Supplementary Information for Strain-induced Topological Phase Transition in Ferromagnetic Janus Monolayer MnSbBiS₂Te₂

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Magnetic Anisotropy Energy of Monolayer MnSbBiS₂Te₂

The magnetocrystalline anisotropy energy (MAE) is used to determine the most favorable spin configuration of the material. It is defined as the difference in energy between the in-plane (100) and out-of-plane (001) spin orientation of magnetic atoms according to the formula

$$MAE = E_{\parallel} - E_{\perp}$$

where E_{\parallel} and E_{\perp} are the total energies associated with in-plane (100) and out-of-plane (001) spin configurations respectively. Negative values of MAE represent preferred in-plane spin configurations whereas positive values represent preferred out-of-plane spin configurations. We find that MnSbBiS₂Te₂ has an MAE of 287 μ eV per Mn atom. Also, it prefers out-of-plane magnetic configuration. The MAE of MnSbBiS₂Te₂ is larger than that of MBT (180 μ eV)¹. Using the same method, we find that the MAE values of MnSbBiTe₄ and MnSbBiSe₂Te₂ monolayers are 146 and -207 μ eV respectively, which are very close to the previously reported values^{2,3}.



Figure 1 (a) Variation of MAE with strain in monolayer MnSbBiS₂Te₂ (b) Variation of band gap with strain in monolayer MnSbBiS₂Te₂

The variation of MAE as a function of applied strain in monolayer $MnSbBiS_2Te_2$ is shown in Figure 1(a). In the range of strain between -8% and 8%, we find that the monolayer $MnSbBiS_2Te_2$ prefers in-plane magnetic orientation at -3% and at -4%. In all other cases, it prefers out-of-plane magnetic orientation.

Magnetic Exchange Energy of Monolayer MnSbBiS₂Te₂

We calculate the magnetic exchange coupling parameters of monolayer $MnSbBiS_2Te_2$ by following the methods described in the paper by Torelli et al.⁴ Since we consider the first, second, and third nearest neighbor exchange interactions, the calculations require a larger supercell of size $3 \times 2 \times 1$. They are calculated without using non-collinear configurations. This is because the calculations involving non-collinear configurations have convergence issues. Similar concerns have been reported in the literature⁴. In fact, before doing the calculations for the monolayer $MnSbBiS_2Te_2$, we first validate our method by reproducing the results of the other MBT-type materials from the literature^{2,3}.

Band gap of Monolayer MnSbBiS₂Te₂

Figure 1(b) shows the variation of the band gap of monolayer $MnSbBiS_2Te_2$ with the applied strain. The band gap is calculated assuming both in-plane (M//X) and out-of-plane (M//Z) magnetic orientations shown using black (circle) and red (triangle) markers

respectively. Between -3% and 8% strain, $MnSbBiS_2Te_2$ is metallic for both magnetic orientations. For M//Z, the band gap opens at -4% with a value of 22.7 meV that increases with increasing compressive strain. At -6%, the band gap becomes 57.5 meV. If the strain is further increased, the gap starts decreasing to a value of 21.7 meV at -7%. The metallic phase returns at -8%. For M//X, the band gap opens only at -6% and -7% strain. The corresponding band gap values are 16.8 meV and 21.1 meV respectively.

Chemical Stability of Monolayer MnSbBiS₂Te₂

We first calculate the formation energy and use it as a proxy for the chemical stability of monolayer $MnSbBiS_2Te_2$. The formation energy of $MnSbBiS_2Te_2$ is calculated as,

$$E_f = E(\text{MnSbBiS}_2\text{Te}_2) - E(Mn) - E(Sb) - E(Bi) - 2 \times E(S) - 2 \times E(Te)$$

We find that $MnSbBiS_2Te_2$ has a formation energy of -1.063 eV per unit cell. Next, the phonon calculations are performed in a large $4 \times 4 \times 1$ supercell containing 112 atoms using the finite displacement method (FDM) implemented in the Phonopy package⁵. The FDM produces 42 structures with displaced atoms. The electronic relaxations are then performed in VASP⁶ with tight convergence criteria. The resulting phonon dispersion spectrum is shown in Figure 2(a). No imaginary phonon modes (or soft phonon modes) are found in the phonon spectrum. This indicates the dynamic stability of the MnSbBiS_2Te_2 monolayer. Small negative frequencies near the Γ point are due to the numerical artifacts in the calculations.



Figure 2 Phonon dispersion of monolayer $MnSbBiS_2Te_2$. (b) Energy profile of monolayer $MnSbBiS_2Te_2$ during the ab initio molecular dynamics simulation at 500 K over 30 ps.

We further test its stability by performing ab initio molecular dynamics simulations at 500 K. The energy profile of 30 ps with a time step of 2 fs is shown in Figure 2(b). No structural reconstruction is observed over the entire simulation period. This implies that the $MnSbBiS_2Te_2$ monolayer is chemically stable.

Band Structures of Monolayer MnSbBiS₂Te₂

The band gap evolution can be explained by plotting the band structure for different values of strain along the high-symmetry path of the Brillouin zone $(M - \Gamma - K)$ as shown in Figure 3.

The band structure at 0% strain (Figure 3(d)) features a small overlap of the valence band (VB) and conduction band (CB) near the Fermi level, indicating a metallic phase. The band overlap occurs at both sides of the Γ point although there is a clear separation of VB and CB at Γ . We highlight five different points at the band edges near the Γ point (A, B, C, D, E in Fig. 3(d)) and follow their evolution with applied strain. When a compressive strain of -4% is applied (Figure 3(c)), it pushes A and E down towards the VB and B, C, and D up towards the CB so that the region of overlap decreases. At -6% strain, A and E lie on the VB while B, C and D lie on the CB. This results in a band gap of 57.5 meV near the Γ point as shown in Figure 3(b). If the compressive strain is further applied (Figure 3(a)), it pushes all the aforementioned points up towards the CB region. At -8%, they lie entirely in the CB. Also, both the VB and CB cross the Fermi level at different points in the Brillouin zone. This closes the gap between VB and CB, hence the material changes to the metallic phase. When the strain is applied in the positive direction, A goes down to VB, E is slightly shifted up, also B and D disappear. Meanwhile, C goes down and crosses the Fermi level. Thus, the band overlap occurs at only one point towards the right from the Γ point (Figure 3(e)). At +8%, the strain again pushes C up towards the CB and E down towards the VB. This pattern suggests that the gap might open if the tensile strain is further applied beyond +8%.



Figure 3 Band structure of monolayer MnSbBiS₂Te₂ at different values of biaxial strain.

Charge Density Plots of Monolayer MnSbBiS₂Te₂

The charge density plots at different values of applied strain are shown in Figure 4. Figure 4(a-f) represents the charge density plots at the highest occupied energy states in valence band (VB). The corresponding plots at the lowest unoccupied conduction band (CB) are shown in Figure 4(g-l). We find that the atoms in the upper half of the structure (i.e. above the plane of Mn atoms) have a major contribution to VB. Similarly, the atoms in the lower half of the structure have a major contribution to CB. As the strain is applied, the shape of the orbitals in Te, Bi, and S atoms undergo gradual changes for both tensile and compressive strain. In addition, their contribution to the VB and CB changes with applied strain.



Figure 4 Charge density plots at the highest occupied valence band states (a-f) and lowest unoccupied conduction band states (g-l) in monolayer $MnSbBiS_2Te_2$ at different strains. The isosurface level is set between 0.007 and 0.008 e/Å³.

Band Structure of Monolayer MnSbBiS₂Te₂ at -8% Strain

Figure 5 shows the electronic band structure of monolayer MnSbBiS₂Te₂ at -8% strain representing the metallic phase.



Figure 5 Band structure of the monolayer $\mathsf{MnSbBiS}_2\mathsf{Te}_2$ at -8% strain.

Notes and references

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