Supporting Information

Two-dimensional topological insulators exfoliated from Na_3Bi-like Dirac semimetals

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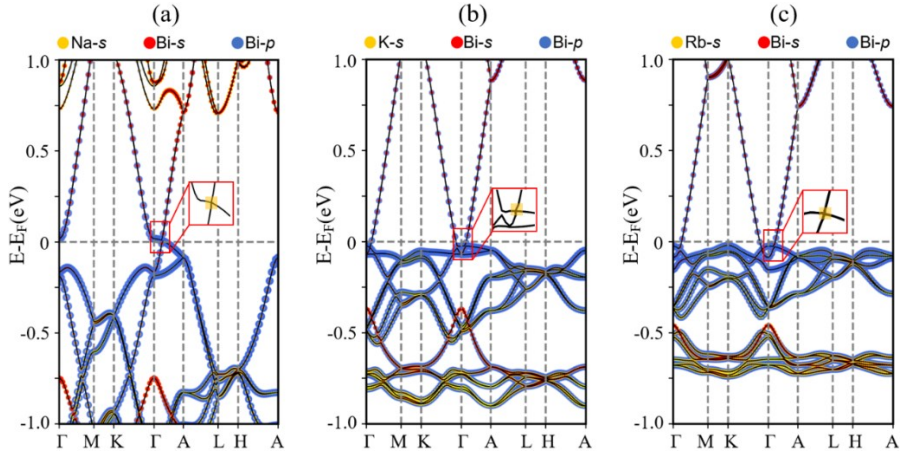

Fig. S1 Electronic structures of bulk $P\bar{3}c1$ phase of (a) Na_3Bi, (b) K_3Bi and (c) Rb_3Bi from PBE + SOC. The dotted lines with different colors denote the band projection onto different atomic orbitals. The yellow squares indicate the positions of Dirac cones.

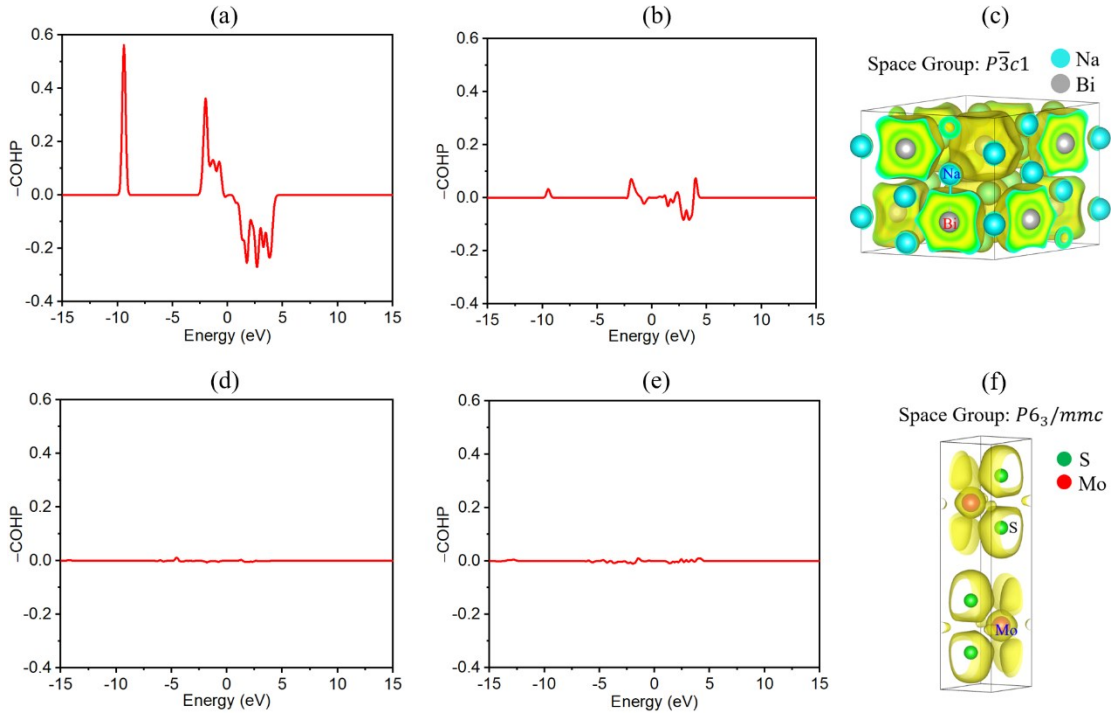

Fig. S2 Bonding/antibonding characters of the states of interlayer (a) Na-Bi, (b) Na-Na pairs in Na_3Bi and interlayer

(d) Mo-S, (e) S-S pairs in MoS_2, as indicated by the calculated Crystal Orbital Hamiltonian Populations (COHPs). Electron localization function of (c) Na_3Bi and (f) MoS_2 with isosurfaces of 0.2. −COHP > 0, −COHP = 0 and −COHP < 0 mean the bonding, non-bonding and anti-bonding characters of each electronic state over the chemical bonds in the system, respectively.

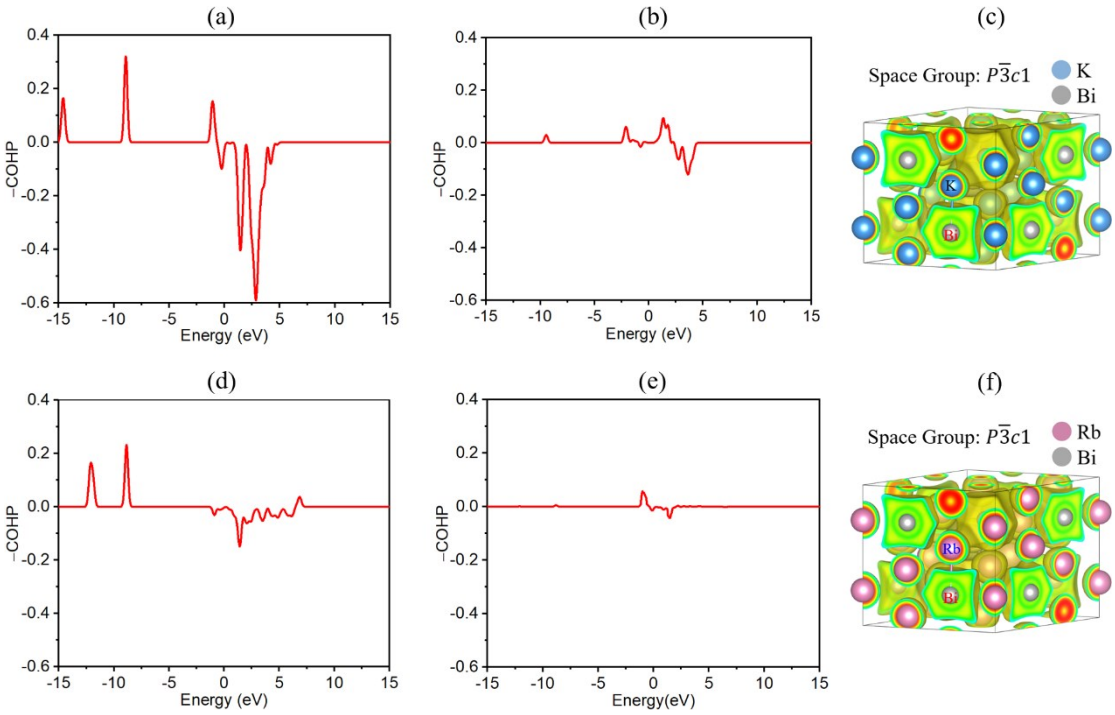

Fig. S3 Bonding/antibonding characters of the states of interlayer (a) K-Bi, (b) K-K pairs in K_3Bi and interlayer (d) Rb-Bi, (e) Rb-Rb pairs in Rb_3Bi, as indicated by the calculated COHPs. Electron localization function of (c) K_3Bi and (f) Rb_3Bi with isosurfaces of 0.2. −COOP > 0, −COOP = 0 and COOP < 0 mean the bonding, non-bonding and antibonding characters of each electronic state over the chemical bonds in the system, respectively.

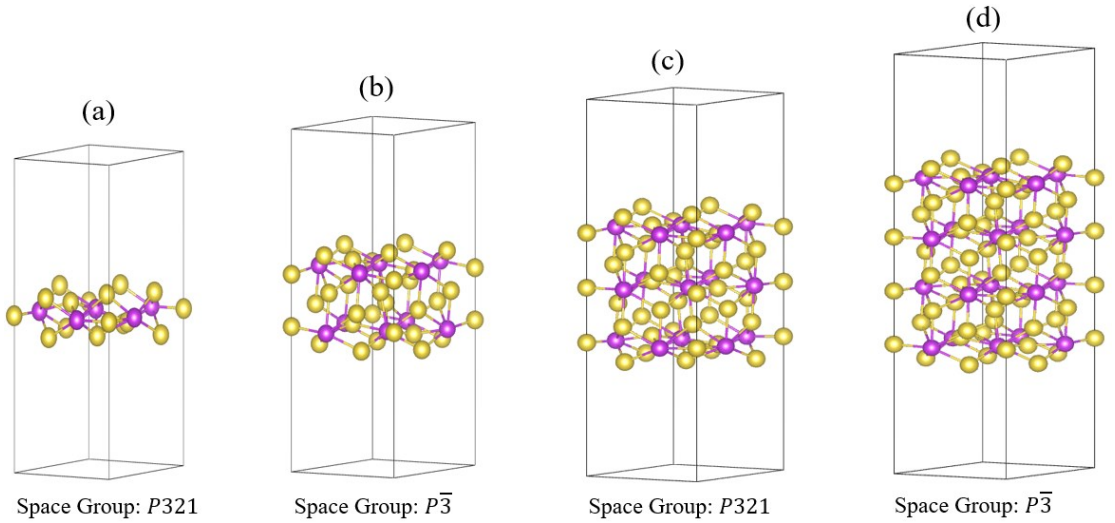

Fig. S4 Optimized structures of n-layer (n=1-4) Na_3Bi. The space group of monolayer, trilayer is $P321$, and that of bilayer, tetralayer is $P\bar{3}$.

Table S1 Topological invariant (Z_2) of n-layer (n = 1-4) Na_3Bi derived from the $P6_3/mmc$ phase of Na_3Bi.

Z_2	Na_3Bi
monolayer	1
bilayer	1
trilayer	1
tetralayer	1

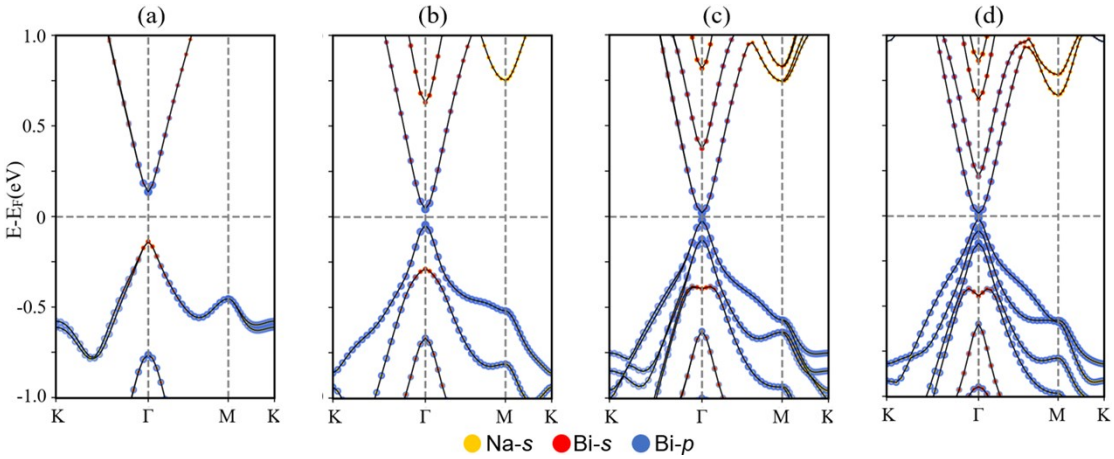

Fig. S5 Electronic structures of (a) monolayer, (b) bilayer, (c) trilayer and (d) tetralayer Na_3Bi derived from the $P6_3/mmc$ phase of Na_3Bi, from PBE + SOC.

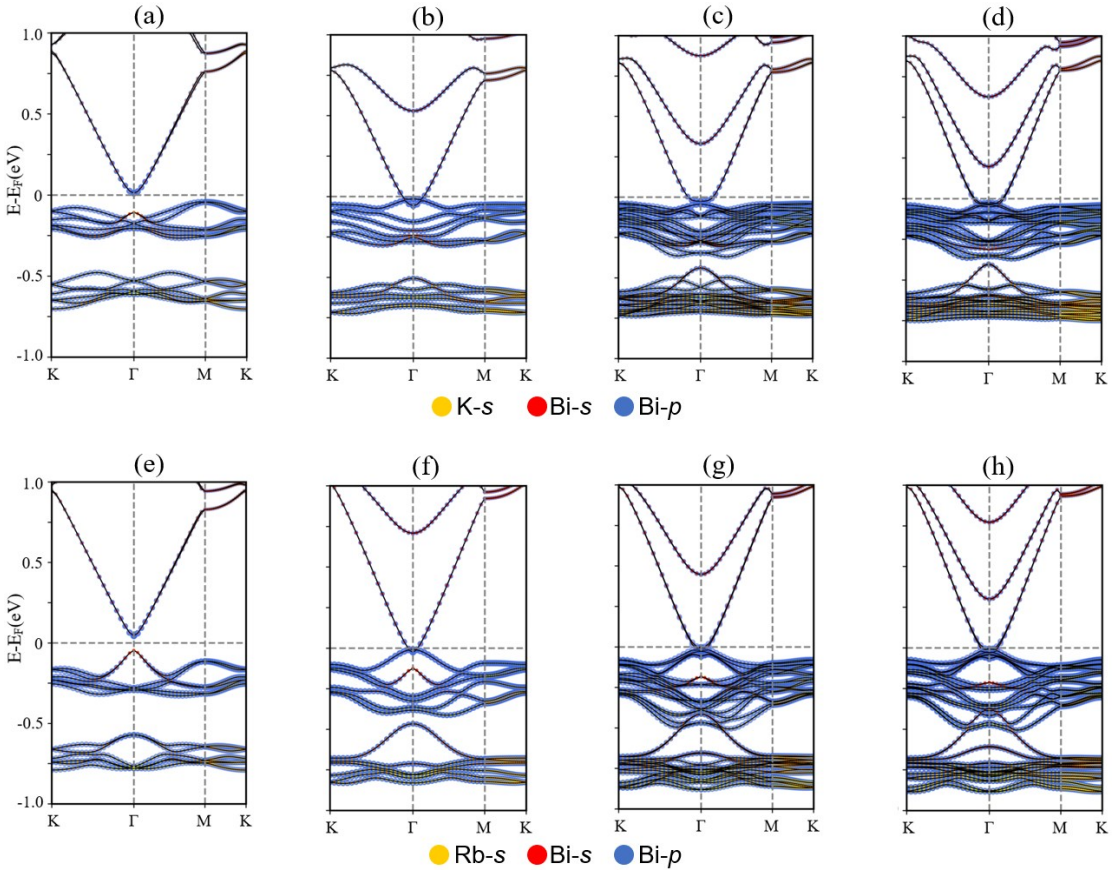

Fig. S6 Electronic structures of n-layer (n=1-4) of (a-d) K_3Bi and (e-h) Rb_3Bi derived from the $P\bar{3}c1$ phase, from PBE + SOC. The dotted lines with different colors denote the band projection onto different atomic orbitals.

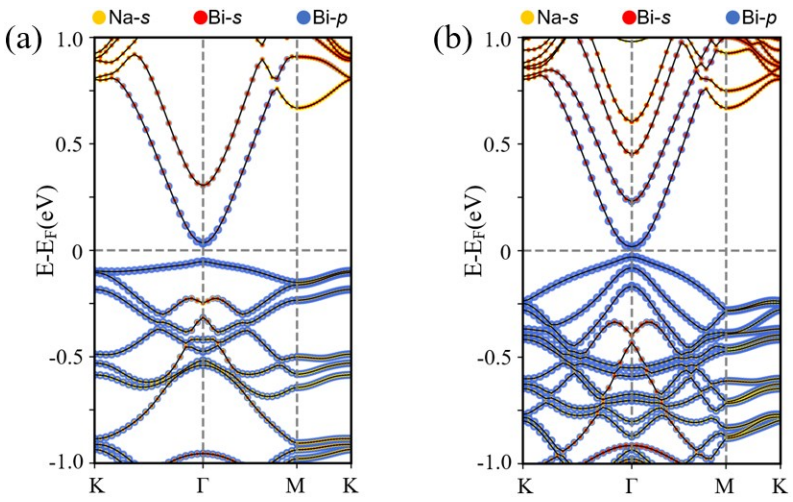

Fig. S7 Electronic structures of Na_3Bi ($P\bar{3}c1$) with the maximum band gap under strain. The maximum band gaps of bilayer and tetralayer Na_3Bi under strain are 9.4% and 84% higher than the unstrained case. (a) Maximum band gap is achieved in bilayer at 14% expansion, (b) in tetralayer at 10% expansion.