Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Table S1 Valley polarization Values of different materials and stacking conditions

	I	II	III	IV	V	VI
CrSe2	48	42	30	28	17	32
CrS2	13	19	15	14	12	22
S	18	8	13	18	23	23
Se	29	80	60	71	2	32

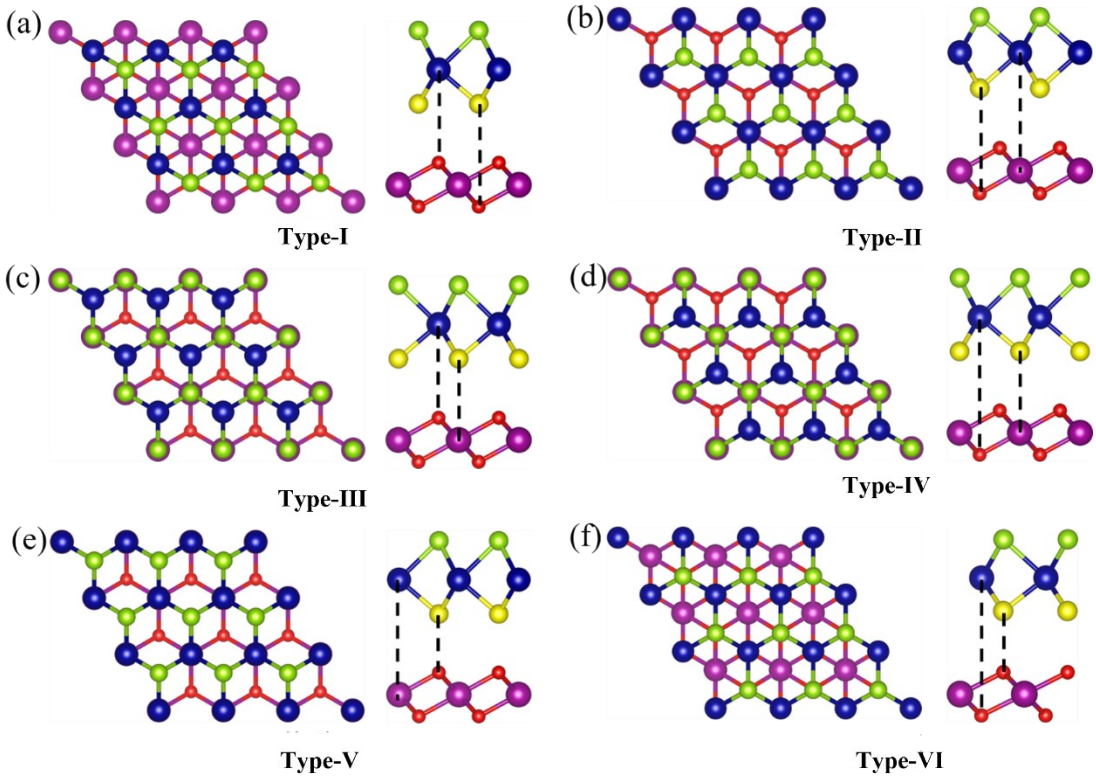

Fig. S1 Structure of CrSSe/MnO2 under different stacks on the contacting interface is S atom.

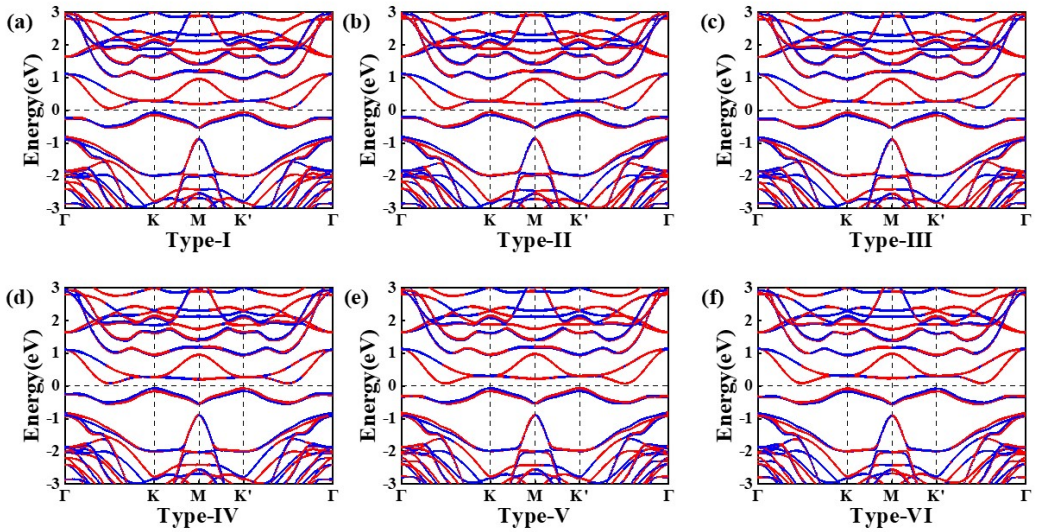

Fig. S2 Band structures corresponding to each structure in the **Fig. S1**.

Table S2 Berry curvature of different materials in papers.

Materials	Berry curvature (Bohr^2)
SnS [1]	4
$TcIrGe_2S_6$ [2]	9.48
CrSSe (our materials)	30.83
MoS_2 [3]	62
TiBrI [4]	106

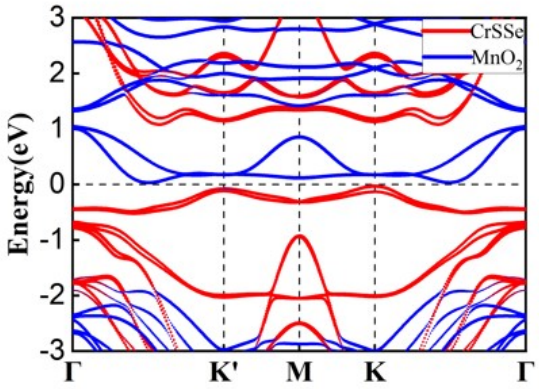

Fig. S3 The layer-resolved band structure of CrSSe/MnO2.

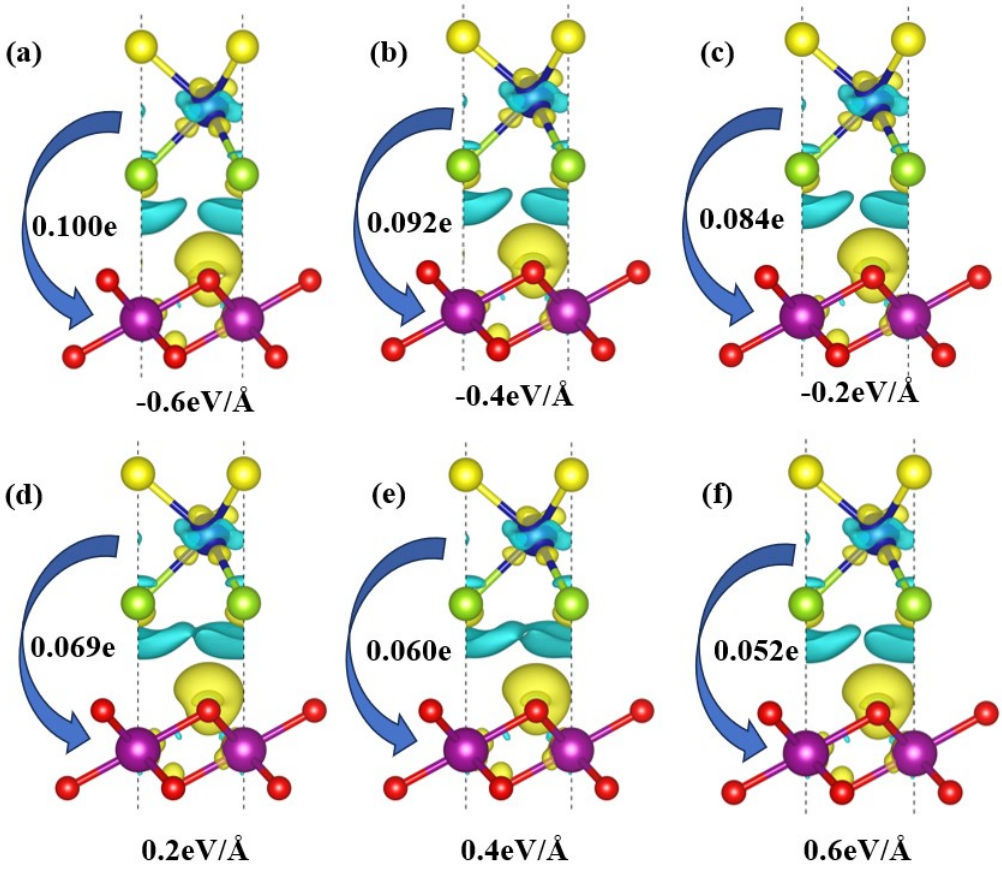

Fig. S4 The plot of charge density difference under different magnitudes of applied electric field.

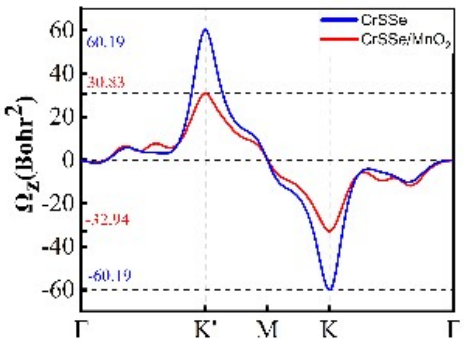

Fig. R1 Berry curvature of CrSSe (blue line) and $CrSSe/MnO_2$ (red line).

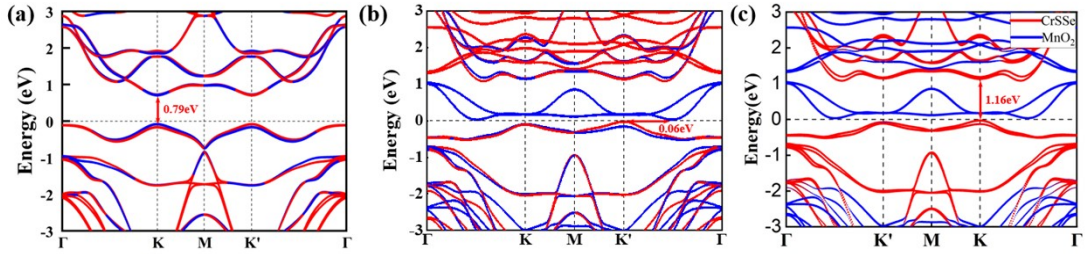

Fig. R2 The spin projected band structure of (a) CrSSe and (b) $CrSSe/MnO_2$, and the material projected band structure of $CrSSe/MnO_2$.

Table R1 Berry curvature of different materials in papers.

Materials	Berry curvature ($Bohr^2$)
SnS [1]	4
$TcIrGe_2S_6$ [2]	9.48
CrSSe(our materials)	30.83
MoS_2 [3]	60
TiBrI [4]	106

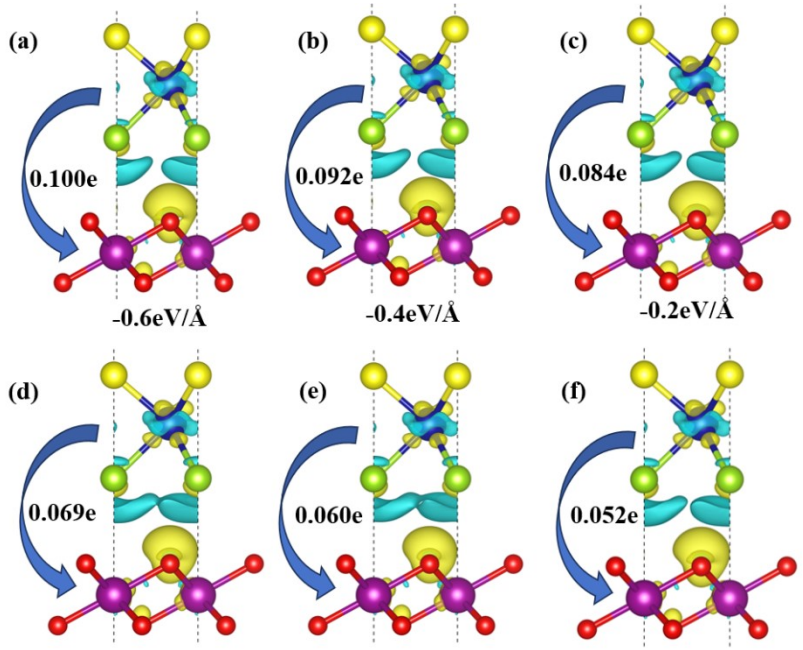

Fig. R3 The plot of charge density difference under different magnitudes of applied electric field.

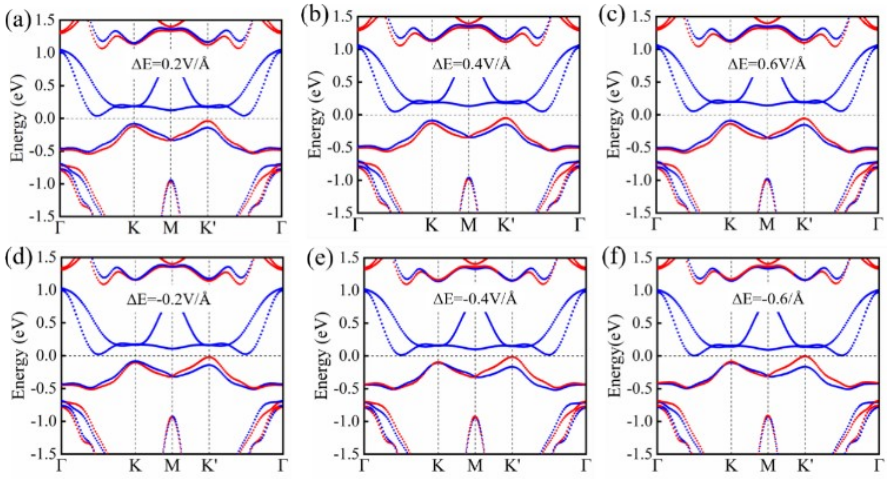

Fig. R4 Spin projection energy bands of CrSSe/MnO_2 heterojunction under different electric field strengths (a) 0.2 V/Å, (b) 0.4 V/Å, (c) 0.6 V/Å, (d) -0.2 V/Å, (e) -0.2 V/Å, (f) -0.2 V/Å.

Table R2 Valley polarization Values of different materials and stacking conditions

	I	II	III	IV	V	VI
CrSe2	48	42	30	28	17	32
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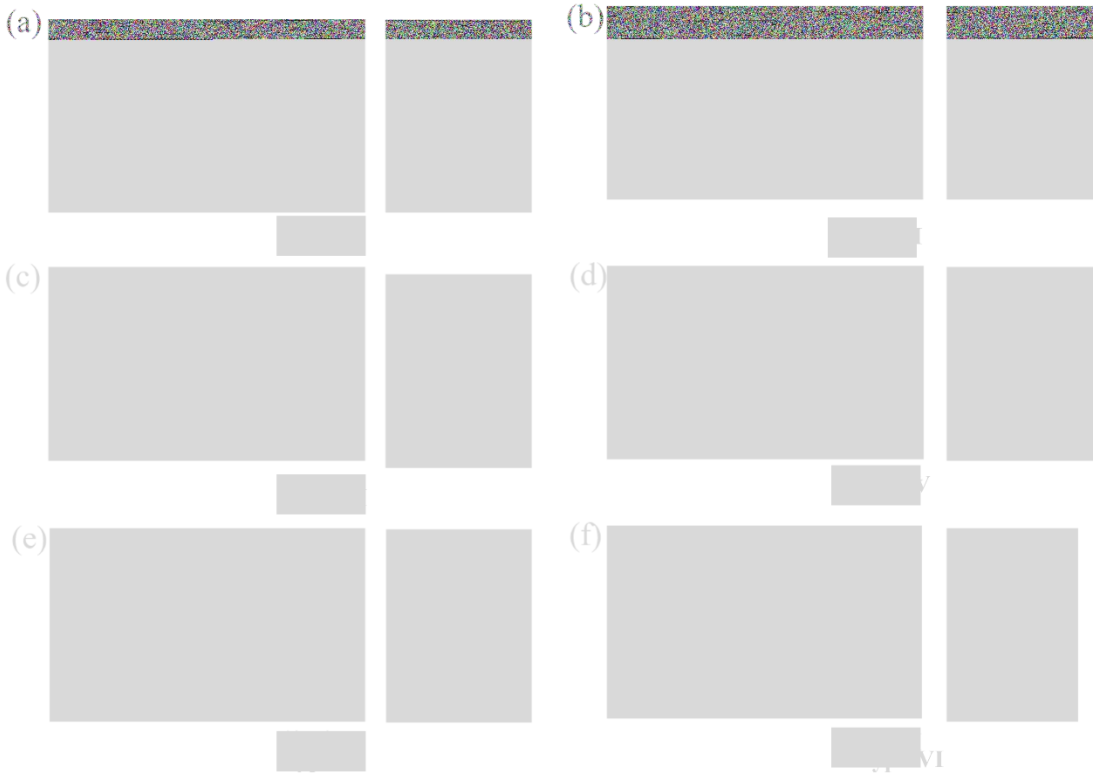

Fig. R5 The Structure of the contacting interface is S atoms in heterojunctions.

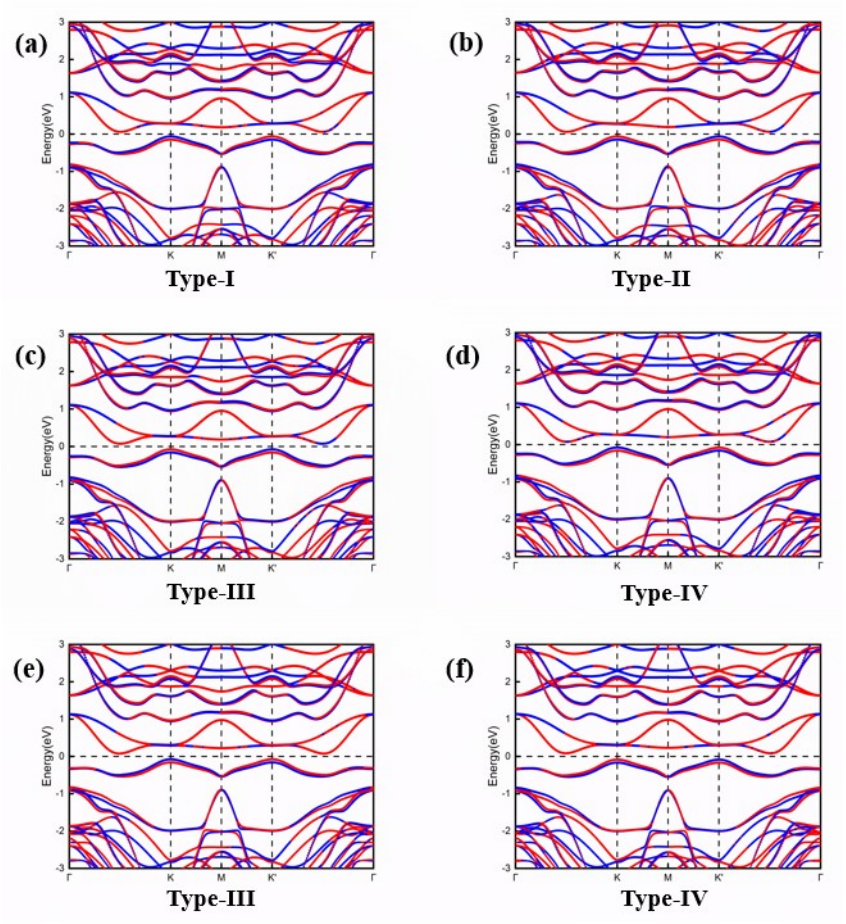

Fig. R6 The band structure of the contacting interface is S atoms in heterojunctions.