

Electronic supplementary information

First-Principles Decoding of Spin-Valley-Polarized H-Phase TMDs: Formation Energies, Magnetic Ground States, and Band Engineering

Shijie Guo,^a Kai Cheng,^{a,*} Guotai Zhang,^a Peng Wu,^a Jianpei Xing,^{b,*} Sandong Guo^a and Yan Su,^c

^a*School of Electronic Engineering, XI'AN University of Posts and Telecommunications, Xi'an 710121, China*

^b*Shaanxi University Key Laboratory of Photonic Power Devices and Discharge Regulation, Xi'an University of Technology, Xi'an 710048, China*

^c*Key Laboratory of Materials Modification by Laser, Ion and Electron Beams (Dalian University of Technology), Ministry of Education, Dalian 116024, China*

**Correspondence and requests for materials should be addressed to Kai Cheng (chengkai_xiyou@163.com) or Jianpei Xing (xingjianpei@xaut.edu.cn)*

Table S1 Calculated valley polarization $\Delta_{KK'}$ (meV) for VX_2 , NbX_2 and TaX_2 ($X = S, Se, Te$) upon enabling spin-orbit coupling (SOC).

Material	$\Delta_{KK'}$ (meV)	Material	$\Delta_{KK'}$ (meV)	Material	$\Delta_{KK'}$ (meV)
VS ₂	78	NbS ₂	137	TaS ₂	68
VSe ₂	137	NbSe ₂	173	TaSe ₂	264
VTe ₂	229	NbTe ₂	251	TaTe ₂	318

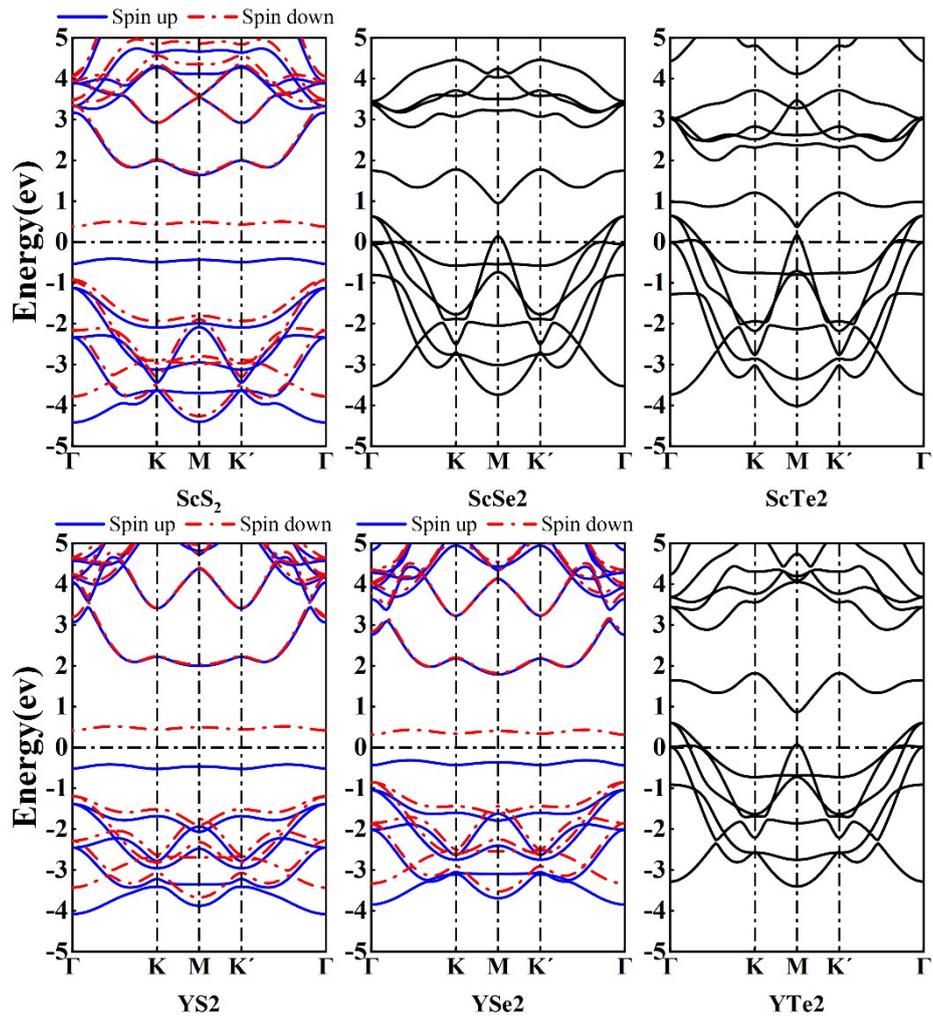


Fig. S1 Schematic illustration of the electronic band structures and spin characteristics of Group-IIIB (Sc, Y) TMDs.

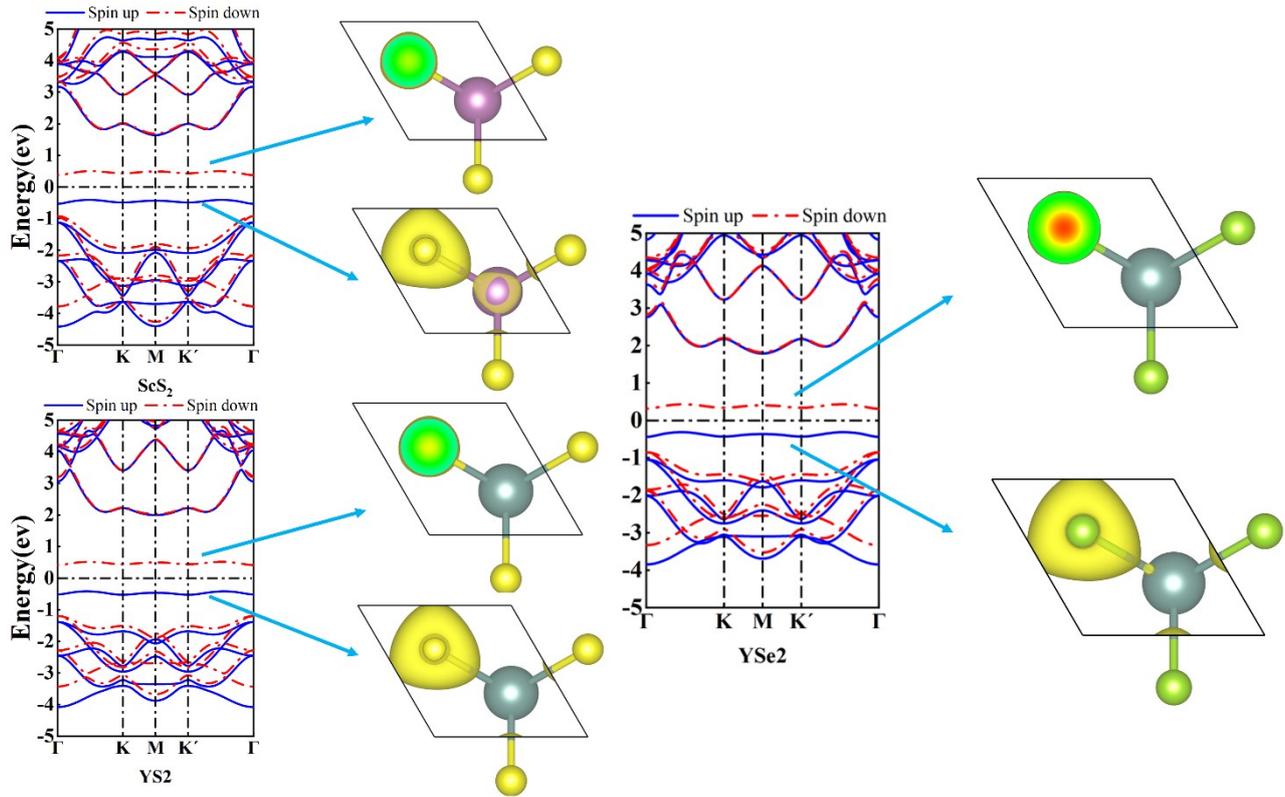


Fig. S2 Charge density distributions of flat-band states near the Fermi level in ScS₂, YS₂, and YSe₂.

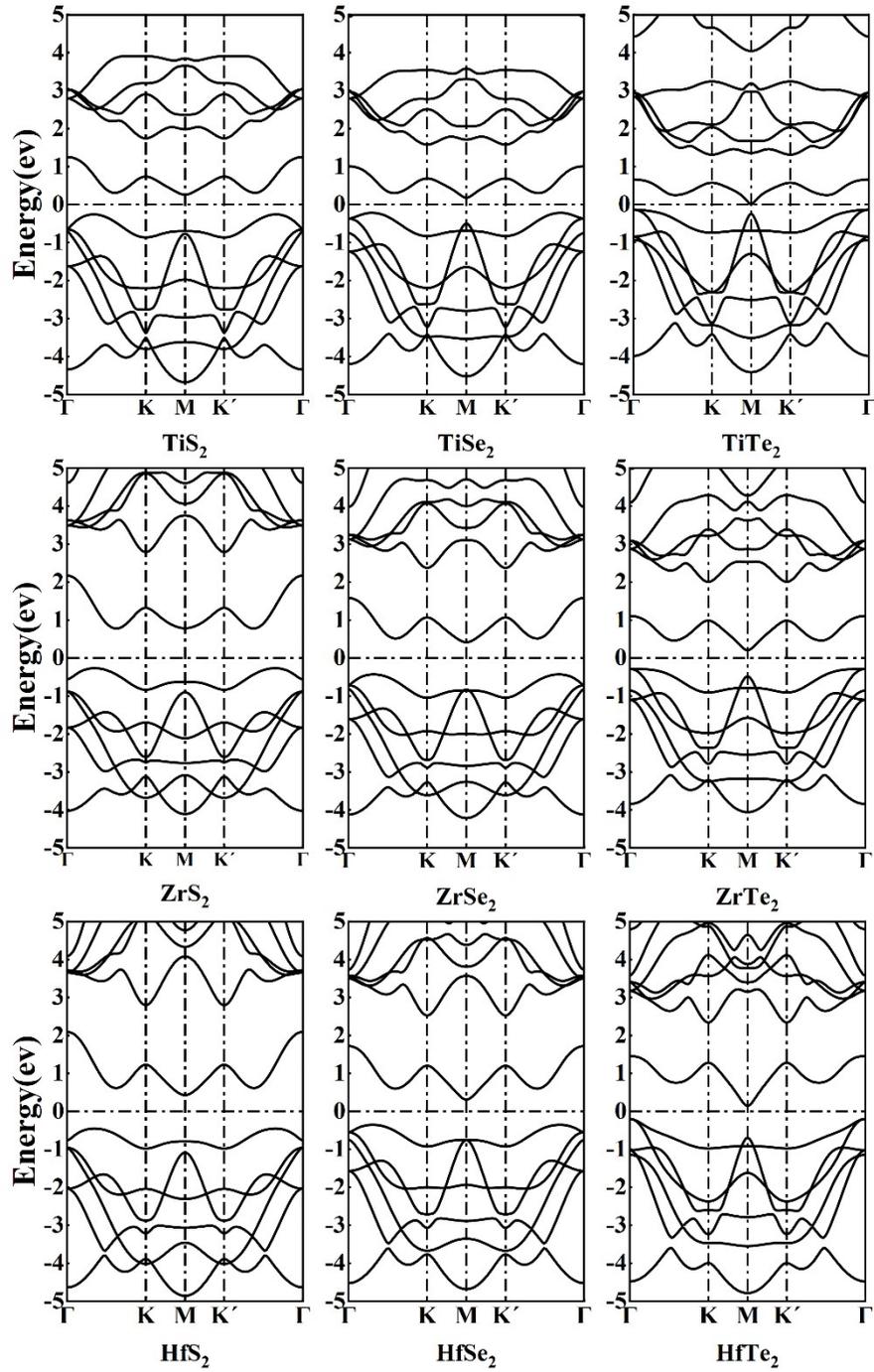


Fig. S3 Schematic illustration of the electronic band structures and spin characteristics of Group-IVB (Ti, Zr, and Hf) TMDs.

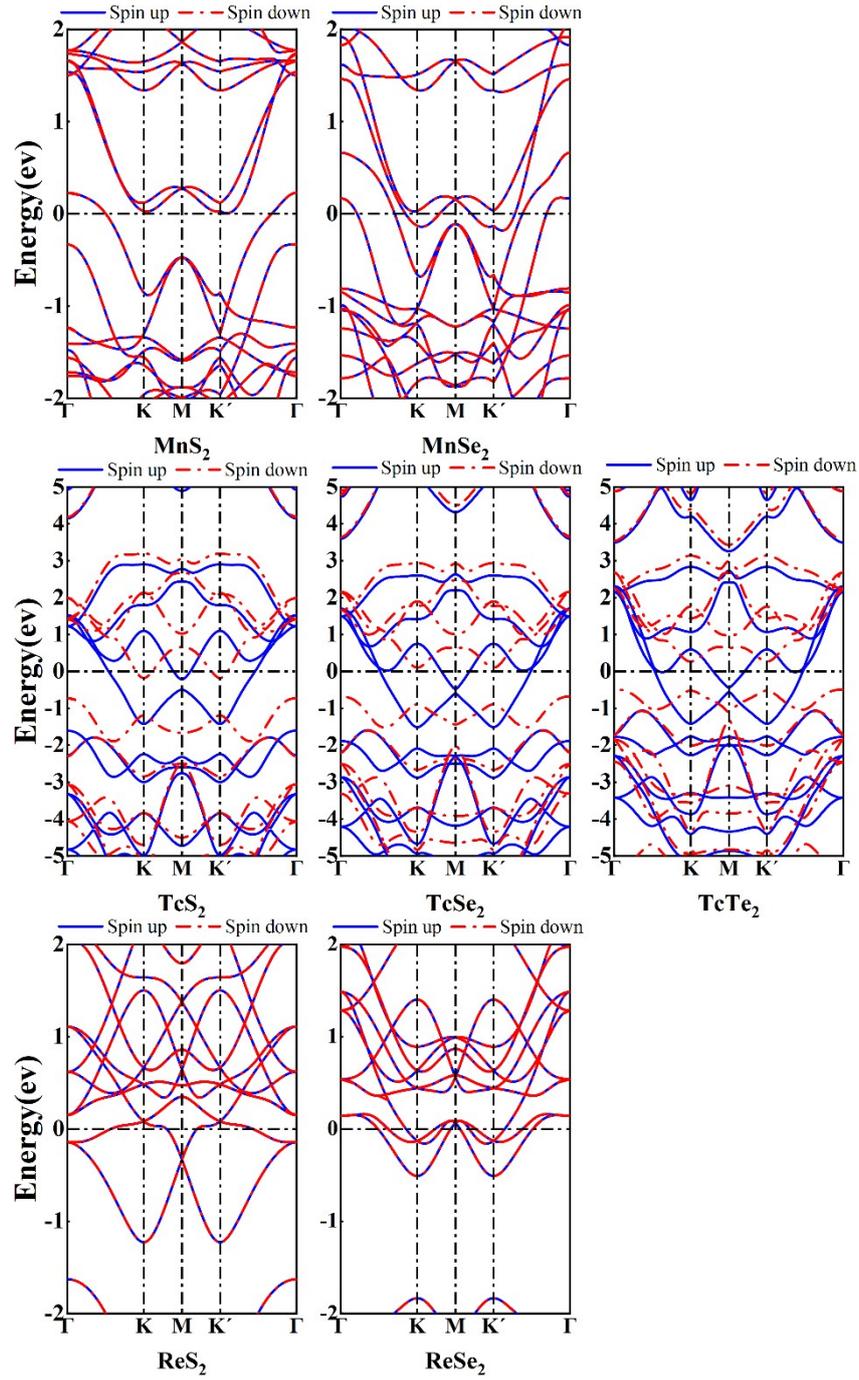
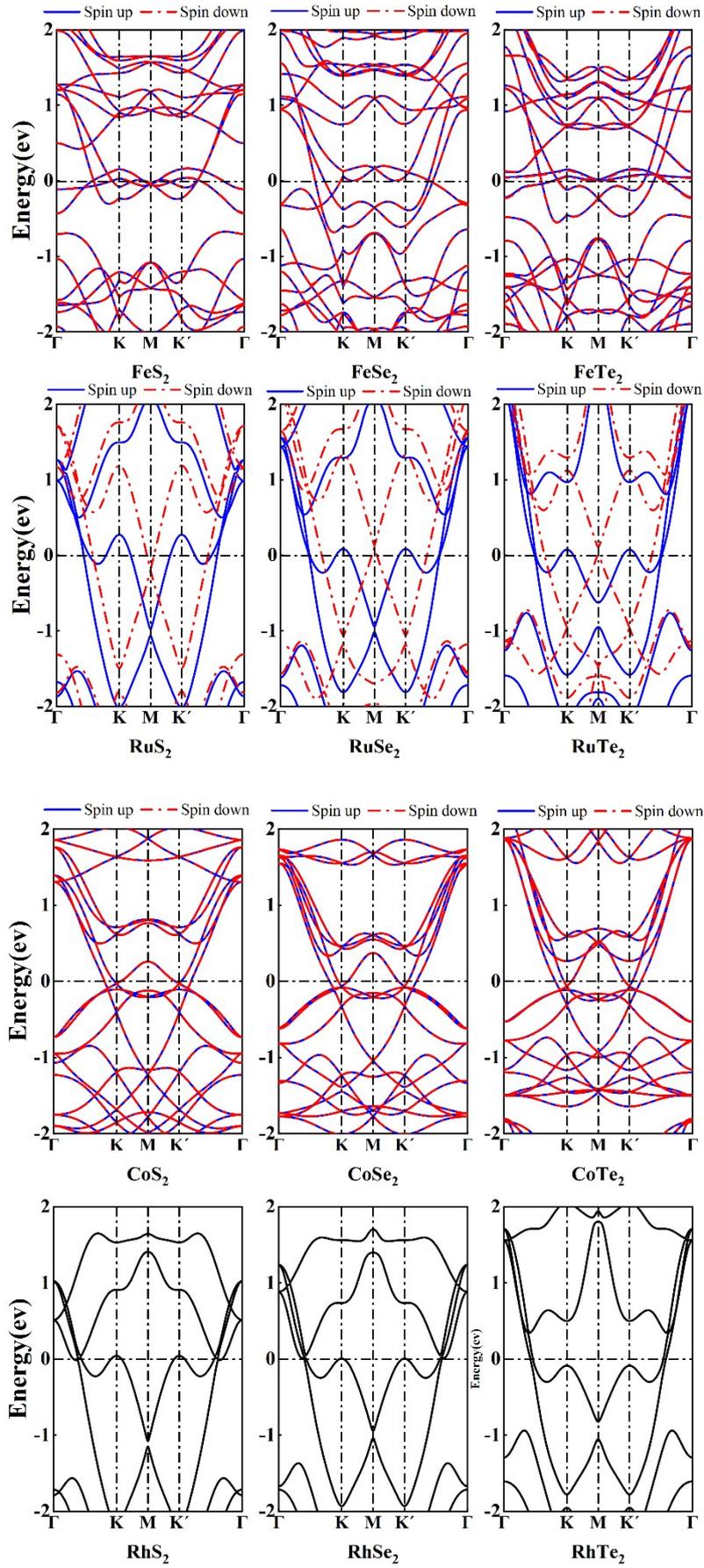


Fig. S4 Schematic illustration of the electronic band structures and spin characteristics of Group-VIIB (Mn, Tc, and Re) TMDs.



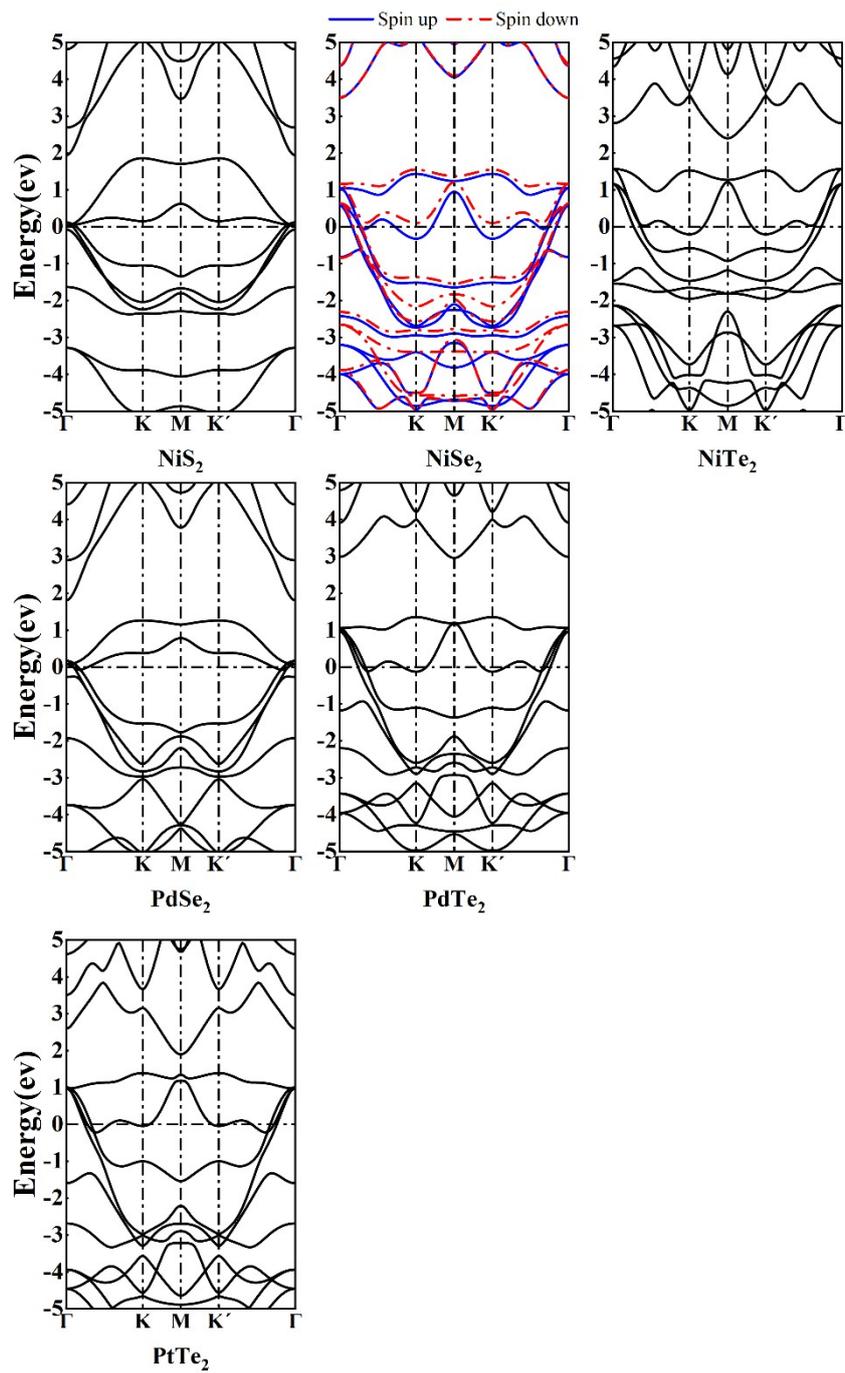


Fig. S5 Schematic illustration of the electronic band structures and spin characteristics of Group-VIII (Fe, Ru, Co, Rh, Ni, Pd, Pt) TMDs.

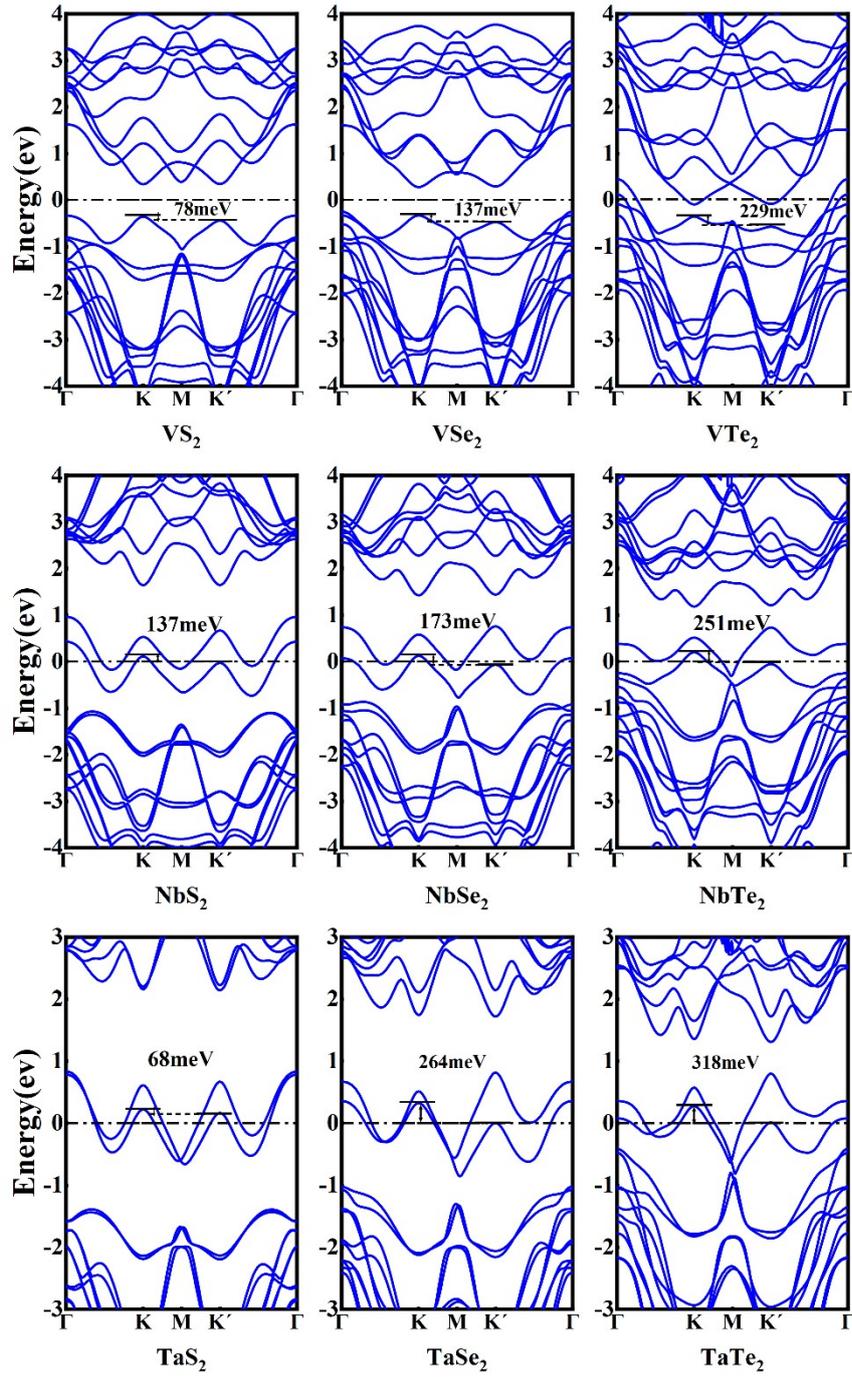


Fig. S6 Schematic illustration of the electronic band structures and spin characteristics of Group-VB (V, Nb, and Ta) TMDs with SOC included; the magnitudes of the valley polarization are annotated.

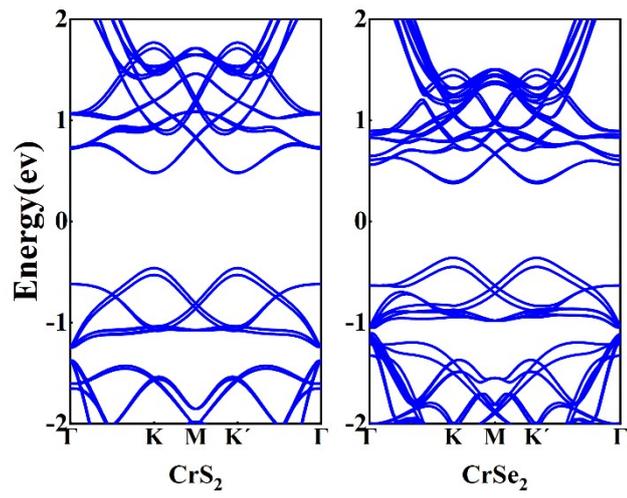


Fig. S7 Schematic illustration of the electronic band structures and spin characteristics of CrS₂ and CrSe₂ with SOC included.