Exploring Strong and Weak Topological States on Isostructural Substitutions in TlBiSe$_2$

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Band profile of TlBiSe$_2$ using PBE+SOC and G$_0$W$_0$@PBE+SOC $\epsilon_{xc}$ functionals.

Figure S1: Band structure of TlBiSe$_2$ calculated using (a) PBE+SOC (b) G$_0$W$_0$@PBE+SOC.
Band inversion mechanism for GaBiSe$_2$ and PbBiSe$_2$.

Band inversion is an essential condition for topological insulators, in which the usual ordering of the conduction and valence band is inverted by spin-orbit coupling (SOC). For instance, the band inversion in GaBiSe$_2$ takes place between the $p$ orbitals of Bi and Se. The $p$ states of Bi lie above the fermi level, whereas $p$ states of Se lie below the fermi level as seen in Figure S2(a). The inclusion of SOC brings band inversion at the Γ point where the Bi $p$ states lies below the Se $p$ states (see Figure S2(b)). This band ordering at Γ is opposite to that away from the zone center. In the case of PbBiSe$_2$, the switching in this band order takes place at two $k$-points, i.e., Γ and F. At these $k$-points, the band ordering becomes opposite on including SOC, i.e., the Se $p$ orbitals become occupied and Bi $p$ orbitals become unoccupied (see Figure S2 (c),(d)).

![Figure S2](image-url)

Figure S2: The band structure for GaBiSe$_2$ at Γ in (a) natural order, (b) inverted order and for PbBiSe$_2$ at F and Γ in (c) natural order and (d) inverted order, showing the band inversion.
Band structures of SnBiSe$_2$, SbBiSe$_2$, Bi$_2$Se$_2$, TlSnSe$_2$ and PbSbSe$_2$.

The band structure of SnBiSe$_2$ without and with SOC is shown in Figure S3(a) and S3(b), respectively. The band inversion occurs at two TRIMs i.e., $\Gamma$ and $F$ and it involves the $p$ orbital of Bi and Se. Similar trend has been observed in SbBiSe$_2$, Bi$_2$Se$_2$, TlSnSe$_2$ and PbSbSe$_2$. In all of them, parity inversion is between the $p$ orbital of Bi and Se at even number of TRIMs.
Figure S3: The band structures for SnBiSe$_2$, SbBiSe$_2$ and Bi$_2$Se$_2$ without SOC are shown in (a), (c), (e) and with SOC are shown in (b), (d), (f), respectively.
Figure S4: The band structures for TlSnSe$_2$ and PbSbSe$_2$ without SOC are shown in (a), (c) and with SOC are shown in (b), (d), respectively.
Optimized lattice parameters of SnBiSe$_2$, SbBiSe$_2$, Bi$_2$Se$_2$, TlSnSe$_2$ and PbSbSe$_2$.

Table S1: Optimized lattice parameters

| Material   | Lattice Parameters(Å) |
|------------|-----------------------|
| SnBiSe$_2$ | 8.254                 |
| SbBiSe$_2$ | 8.059                 |
| Bi$_2$Se$_2$ | 8.163              |
| TlSnSe$_2$ | 7.747                 |
| PbSbSe$_2$ | 8.166                 |
Phonon band structures of different configurations showing band inversion. To analyze the dynamical stability, we have plotted the phonon band structures for SnBiSe$_2$, SbBiSe$_2$, Bi$_2$Se$_2$, TlSnSe$_2$ and PbSbSe$_2$. Figure S5 shows the phonon band structures calculated along the high-symmetry points of the BZ. Notably, the absence of negative frequencies in all of them confirm the dynamical stability of these materials.

Apart from the materials discussed in manuscript, InBiSe$_2$, TlSbSe$_2$, TlHgSe$_2$, TlBiI$_2$ and GaHgSe$_2$ also showed the band inversion. We have screened all of these materials based on their dynamical stability. As shown in Figure S6, none of these is dynamically stable, hence no further analysis is carried out on InBiSe$_2$, TlSbSe$_2$, TlHgSe$_2$, TlBiI$_2$ and GaHgSe$_2$. The other materials do not show parity inversion anywhere in the Brillouin Zone, therefore, they are characterized as trivial.
Figure S5: Phonon band structures of (a) SnBiSe$_2$, (b) SbBiSe$_2$, (c) Bi$_2$Se$_2$, (d) TlSnSe$_2$ and (e) PbSbSe$_2$. 
Figure S6: Phonon band structures of (a) InBiSe$_2$, (b) TlHgSe$_2$, (c) TlBiI$_2$ and (d) GaHgSe$_2$. 
Surface state band structures of SnBiSe\textsubscript{2}, SbBiSe\textsubscript{2}, Bi\textsubscript{2}Se\textsubscript{2}, TlSnSe\textsubscript{2} and PbSbSe\textsubscript{2}.

Figure S7: Surface states for the left surface of SnBiSe\textsubscript{2}, SbBiSe\textsubscript{2} and Bi\textsubscript{2}Se\textsubscript{2} are shown in (a), (c), (e) and for the right surface are shown in (b), (d), (f), respectively.
Figure S8: Surface states for the left surface of TlSnSe$_2$ and PbSbSe$_2$ are shown in (a), (c) and for the right surface are shown in (b), (d), respectively.
Band gap (in meV) of different materials using \( G_0W_0@\text{HSE06}+\text{SOC} \) \( \epsilon_{xc} \) functional.

**Table S2: Band gap using \( G_0W_0@\text{HSE06}+\text{SOC} \) \( \epsilon_{xc} \) functional.**

| Material    | Indirect Band Gap (meV) |
|-------------|--------------------------|
| GaBiSe\(_2\) | 135                      |
| PbBiSe\(_2\) | 62                       |
| SnBiSe\(_2\) | 11                       |
| SbBiSe\(_2\) | 85                       |
| Bi\(_2\)Se\(_2\) | 122                      |
| TlSnSe\(_2\) | 43                       |
| PbSbSe\(_2\) | 166                      |