Supporting Information

Intrinsic spin-valley-coupled Dirac state in Janus functionalized *β*-BiAs monolayer

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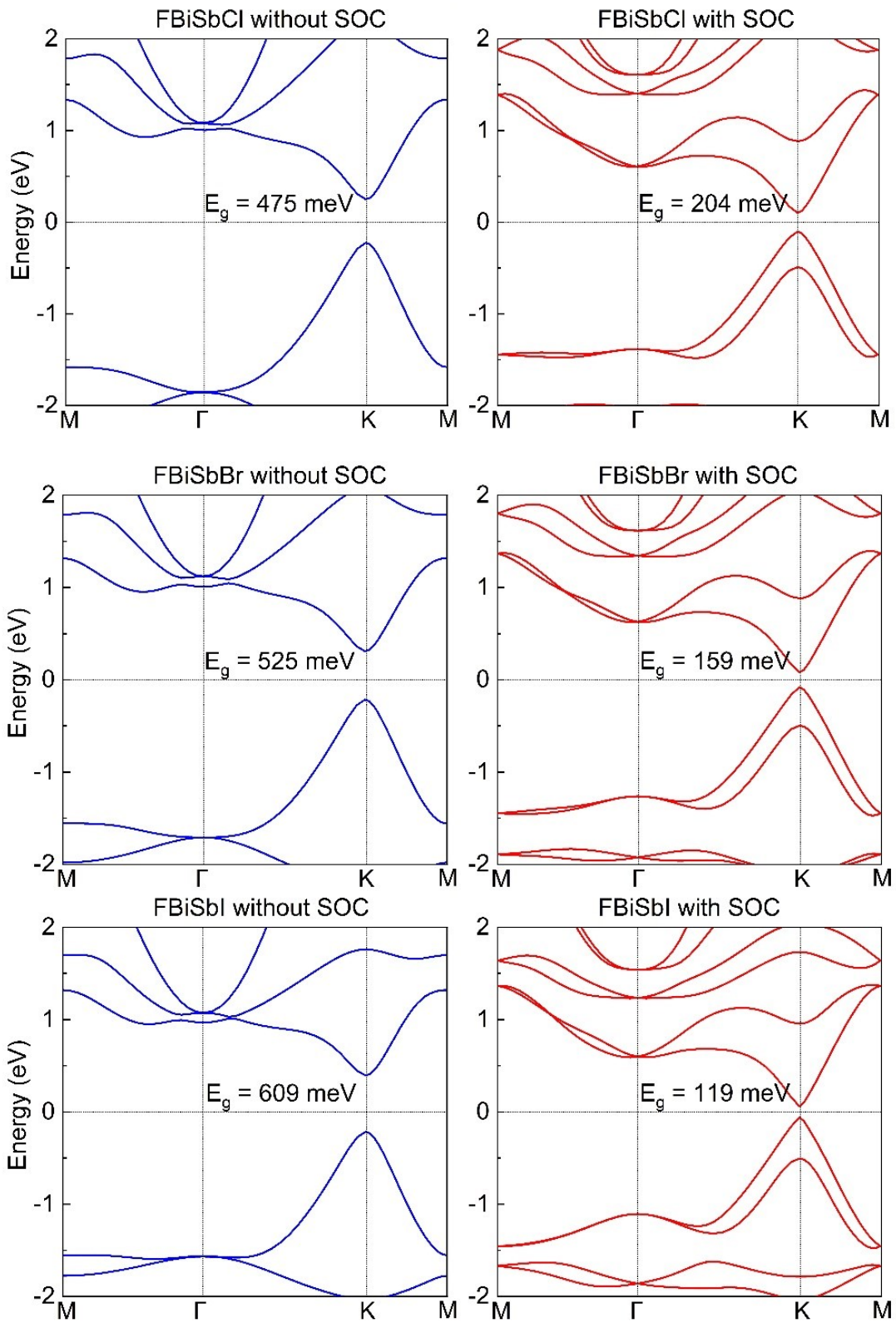

Figure S1-a. Electronic band structures of FBiSbY (Y=Cl, Br and I) MLs without SOC and with SOC.

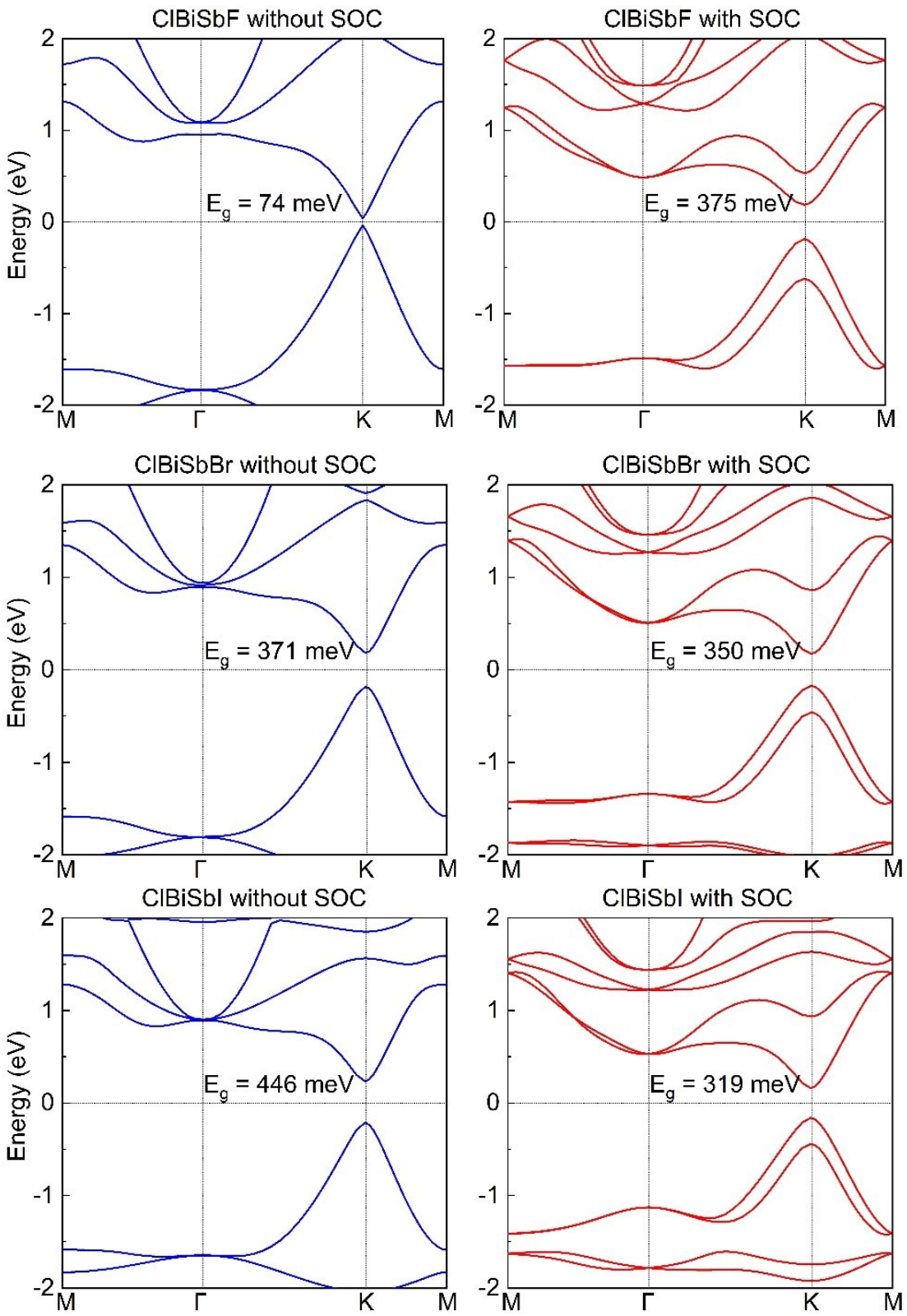

Figure S1-b. Electronic band structures of ClBiSbY (Y=F, Br and I) MLs without SOC and with SOC.

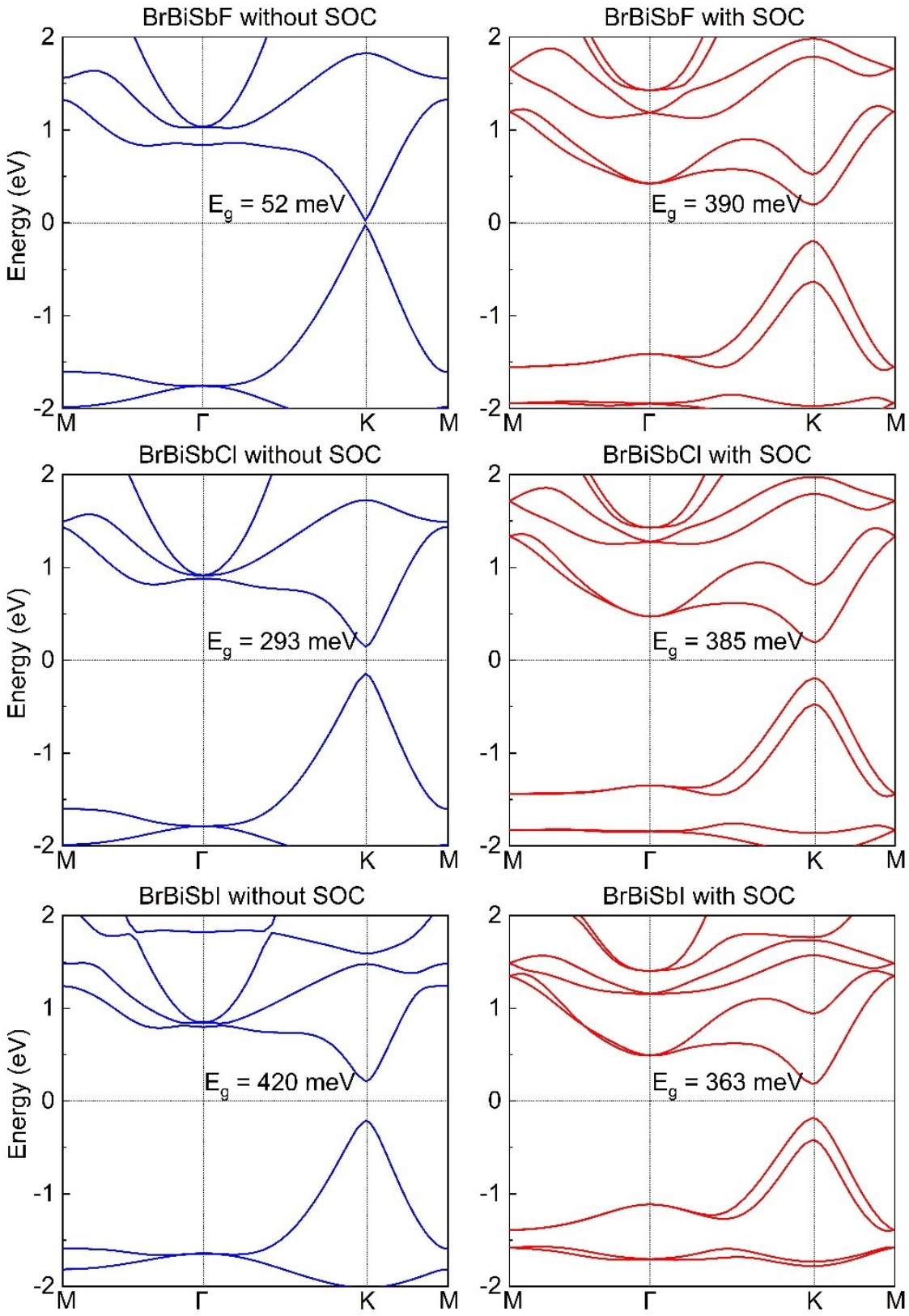

Figure. S1-c. Electronic band structures of BrBiSbY (Y=F, Cl and I) MLs without SOC and with SOC.

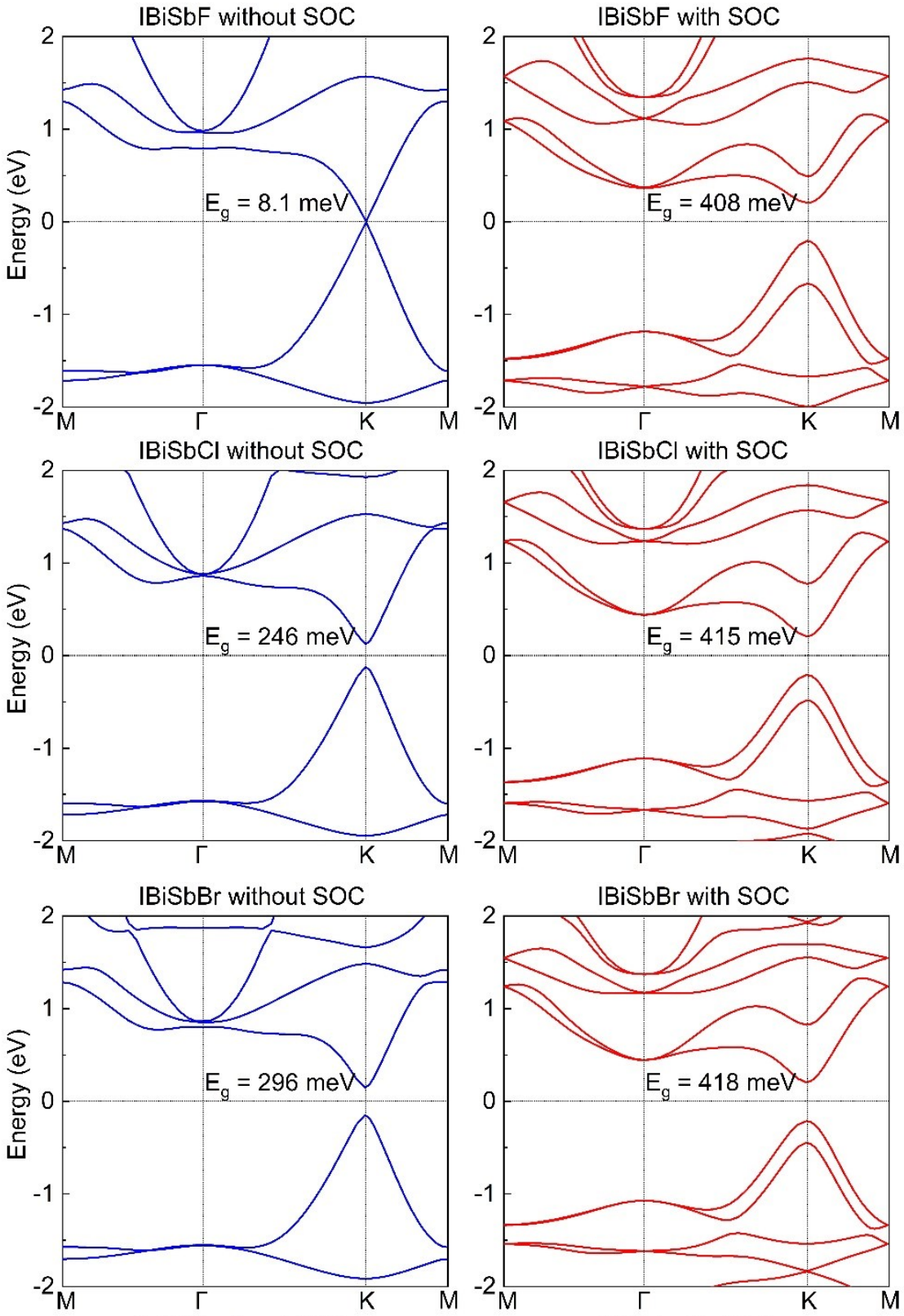

Figure. S1-d. Electronic band structures of IBiSbY (Y=F, Cl, and Br) MLs without SOC and with SOC.

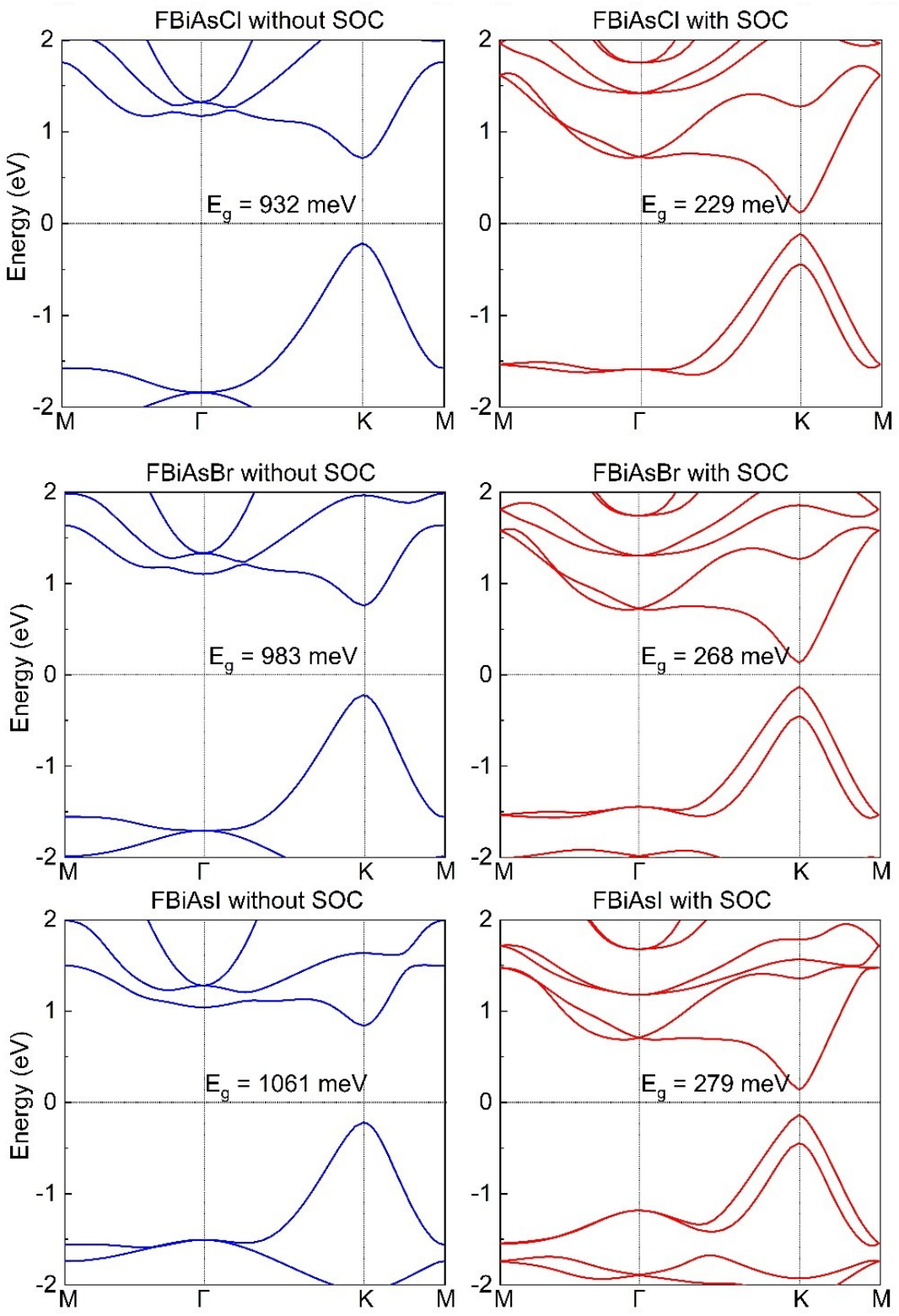

Figure. S2-a. Electronic band structures of FBiAsY (Y= Cl, Br and I) MLs without SOC and with SOC.

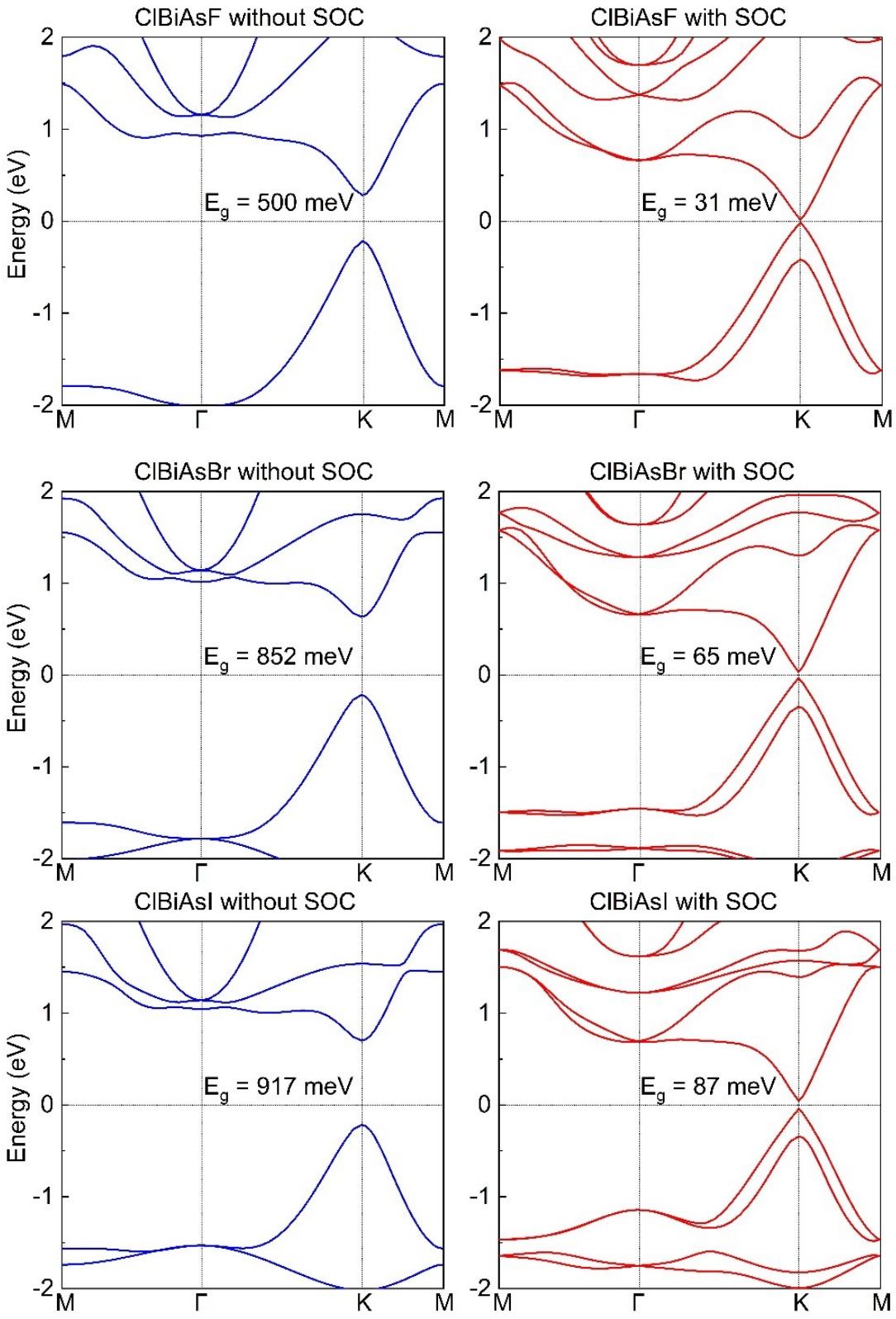

Figure. S2-b. Electronic band structures of ClBiAsY (Y=F, Br and I) MLs without SOC and with SOC.

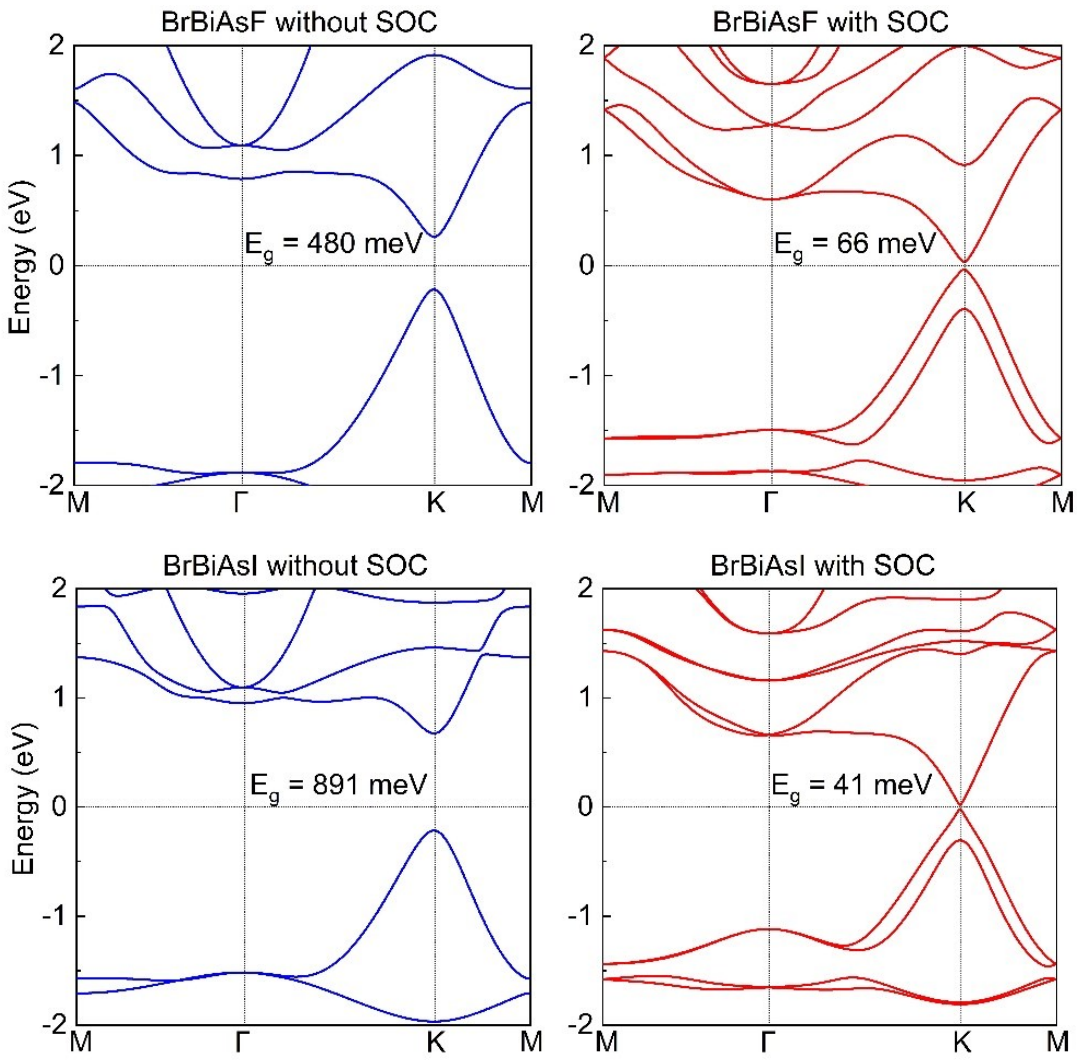

Figure. S2-c. Electronic band structures of BrBiAsY (Y=F and I) MLs without SOC and with SOC.

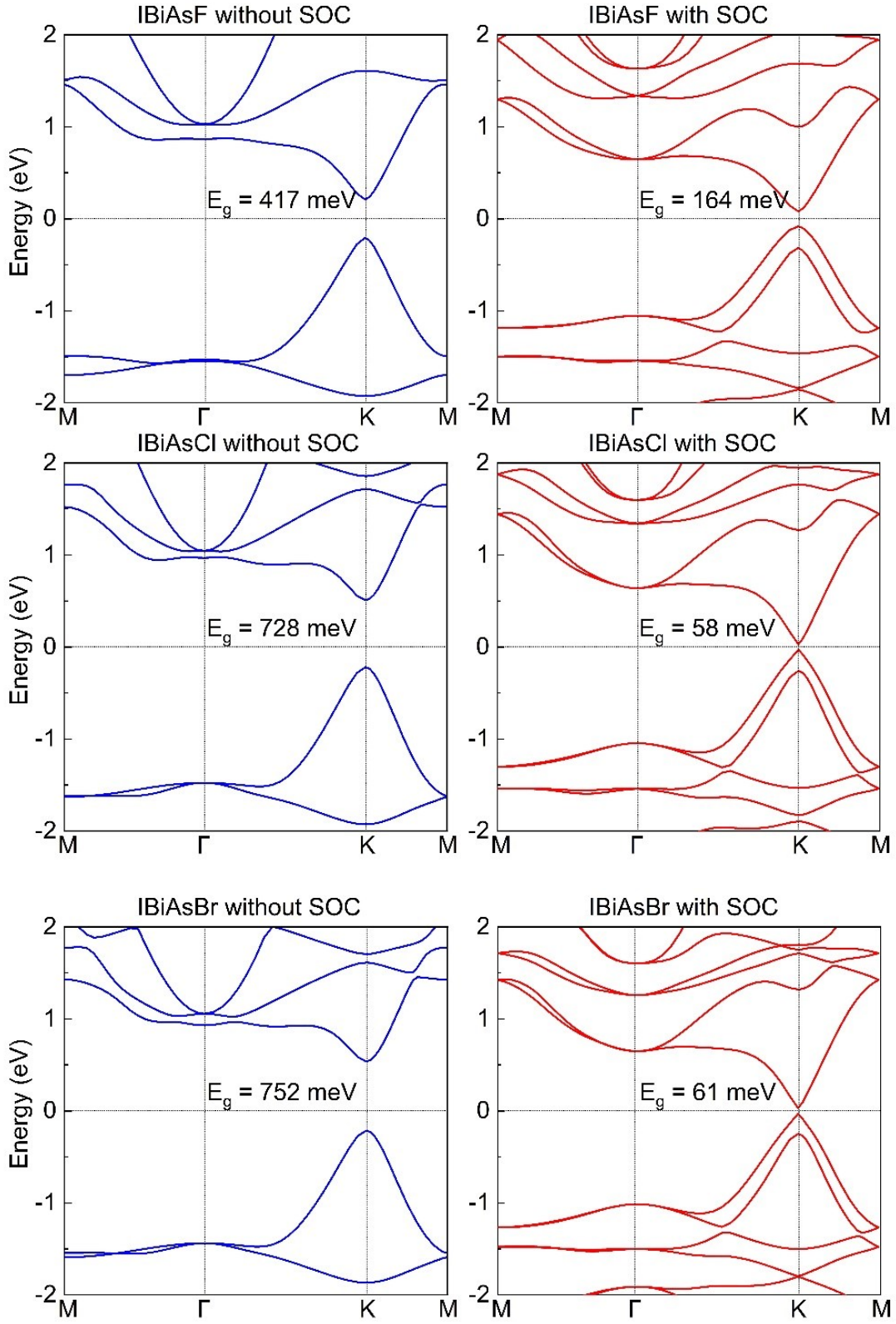

Figure. S2-d. Electronic band structures of IBiAsY (Y=F, Cl and Br) MLs without SOC and with SOC.

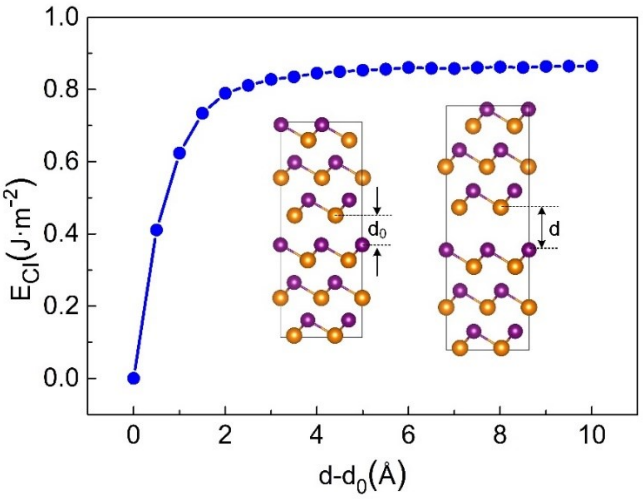

Figure S3. Cleavage energy E_{Cl} as a function of the separation distance $d–d_0$ between two fractured parts of 3D layered BiAs. It can be seen that the cleavage energy increases quickly with the increasing separation distance and then saturates to a value corresponding to the exfoliation energy of about 0.88 J/m^2, which is comparable with that of *h*-BN (~0.51 J/m^2). Thus, it is feasible to obtain the BiAs ML by mechanical exfoliation from the layered bulk, like *h*-BN.

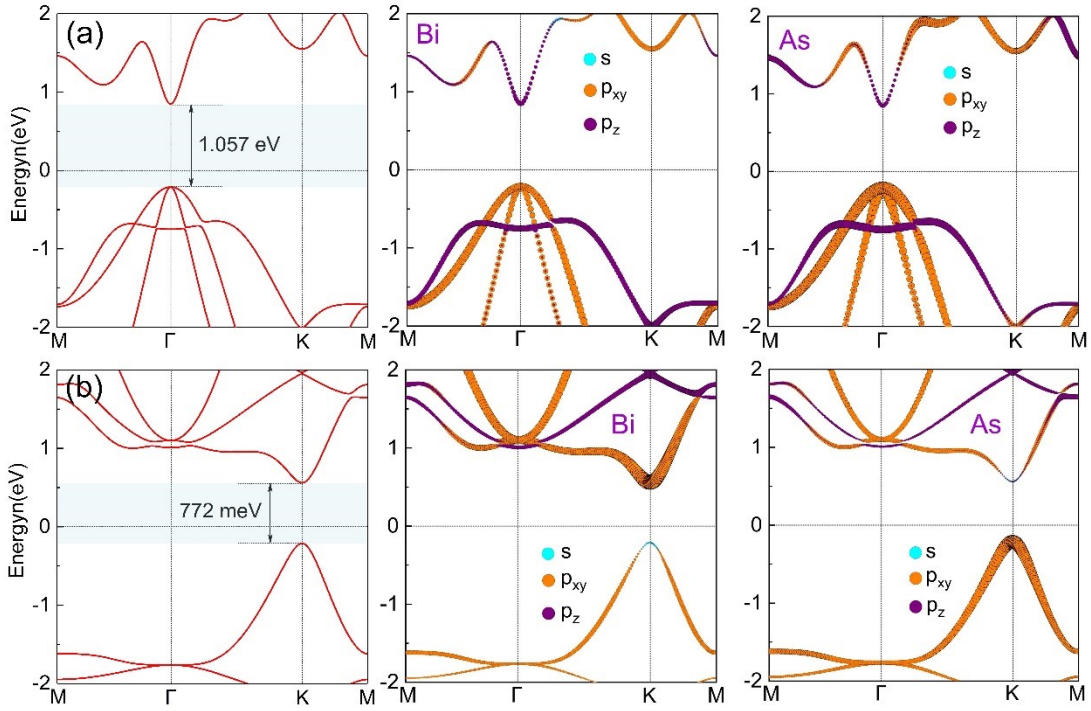

Figure S4. Electronic band structures of (a) *β*-BiAs and (b) BrBiAsCl MLs without SOC. The corresponding orbital-resolved electronic band structures for Bi and As atoms are displayed at the same row. It can be seen that *β*-BiAs ML is a direct semiconductor. The valence band maximum (VBM) at Γ point is mainly contributed by hybridization of Bi-p_{xy} and As-p_{xy} states, while the conduction band minimum (CBM) originates from Bi-p_z and As-p_z states, as well as few contributions of *s* state. Note that at the higher-lying valence band, the contribution of p_z for both Bi and As is obvious. Unlike the case of *β*-BiAs ML, BrBiAsCl ML is a direct semiconductor with greatly reduced band gap (0.772 eV) at K point. Since the electronegativity of halogen atoms is larger than that of both Bi and As atoms, the p_z electrons of the *β*-BiAs ML transfer to the halogen atoms. Therefore, the Fermi level shifts down, and the p_z states of *β*-BiAs ML strongly couple with the *s* states of halogen atoms at the deep energy level. It can be seen that the p_z state has almost no contribution to both valance and conduction bands in the vicinity of Fermi level for BrBiAsCl ML. The VBM is mainly contributed by As-p_{xy} and Bi-*s* orbitals, while the main contribution of CBM is As-s and Bi-p_{xy} orbitals.

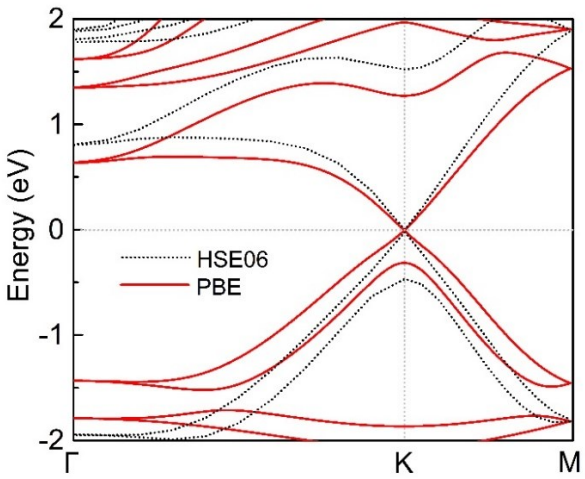

Figure S5. The electronic band structures calculated from both HSE06 (black dashed line) and GGA/PBE (red solid line) with SOC for BrBiAsCl ML. While the bands from HSE06 show some shifts with respect to the bands of PBE, the spin-splitted Dirac state is well preserved except for the change of dispersion slope. This further confirms that BrBiAsCl ML is an intrinsic svc-DSM.

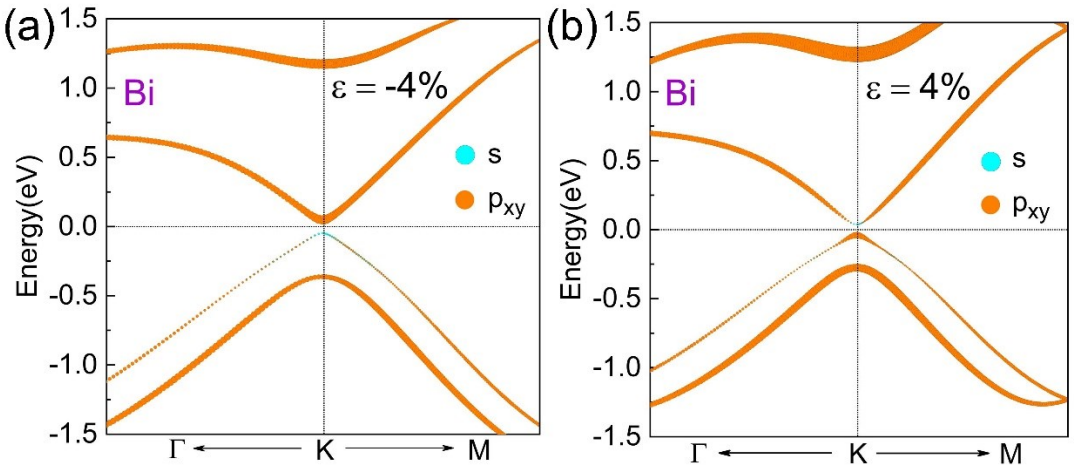

Figure S6. The orbital-resolved band structures in the vicinity of Fermi level for Bi atoms of BrBiAsCl ML under −4% and 4% external strain. It can be seen that the energy order between *s* and p_{xy} states have been changed under strain. This means that there exists the strain induced band inversion, namely topological phase transition should occur under external strains from −4% to 4%.

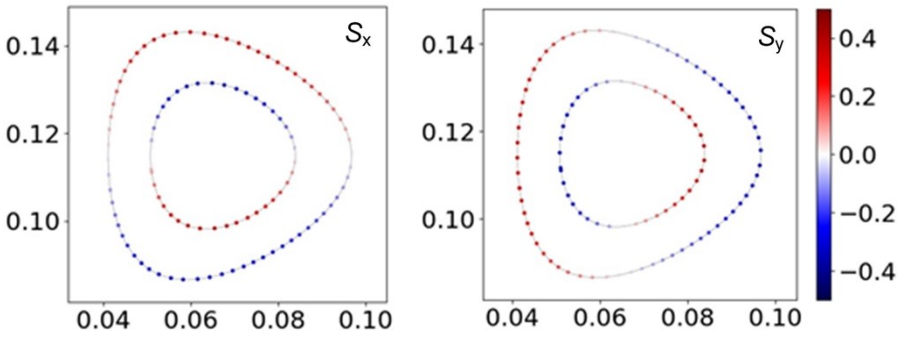

Figure S7. In-plane spin textures (S_x and S_y) calculated at the iso-energy surface of 0.6 eV blow the Fermi level for BrBiAsCl@*h*-BN heterostructure. From the spin projected constant energy contour plots of the spin textures in the k_x-k_y plane centred at the K point, one can see the similar Rashba SOC effect like in pristine BrBiAsCl ML. Firstly, the pair of spin-splitting bands have opposite spin orientation for both S_x and S_y spin component. Secondly, the in-plane spin moments at the two rings have opposite chirality. This indicates the existence of spin-momentum locking, namely the spin orientation is perpendicular to the electron momentum.