

Supplementary Information

Xinge Zhang,¹ Yuqian Jiang,¹ Yuping Tian,¹ Binyuan Zhang,¹
Linyang Li,² Wei-jiang Gong,^{1,*} and Xiangru Kong^{1,†}

¹College of Sciences, Northeastern University, Shenyang 110819, China

²School of Science, Hebei University of Technology, Tianjin 300401, China

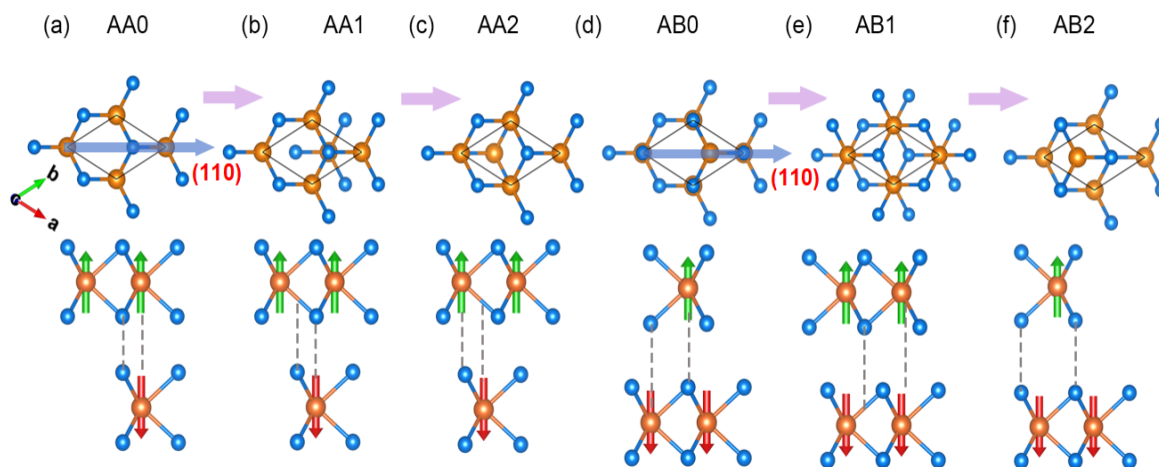


Fig. S1. Two kinds of stacking patterns of RuCl_2 bilayers: AA-type for (a) AA0, (b) AA1 and (c) AA2; AB-type for (d) AB0, (e) AB1 and (f) AB2. The top panel demonstrates the top view, and the bottom panel indicates the side view. The red and green arrows indicate the magnetic order. The blue arrows along (110) direction indicate the direction of interlayer sliding translation, and the purple arrows indicate the sliding process.

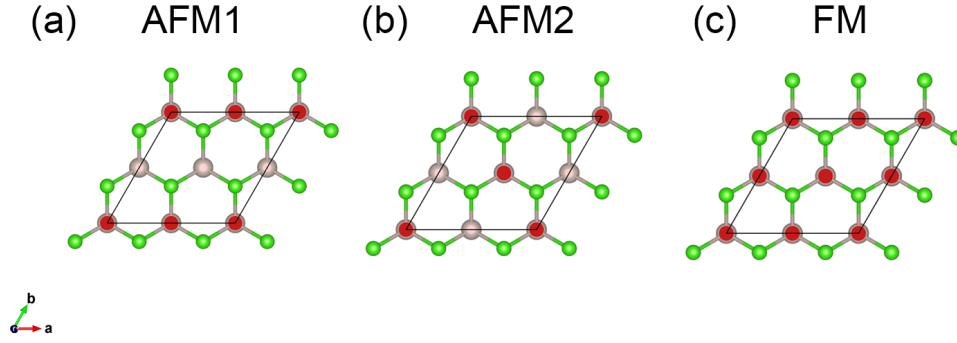


Fig. S2. Three magnetic configurations: (a) AFM1, (b) AFM2, and (c) FM of monolayer RuCl_2 . The green symbols indicate the Cl atoms, the red symbols indicate Ru atoms with spin up, grey symbols indicate Ru atoms with spin down.

Table S1. Energy calculated in different magnetic states of monolayer RuCl_2 .

magnetic configurations	AFM1	AFM2	FM
Energy (eV)	0.883	0.873	0

Table S2. Energy calculated in different magnetic states of bilayer RuCl_2 .

magnetic configurations	AFM1	AFM2	AFM	FM
Energy (eV)	1.77	1.76	0	0.03

* Contact author: gwj@mail.neu.edu.cn

† Contact author: kongxiangru@mail.neu.edu.cn

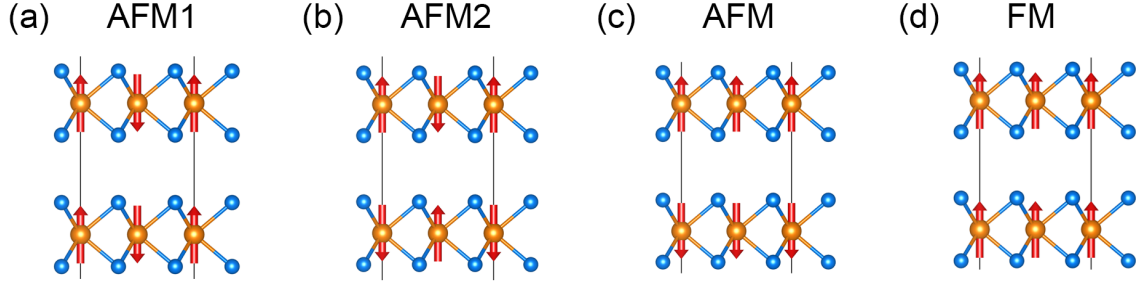


Fig. S3. Four magnetic configurations of RuCl_2 bilayers: (a) AFM1, (b) AFM2, (c) AFM and (d) FM. The blue and orange symbols indicate the Cl and Ru atoms, the red arrows indicate the spin direction.

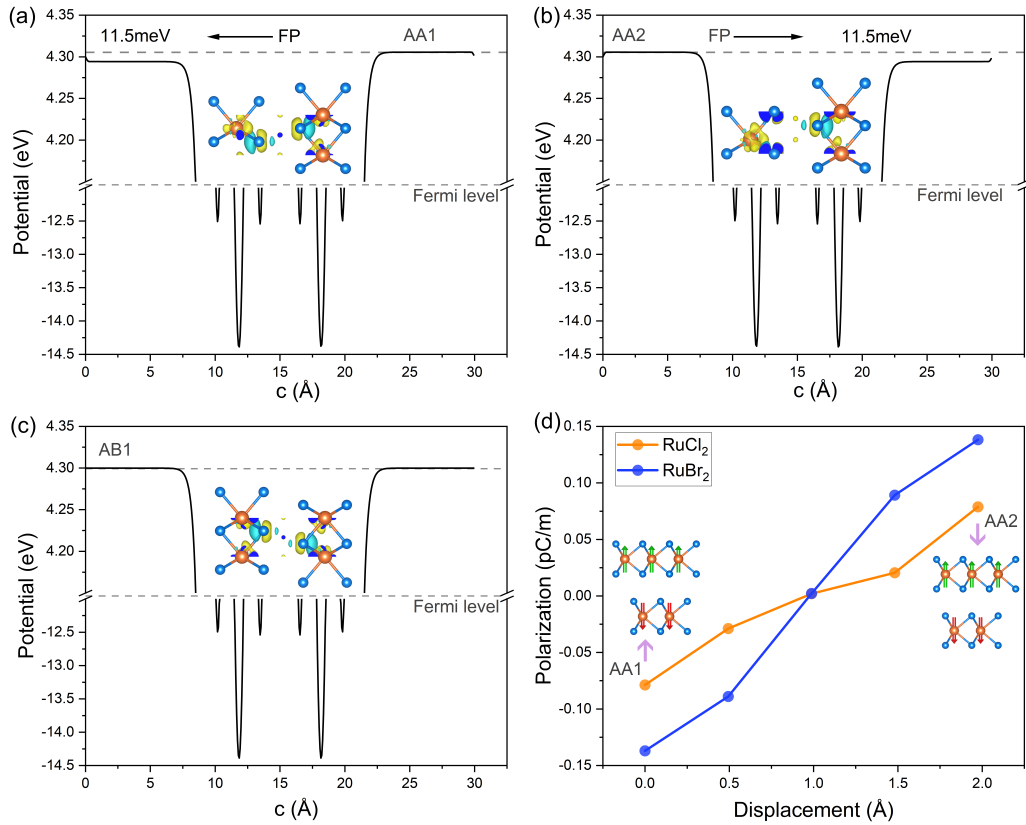


Fig. S4. The plane-averaged electrostatic potential and charge density difference for the AA1-type, AA2-type and AB1-type stacking RuCl_2 bilayers. (d) The evolution of the ferroelectric polarization from AA1-type to AA2-type on the interlayer sliding displacement for bilayer RuCl_2 and RuBr_2 .

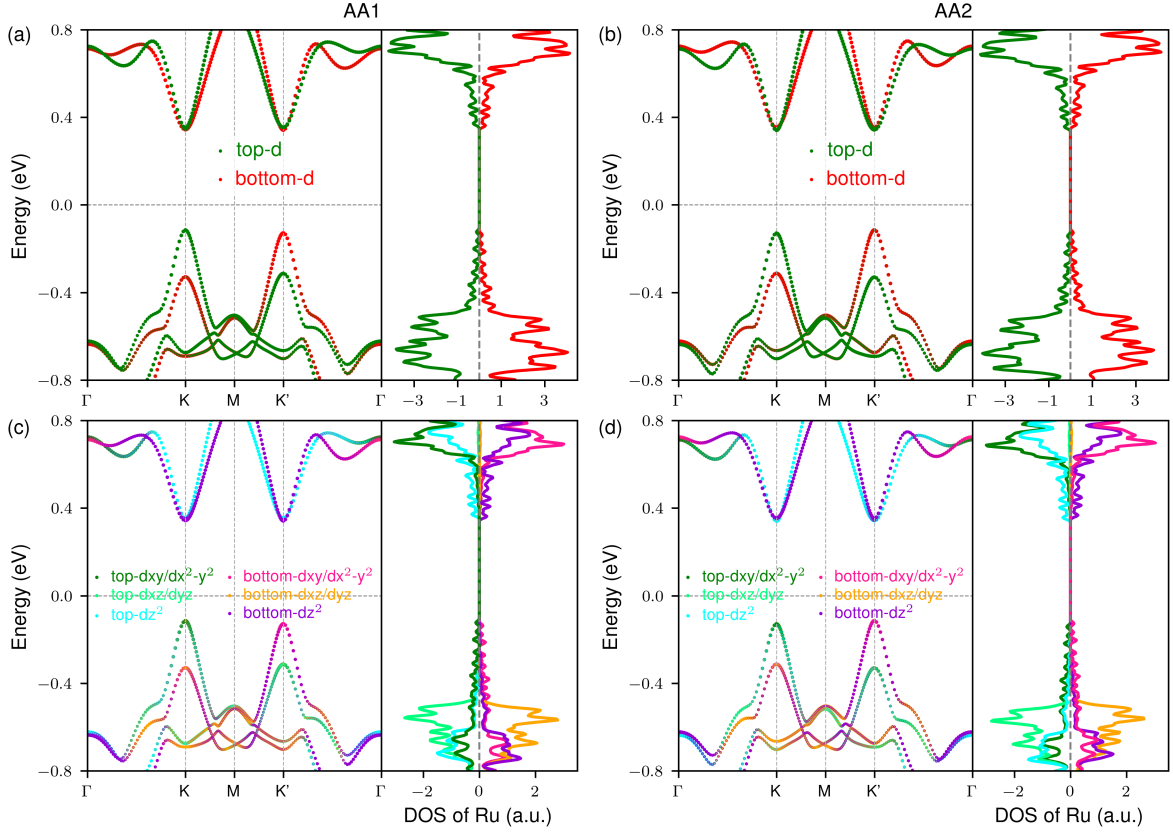


Fig. S5. The orbital-resolved electronic structures of (a,c) AA1-type and (b,d) AA2-type. The left panels in (a,b) describe the d orbitals of Ru atoms from the top (green) and bottom (red) layers, and the right panels in (a,b) describe the corresponding orbital-projected density of states. The left panels in (c,d) describe the five sub-orbitals of d orbitals in Ru atoms, and the right panels in (a,b) describe the corresponding orbital-projected density of states (DOS). The Fermi level is set to zero.

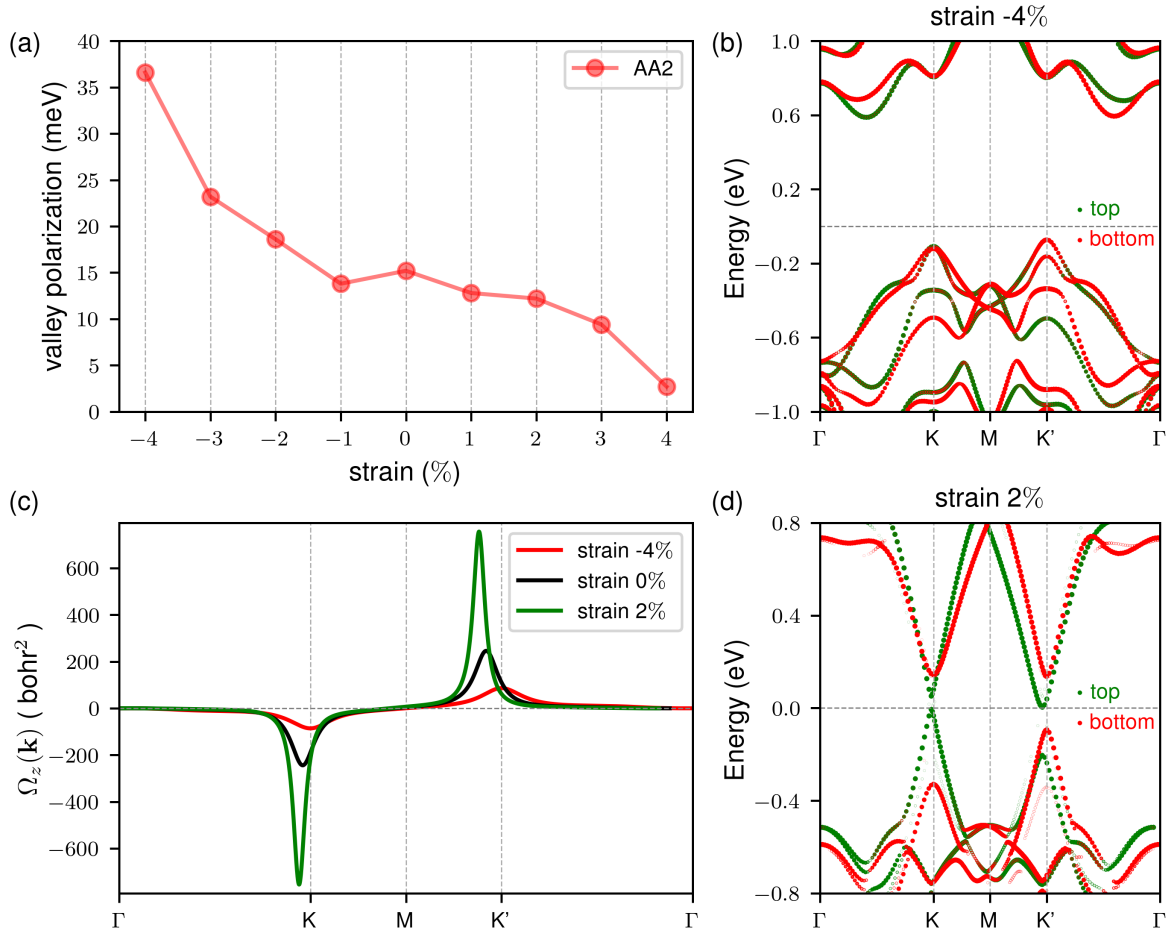


Fig. S6. (a) Valley polarization for biaxial strain from -4% to 4% for the AA2-type stacking. (b,d) The layer-resolved bands structures at strain -4% and 2% with SOC , red color indicates the bottom layer and green color indicates the top layer. (c)The Berry curvature at strain -4%, 0% and 2%.

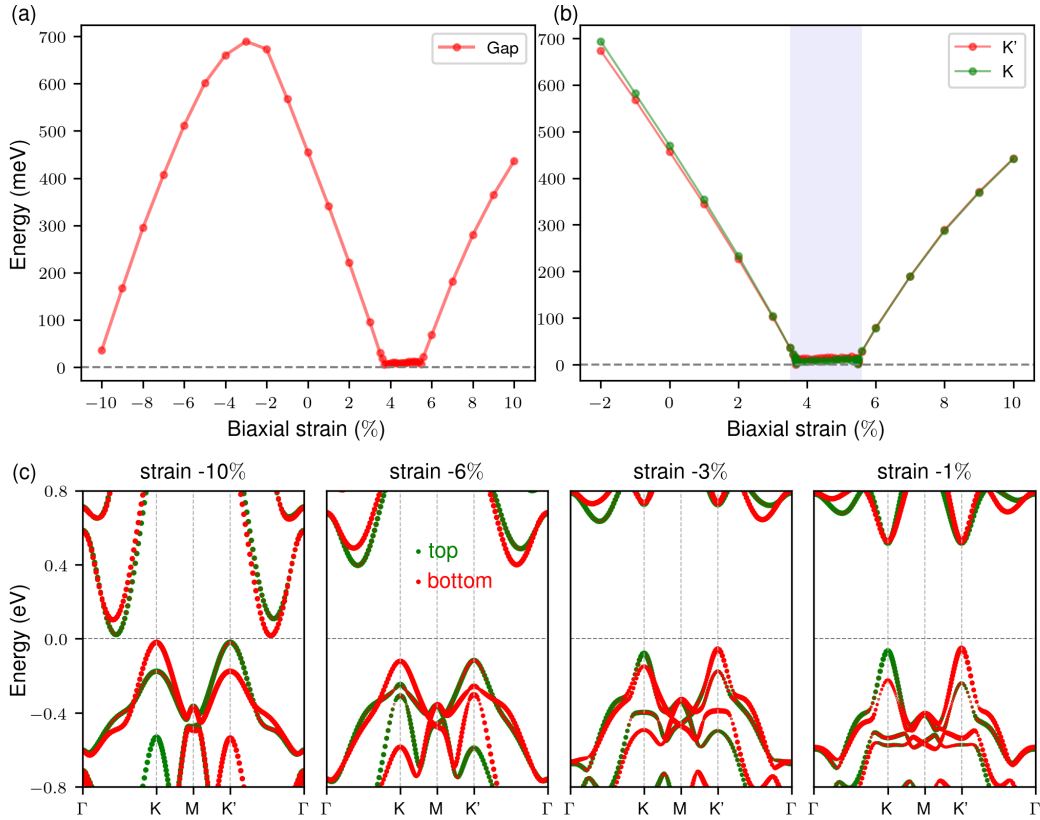


Fig. S7. (a) The evolution of global band gap across the strain from -10% compressive strain to 10 % (tensile strain). (b) The evolution of band gaps at K and K' valleys from -2% to 10% strain. The shaded area in (b) is presented in Fig. 3 (a) in detail. (c) The band structures under the strain of -10%, -6%, -3% and -1%, respectively.

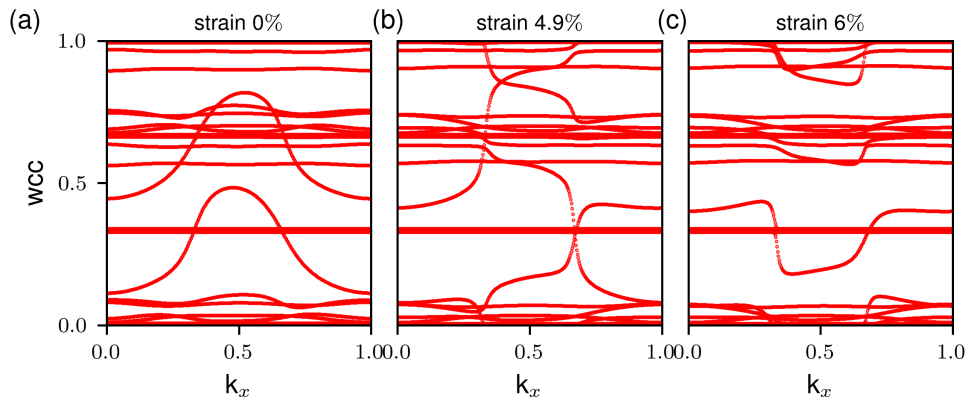


Fig. S8. Results in AA2-type stacking bilayer. The evolution of WCCs under the strain of (a) 0%, (b) 4.9%, and (c) 6%.

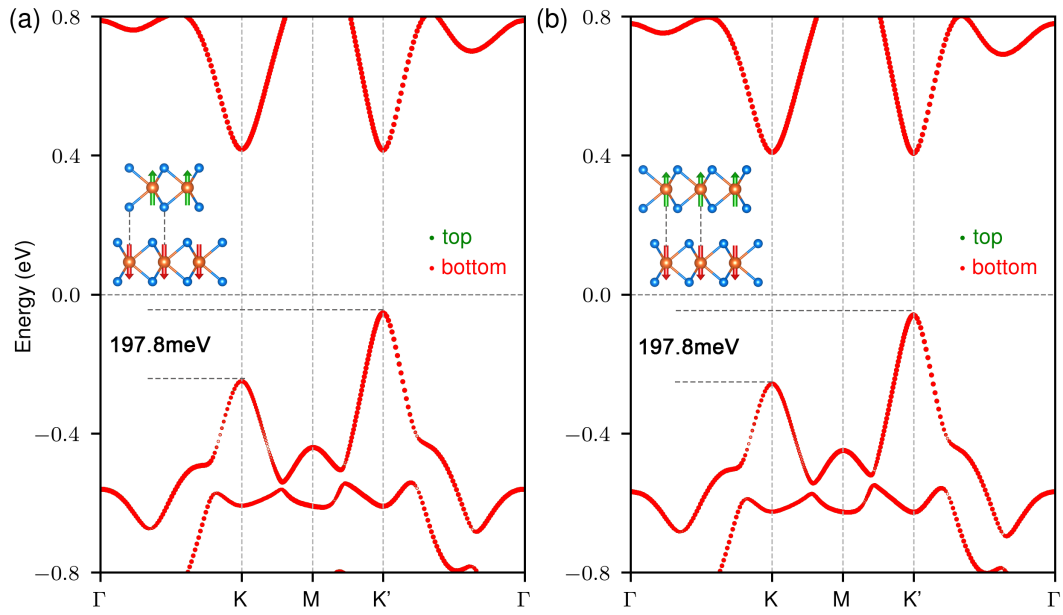


Fig. S9. The layer-resolved (also spin-resolved) band structures for (a) AB0-type and (b) AB1-type stacking patterns of RuCl₂ bilayers with SOC effect.

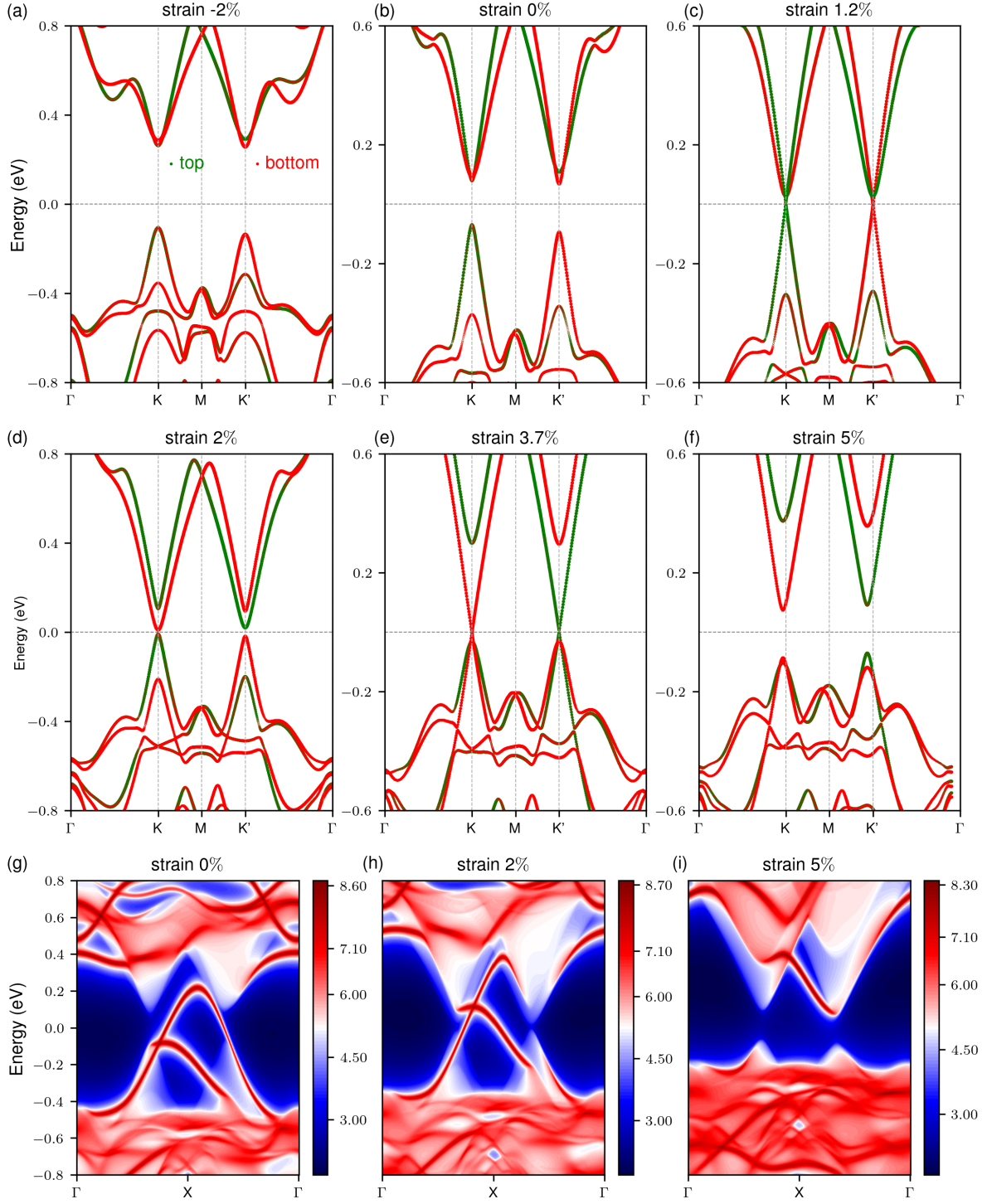


Fig. S10. Results in AA1-type stacking of RuBr₂ bilayer. The electronic band structures in the process of band inversions under the strain of (a) -2%, (b) 0%, (c) 1.2%, (d) 2%, (e) 3.7% and (f) 5%. The evolution of edge states under the strain of (g) 0%, (h) 2% and (i) 5%.

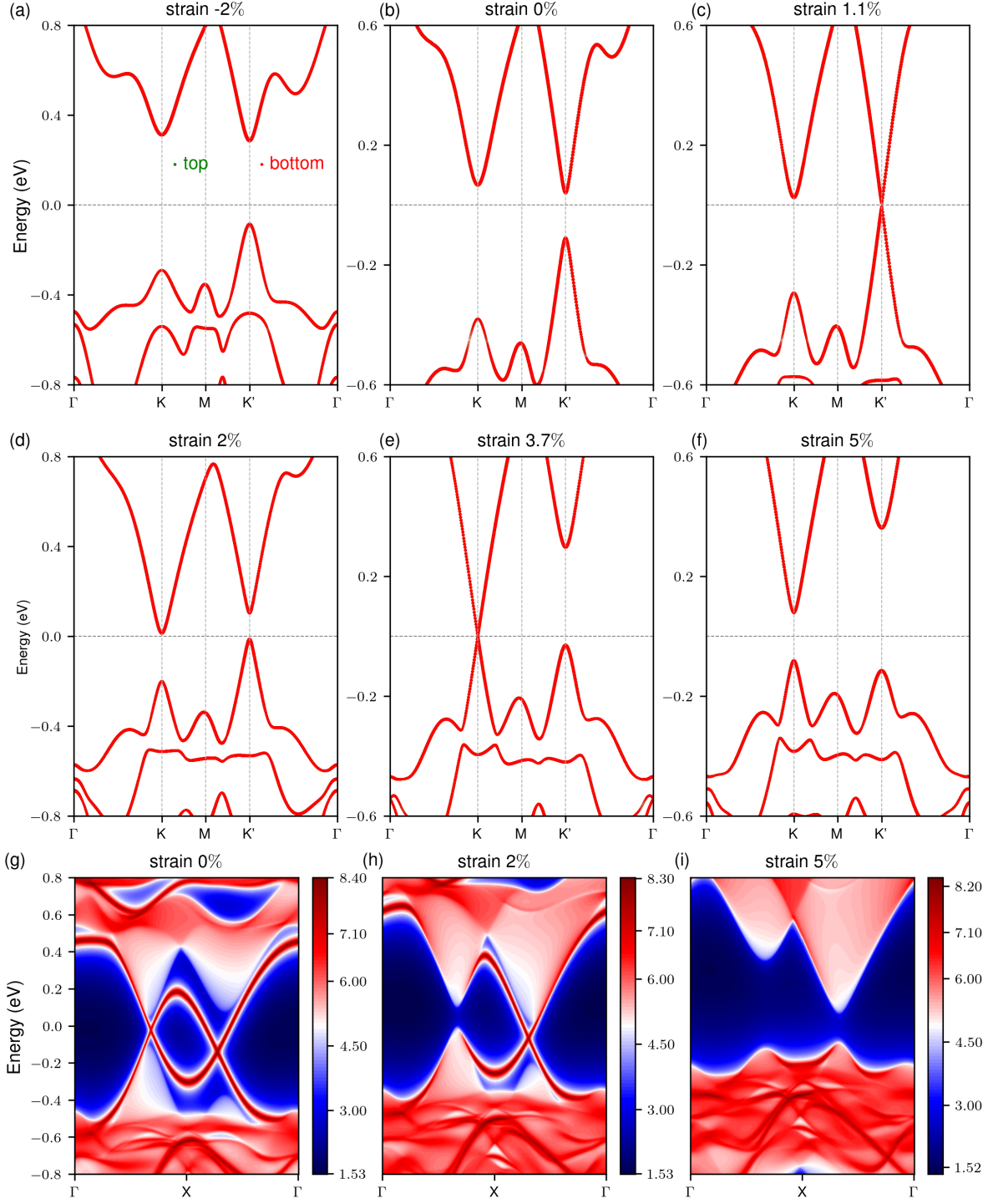


Fig. S11. Results in AB1-type stacking of RuBr₂ bilayer. The electronic band structures in the process of band inversions under the strain of (a) -2%, (b) 0%, (c) 1.1%, (d) 2%, (e) 3.7% and (f) 5%. The evolution of edge states under the strain of (g) 0%, (h) 2% and (i) 5%.

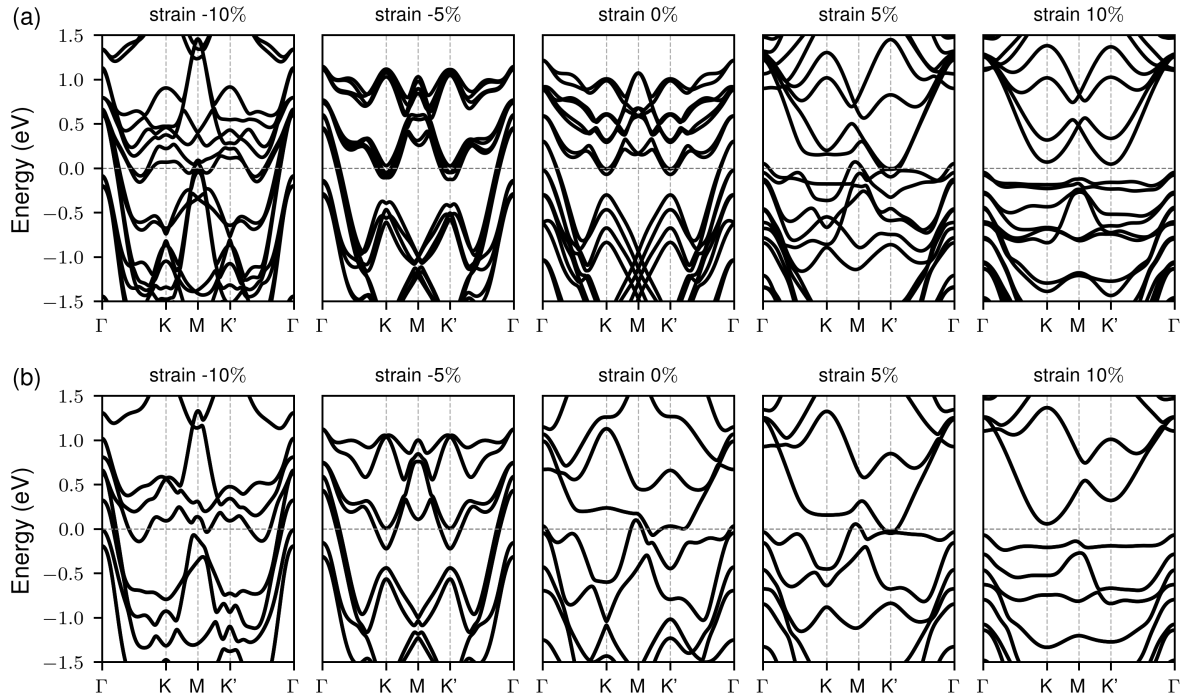


Fig. S12. Results in (a) AA2-type and (b) AB0-type stacking of RuI₂ bilayers. The electronic band structures under the strain of -10%, -5%, 0%, 5% and 10%.