BRIEF COMMUNICATION

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Intertwined ferroelectricity and topological state in two-dimensional multilayer

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The intertwined ferroelectricity and band topology will enable the non-volatile control of the topological states, which is of importance for nanoelectrics with low energy costing and high response speed. Nonetheless, the principle to design such system is unclear and the feasible approach to achieve the coexistence of two parameter orders is absent. Here, we propose a general paradigm to design 2D ferroelectric topological insulators by sliding topological multilayers on the basis of first-principles calculations. Taking trilayer Bi_2Te_3 as a model system, we show that in the van der Waals multilayer based 2D topological insulators, the in-plane and out-of-plane ferroelectricity can be induced through a specific interlayer sliding, to enable the coexistence of ferroelectric and topological orders. The strong coupling of the order parameters renders the topological states sensitive to polarization flip, realizing non-volatile ferroelectric control of topological properties. The revealed design-guideline and ferroelectric-topological coupling not only are useful for the fundamental research of the coupled ferroelectric and topological physics in 2D lattices, but also enable innovative applications in nanodevices.

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INTRODUCTION

Ferroelectricity and band topology are two intensively investigated yet distinct properties of insulators^{1–12}. Physically, there is no inherent exclusion between them due to different origins of polarization and band inversion, their coexistence in a single material leads to the concept of ferroelectric topological insulator (FETI)^{3,13–15}. The past years have witnessed the discovery of FETIs, especially in three dimensional, including strained CsPbI_3 ¹⁶, strained LiZnSb ¹⁷, pressured or strained AMgBi ¹⁴, and alloyed KMgBi ¹⁸, and the induced intriguing electronic properties such as ferroelectric controlled spin vortex. The strongly coupled ferroelectric and topological orders render them both fundamentally intriguing and practically appealing to be used in potential devices.

Unlike 3D FETIs, two-dimensional (2D) lattices with intertwined ferroelectric and topological orders are rather scarce^{3,19–21}, and the coupling of the order parameters are also quite weak in several existing cases^{15,22,23}. This is partly due to that, the ferroelectricity in 2D materials has been mainly established in the single-layer asymmetric structures^{24,25}, while band topology is commonly seen in the materials with heavy elements and strong spin orbital coupling. There, the requirements of symmetrical structure with switchable polarization and band inversion with different parity in the revealed 2D material family have to be simultaneously satisfied, to build the 2D FETIs, which significantly restrict the possible realization of 2D FETIs^{9,26–28}. So far, how to expand the scope for material candidates of 2D FETIs, especially with intertwined ferroelectric and topological physics, remains an open question.

The intriguing model of sliding switchable interfacial ferroelectricity has been proposed theoretically²⁹ and recently confirmed experimentally^{30–32}, which serves as a good starting point for realizing 2D FETI. Here, based on first-principles calculations, we fill aforementioned outstanding gap by introducing a general and

simple scheme to realize 2D FETIs with intertwined ferroelectric and topological physics. By studying an example system of trilayer Bi_2Te_3 , we discover that through a specific interlayer sliding, the charge rearrangement brings the spatial electron-hole separation in van der Waals multilayer based 2D topological insulators. The resultant reversal separation leads to the appearance of both in-plane and out-of-plane ferroelectricity, while the nontrivial topological phase is well reserved, thus enabling the coexistence of ferroelectric and topological orders. The strong coupling between ferroelectric and topological orders is also observed, distinct different topological physics can be induced in such multilayer systems when the ferroelectric polarization is reversed, suggesting the ferroelectric controlled topological properties.

RESULTS

Ferroelectric and topological orders

The proposed scheme to realize 2D FETIs with intertwined ferroelectric and topological physics is schematically presented in Fig. 1. Without losing generality, we start from 2D van der Waals multilayers with nontrivial topological properties, the time-reversal symmetry is preserved. Unlike band topology that links with electronic properties^{1,4,10,33}, ferroelectricity relates to crystal structure symmetry and electric dipole induced by electron distribution^{34–38}. To realize ferroelectricity, the polarization has to be switchable. As illustrated in the upper part of Fig. 1, if the in-plane (IP) and out-of-plane (OPP) mirror symmetries ($M_{\text{IP/OOP}}$), as well as the inversion symmetry (I), of the 2D multilayer are broken, the ferroelectricity occurs as long as the polarization is switchable, yielding the 2D FETI. The polarization switching is obtained via interlayer translation. If the polarization is unswitchable, it is just a normal 2D TI, without showing ferroelectric order. On the other hand, when the 2D multilayer possesses $M_{\text{IP/OOP}}$ or I symmetry, as illustrated in the lower part of Fig. 1, two different

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Fig. 1 Schematic diagram for screening 2D FETI. Schematic diagram for designing 2D FETIs from 2D van der Waals multilayers with nontrivial topological properties. Both topological and ferroelectric phases are determined by the mirror and inversion symmetry ($M_{IP/OOP}&I$) of multilayer. Blue cross denotes the situation that is not under consideration.

cases can be induced by the interlayer sliding. In first case, the systems like bilayer possess the spatial I symmetry there is no polarization, this configuration is out of our consideration. In second case, the $M_{IP/OOP}$ and I symmetry can be broken by interlayer sliding, the polarization thus appears, and obviously such polarization is electrically switchable. In the latter case, considering that the topological property of the multilayer may be disturbed by the sliding^{39,40}, if the topological property is preserved, the ferroelectric-topological phases can be achieved; otherwise, it is only a trivial 2D ferroelectric material. As we will show below, the obtained ferroelectric and topological orders in such systems exhibits a strong coupling. This design scheme suggests that the crystal symmetry can be utilized as one screening factor to identify 2D FETIs with intertwined ferroelectric and topological physics.

Following the design scheme, we study the coexistence of FE and band topology in a real material of trilayer Bi_2Te_3 . Our first-principles calculations are performed based on density functional theory as implemented in the Vienna Ab Initio Simulation Package (VASP)⁴¹. Figure 2a shows the crystal structure of trilayer Bi_2Te_3 ($\alpha\text{-Bi}_2\text{Te}_3$), which are obtained by direct exfoliating from the bulk phase. It shows a space group of D_{3d} with symmetry elements ($E, 2C_3, 3C_2, i, 2S_6, 3\sigma_d$). Clearly, the inversion symmetry prevents it from hosting any polarization. We thus slide the upper and lower quintuple layer (QL) along the $[1\bar{1}0]$ and $[\bar{1}10]$ directions, respectively, which are referred to as $\beta_1\text{-Bi}_2\text{Te}_3$ and $\beta_2\text{-Bi}_2\text{Te}_3$, respectively, as shown in Fig. 2b and Supplementary Note 1. Such interlayer sliding reduces the space group of trilayer Bi_2Te_3 to C_{3v} with symmetry elements ($E, 3C_3$ and $3\sigma_d$). Due to the simultaneous absence of I and M_z symmetries, $\beta_1\text{-Bi}_2\text{Te}_3$ and $\beta_2\text{-Bi}_2\text{Te}_3$ host a spontaneous electric polarization of $-5.1 \times 10^9 \text{ e cm}^{-2}$ and $5.1 \times 10^9 \text{ e cm}^{-2}$, respectively, along the out-of-plane (OOP) direction. Obviously, these two polarized configurations can be switched to each other by electric field triggered middle QL sliding [(Fig. 2b)], and thus correlate to two ferroelectric states, suggesting the OOP ferroelectricity.

To get more insight into the OOP ferroelectricity, we investigate the underlying physics for the electric polarization. In $\beta_1\text{-Bi}_2\text{Te}_3$, as displayed in Fig. 2c, the Te_u atom sits above the Bi_l atom, while the Te_l atom lies right below the Te_c atom. The inequivalent distribution of these atoms gives rise to the spatial electron-hole separation along the OOP direction [Fig. 2d], yielding an electric polarization pointing $-z$ direction. The resultant polarization is also suggested by the calculated planar average electrostatic potential

of $\beta_1\text{-Bi}_2\text{Te}_3$ along the $[001]$ direction. As shown in Fig. 2e, there is a discontinuity (ΔV) of 34 meV between the vacuum levels of the upper and lower QL layers, confirming the formation of electric polarization pointing $-z$ direction. While in $\beta_2\text{-Bi}_2\text{Te}_3$, the Te_u atom shifts to above the Te_l atom, while the Te_l atom shifts right below the Bi_2 atom; see Fig. 2b. Accordingly, the distribution of these atoms, as well as the spatial electron-hole separation, in $\beta_2\text{-Bi}_2\text{Te}_3$ is reversed with respect to that of $\beta_1\text{-Bi}_2\text{Te}_3$. Such reversal produces an electric polarization pointing to $+z$ direction for $\beta_2\text{-Bi}_2\text{Te}_3$, which are confirmed by the calculated planar average electrostatic potential ($\Delta V = -34 \text{ meV}$). To evaluate the feasibility of the OOP ferroelectricity in trilayer Bi_2Te_3 , we study the minimum energy path for the ferroelectric switching, which are shown Fig. 2f. The energy barrier is estimated to be 69.46 meV per unit cell, which is comparable to the values of other ferroelectrics^{37,42–45}, indicating its feasibility.

By further examining the distribution of these atoms in the (110) plane [Fig. 2c], we can see that, for $\beta_1\text{-Bi}_2\text{Te}_3$, the distance between Te_u and Te_c atoms in the $[1\bar{1}0]$ direction is larger than that between Te_l and Te_c atoms. Such imbalance distribution also generates the spatial electron-hole separation along the $[1\bar{1}0]$ direction, yielding an in-plane (IP) electric polarization of $3.1 \times 10^{10} \text{ e cm}^{-2}$ pointing to $[1\bar{1}0]$ direction. When transforming $\beta_1\text{-Bi}_2\text{Te}_3$ into $\beta_2\text{-Bi}_2\text{Te}_3$, the Te_c atom moves close to Te_u in the $[1\bar{1}0]$ direction. In this regard, the spatial electron-hole separation in the $[1\bar{1}0]$ direction is reversed, inducing an IP electric polarization of $-3.1 \times 10^{10} \text{ e cm}^{-2}$ pointing $[\bar{1}10]$ direction. The reversal of IP electric polarization shares the same energy path as the OOP case. It should be emphasized that, similar to single-layer In_2Se_3 ^{46,47}, there are three equivalent IP polarizations along the $[1\bar{1}0]$, $[\bar{1}20]$ and $[2\bar{1}0]$ directions, which leads to a zero net IP polarization. However, introducing the substrate proximity effect can readily break the threefold rotation symmetry, realizing the IP ferroelectricity, which has been well demonstrated in experiments^{38,48–50}. Accordingly, both IP and OOP ferroelectricity can be expected in trilayer Bi_2Te_3 .

It is important to emphasize that, it is feasible to engineer ferroelectricity in trilayer Bi_2Te_3 . First, the phonon dispersions without imaginary phonon modes (Supplementary Fig. 1) and negative cohesive energy of -2.97 eV/atom confirm the stability of $\beta_1\text{-Bi}_2\text{Te}_3$. And secondly, sliding ferroelectricity and stacking manipulation have been well established in experiments^{30–32}. The tear-and-stack method, for example, that pick up one layer and then stamp it on top of the remaining part can be used to precisely fabricate the large-scale commensurate trilayer Bi_2Te_3 with intercorrelated ferroelectricity^{30,31}.

Next, we study the electronic properties of trilayer Bi_2Te_3 in the ferroelectric phase. As $\beta_1\text{-}$ and $\beta_2\text{-Bi}_2\text{Te}_3$ are linked as two equivalent ferroelectric states, here we take $\beta_1\text{-Bi}_2\text{Te}_3$ as an example. Supplementary Fig. 2a shows the band structure of $\beta_1\text{-Bi}_2\text{Te}_3$ without including spin-orbit coupling (SOC), from which we see that it is an indirect gap semiconductor with a global gap of 0.51 eV near the Γ point. By analyzing orbital contributions, we find the highest valence bands (VB) near the Fermi level is mainly contributed by Te-p orbital, while Bi-p orbital makes the dominate contribution to the lowest conduction bands (CB). Upon taking SOC into account, the VB and CB bands near the Γ point experience a significant Rashba spin splitting [Supplementary Fig. 2b], which can be attributed to the existence of OOP electric polarization in $\beta_1\text{-Bi}_2\text{Te}_3$. The corresponding Rashba parameter is calculated to be $\alpha_R = \frac{2E_R}{k_0} = 0.67 \text{ eV\AA}$. When SOC effect is considered, it is interesting to notice that the CBM and VBM move closer and the band gap is reduced to 9 meV. Such band narrowing and M-shaped VBM normally indicates a nontrivial topological phase.

To confirm the nontrivial topological order in $\beta_1\text{-Bi}_2\text{Te}_3$, we calculate the topological invariant Z_2 . Due to its broken inversion symmetry, the Z_2 invariant is calculated by tracing the Wannier

Fig. 2 Structural configurations and ferroelectricity. Crystal structures of trilayer Bi_2Te_3 with (a) inversion symmetry and (c) broken inversion symmetry. Te_u and Te_l represent the bottom Te sublattice of upmost Bi_2Te_3 QL and top Te sublattice of lowest Bi_2Te_3 QL, respectively; Te_c denotes the middle Te sublattice of central Bi_2Te_3 QL. (b) Crystal structures of β_1 - and β_2 - Bi_2Te_3 as well as the schematic diagram of ferroelectric switching. Blue arrows denote the location of interfacial Te atoms of outmost layers. Right parts in (a), (b) show the zoomed-in interfacial atomic layers and the equatorial plane projection of point group. (d) Charge density difference and (e) planar average electrostatic potentials along the [001] direction for β_1 - Bi_2Te_3 . (f) Minimum energy path for ferroelectric polarization reversal between β_1 - and β_2 - Bi_2Te_3 .

charge center (WCC) using non-Abelian Berry connection⁵¹. The Wannier functions (WFs) related with lattice vector R can be written as:

$$|u_{nk}\rangle = \frac{1}{2\pi} \int_{-\pi}^{\pi} d\mathbf{k} e^{-i\mathbf{k}(R-x)} |u_{nk}\rangle \quad (1)$$

Here, a WCC is defined by the mean value of $\langle 0_n | \hat{X} | 0_n \rangle$, where the $\hat{X}$ represent the position operator and $| \cdot \rangle$ is the state corresponding to a WF in the cell with $R = 0$. Then we obtain:

$$\bar{x}_n = \frac{i}{2\pi} \int_{-\pi}^{\pi} d\mathbf{k} \langle u_{nk} | \nabla_{\mathbf{k}} | u_{nk} \rangle \quad (2)$$

Assuming $\sum_a \bar{x}_a^S = \frac{1}{2\pi} \int_{\text{BZ}} A^S$ with $S = I$ or II , the summation in a is the occupied states and A is Berry connection. So we get the $\mathbf{Z}_2$ invariant following

$$\mathbf{Z}_2 = \sum_a \left[\bar{x}_a^I \left(\frac{T}{2} \right) - \bar{x}_a^{II} \left(\frac{T}{2} \right) \right] - \sum_a [\bar{x}_a^I(0) - \bar{x}_a^{II}(0)] \quad (3)$$

The calculated evolution of WCC is shown in Fig. 3a. As expected, the WCC is crossed by any arbitrary horizontal reference lines an odd number of times, indicating $\mathbf{Z}_2 = 1$. This firmly confirms the nontrivial topological phase of β_1 - Bi_2Te_3 . As the existence of the localized metallic helical edge channels is the prominent feature for 2D TI, we calculate the armchair edge states by using a tight-binding (TB) Hamiltonian in the maximally localized WF. As shown in Fig. 3b, a pair of edge states around the edge projected Γ -point are observed within the bulk gap. And these states are robust and spin helical, where opposite spin polarizations are propagated along the different directions. The topological edge states further manifest the nontrivial properties. As a result, the coexistence of ferroelectric and topological orders is obtained in trilayer Bi_2Te_3 .

Ferroelectric-topological coupling

In the following, we discuss the coupling of ferroelectricity and topological orders in trilayer Bi_2Te_3 . Different from 2D TI with inversion symmetry, due to the existence of IP electric polarization, the characters of the nontrivial edge states contributed by two opposite zigzag edges would be remarkably anisotropic. Taking the edge states along $[1\bar{1}0]$ and $[\bar{1}10]$ as examples, we show them in Supplementary Fig. 3. As expected, these two

edge states are distinctly different. Under the ferroelectric switching, these two different nontrivial edge states would be exchanged. This means that the character of nontrivial edge state, such as the position of Dirac point, at an assigned edge can be precisely controlled by ferroelectricity. Moreover, because of the coupling between IP and OOP ferroelectricity, either IP or OOP external electric field can trigger such modulation. This results in the coupled ferroelectric and topological physics in such multilayer systems. Benefit from such ferroelectric-topological coupling, the fascinating topological p - n junctions can be easily obtained when forming a side-by-side ferroelectric domain walls with opposite polarizations^{52–54}. In addition, utilizing the either IP or OOP external electric field, such topological p - n junctions are controllable.

Meanwhile, for such multilayer exhibiting weakly coupled vdW interface, the electric field induced transition between two equivalent structural variants with opposite electric polarization could be regarded as a lateral sliding of central Bi_2Te_3 QL with respect to the outmost Bi_2Te_3 QLs. The ferroelectric reversal operation acts as a 180° rotation with respect to the direction perpendicular to both IP and OOP polarizations. That's to say, the two ferroelectric states with opposite polarizations as well as the boundary morphology are linked together through an inversion operation. Besides, IP electric dipole reversal also switches the spin polarization of electronic states^{34,55}, which switches the spin currents at the boundaries. As a result, as shown in Fig. 3c, the chirality as well as the direction of the spin-locked currents at boundaries are closely associated with the direction of ferroelectric polarization, and the direction of topological spin current can be fully controlled by ferroelectricity, which would promote exotic applications in conceptually multifunctional devices. Moreover, due to the direction of spin-locked current can be viewed as a ferroic order, such multilayers can also be treated as multiferroic systems, see Fig. 3d, holding potential for highly efficient multiferroic devices.

DISCUSSION

It should be noted that, the coupled properties are robust in ferroelectric multilayer TIs with different layer number, as long as the polarization reversal in multilayers could be regarded as an

Fig. 3 Topological properties and coupled ferroelectricity and band topology. **a** Evolution of the WCC along k_y for β_1 -Bi₂Te₃. **b** TB band structure of armchair-edged nanoribbon of β_1 -Bi₂Te₃ obtained by MLWFs, showing edge states inside the gap of bulk bands. **c** Proposed electrical switch of the chirality and direction of the spin-locked current at boundary. **d** Schematic representation of 2D FETI with two polar TI states.

inversion operation. We also wish to stress that these coupled ferroelectric and topological physics are not limited to trilayer Bi₂Te₃, but applicable for all 2D FETIs designed by this scheme.

In summary, we introduce a general scheme to realize coupled ferroelectric and topological physics in multilayer systems. Taking trilayer Bi₂Te₃ as an example, we show that through a specific interlayer sliding, both IP and OOP ferroelectricity can be realized in van der Waals multilayer based 2D topological insulators, resulting in the coexistence of ferroelectric and topological orders. We further show that the ferroelectric and topological orders exhibit a strongly coupling. Under the ferroelectric switching, distinct different topological physics can be induced in such multilayer systems.

METHODS

Density functional theory calculations

First-principles calculations are performed based on density functional theory as implemented in the Vienna Ab Initio Simulation Package (VASP)⁴¹. The generalized gradient approximation (GGA) in the scheme of Perdew, Burke, and Ernzerhof (PBE)⁵⁶ is used to describe the exchange correlation. PBE-D3 method is employed for taking van der Waals interaction into account⁵⁷. A 500 eV is adopted for the cut-off energy. A vacuum space larger than 18 Å is employed to eliminate the periodic interactions. Convergence criteria of 10⁻⁵ eV and 0.01 eV Å⁻¹ for energy and forces, respectively, are used. The Brillouin zone integration is sampled with Monkhorst-Pack grids of 9 × 9 × 1. A tight binding (TB) method, based on maximally localized Wannier functions (MLWFs)⁵⁸, is used to calculate

the edge states. Energy barriers of ferroelectric switching are obtained by using nudged elastic band (NEB) method⁵⁹.

DATA AVAILABILITY

All data generated or analysed during this study are included in this published article (and its supplementary information files).

CODE AVAILABILITY

The central codes used in this paper are VASP and WANNIER90. Detailed information related to the license and user guide are available at <http://www.wannier.org> and <https://www.vasp.at>.

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AUTHOR CONTRIBUTIONS

Y.M conceived the project. Y.L performed DFT calculations. All authors commented on the manuscript and contributed to its final version.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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