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

Supplementary Information for:

Prediction of Flatness-driven Quantum Spin Hall Effect in Functionalized Germanene and Stanene

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Section I:

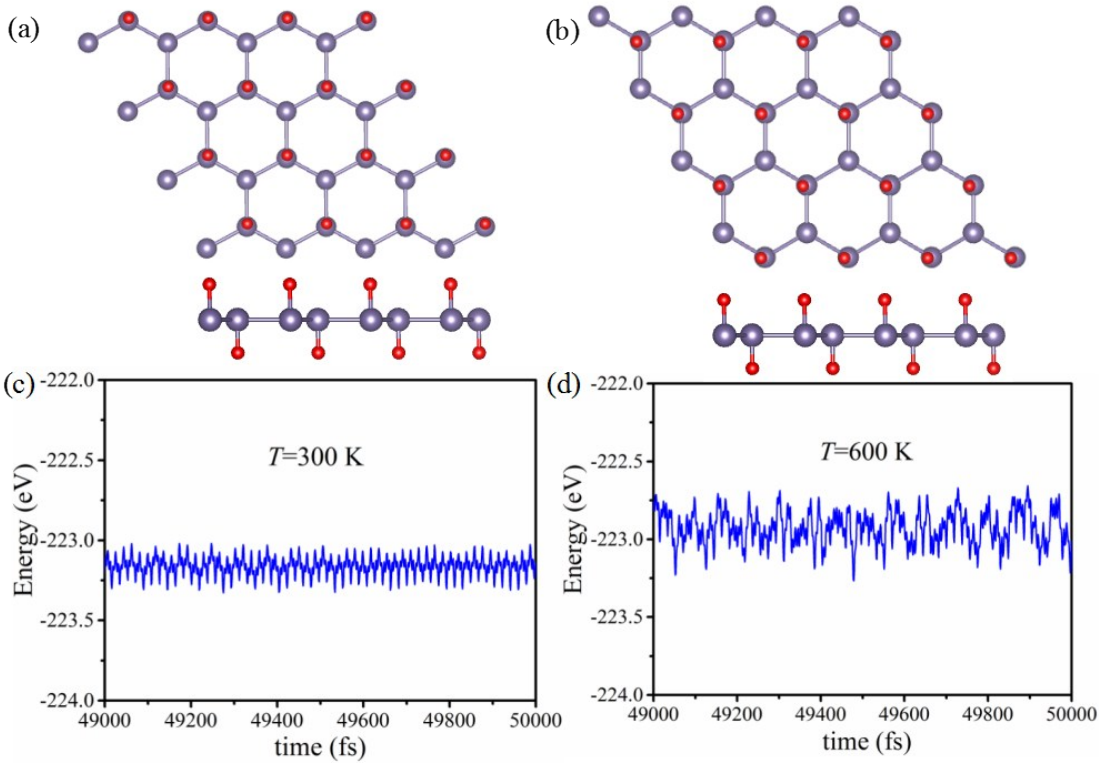

Fig. S1 (a) and (b) present snapshots of the molecular dynamics (MD) simulation of the structures for *f*-GeH_2 at the temperatures of 300 K and 600 K. (c) and (d) show variation of free energies at 10 ps during ab initio MD simulations at the temperatures of 300 K and 600 K for *f*-GeH_2, respectively.

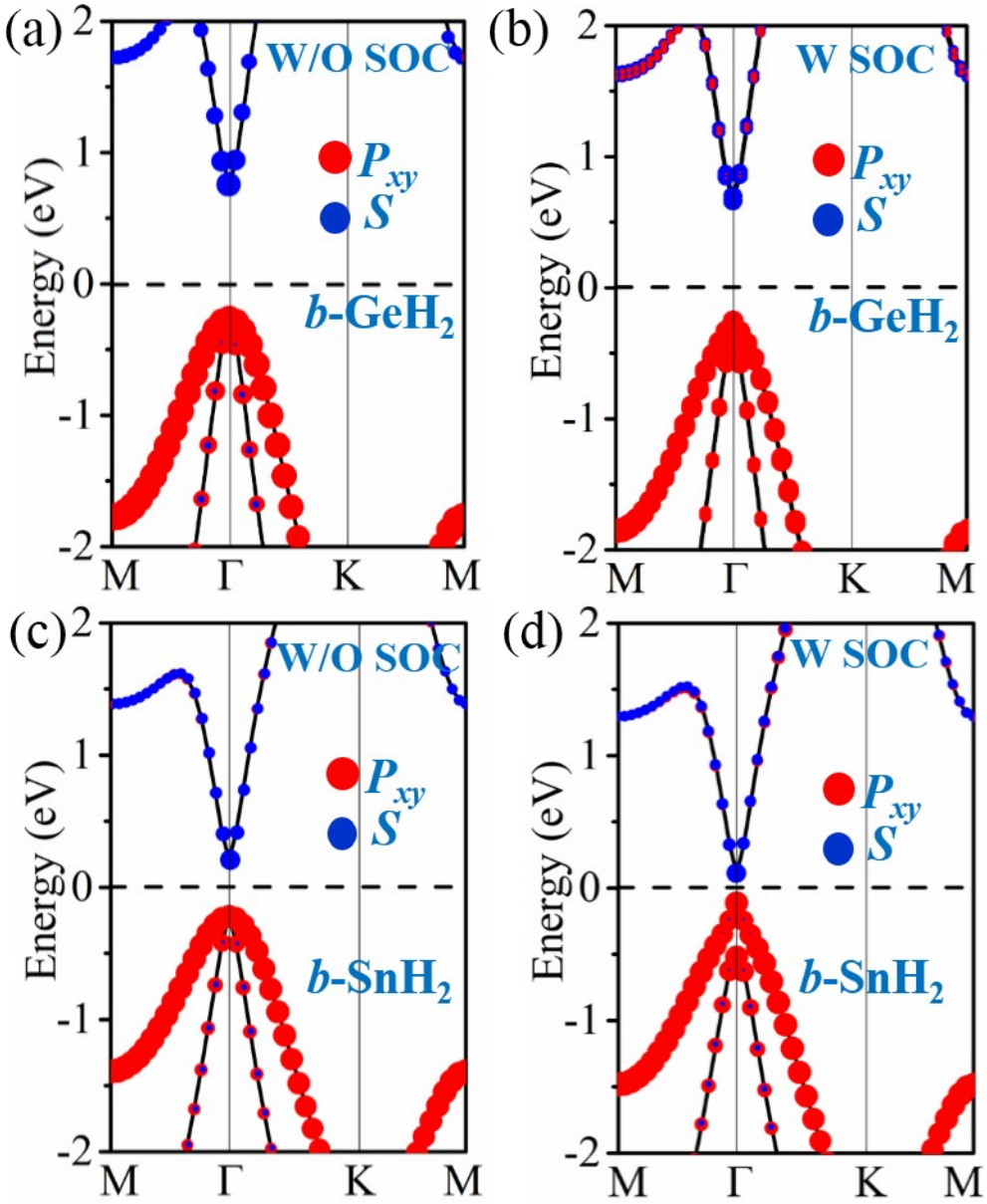

Fig. S2 Orbital-resolved band structures without SOC and with SOC of b-GeH_2 (a-b) and b-SnH_2 (c-d) under equilibrium state, respectively. The blue dots represent the contributions from the *s* atomic orbital of Ge(Sn) atom, while red dots represent contributions from $p_{x,y}$ orbitals of Ge(Sn) atom.

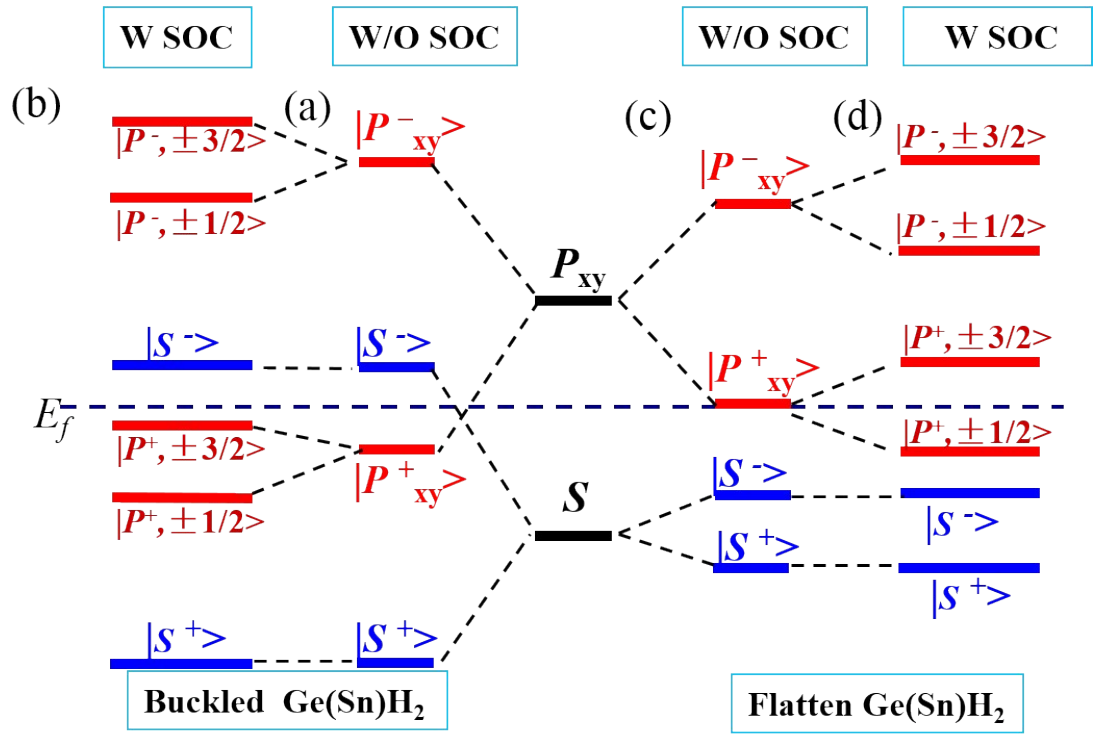

Fig. S3 The evolution of atomic *s* and $p_{x,y}$ orbitals without SOC and with SOC of (a) and (b) belong to *b*-Ge(Sn)H_2, (c) and (d) refer to *f*-Ge(Sn)H_2. The horizontal blue dashed lines indicate the Fermi level.

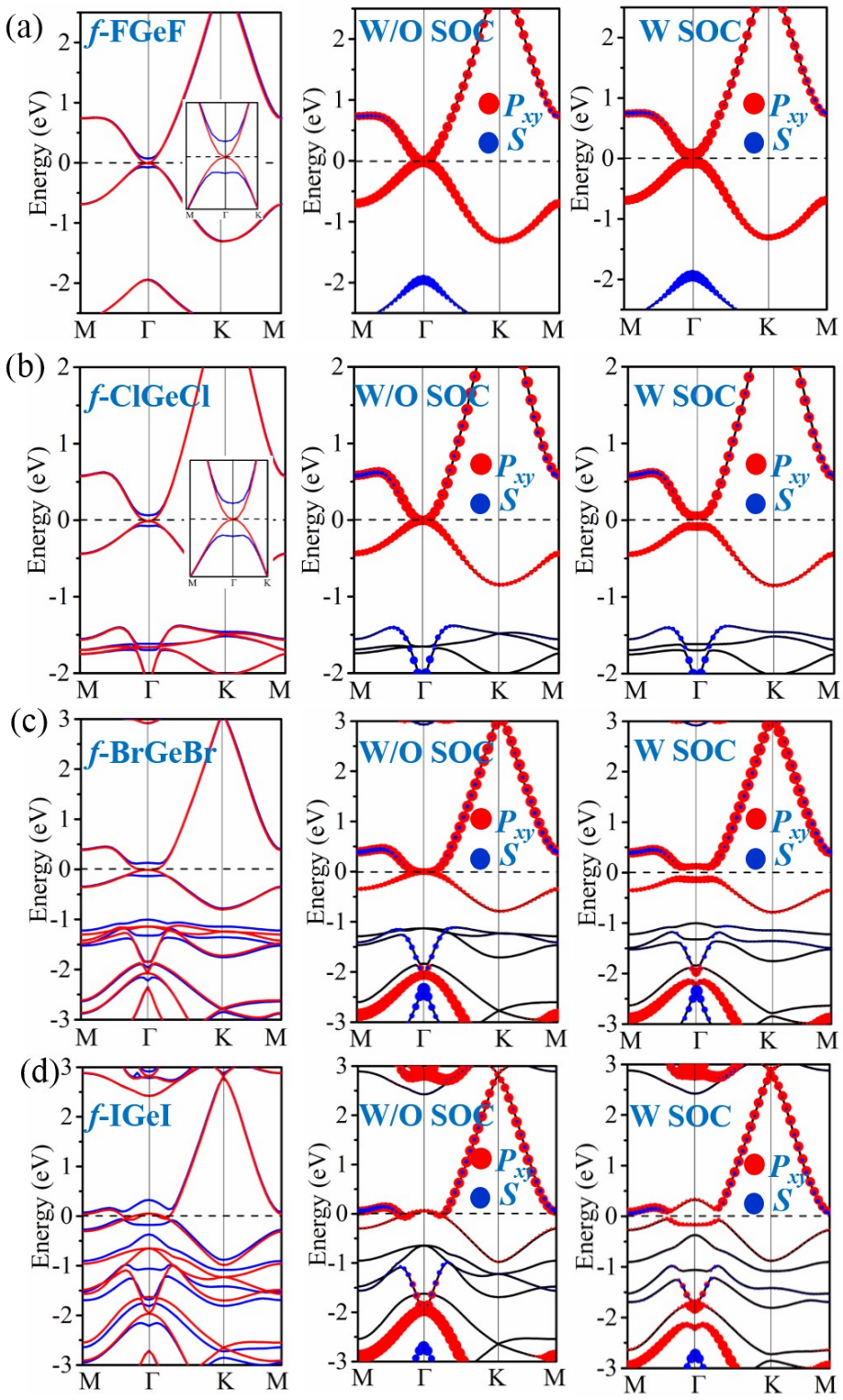

Fig. S4 Orbital-resolved band structures without SOC and with SOC of (a) *f*-GeF_2, (b) *f*-$GeCl_2$, (c) *f*-$GeBr_2$, (d) *f*-GeI_2, respectively. The blue dots represent the contributions from the *s* atomic orbital of Ge atom and the red dots represent contributions from the $p_{x,y}$ atomic orbitals of Ge atom.

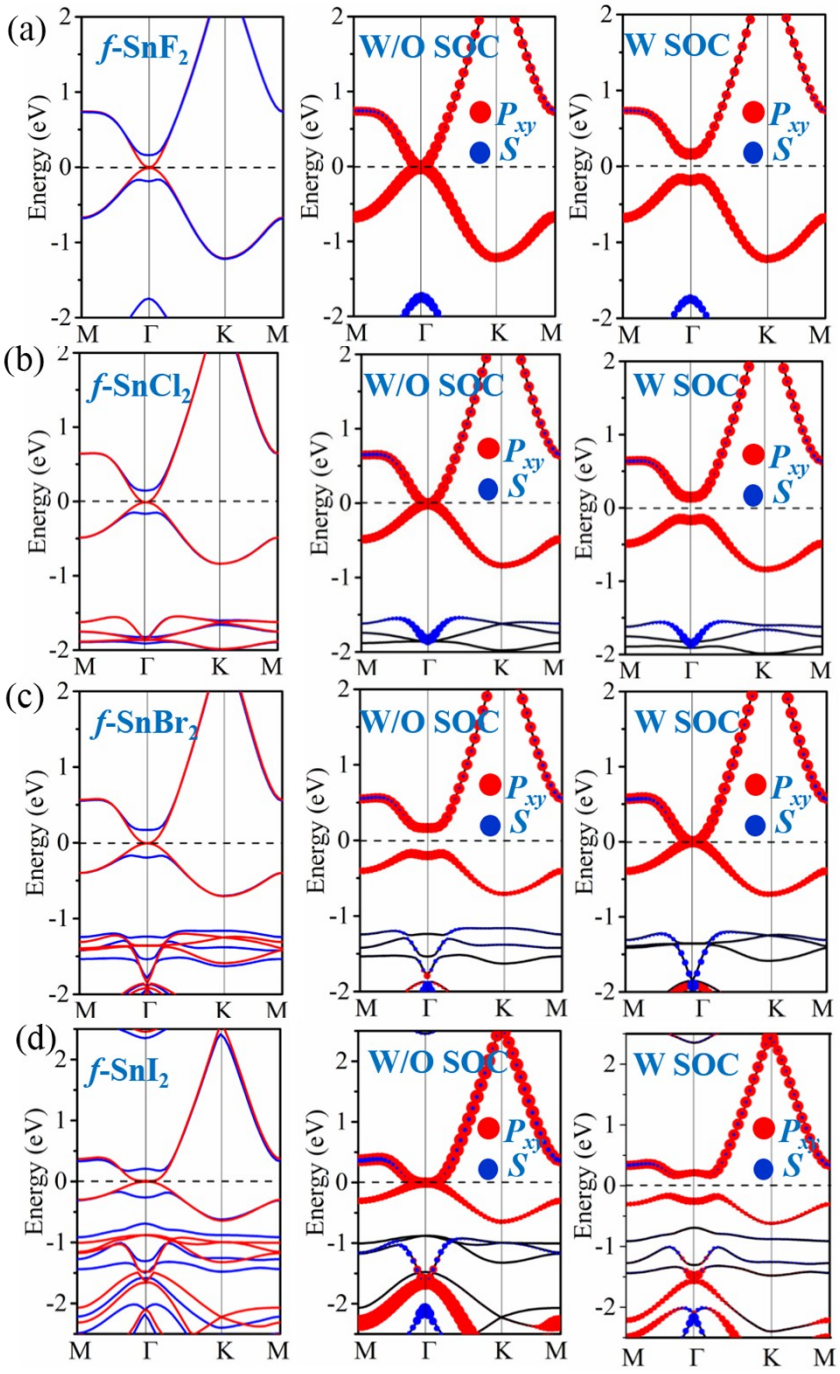

Fig. S5 Orbital-resolved band structures without SOC and with SOC of (a) *f*-SnF_2, (b) *f*-$SnCl_2$, (c) *f*-$SnBr_2$, (d) *f*-SnI_2, respectively. The blue dots represent the contributions from the *s* atomic orbital of Sn atom and the red dots represent contributions from the $p_{x,y}$ atomic orbitals of Sn atom.

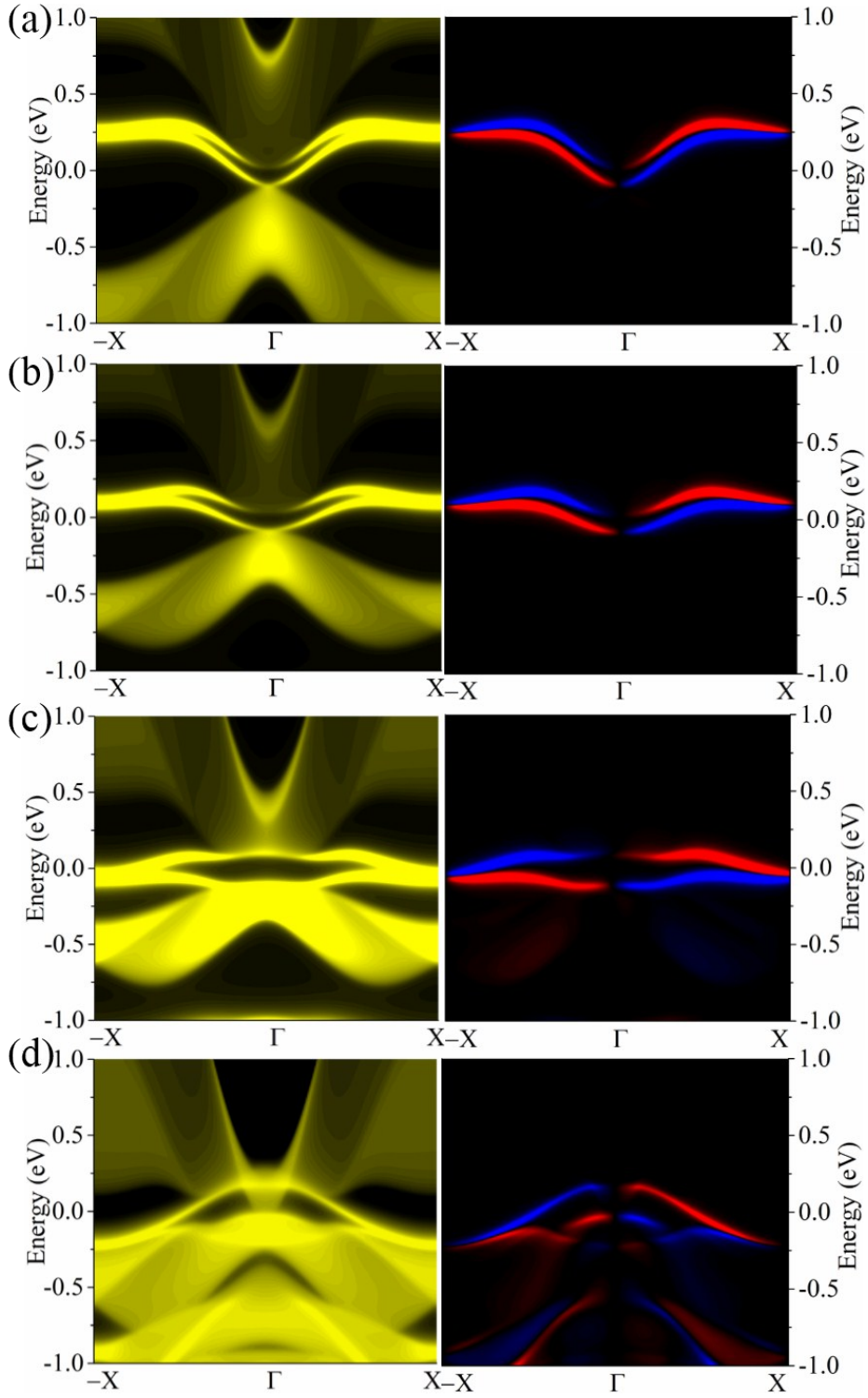
(a)
(b)
(c)
(d)
Energy (eV)
1.0
0.5
0.0
-0.5
-1.0
−X
Γ
X

Fig. S6 Total (left panel) and spin (right panel) edge density of states for (a) *f*-GeF_2, (b) *f*-$GeCl_2$, (c) *f*-$GeBr_2$ and (e) *f*-GeI_2. In the spin edge plot, red/blue lines denote the spin up/down polarization.

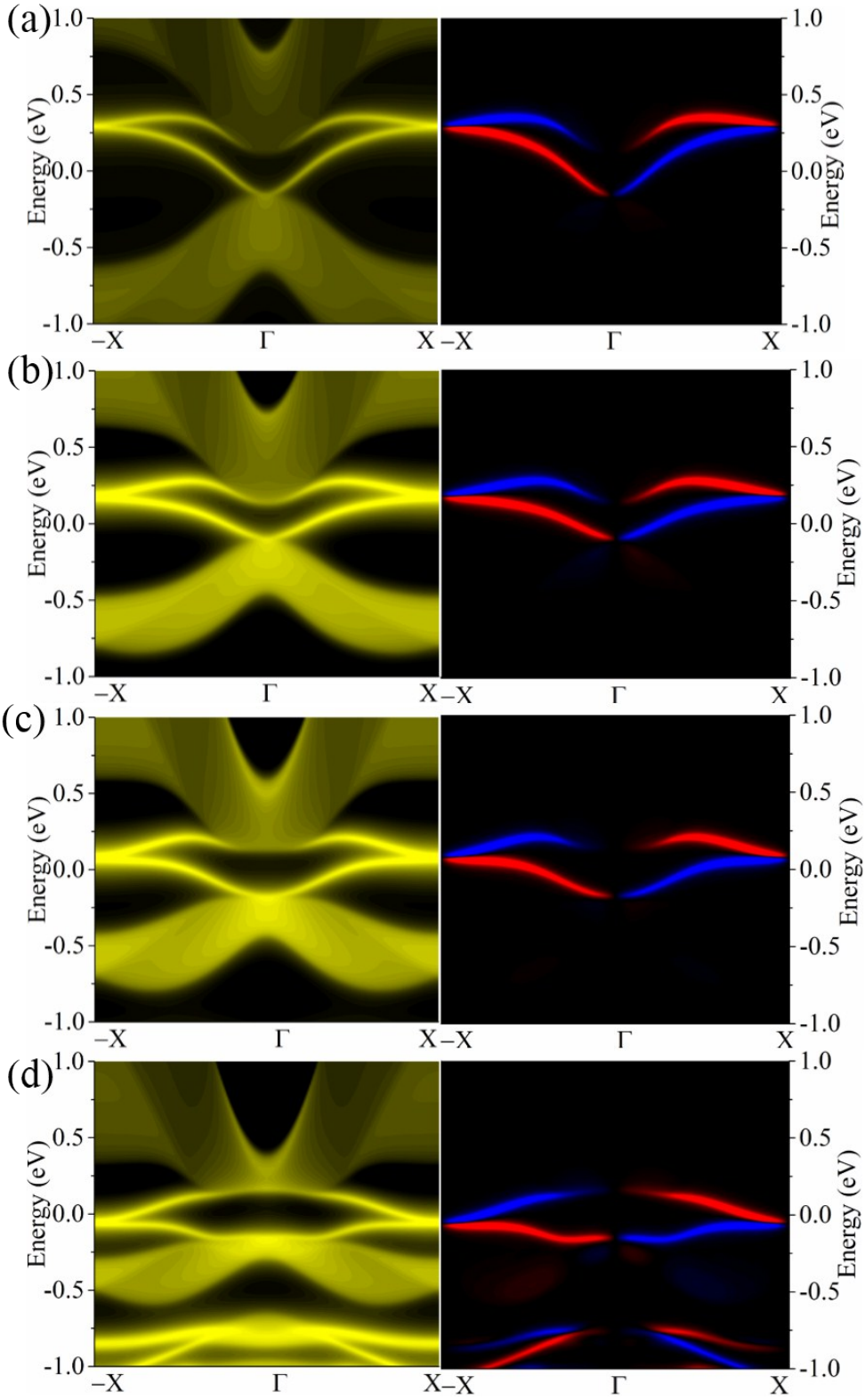

Fig. S7 Total (left panel) and spin (right panel) edge density of states for or (a) *f*-SnF_2, (b) *f*-$SnCl_2$, (c) *f*-$SnBr_2$ and (e) *f*-SnI_2. In the spin edge plot, red/blue lines denote the spin up/down polarization.

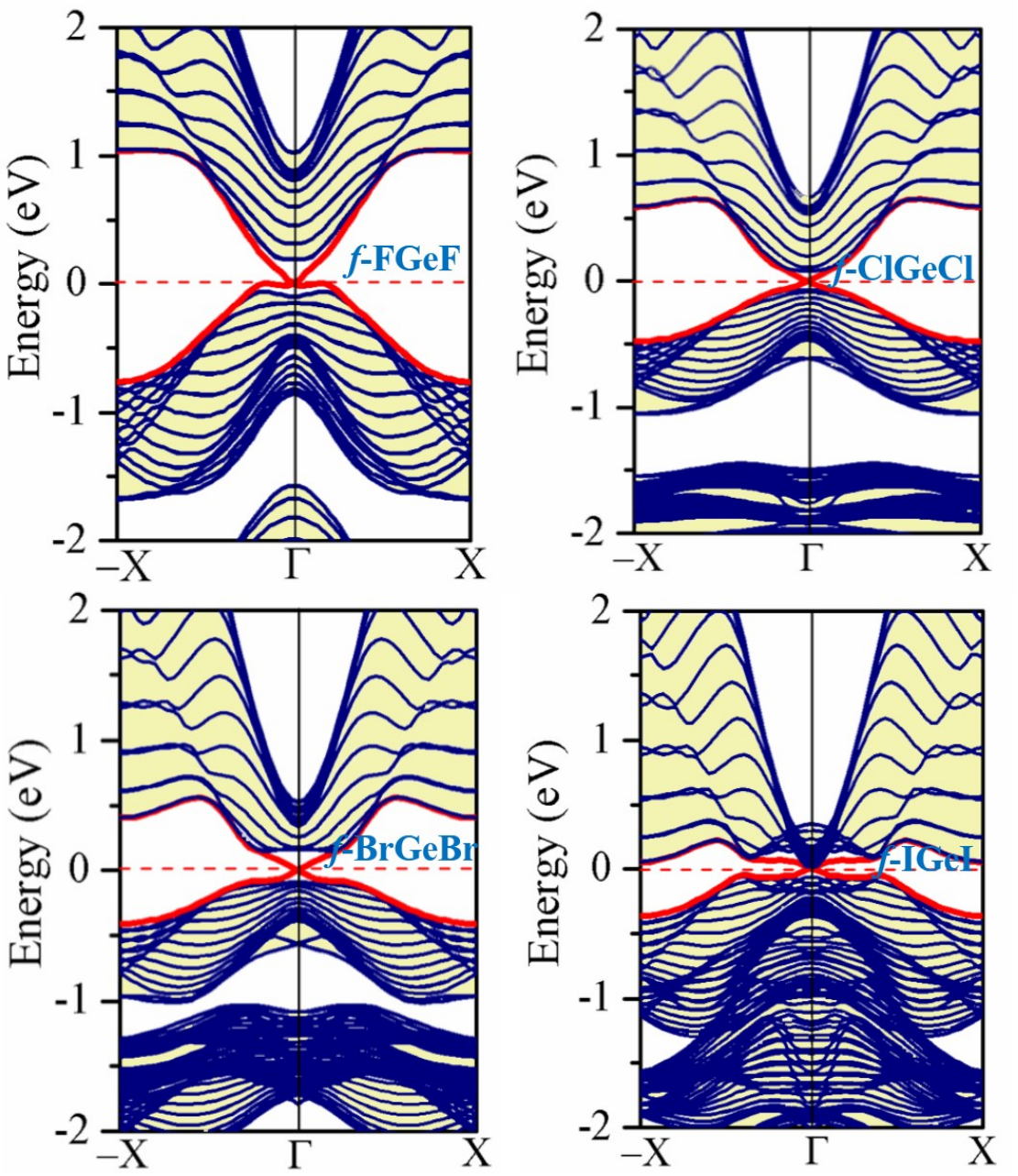

Fig. S8 (a) *f*-GeF_2, (b) *f*-$GeCl_2$, (c) *f*-$GeBr_2$ and (d) *f*-GeI_2 indicate the band structures of zigzag-type edge states in QSH phase. The helical edge states are indicated by the red lines.

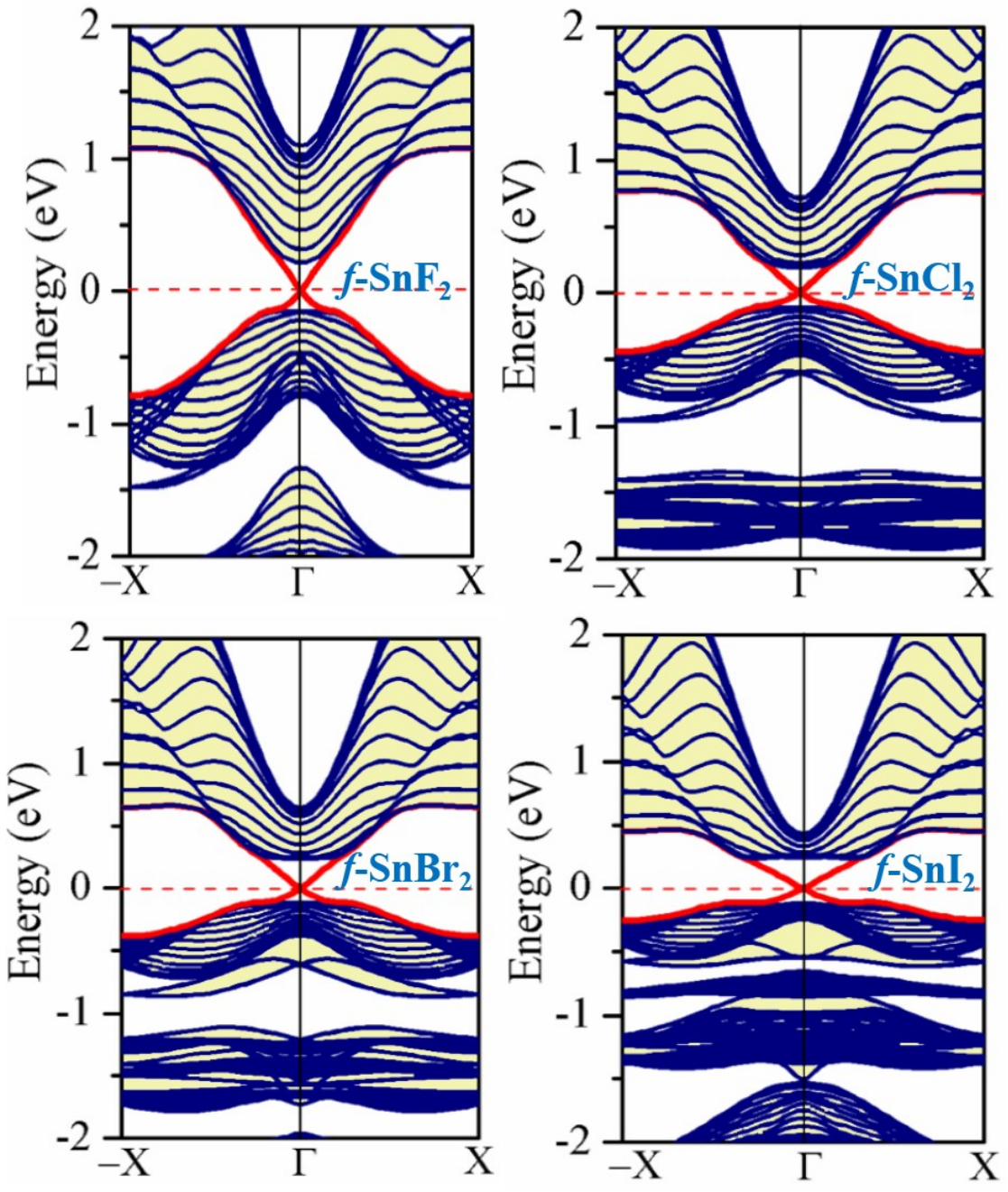

Fig. S9 (a) *f*-SnF_2, (b) *f*-$SnCl_2$, (c) *f*-$SnBr_2$ and (d) *f*-SnI_2 indicate the band structures of zigzag-type edge states in QSH phase. The helical edge states are indicated by the

red lines.

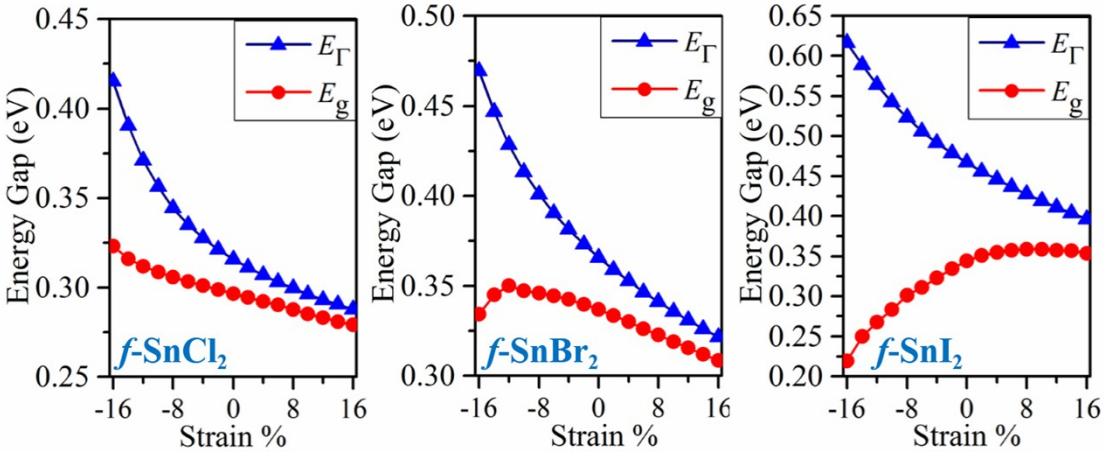

Fig. S10 The calculated energy gaps at Γ point (E_Γ) and the global energy gap (E_g) of (a) *f*-$SnCl_2$, (b) *f*-$SnBr_2$ and (d) *f*-SnI_2 with SOC as a function of external strain.

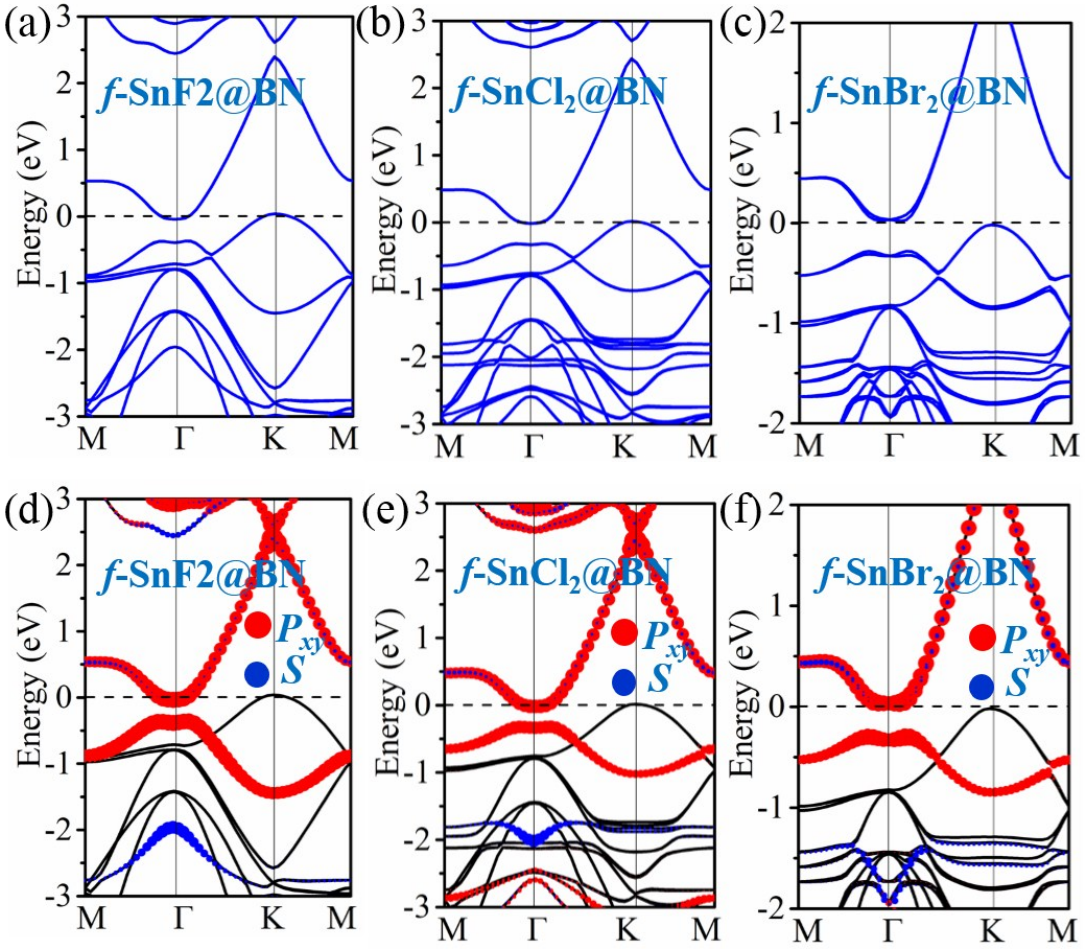

Fig. S11 The band structures with SOC for (a) *f*-SnF_2@BN, (b) *f*-$SnCl_2$@BN and (c) *f*-$SnBr_2$@BN, respectively. Orbital-resolved band structures with SOC of (d) *f*-SnF_2@BN, (e) *f*-$SnCl_2$@BN and (f) *f*-$SnBr_2$@BN, respectively. The blue dots represent the contributions from the *s* atomic orbital of Sn atom and the red dots represent contributions from the $p_{x,y}$ atomic orbitals of Sn atom.

Section II:

Here, we take the *f*-SnH as typical example:

POSCAR:

f-SnH

1.00000000000000

4.3771849042026743 -2.5162743277888593 0.0000000000000000

0.0000000000000000 5.0325774383195130 0.0000000000000000

0.0000000000000000 0.0000000000000000 17.5202999114990234

H Sn

2 2

Selective dynamics

Direct

0.6666666666666667 0.3333333333333333 0.6008614058421329 T T T

0.3333333333333333 0.6666666666666667 0.3931583104073795 T T T

0.3333333333333333 0.6666666666666667 0.5000000000000000 T T F

0.6666666666666667 0.3333333333333333 0.5000000000000000 T T F

INCAR (self-consistent)

!.....

ISTART = 0

ICHARG = 2

NSW = 0

IBRION = -1

ISPIN = 2

!.....

ENCUT = 500

ISMEAR = 0

SIGMA = 0.05

ISIF = 2

EDIFF = 1E-5

EDIFFG = -0.02

LORBIT = 11

POTIM = 0.4

LREAL=Auto

!.....

SYSTEM = *f*-HSn

PREC = Accurate

LPLANE = .TRUE.

LSCALU = .FALSE.

NSIM = 12

ALGO = Fast

!NPAR = 1

SYMPREC = 1E-4

ISYM = 0

LMAXMIX = 4

KPOINTS :

k-points along high symmetry lines

52

Reciprocal

0.000000 0.500000 0.000000 1.00

0.000000 0.470588 0.000000 1.00

0.000000 0.441176 0.000000 1.00

0.000000 0.411765 0.000000 1.00

0.000000 0.382353 0.000000 1.00

0.000000 0.352941 0.000000 1.00

0.000000 0.323529 0.000000 1.00

0.000000 0.294118 0.000000 1.00

0.000000 0.264706 0.000000 1.00

0.000000 0.235294 0.000000 1.00

0.000000 0.205882 0.000000 1.00

0.000000 0.176471 0.000000 1.00

0.000000 0.147059 0.000000 1.00

0.000000 0.117647 0.000000 1.00

0.000000 0.088235 0.000000 1.00

0.000000 0.058824 0.000000 1.00

0.000000 0.029412 0.000000 1.00

0.000000 0.000000 0.000000 1.00

0.017544 0.017544 0.000000 1.00

0.035087 0.035087 0.000000 1.00

0.052631 0.052631 0.000000 1.00

0.070175 0.070175 0.000000 1.00

0.087718 0.087718 0.000000 1.00

0.105262 0.105262 0.000000 1.00

0.122806 0.122806 0.000000 1.00

0.140349 0.140349 0.000000 1.00

0.157893 0.157893 0.000000 1.00

0.175437 0.175437 0.000000 1.00

0.192981 0.192981 0.000000 1.00

0.210524 0.210524 0.000000 1.00

0.228068 0.228068 0.000000 1.00

0.245612 0.245612 0.000000 1.00

0.263155 0.263155 0.000000 1.00

0.280699 0.280699 0.000000 1.00

0.298243 0.298243 0.000000 1.00

0.315786 0.315786 0.000000 1.00

0.333330 0.333330 0.000000 1.00

0.311108 0.344441 0.000000 1.00

0.288886 0.355553 0.000000 1.00

0.266664 0.366664 0.000000 1.00

0.244442 0.377775 0.000000 1.00

0.222220 0.388887 0.000000 1.00

0.199998 0.399998 0.000000 1.00

0.177776 0.411109 0.000000 1.00

0.155554 0.422221 0.000000 1.00

0.133332 0.433332 0.000000 1.00

0.111110 0.444443 0.000000 1.00

0.088888 0.455555 0.000000 1.00

0.066666 0.466666 0.000000 1.00

0.044444 0.477777 0.000000 1.00

0.022222 0.488889 0.000000 1.00

0.000000 0.500000 0.000000 1.00