Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.
This journal is © the Owner Societies 2021

Supplementary Information

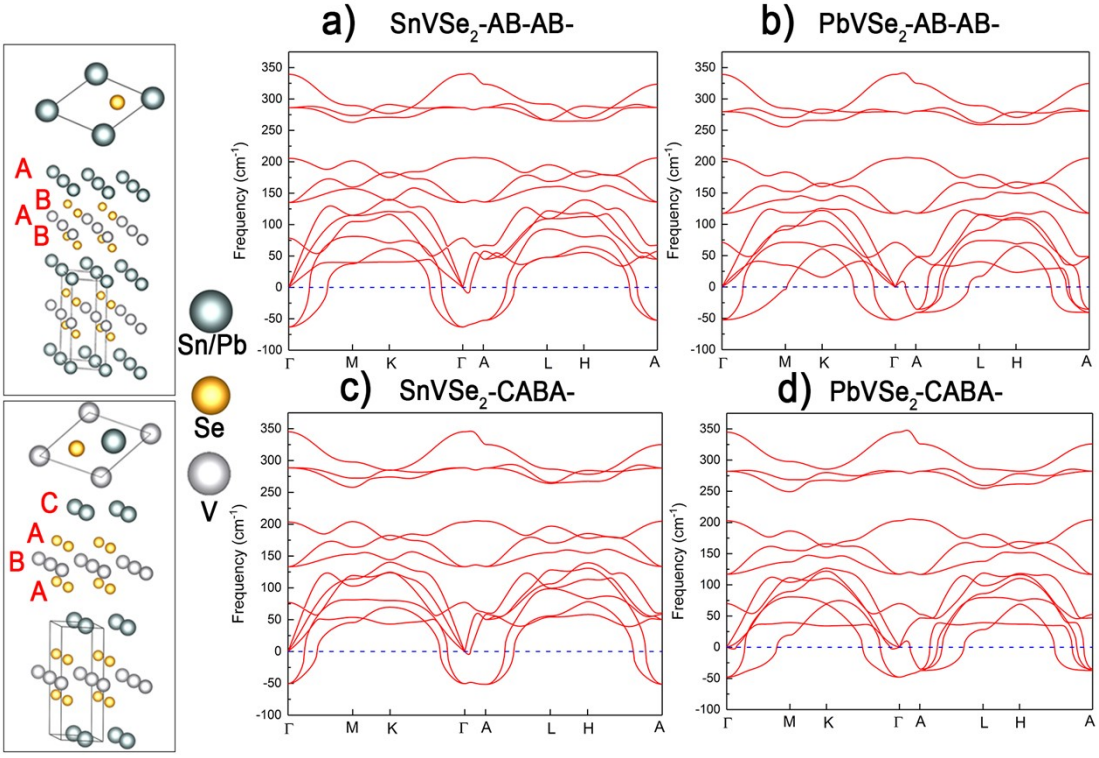

Fig. S1 The structure diagrams and related phonon spectra of $SnVSe_2$ and $PbVSe_2$ with -ABAB- and -CABA- stacking. The results indicate that these configurations are unstable for existing imaginary frequencies.

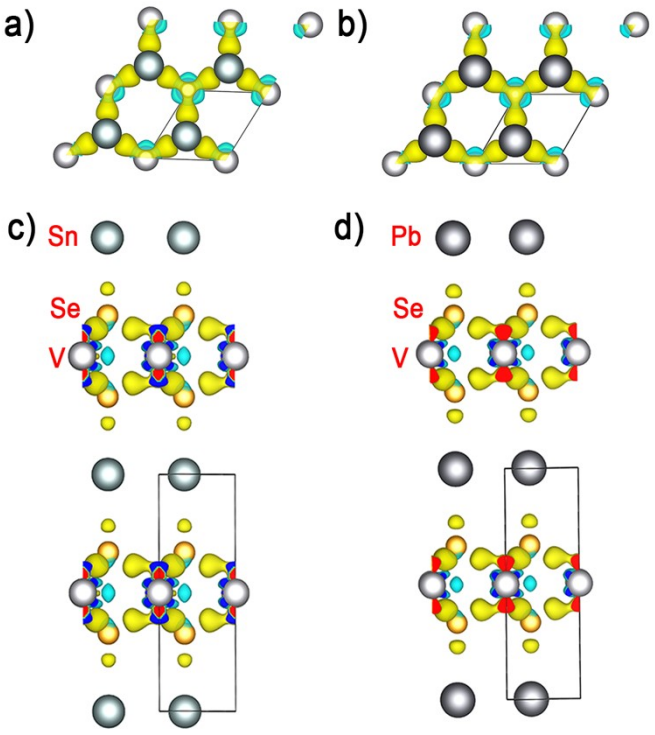

Fig. S2 The charge density differences of $SnVSe_2$: **a)** in top view and **c)** side view, of $PbVSe_2$ in **b)** top view and **d)** side view. The cyan and the yellow parts represent the depletion and accumulation of electrons, respectively. The value of isosurface is set to 0.01 eV. The results clearly depict the ionicity between V-Se atoms and the weak covalent trend between Sn-Se and Pb-Se atoms.

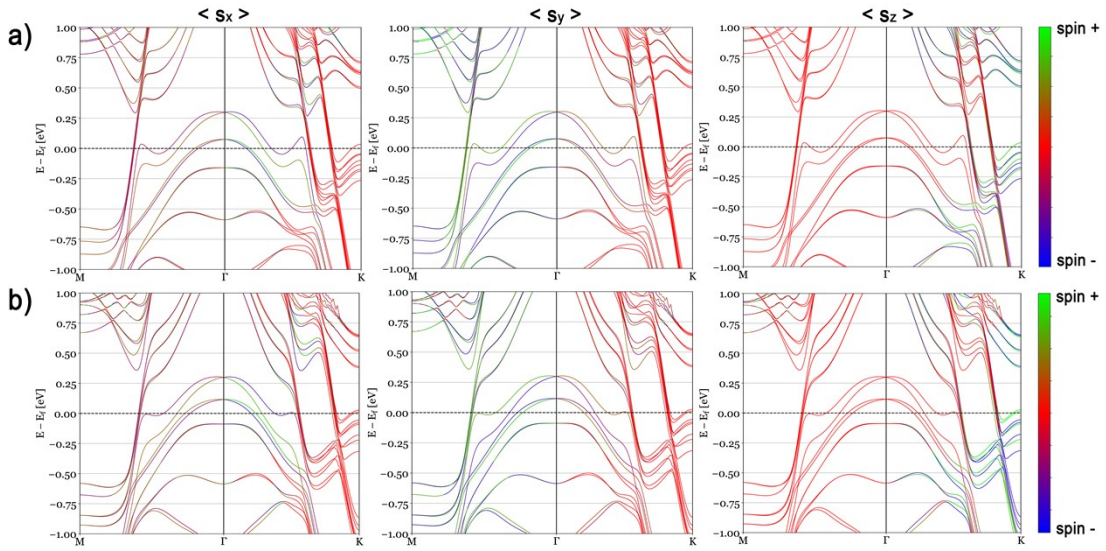

Fig. S3 The calculated spin-decomposed surface band structures by using a slab model containing 4 Sn/Pb-Se-V-Se layers of **a)** $SnVSe_2$ and **b)** $PbVSe_2$. One can clearly observe the spin-momentum lock feature of their surface states of $SnVSe_2$ and $PbVSe_2$ at Γ point, which demonstrates their topological properties, as shown in $< S_x >$ and $< S_y >$.

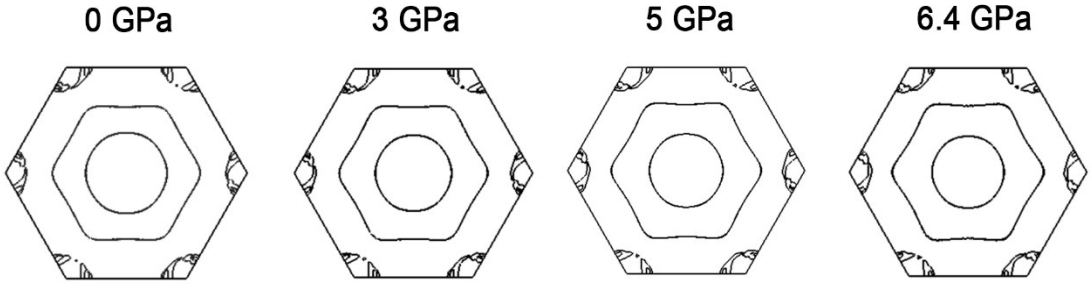

Fig. S4 The sections of Fermi surface contours of $VPbSe_2$ at kz = 0 with external pressures a) 0 GPa, b) 3 GPa, c) 5 GPa and d) 6.4 GPa.