Interlayer ferromagnetic coupling and enhanced magnetism by biaxial strain in MnBi$_2$Te$_4$/VBi$_2$Te$_4$ heterojunction

Zebin Wu$^1$, Changsheng Song$^{1*}$ and Chaorong Li$^{1*}$

$^1$Key Laboratory of Optical Field Manipulation of Zhejiang Province, Department of Physics, Zhejiang Sci-Tech University, Hangzhou, 310018, China

$^*$Corresponding author’s e-mail: cssong@zstu.edu.cn and crli@zstu.edu.cn

Abstract: The pristine anti-ferromagnetic interlayer coupling of even layers MnBi$_2$Te$_4$ greatly restricts the possibility of realizing quantum anomalous Hall (QAH) effect. In this work, based on first-principles calculations, we find that the interlayer coupling can be transformed into ferromagnetic order from anti-ferromagnetic by substituting cations with V atoms. Furthermore, when applying biaxial strain, a strong band inversion occurs around Fermi level, achieving quantum anomalous Hall effect. A gapless surface states can further confirm it. Meanwhile, the magnetic moment of Mn and V atoms increases with the increase of strain. We attribute this to the electron hopping to magnetic atoms from p orbitals of nonmagnetic Bi and Te atoms. Our work extends the range for designing and applying for spintronic devices with QAH effect.

1. Introduction

MnBi$_2$Te$_4$ (MBT)-family are novel van der Waals topological insulators, which have attracted considerable attention. Since three dimensional MBT has intrinsic magnetism and can display a variety of topological phases, it provides an excellent platform to explore the magnetic topological phases. MBT has a special septuple layer (SL) structure which is composed of Te-Bi-Te-Mn-Te-Bi-Te sequences. And it shows intralayer ferromagnetic (FM) and interlayer anti-ferromagnetic (AFM) exchange coupling.

Different from bulk case, the thin films will exhibit various topological states with various number of SLs. For the even SLs of MBT, the spin magnetic moment was counteracted by another layer due to AFM interlayer coupling. Consequently, there is no Hall current in the band gap. A large external magnetic field is needed to realize the quantum Hall effect\cite{1}. This is also called axion insulator\cite{2}. However, in the Odd SLs, since the net magnetic moment is always greater than zero, this material has enough magnetism to harbor a QAH effect without external magnetic field.

In some recent research, Otrokov et al.\cite{1} reported that a 2 SLs AFM MBT always yields a zero Chern number because of the inversion symmetry. When an artificial FM phase is used in calculation, the Chern number becomes to -1, indicating a QAH insulator. Fu et al.\cite{3} modulated the interlayer coupling of 4 SLs MBT slab from AFM to FM by constructing a sandwiched CrI$_3$/MBT/CrI$_3$ heterojunction, where the QAH effect was achieved. Therefore, FM coupling is one of the essential prerequisites for the realization of QAH effect.

In this work, we substituted cation with V to form MBT/VBT heterojunction, which can tune the interlayer coupling from AFM to FM. In addition, we have studied the influence of biaxial strain on band structures of bilayer MBT and MBT/VBT heterojunction by first-principles calculations. The results show that biaxial strain has a great influence on band structure, even inducing MBT/VBT into metallic phase. When the tensile strain increases to 4%, a band inversion phenomenon occurs. As a typical character of QSH insulator, the helical surface states are found in MBT/VBT heterojunction,
which connects the bulk conduction and valence bands. Moreover, we calculated the magnetic moment of both MBT-based materials, and gave a reasonable analysis through electron hopping theory. Our work can provide a useful guideline for design of topological spintronics devices with dissipation-less QAH effect.

2. Computational Details
Density functional theory (DFT) calculations are carried out by Vienna ab initio Simulation Package (VASP)\(^4\) with projector augmented wave (PAW) method\(^5\). The exchange correlation interaction of electrons is treated within the generalized gradient approximation (GGA)\(^6\) by using the Perdew-Burke-Ernzerhof (PBE) functional. The energy cut-off for plane-wave basis was set by 350 eV. The Monkhorst-Pack \(k\) mesh of \(5\times5\times1\) was adopted for the structural optimization and self-consistent calculations. The convergence criteria for total energy and force on each atom are less than \(10^{-5}\) eV per atom and \(0.01\)V/Å, respectively. Moreover, to avoid unnecessary interactions between adjacent lattices, a vacuum space larger than \(15\) Å is set alone z-axis. To improve the description of the on-site Coulomb interaction of localized 3\(d\) states of V and Mn atoms, the GGA + \(U\) method\(^7\) was implemented with an effective Hubbard term \(U\) set to 3.0eV and 4.0 eV, respectively. And DFT-D2\(^8\) potential was used to describe the interlayer interactions of bilayer MBT and MBT/VBT. The surface states are calculated with the method of surface Green’s function\(^9\) by WANNIER90\(^{10}\) and WANNIERTOOLS package\(^{11}\).

3. Results and Discussion
We first investigate the total energy of both MBT-based materials. After the atomic position was fully optimized, the lattice constants obtained of bilayer MBT and MBT/VBT was 4.30Å and 4.21Å, respectively. Then we applied biaxial strain on both materials. As can be seen in Fig. 1(b-c), in the absence of strain, both materials have the lowest energy, which indicates that the lattice constant we selected is in the ground state. The blue line presents the energy difference between FM and AFM order (\(\Delta E\)). In bilayer MBT, \(\Delta E\) is always greater than zero, which means that bilayer MBT has an AFM interlayer coupling. However, the energy difference of MBT/VBT is negative value, presenting FM order, in agreement with the recent work\(^{12}\). Therefore, we conclude that substituted cation with V can turn the interlayer coupling to FM ground state.

Figure 1. (a) The side and top view of atomic structure of MBT/VBT heterojunction. The violet, red, blue and gray balls represent Mn, V, Bi and Te atoms, respectively. (b-c) Total energy and energy difference of interlayer magnetic coupling under different strain \(\varepsilon\) of (b) bilayer MBT and (c) MBT/VBT heterojunction.
Applying biaxial strain, the shape of band structure changes and shifts. A compressive strain can lift both Te-p orbitals, while the conduction band minimum (CBM) consists of Bi-p orbitals. One can notice a nontrivial band structure with band inversion, which is consistent with recent works of MBT\cite{2, 13}. When inverting and gapless surface states at pristine MBT/VBT. We believe this difference can be explained in terms of the differences of lattice constants. In Zhu’s work, the selection of lattice constants is closer to experimental values. As illustrated in Fig. 3, band inversion occurs when the tensile strain reaches up to 4%. Although the tendency of MBT/VBT band movement is the same as that of MBT, the band gap variation shows a different trend with MBT. The band gap decreases with increasing tensile strain.

![Band structure of bilayer MBT under different strain](image1)

**Figure 2.** (a-g) Band structure of bilayer MBT under different strain $\varepsilon$. Red and blue ball line represent projectors of Te and Bi atoms. The Fermi level are set to be zero. (h) Band gap at $\Gamma$ point versus biaxial strain $\varepsilon$.

We next investigate the electronic band structure and the gap at $\Gamma$ point. Fig. 2 shows band structures for bilayer MBT under different biaxial strain. We find the valence band maximum (VBM) is mainly composed of Te-p orbitals, while the conduction band minimum (CBM) consists of Bi-p orbitals. One can notice a nontrivial band structure with band inversion, which is consistent with recent works of MBT\cite{2, 13}. When applying biaxial strain, the shape of band structure changes and shifts. A compressive strain can lift both VBM and CBM at $\Gamma$ point, while tensile strain can push them down. Note that on the path of $\Gamma$ to K has the opposite moving.

![Band structure of MBT/VBT heterojunction under different strain](image2)

**Figure 3.** (a-g) Band structure of MBT/VBT heterojunction under different strain $\varepsilon$. Red and blue ball line represent projectors of Te and Bi atoms. The Fermi level are set to be zero. (h) Band gap at $\Gamma$ point versus biaxial strain $\varepsilon$.

Different with bilayer MBT, in the absence of strain, the MBT/VBT heterojunction does not host band inversion in our calculation. However, Zhu et al.\cite{13} performed calculations and observed the band inversion and gapless surface states at pristine MBT/VBT. We believe this difference can be explained in terms of the differences of lattice constants. In Zhu’s work, the selection of lattice constants is closer to experimental values. As illustrated in Fig. 3, band inversion occurs when the tensile strain reaches up to 4%. Although the tendency of MBT/VBT band movement is the same as that of MBT, the band gap variation shows a different trend with MBT. The band gap decreases with increasing tensile strain.
Figure 4. (a)(c) Magnetic moment ($\mu_B$) of Mn and V atoms in (a) bilayer MBT and (b) MBT/VBT as the function of strain $\varepsilon$. (b)(d) Schematics of super-exchange interaction between (b) Mn1-p-Mn2 in bilayer MBT and (d) Mn-p-V in MBT/VBT.

Fig. 4(a) and (c) exhibits the magnetic moment of Mn and V atoms. One can notice that the magnetic moment increases with the tensile strain in both materials. We attribute this to electron hopping to magnetic atoms. According to Hund’s rule, the d orbitals of each spin state will split into a triply degenerate $t_{2g}$ states and a doubly degenerate $e_g$ states. For MBT and VBT, Mn has a $d^5$ configuration as $t_{2g}^3 e_g^2$, while V has a $t_{2g}^3 e_g^0$ configuration. With the empty $e_g$ states of V and half-filled $t_{2g}$ of Mn, electrons from p orbital of nonmagnetic atoms hopping to d orbital of magnetic atoms are allowed. Therefore, the magnetic atoms gain more electrons and the magnetic moment grows up.

Figure 5. Topological surface states on (010) surface of MBT/VBT heterojunction under 4% tensile strain. The regions with a continuous spectrum correspond to the 2D bulk stats projected onto a 1D Brillouin zone.

We also calculated the surface states of MBT/VBT heterojunction to examine its topological property. As can be shown in Fig.5, (a) presents the band structure on its bulk, while (b) is that on the 010 surface. The former is obviously gapped, but the latter appears gapless surface states, which connects the conduction and valence bands. This further confirms the QAH effect of MBT/VBT heterojunction.
4. Conclusion

Based on first-principles calculations, we have investigated the fundamental properties of bilayer MBT and MBT/VBT heterojunction. Our results show that the interlayer coupling of bilayer MBT is AFM, by contrast, that of MBT/VBT is FM order. Moreover, biaxial strain can effectively modulate the band structure. Remarkably, the QAH state with a gapless surface states in MBT/VBT appears at tensile strain of 4%. The magnetic moment of both bilayer MBT and MBT/VBT are enhanced by applying tensile strain. Our work provides references for experimental control of interlayer coupling to realize QAH effect, especially for MBT family materials.

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