

## Supplementary Information

### Enhancement and Modulation of Valley Polarization in Janus CrSSe with Internal and External Electric Fields

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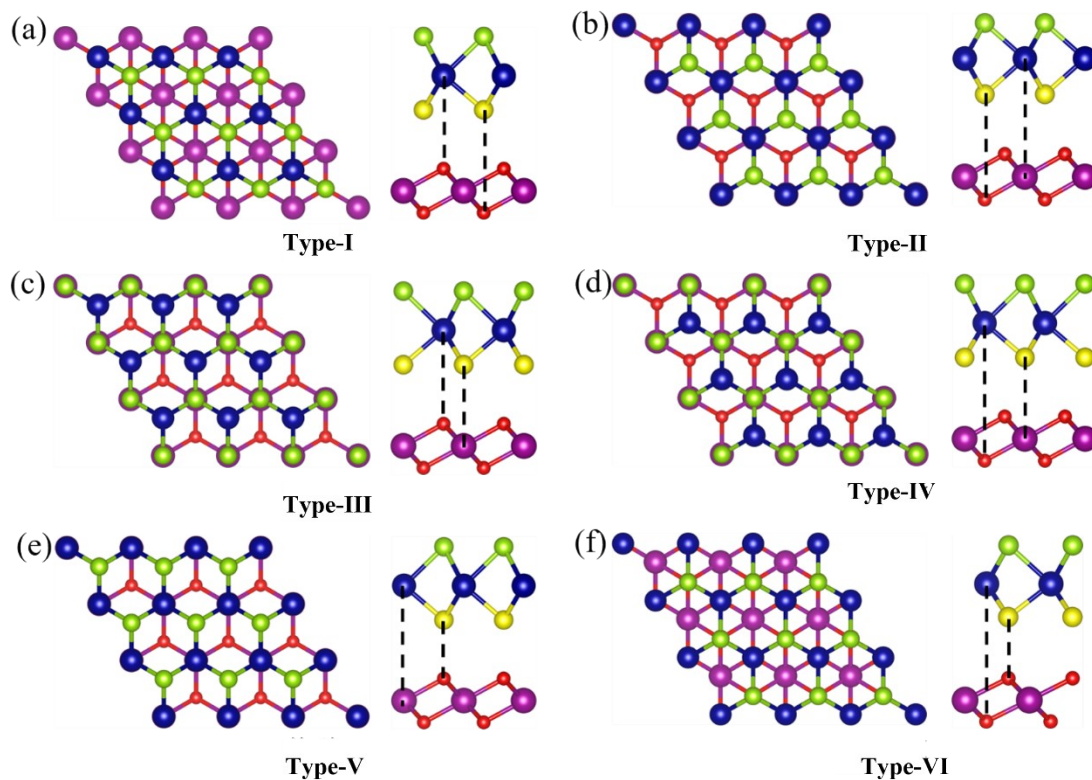
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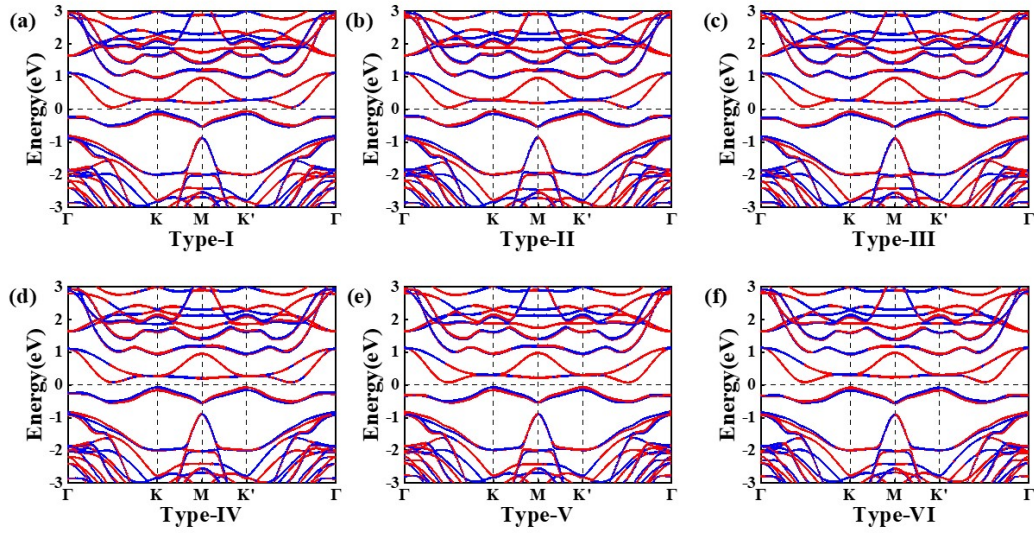
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**Table S1 Valley polarization Values of different materials and stacking conditions**

	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



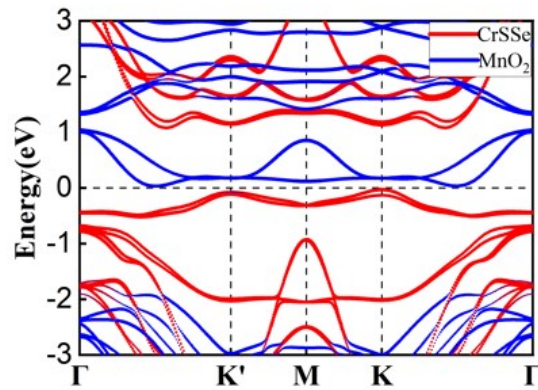
**Fig. S1** Structure of CrSSe/MnO2 under different stacks on the contacting interface is S atom.



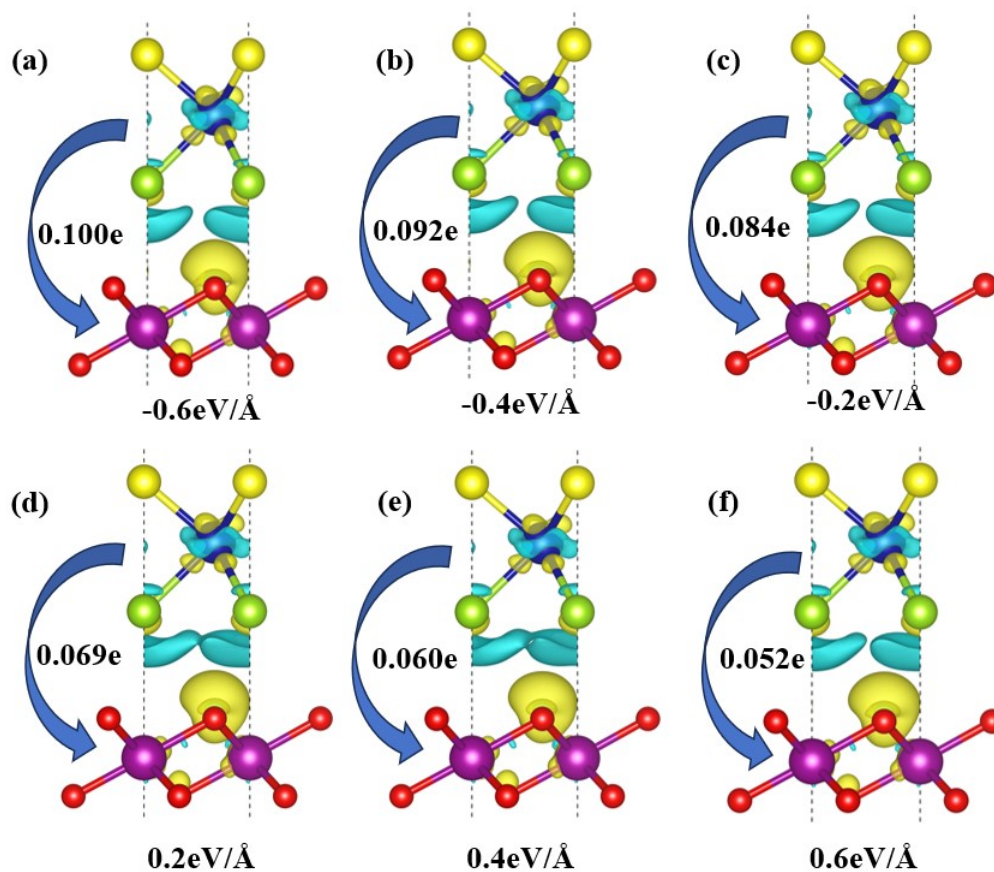
**Fig. S2** Band structures corresponding to each structure in the **Fig. S1**.

**Table S2** Berry curvature of different materials in papers.

Materials	Berry curvature (Bohr <sup>2</sup> )
SnS <sup>1</sup>	4
TcIrGe <sub>2</sub> S <sub>6</sub> <sup>2</sup>	9.48
CrSSe (our materials)	30.83
MoS <sub>2</sub> <sup>3</sup>	62
TiBrI <sup>4</sup>	106



**Fig. S3** The layer-resolved band structure of CrSSe/MnO<sub>2</sub>.



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

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