe	
Se-Se	3.22	2.41	2.52	82.08	83.83	79.25	3.73
S-S	3.22	2.41	2.52	82.01	83.94	79.25	3.55
S-Se	3.22	2.41	2.52	82.04	83.92	79.22	3.64

Table S2. The energy difference of AB stacking for S-S, Se-Se, and S-Se configurations. The parameter D represents the interlayer distance between the nearest neighboring atomic layers of the upper and lower layers.

Structure	AB stacking		
	Se-Se	S-S	S-Se
ΔE (meV)	0	13.78	0.30
D (Å)	3.24	2.99	3.08

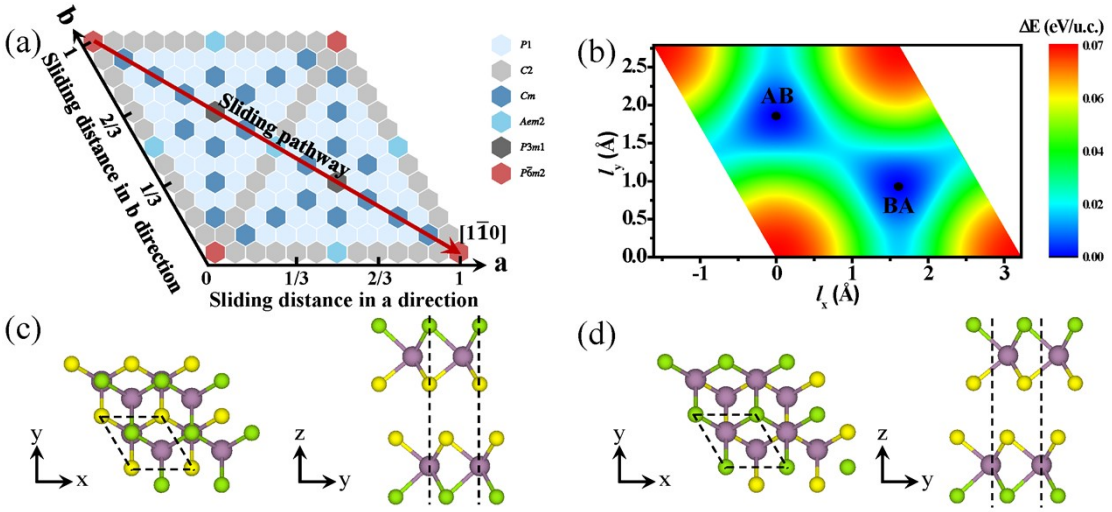

Fig. S1. (a) Illustration of the structural space group of the S-S configuration sliding in the ab-plane. Each honeycomb data point represents a slided structure. Different colors indicate different space groups. (b) The energy contour plot of bilayer Se-Se configuration versus the sliding distance (l_x, l_y). The contour colors illustrate the energy difference of the unit cell relative to the energy of ground state (AB stacking and BA stacking). (c)-(d) The top and side views of the two lowest-energy ground-state structures in Fig. S1(b), the AB stacking and the BA stacking, respectively.

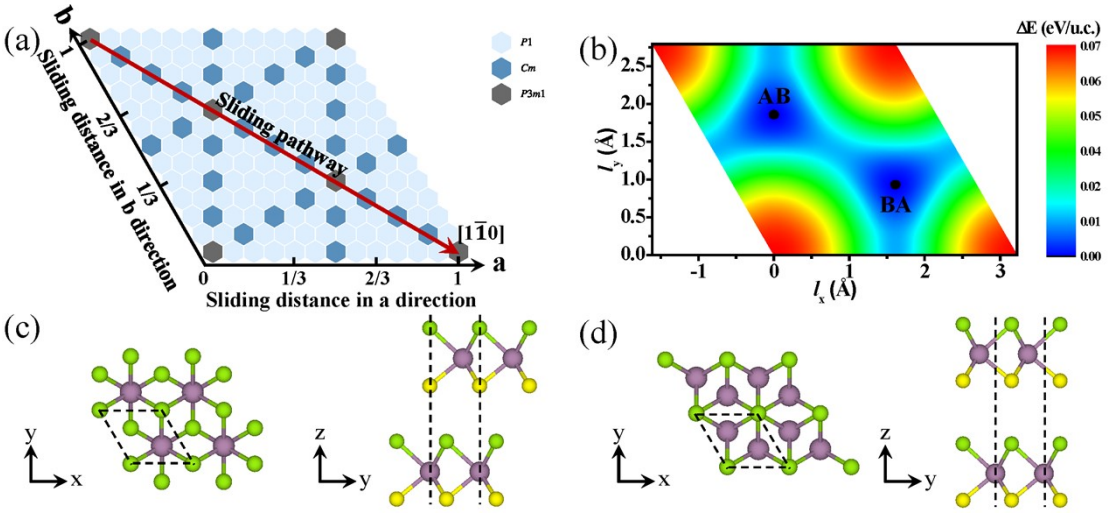

Fig. S2. (a) Illustration of the structural space group of the S-Se configuration sliding in the ab-plane. Each honeycomb data point represents a slided structure. Different colors indicate different space groups. (b) The energy contour plot of bilayer Se-Se configuration versus the sliding distance (l_x, l_y). The contour colors illustrate the energy difference of the unit cell relative to the energy of ground state (AB stacking and BA stacking). (c)-(d) The top and side views of the two lowest-energy ground-state structures in Fig. S2(b), the AB stacking and the BA stacking, respectively.

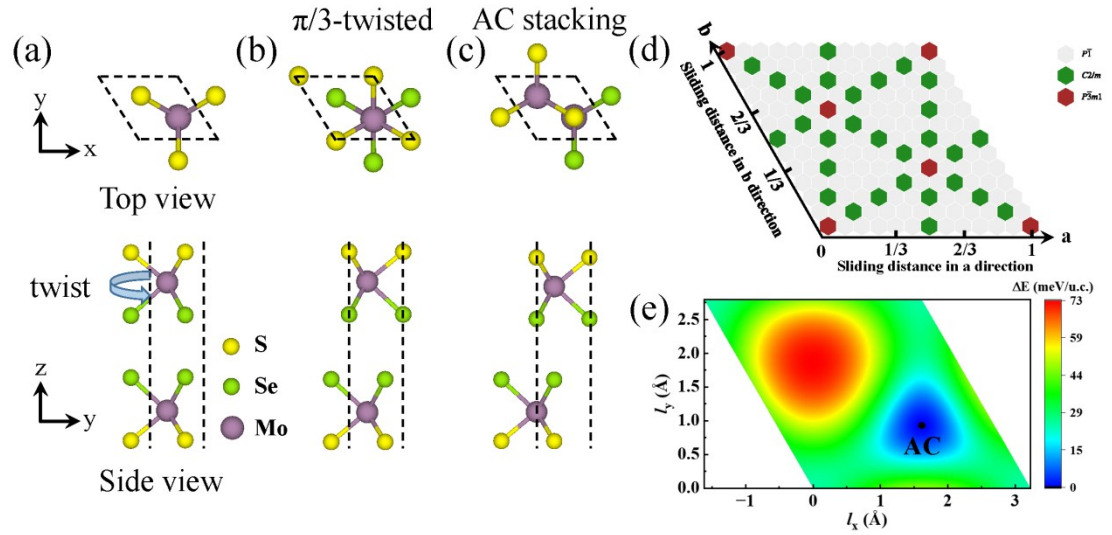

Fig. S3. (a)-(b) The top and side views of untwisted and π/3-twisted of AA stacking structures in Se-Se configuration. (c) The top and side views of AC stacking. (d) Illustration of the structural space group of the π/3-twisted Se-Se configuration sliding in the ab plane. Each honeycomb data point represents a slided structure. Different colors indicate different space groups. (e) The energy contour plot of bilayer Se-Se configuration versus the sliding distance (l_x, l_y). The contour colors illustrate the energy difference of the unit cell relative to the energy of ground state (AC stacking).

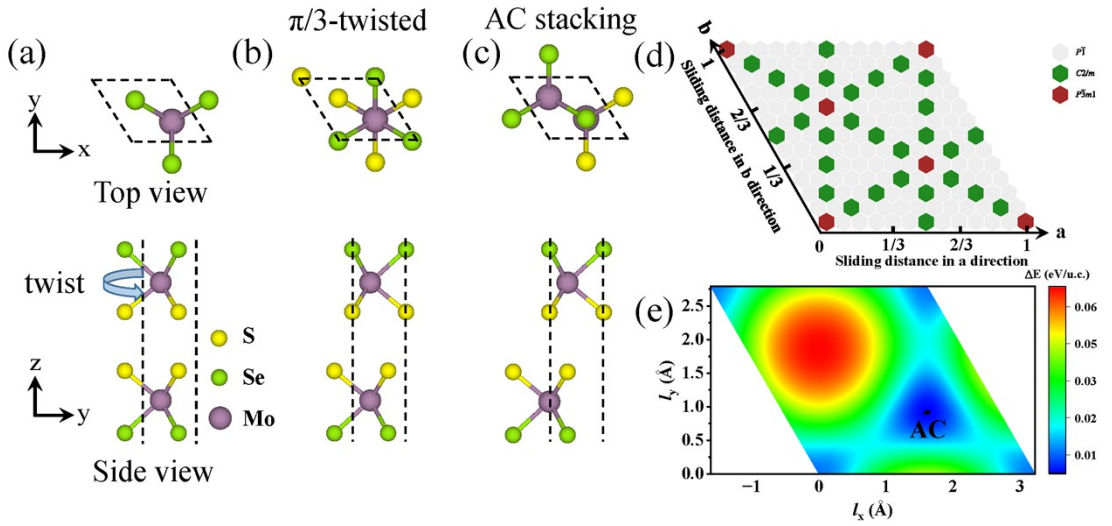

Fig. S4. (a)-(b) The top and side views of untwisted and π/3-twisted of AA stacking structures in S-S configuration. (c) The top and side views of AC stacking. (D) Illustration of the structural space group of the π/3-twisted S-S configuration sliding in the ab plane. Each honeycomb data point represents a slided structure. Different colors indicate different space groups. (E) The energy contour plot of bilayer S-S configuration versus the sliding distance (l_x, l_y). The contour colors illustrate the energy difference of the unit cell relative to the energy of ground state (AC stacking).

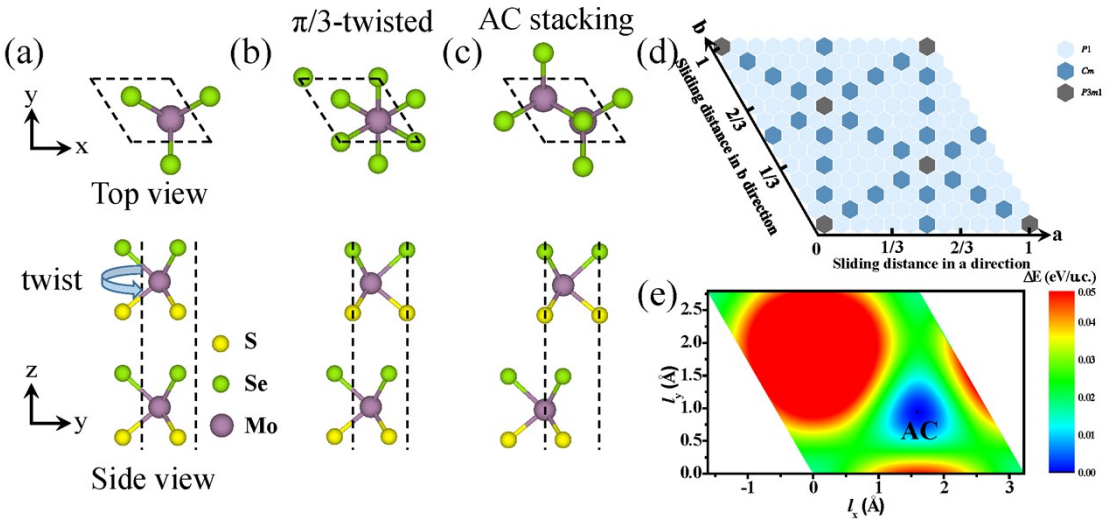

Fig. S5. (a)-(b) are the top and side views of untwisted and π/3-twisted of AA stacking structures in S-Se configuration. (c) The top and side views of AC stacking. (d) Illustration of the structural space group of the π/3-twisted S-Se configuration sliding in the ab plane. Each honeycomb data point represents a slided structure. Different colors indicate different space groups. (e) The energy contour plot of bilayer S-Se configuration versus the sliding distance (l_x, l_y). The contour colors illustrate the energy difference of the unit cell relative to the energy of ground state (AC stacking).

Table S3. The interlayer distance (D) and out-of-plane polarization (OP) of the optimized AB stacking are corrected with different vdW functionals.

Configuration	OP & D (pC/m) & (Å)	vdW functional					
		DFT-D3	DFT-D2	vdW-DF2	optB86b-vdW	optB88-vdW	optPBE-vdW
Se-Se	OP	-0.42	-0.45	-0.25	-0.45	-0.40	-0.28
	D	3.24	3.23	3.49	3.22	3.27	3.422
S-S	OP	-0.84	-0.77	-0.55	-0.87	-0.79	-0.58
	D	3.00	3.04	3.20	2.98	3.02	3.16

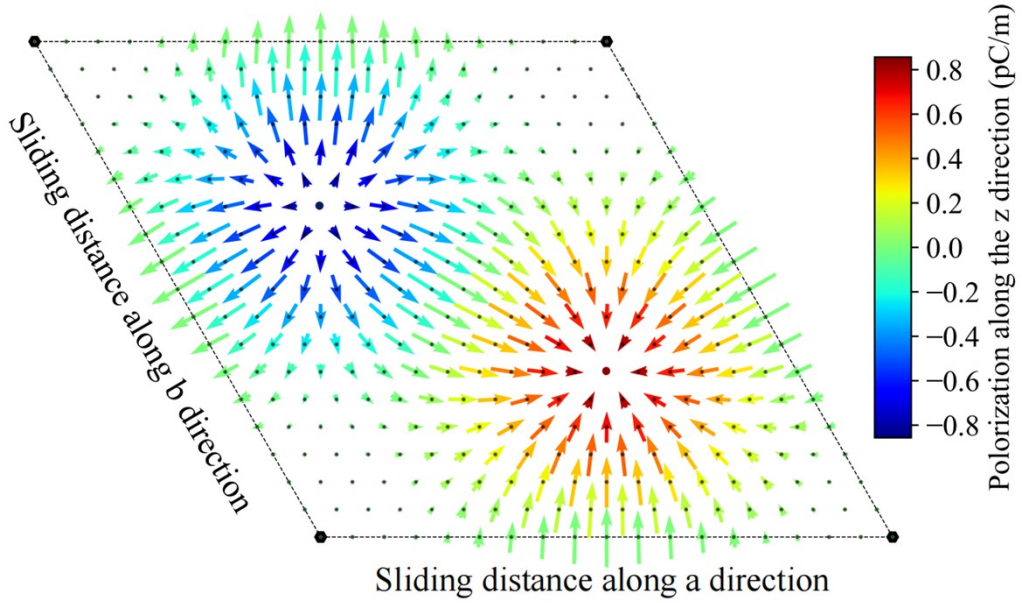

Fig. S6. Diagram of in-plane and out-of-plane polarization of sliding structures for S-S configuration. The color contour map shows the magnitude and direction of out-of-plane polarization. The arrow indicates the direction of in-plane polarization. The length of the arrows in the diagram represents the magnitude of the in-plane polarization, with longer arrows indicating a greater polarization. The longest arrow signifies the maximum in-plane polarization of 1.28 pC/m.

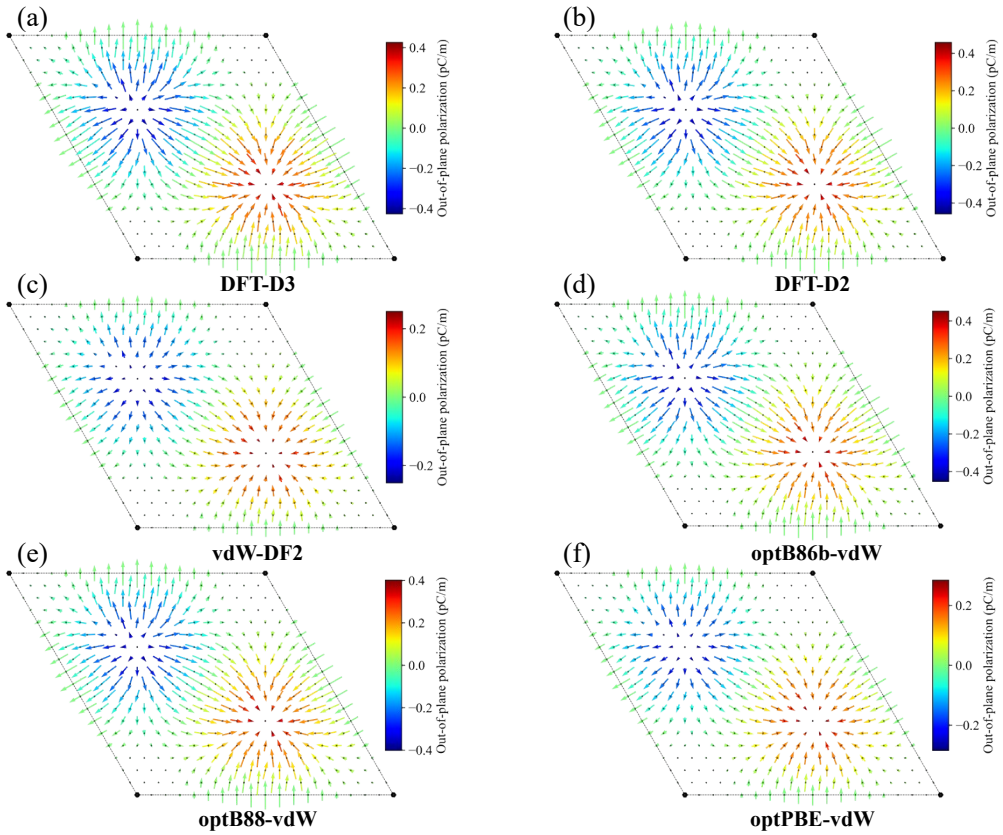

Fig. S7. The in-plane and out-of-plane polarization of the sliding structure in the Se-Se configuration, were computed using six different vdW functionals, specifically (a) DFT-D3, (b) DFT-D2, (c) vdW-DF2, (d) optB86b-vdW, (e) optB88-vdW, and (f) optPBE-vdW, respectively. The color contour map shows the magnitude and direction of out-of-plane polarization. The arrow indicates the direction of in-plane polarization. The length of the arrows in the diagram represents the magnitude of the in-plane polarization, with longer arrows indicating a greater polarization.

Table S4. Predicted 2D sliding ferroelectric. The "$P_{\perp}$" symbol denotes out-of-plane polarization. And the "P∥" denotes in-plane polarization.

Materials	$P_{\perp}$	$P_{\parallel}$	Thickness
BN [1]	2.08 pC/m	-	Bilayer
InSe [1]	0.24 pC/m	-	Bilayer
MoS_2 [1]	0.97 pC/m	-	Bilayer
GaSe [1]	0.46 pC/m	-	Bilayer
GaN [1]	9.72 pC/m	-	Bilayer
$MoSi_2N_4$ [2]	3.36 pC /m	-	Bilayer
$MoGe_2N_4$ [2]	3.05 pC/m	-	Bilayer
$CrSi_2N_4$ [2]	2.49 pC/m	-	Bilayer
WSi_2N_4 [2]	3.44 pC/m	-	Bilayer
BP [3]	1.07 pC/m	-	Bilayer
BAs [3]	0.96 pC/m	-	Bilayer
BSb [3]	3.70 pC/m	-	Bilayer
AlN [3]	7.19 pC/m	-	Bilayer
GaN [3]	5.76 pC/m	-	Bilayer
InN [3]	13.83 pC/m	-	Bilayer
SiC [3]	2.89 pC/m	-	Bilayer
GeC [3]	2.78 pC/m	-	Bilayer
SnC [3]	4.41 pC/m	-	Bilayer
MnSe [4]	2.70 pC/m	-	Bilayer
$MoSe_2$ [5]	0.59 pC/m	-	Bilayer
WS_2 [5]	0.69 pC/m	-	Bilayer
WSe_2 [5]	0.73 pC/m	-	Bilayer
MoSSe-SS (AB stacking)	0.86 pC/m	-	Bilayer
MoSSe-SS (TS stacking)	-	1.2 pC/m	Bilayer

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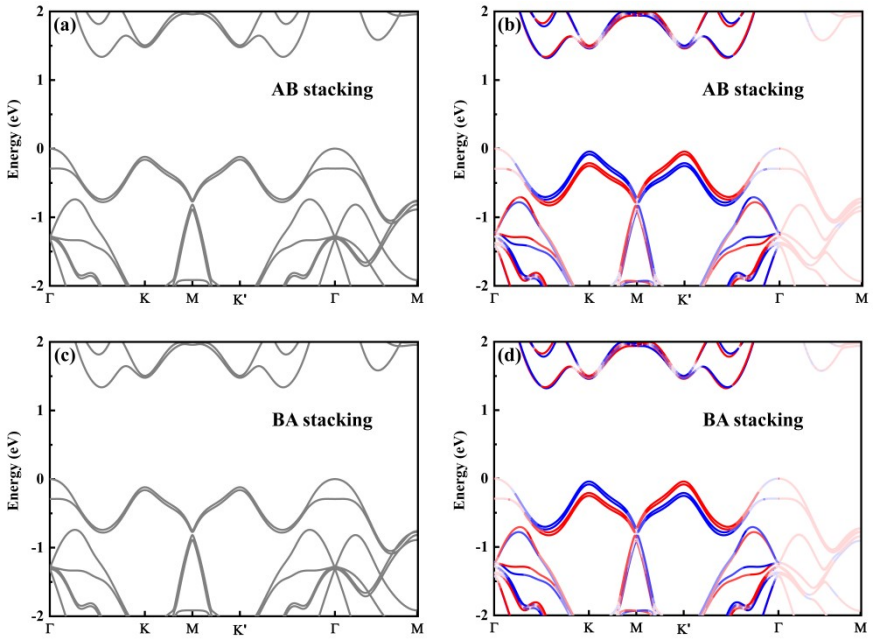

Figure R8. The electronic band structures of Se-Se configuration. (a) and (b) are the electronic band structures for AB stacking with PBE and PBE+SOC, respectively. (c) and (d) are the electronic band structures for BA stacking with PBE and PBE+SOC, respectively. The color depth indicates the quantity of the projected contribution.

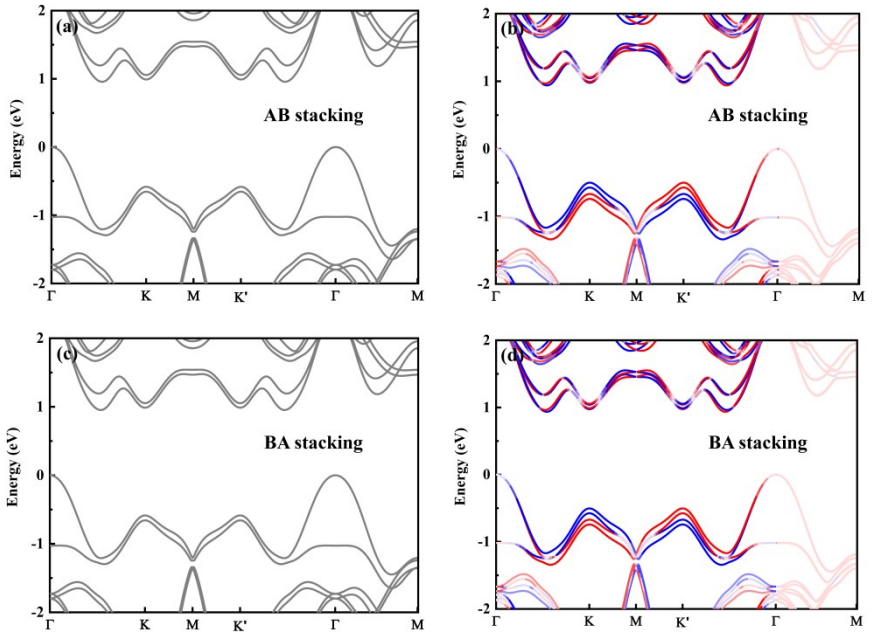

Figure S9. The electronic band structures of S-S configuration. (a) and (b) are the electronic band structures for AB stacking with PBE and PBE+SOC, respectively. (c) and (d) are the electronic band structures for BA stacking with PBE and PBE+SOC, respectively. The color depth indicates the quantity of the projected contribution.