Three-Dirac-fermion approach to unexpected gapless surface states of van der Waals magnetic topological insulators

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A diverse range of topological quantum phenomena and potential applications of three-dimensional topological insulators (TIs) hinge on opening an energy gap of Dirac-cone surface states. Layered van der Waals (vdW) topological materials, especially the recently discovered MnBi₂Te₄-family magnetic TIs, have aroused great attention, where the interlayer vdW gap is expected to play a crucial role in topological surface states. However, it remains a serious controversy whether the surface states are gapped or gapless for magnetic TI MnBi₂Te₄, which is a crucial issue for the prospect of various magnetic topological states. Here, a 3-Dirac-fermion approach is developed to generally describe surface states of nonmagnetic/magnetic vdW TIs under the interlayer vdW gap modulation. In particular, we apply this approach to solving controversial issues in the surface states of vdW antiferromagnetic (AFM) TIs. Remarkably, unexpected topologically protected gapless Dirac-cone surface states are found to arise due to the interlayer vdW gap expansion on the surface, when the surface ferromagnetic layer has a zero Chern number, while the surface states remain gapped for all other cases. These results are further confirmed by first-principles calculations on AFM TI MnBi₂Te₄. The unexpected gapless Dirac-cone states are invaluable in solving the puzzle of the observed gapless surface states in MnBi₂Te₄. This work also provides a promising way for experiments to realize intrinsic magnetic quantum anomalous Hall effect with a large energy gap in MnBi₂Te₄ films.

INTRODUCTION

Three-dimensional topological insulators (TIs) are featured by topologically protected gapless spin-momentum locked surface states crossing the bulk band gap [1, 2]. When the time-reversal symmetry is broken, an energy gap will be induced at the Dirac point of surface states [3]. The gapped Dirac-cone surface states are essential for a variety of exotic topological quantum phenomena [4], such as, the topological magnetoelectric effect [5, 6] and quantum anomalous Hall effect (QAHE) [7, 8], which are important for technological applications including low-dissipation spintronics and topological quantum computation. A conventional way to achieve the gapped Dirac-cone surface states is through the magnetic doping in TIs, as adopted in the first experimental realization of QAHE [8], but there will inevitably be complex and detrimental effects, for example, magnetic inhomogeneity and disorder effects. Recently discovered MnBi₂Te₄-based intrinsic magnetic TIs [9–43] offer a promising alternative approach, not only in raising the QAHE temperature [44], but also in facilitating the study of axion electrodynamics [45–50]. Nevertheless, there still exists a serious controversy on the surface states of MnBi₂Te₄, since both gapped and gapless surface states are experimentally observed for MnBi₂Te₄ [10, 51–57].

Meanwhile, the most studied nonmagnetic/magnetic TIs are layered materials with an interlayer van der Waals (vdW) gap, such as, Bi₂Se₃ family TIs [58] and MnBi₂Te₄ family magnetic TIs [9–12]. All these materials consist of covalent-bonding layers as building units, which are linked by the weak vdW force, but the interlayer vdW gap plays a significant role in their electronic structures of both bulk and surface states. The vdW gap is sensitive to impurities or intercalated atoms in fabrication processes, and even a small concentration of impurities could cause a sizable expansion of the vdW gap. Especially, the topmost vdW gap on the surface is expected to have the prominent change because of the sudden symmetry breaking on the surface. It has been shown that an expansion of the topmost vdW gap can lead to a re-location of surface states in Bi₂Se₃ family TIs [59].

In this work, we develop a 3-Dirac approach to generally describe the surface states of layered nonmagnetic/magnetic TIs under the interlayer vdW gap modulation. We apply this model to layered A-type AFM TIs [60] and the evolution of surface states under the topmost vdW gap expansion from the bulk vdW gap (d = d₀) to the surface layer decoupled limit (d = ∞), as illustrated in Fig. 1. The surface layer may include one FM unit [e.g. 1 septuple layer of MnBi₂Te₄] [11], schematically shown in Fig. 1(i,ii)] or an AFM unit [e.g. 1 nonuple layer of Mn₂Bi₂Te₅] [45], schematically shown in Fig. 1(iii)]. For the FM surface layer, we find that if the surface layer has a zero Chern number, unexpected topologically protected gapless Dirac-cone surface states will arise at the topological transition point [see (ii) in Fig. 1], while the gapped surface states persist if the surface layer
has a nonzero Chern number [see (i) in Fig. 1]. On the other hand, for the AFM surface layer, the energy gap of surface states is also always maintained in the interlayer vdW gap expansion process [see (iii) in Fig. 1]. Furthermore, these results are confirmed by first-principles calculations for AFM TI MnBi$_2$Te$_4$, which intriguingly uncover the puzzle of the angle resolved photoelectron spectroscopy (ARPES) observations of gapless or gapped surface states of MnBi$_2$Te$_4$. The 3-Dirac approach provides a unified description of surface states of layered TIs, indicating that the interlayer vdW gap engineering would be a new route to facilitate nonmagnetic/magnetic TIs and their applications.

FIG. 1: Evolution of surface states of an AFM TI with a vdW gap expansion. The AFM TI consists of ferromagnetic (FM) or AFM unit layers (colored in blue), which are linked to each other by the weak vdW force. In the expanded case ($d_0$), the AFM TI has gapped Dirac-cone surface states. With increasing the topmost vdW gap ($d = d_0 + \delta d$) between a surface layer (colored in pink) and the below bulk (colored in blue), if the surface layer is FM, depending on its Chern number $C$, two cases will occur: (i) When $|C| = 1$, the surface states remain gapped. (ii) When $C = 0$, the energy gap of surface states has a topologically protected gapless transition. In contrast, (iii) if the surface layer is AFM, the surface states always keep gapped.

EFFECTIVE MODEL ANALYSIS

The surface states of the nonmagnetic/magnetic TIs with an interlayer vdW gap expansion can be captured by a low-energy effective model composed of three helical Dirac fermions. As illustrated in Fig. 2(a), two of the Dirac fermions, labelled as $D_1$ and $D_2$, respectively, come from the top and bottom surface states of the surface layer. The third Dirac fermion labelled $D_3$ comes from the top surface state of the remaining bulk. In the presence of magnetic moments along the out-of-plane direction ($z$-direction), each Dirac fermion can be described by the low-energy Hamiltonian

$$H_{D_i} = sv(k_x \sigma_y - k_y \sigma_x) + h_i \sigma_z,$$

where $i = 1, 2, 3$, $v$ is the Fermi velocity, $s = +1(-1)$ for $D_1$ and $D_3$ ($D_2$), the Pauli matrices act on the spin subspace, and $h_i$ indicates the Zeeman coupling. We further consider two coupling energies: $\Delta_{12}$ between $D_1$ and $D_2$, $\Delta_{23}$ between $D_2$ and $D_3$. Note that $\Delta_{12}$ depends on the thickness of the surface layer, while $\Delta_{23}$ is dependent on the topmost interlayer vdW gap $d$ between the surface layer and the below bulk. In the ordered basis of $(|D_{1\uparrow}⟩, |D_{1\downarrow}⟩, |D_{2\uparrow}⟩, |D_{2\downarrow}⟩, |D_{3\uparrow}⟩, |D_{3\downarrow}⟩)$, the total Hamiltonian of the coupled 3-Dirac-fermion system is then given by

$$H = \begin{pmatrix}
    h_1 & -ivk^− & \Delta_{12} & 0 & 0 & 0 \\
    ivk^+ & -h_1 & 0 & \Delta_{12} & 0 & 0 \\
    \Delta_{12} & 0 & h_2 & ivk^− & \Delta_{23} & 0 \\
    0 & \Delta_{12} & -ivk^+ & -h_2 & 0 & \Delta_{23} \\
    0 & 0 & \Delta_{23} & 0 & h_3 & -ivk^− \\
    0 & 0 & 0 & \Delta_{23} & ivk^+ & -h_3
  \end{pmatrix}. $$

(2)

We first consider the nonmagnetic TIs to see how the surface states change with the topmost vdW gap expansion. With increasing $d$ from the bulk value ($d = d_0$), the coupling strength $\Delta_{23}$ gradually decreases to zero, while $\Delta_{12}$ remains unchanged. Therefore, we treat $\Delta_{12}$ as the energy unit hereinafter. In the absence of the Zeeman term, the nonmagnetic 3-Dirac system exhibits two linear bands with the dispersion $\pm ivk$, and two doubly degenerate bands with the dispersion $\pm \sqrt{k^2 + \Delta_{12}^2 + \Delta_{23}^2}$, where $k \equiv \sqrt{k_x^2 + k_y^2}$. Regardless of the value of $\Delta_{23}/\Delta_{12}$, the two linear bands always meet at $\Gamma$, forming a massless Dirac cone. This can be seen from the doubly degenerate energy level at $\Gamma$ ($k = 0$), as shown in Fig. 2(b). However, the location of the gapless Dirac cone changes from the top of the surface layer to the top of the below bulk [see Fig. 2(a)].

In the presence of A-type AFM order, each of the three Dirac fermions becomes gapped due to a mass term induced by the Zeeman coupling. However, the coupling between them leads to interesting results. First, we consider the surface layer including one FM unit with $h_1 = h_2 = -h_3 = h$ ($h$ is set to be positive, assumed as the Zeeman coupling strength), when the condition $\Delta_{12} > h$ is satisfied, e.g., for a sufficiently thin surface layer, there always appears an energy-gap-closing-and-reopening process with tuning the vdW gap. This can be clearly seen in Fig. 2(e), a gapless Dirac-cone surface state arising at the energy-gap-closing point. Such a gapless surface state is topologically protected by the Chern
number transition of the 3-Dirac system between the unexpanded case of $\Delta_{23} \gg \Delta_{12}$ and the expanded case of $\Delta_{23} \to 0$. The Chern number in the $\Delta_{23} \gg \Delta_{12}$ case can be simply obtained in the limit of $\Delta_{23}/\Delta_{12} \to \infty$ or $\Delta_{12} \to 0$, since the topological properties in both cases can be adiabatically connected to each other without gap closing, so that they have the same Chern numbers. In fact, in the $\Delta_{12} \to 0$ limit, only $D_2$ and $D_3$ are coupled. Because of the $PT$ symmetry combining spatial inversion ($P$) and time reversal ($T$) [see the supplementary material (SM) [61] for more details], the coupled $D_2$ and $D_3$ gives a zero Chern number. Consequently, the total Chern number of the 3-Dirac system in the $\Delta_{12} \to 0$ limit is equivalent to that of the isolated massive Dirac fermion $D_1$, which is given by $C_1 = \text{sgn}(h_1)/2 = 1/2$. In the opposite limit of $\Delta_{23} \to 0$, only $D_1$ and $D_2$ are coupled, while $D_3$ is isolated with the Chern number $C_3 = \text{sgn}(h_3)/2 = -1/2$. For the coupled $D_1$ and $D_2$, its Chern number $C_{12} = 0$ [$C_{12} = \text{sgn}(h)$], when $\Delta_{12} > h$ ($\Delta_{12} < h$) [7]. Correspondingly, the total Chern number in the surface decoupled limit becomes $-1/2$ [$1/2$]. It follows that once $\Delta_{12} > h$ is satisfied, the total Chern number of the 3-Dirac system changes by one in the interlayer vdW gap expansion process, as shown in Fig. 2(e), resulting in the existence of a gapless transition. On the contrary, for $\Delta_{12} < h$ in the surface layer, the total Chern number remains unchanged and the energy gap of surface states keeps open in the vdW gap expansion process [see Fig. 2(f)]. It should be emphasized that the gapless point is topologically protected and comes from the competition between the Zeeman coupling and the Dirac fermion coupling.

Second, we consider an AFM surface layer with $h_1 = -h_2 = h_3 = h$. In this case, there is no gapless transition within the interlayer vdW gap expansion process, as shown in Fig. 2(c). Instead, we see that there is an energy gap of almost constant magnitude $2h$ for the surface state, irrespective of the ratio $\Delta_{23}/\Delta_{12}$. In addition, the total Chern number keeps unchanged as $\text{sgn}(h)/2$, which can be obtained through similar arguments as above by taking two limits of $\Delta_{12} \to 0$ and $\Delta_{23} \to 0$ into account. Furthermore, it is worth mentioning that the 3-Dirac model can also be used to describe the FM TIs, for example, by setting $h_1 = h_2 = h_3 = h$. However, in contrast to the $A$-type AFM TIs, it is found that the gapless transition point appears only when $\Delta_{12} < h$ is satisfied (see the SM for more details of the FM case [61]).

FIG. 2: Coupled 3-Dirac-fermion model. (a, d) Schematic of the 3-Dirac model of the nonmagnetic case (a), and the AFM case (d), where Dirac states ($D_1$ and $D_2$) correspond to the top and bottom surface states of the surface layer, and $D_3$ comes from the top surface states of the below bulk. The coupling energies: $\Delta_{12}$ between $D_1$ and $D_2$; $\Delta_{23}$ between $D_2$ and $D_3$. $\Delta_{12}$ depends on the thickness of the surface layer and is fixed in the interlayer vdW gap expansion, while $\Delta_{23}$ gradually decreases to zero with increasing the interlayer vdW gap $d$. (b, c, e, f) We plot bands at the $\Gamma$ point as a function of $\Delta_{23}/\Delta_{12}$, corresponding to increasing the interlayer vdW gap for the nonmagnetic case (b) and the AFM case with the AFM surface layer (c) and the FM surface layer with $\Delta_{12} > h$ (e) and with $\Delta_{12} < h$ (f). $h$ represents the Zeeman coupling strength. The Chern number changes by one across the gapless transition point in (e).
FIG. 3: AFM TI MnBi$_2$Te$_4$ and nonmagnetic TI CaBi$_2$Te$_4$ with the topmost interlayer vdW gap expansion. (a-d) Energy bands at Γ for (7 + 1)-SL CaBi$_2$Te$_4$ (a), (7 + 1)-SL MnBi$_2$Te$_4$ (b), (7 + 2)-SL MnBi$_2$Te$_4$ (c), and (7 + 3)-SL MnBi$_2$Te$_4$ (d). The bands on the surface layer and the top SL of the below bulk are highlighted in blue, red, orange, and green, respectively. (e) The corresponding band gaps are presented. A gapless transition of the band gap arises at the vdW gap expansion. Cancies might lead to significant changes of the vdW gap between neighboring SLs. (f) The surface LDOS of (7 + 1)-SL MnBi$_2$Te$_4$ at the critical vdW gap expansion δd = 0.18 Å. It exhibits the gapless Dirac-cone surface states.

MATERIAL REALIZATION

As a concrete example of the 3-Dirac model, in what follows we study the layered AFM TI MnBi$_2$Te$_4$, which is built up of FM septuple layers (SLs) coupled to each other through the vdW force. Its magnetic ground state exhibits an A-type AFM order, with the out-of-plane FM coupling within each SL and AFM coupling between neighboring SLs [11]. Notably, the existence of unavoidable MnBi and BiMn antisite defects and Mn vacancies might lead to significant changes of the vdW gap [18, 30, 63, 64]. In addition, the mechanical cleavage and exfoliation processes of the surface preparation, or the sudden symmetry breaking on the surface may also result in the topmost interlayer vdW gap expansion [63]. Now, based on first-principles calculations, we focus on the expansion effects of the vdW gap between a surface layer and the below bulk of MnBi$_2$Te$_4$ by increasing δd = d − d$_0$.

In accordance with the above model analysis, we start from the nonmagnetic case CaBi$_2$Te$_4$ by replacing Mn atoms with Ca atoms in MnBi$_2$Te$_4$. For our calculations, we take a (7 + 1)-SL CaBi$_2$Te$_4$ thin film with gradually expanding the topmost vdW gap between the first and second SL. The evolution of energy bands at Γ with increasing δd = d − d$_0$ is presented in Fig. 3(a), where the bands located at the first and second SLs are highlighted in blue. The blue lines at the Fermi level exhibit doubly degenerate bands in Fig. 3(a). This indicates the persistent existence of the gapless Dirac-cone surface states in the first and second SLs, which are well consistent with the above model analysis, as shown in Fig. 2(b).

We now turn to studying the AFM TI MnBi$_2$Te$_4$. We take (7 + 1)-SL and (7 + 3)-SL MnBi$_2$Te$_4$ films for our calculations. Here, 1 SL in (7 + 1)-SL film and 3 SLs in (7 + 3)-SL film are effectively considered as the surface layer, and the 7 SLs are considered as the below bulk. The reason for such choices lies in the following arguments. For the thickness of MnBi$_2$Te$_4$ thin film smaller (greater) than 3 SLs, the coupling ∆$_{12}$ between the two Dirac-cone surface states (D$_1$ and D$_2$) is expected to be greater (smaller) than the Zeeman coupling strength h, thus leading to a Chern number $C_{12}$ of 0 for the 1-SL (3-SL) MnBi$_2$Te$_4$. In Figs. 3(b) and 3(d), we plot the energy levels at Γ with increasing the vdW gap expansion δd for (7 + 1)-SL and (7 + 3)-SL MnBi$_2$Te$_4$, respectively, where red and green bands denote the bands from the surface layer. As clearly seen in Fig. 3(b), with increasing δd, the energy gap first closes around δd = 0.18 Å and then opens (read lines) in Fig. 3(e). In Fig. 3(f), we explicitly plot the surface local density of states (LDOS) at δd = 0.18 Å, the critical value of the vdW gap ex-
pansion, confirming the emergence of gapless Dirac-cone surface state. In contrast, for the case of the \((7 + 3)\)-SL MnBi\(_2\)Te\(_4\) film, the band gap decreases with increasing \(\delta d\), as shown by green lines in Figs. 3(d) and 3(e), but it tends to saturate for large \(\delta d\) and cannot be closed. In addition, we also present corresponding results for \((7 + 2)\)-SL MnBi\(_2\)Te\(_4\) in Figs. 3(c) and 3(e) (orange lines), where the energy gap of the surface states remains almost unchanged in the whole expansion process. Therefore, the present first-principles calculations for the \((7 + 1)\)-SL \((7 + 2)\)-SL , and \((7 + 3)\)-SL MnBi\(_2\)Te\(_4\) films are in agreement with those obtained by the 3-Dirac model, corresponding to Figs. 2(c), 2(e) and 2(f), respectively.

Interestingly, we find that the gapless Dirac-cone surface states due to the topmost vdW gap expansion (e.g. \(\delta d = 0.18\) Å for MnBi\(_2\)Te\(_4\)), revealed by our 3-Dirac model and first-principles calculations, are expected to solve the puzzle of surface states of MnBi\(_2\)Te\(_4\): contrary to gapped surface states predicted by theories \([11, 42]\), unexpected gapless surface states were observed by many ARPES experiments\([52–54]\). The reconstruction of the surface magnetic moments was proposed as a possible explanation for the gapless surface state of MnBi\(_2\)Te\(_4\)\([54]\). Its key deficiency is that the three-fold rotation symmetry would be broken and the surface states correspondingly lose the three-fold rotation symmetry. On the contrary, the gapless surface states induced by the vdW gap expansion can preserve the three-fold rotation symmetry besides the gapless surface states. In Fig. 4, we plot the calculated surface LDOS and Fermi surfaces (FSs) at two selected energy levels for MnBi\(_2\)Te\(_4\) with \(\delta d = 0.18\) Å (first and second columns). We can see that the surface states are gapless, and that their FSs preserve the three-fold rotation symmetry but break the six-fold rotation symmetry due to breaking the time-reversal symmetry. In order to exclude the possibility of gapless surface states from the nonmagnetic TIs, we calculated the surface states and the FSs of CaBi\(_2\)Te\(_4\), as shown in Fig. 4 (third and forth column). Though the surface states are gapless, the surface states of CaBi\(_2\)Te\(_4\) preserves the six-fold rotation symmetry, essentially different from the three-fold rotation symmetry of AFM TI MnBi\(_2\)Te\(_4\) with

FIG. 4: Surface LDOSs and Fermi surfaces of AFM MnBi\(_2\)Te\(_4\) and nonmagnetic CaBi\(_2\)Te\(_4\). (a,b,e,f,i,j) The surface LDOS of MnBi\(_2\)Te\(_4\) (a,b) and FSs at the energy levels E1 (e,f) and E2 (i,j) with the topmost interlayer vdW gap expansion \(\delta d = 0.18\) Å. The surface states are gapless in (a,b). Surface FSs show the three-fold rotation symmetry due to breaking the time-reversal symmetry. In order to compare with ARPES measurements, we also plot bulk FSs (e.g. \(k_z = 1.2\pi\)), some of which interestingly show a like six-fold rotation symmetry, seen in (f,j). This is because of the weak \(k_z\) dependence of bulk bands due to the PT symmetry\([62]\).  (c,d,g,h,k,l) The surface LDOS (c,d) and FSs at E1 (g,h) and E2 (k,l) of unexpanded CaBi\(_2\)Te\(_4\). Because of the nonmagnetism, surface FSs show a six-fold rotation symmetry, while bulk FSs (e.g. \(k_z = 1.2\pi\)) clearly show a three-fold rotation symmetry. In addition, it is notable that there are a set of Rashba-type surface states at the energy level E2 due to breaking the inversion symmetry on the surface.
the vdW gap expansion. It is worth to mention that the recent point contact tunneling spectroscopy on MnBi$_2$Te$_4$ observed the gapped surface states,[55], which indicates that a moderate pressure on the surface may deduce the the vdW gap expansion to obtain gapped surface states. Therefore, we expect that the moderate pressure would provide a promising way to realize intrinsic magnetic QAHE with a large band gap in MnBi$_2$Te$_4$ films.

CONCLUSION

In summary, we have developed a coupled 3-Dirac-fermion approach to describe the tunability of surface states for layered nonmagnetic/magnetic topological insulators with the vdW gap modulation. We apply the model to the layered AFM TI with the vdW gap expansion, and predict the emergence of topologically protected gapless Dirac-cone surface states in the expansion process. This finding is further confirmed by the first-principles calculations for A-type AFM TI MnBi$_2$Te$_4$. Remarkably, the unexpected gapless Dirac-cone surface states, revealed by our 3-Dirac approach and first-principles calculations, can be used to solve the puzzle of observed gapless surface states of MnBi$_2$Te$_4$ in experiments. Our results also contribute to the understanding of various topological phenomena related to gapped Dirac-cone surface states in vdW topological materials, and provide a promising route to promote applications of magnetic TIs, such as, high-temperature QAHE and topological magnetoelcetric effects.

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Supplemental Materials for “Three-Dirac-fermion approach to unexpected gapless surface states of van der Waals magnetic topological insulators”

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A. Coupled two-Dirac-fermion model with PT symmetry

Here, we show that combined symmetry of inversion \( P \) and time-reversal \( T \) is preserved in a coupled two-Dirac-fermion system, where the two Dirac fermions have opposite T-breaking mass terms (induced by opposite magnetic moments) and opposite helicities. For the 3-Dirac-fermion model with \( h_1 = h_2 = -h_3 = h \) in the main text, when \( \Delta_{12} = 0 \), \( D_1 \) is isolated from \( D_2 \) and \( D_3 \), and the coupled \( D_2 \) and \( D_3 \) subsystem can be exactly described by the above model. In the ordered bases of \( (|D_2\uparrow\rangle,|D_2\downarrow\rangle,|D_3\uparrow\rangle,|D_3\downarrow\rangle) \), the four-band model Hamiltonian is given by

\[
H(k) = \begin{pmatrix}
    h & ivk^- & \Delta_{23} & 0 \\
    -ivk^+ & -h & 0 & \Delta_{23} \\
    \Delta_{23} & 0 & -h & -ivk^- \\
    0 & \Delta_{23} & ivk^+ & -h
\end{pmatrix} = -\tau_z(k_x\sigma_y - k_y\sigma_x - h\sigma_z) + \Delta_{23}\tau_x, \tag{S1}
\]

where \( k^\pm = k_x \pm ik_y \), \( v \) is the Fermi velocity, \( h \) represents the Zeeman coupling strength, and the Pauli matrices \( \tau_i \) and \( \sigma_i \) act in the orbital and spin subspaces. The inversion and time-reversal operators are given by \( \tau_x \) and \( i\sigma_yK \) (\( K \) mean complex conjugate), respectively. The combined \( PT \) operator is then simply obtained as \( i\tau_x\sigma_yK \). It is straightforward to show that the above Hamiltonian satisfies the \( PT \) symmetry as

\[
PT H(k)(PT)^{-1} = H(k). \tag{S2}
\]

Since the antiunitary \( PT \) operator satisfies \( PT^2 = -1 \), Kramers degeneracy is ensured at every momentum, leading to doubly degenerate band structures with vanishing total Berry curvatures and thus zero Chern number for the occupied bands.

B. Ferromagnetic 3-Dirac-fermion model

For the coupled 3-Dirac-fermion model with ferromagnetic moments, i.e., \( h_1 = h_2 = h_3 = h \) (\( h > 0 \) is assumed), the Hamiltonian is given by

\[
H = \begin{pmatrix}
    h & -ivk^- & \Delta_{12} & 0 & 0 & 0 \\
    ivk^+ & -h & 0 & \Delta_{12} & 0 & 0 \\
    \Delta_{12} & 0 & h & ivk^- & \Delta_{23} & 0 \\
    0 & \Delta_{12} & -ivk^+ & -h & 0 & \Delta_{23} \\
    0 & 0 & \Delta_{23} & 0 & h & -ivk^- \\
    0 & 0 & 0 & \Delta_{23} & ivk^+ & -h
\end{pmatrix}. \tag{S3}
\]

The energy spectrum can be easily obtained as

\[
E(k) = \pm\sqrt{h^2 + k^2}, \pm\sqrt{k^2 + (\sqrt{\Delta_{12}^2 + \Delta_{23}^2} - h)^2}, \pm\sqrt{k^2 + (\sqrt{\Delta_{12}^2 + \Delta_{23}^2} + h)^2}. \tag{1}
\]

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Fig. S 1: Bands at the Γ point of the coupled 3-Dirac model with ferromagnetic moments and (a) \( \Delta_{12} < h \), (b) \( \Delta_{12} > h \) in the expansion process of the van der Waals gap between surface and layers, where \( \Delta_{23}/\Delta_{12} \) gradually decreases to zero in the surface-detached limit. The gap closing-and-reopening process appears only when \( \Delta_{12} < h \), which is accompanied by a unit change of the Chern number \( (C) \). In the numerical calculation, \( h/\Delta_{12} \) is chosen to be 1.2 and 0.8 in (a) and (b), respectively.

Obviously, the energy gap closes at the Γ point when \( \sqrt{\Delta_{12}^2 + \Delta_{23}^2} = h \) is satisfied.

In the detaching process of the surface system with the van der Waals gap distance between surface and bulk layers changing from \( d_0 \) to \( \infty \), as shown in Fig. 2(a) in the main text, \( \Delta_{12} \) remains unchanged, while \( \Delta_{23}/\Delta_{12} \) gradually decreases from a large value to zero. It follows that, when \( \Delta_{12} < h \), there exists a gap closing-and-reopening process at a critical value of \( \Delta_{23}/\Delta_{12} \), as shown in Fig. S1(a). This is accompanied by a change of the total Chern number from 1/2 to 3/2. In contrast, when \( \Delta_{12} > h \), the gap always keeps open, as shown in Fig. S1(b), and the Chern number keeps unchanged as 1/2. Note that the condition for the emergence of gap closing in the ferromagnetic case is opposite to that in the antiferromagnetic case in the main text.

C. Surface LDOS and Fermi contours by 3-Dirac-fermion model

In this section, to further show the functionality of the 3-Dirac model, we will use it to calculate the surface local density of states (LDOS) and Fermi contours in the vdW gap expansion process for both the nonmagnetic case and antiferromagnetic case exhibiting a gapless transition point, corresponding to the (7+1)-SL nonmagnetic CaBi$_2$Te$_4$ and antiferromagnetic MnBi$_2$Te$_4$ films.

For the purpose of reflecting the trigonal or hexagonal structures of the surface states, we must include symmetry-allowed higher-order corrections up to third-order terms of \( k \) in the Hamiltonian of Eq. (2) in the main text. In the presence of magnetic order, the only remaining symmetry of each surface state are the three-fold rotation symmetry. As a result, we need to take into account the following symmetry allowed third-order correction for each of the three Dirac fermions

\[
H'_{D_i} = \lambda_i (k_+^3 + k_-^3) \sigma_z, \tag{S4}
\]

where \( k_\pm = k_x \pm ik_y \). The full Hamiltonian \( H' \) of the coupled 3-Dirac-fermion system is then obtained by combining the Hamiltonian in Eq. (2) of the main text with the above terms.

To calculate the LDOS and Fermi contours of the gradually detatched surface composed of the two Dirac fermions, \( D_1 \) and \( D_2 \), we resort to the Green’s function method, and the surface LDOS is given by

\[
\rho(\omega) = -\frac{1}{\pi} \text{Im} G^r_{11,\sigma}(\omega) - \frac{1}{\pi} \text{Im} G^r_{22,\sigma}(\omega), \tag{S5}
\]

where the retarded Green’s function \( G^r(\omega) \) is given by \( G^r(\omega) = (\omega + i\eta - H')^{-1} \). In Fig. S2, we present the results of surface LDOS (first column) and three representative Fermi contours in ascending order of the three energy cuts in the LDOS for the nonmagnetic layered topological insulators (last row) and three stages in the interlayer vdW gap expansion process of layered antiferromagnetic topological insulators, namely, without vdW expansion (first row with a representative \( \Delta_{23}/\Delta_{12} = 2 \)), at the critical gap expansion value exhibiting the gapless Dirac cone (second row with \( \Delta_{23}/\Delta_{12} \approx 0.95 \) for \( h/\Delta_{12} = 0.3 \)), and, in the surface-detached limit with \( \Delta_{23}/\Delta_{12} = 0 \) (third row). From the third
Fig. S 2: Surface LDOS (first column) and corresponding Fermi contours in ascending order of the three energy cuts. The upper three rows correspond to the three stages in the interlayer vdW gap expansion process of a layered antiferromagnetic topological insulator, which presents the gapless transition point in the expansion process. (First row) the unexpanded case with a representative value of $\Delta_{23}/\Delta_{12} = 2$. (Second row) at the critical gap expansion value of $\Delta_{23}/\Delta_{12} \approx 0.95$. (Third row) the surface-detached limit with $\Delta_{23}/\Delta_{12} = 0$. The last row describes the nonmagnetic case with a representative $\Delta_{23}/\Delta_{12} = 2$.

In the numerical calculations, the Zeeman field strength is set as $h/\Delta_{12} = 0.3$, and the coefficient $\lambda$ of the third-order terms in Eq. (S4) is chosen as $\lambda_1 = -\lambda_2 = \lambda_3$ with $\lambda/\Delta_{12} = 0.2$.

It can be clearly seen that, with increasing vdW gap expansion, the Fermi contours gradually change from trigonal shape to hexagonal shape, which results from the gradually recovered inversion symmetry of the detached surface system. It should be emphasized that the results predicted by the effective 3-Dirac-fermion model are in qualitative consistence with the first-principles results in Fig. 4 in the main text.
D. First-principles calculation details

First-principles calculations were performed using the Vienna ab initio simulation package (VASP) \[1\]. The Perdew-Burke-Ernzerhof (PBE) functional \[2\], within the projector augmented wave (PAW) \[3\] are used to describe the exchange-correlation potential and energy. Lattice constants are adopted from experimental data, \(a = 4.334 \text{ Å}\) and \(c = 13.637 \text{ Å}\) for each septuple layer. Then we relax atoms positions. A Hubbard-like \(U = 5 \text{ eV}\) is used to account for strongly localized Mn 3d orbitals. A kinetic cutoff energy of 410 eV and a \(10 \times 10 \times 1\) \(\Gamma\)-centered \(k\) points mesh is used in a self-consistent field. The unit cells are stacked along \(c\) direction to generate multi-septuple layers films with a 20 Å vacuum space for the thick slab system. In all calculations, we keep G-type AFM with a magnetic axis along the \(c\) direction. By replacing Mn in MnBi\(_2\)Te\(_4\) with Ca, and then relaxing the atoms positions, we simulate the non-magnetic case in CaBi\(_2\)Te\(_4\). To account for the surface layer vdW expansion effect, we construct the maximally localized Wannier function (MLWF) \[4\] from first-principles calculations with Mn \(d\), Bi \(p\), and Te \(p\) orbitals. The tight-binding Hamiltonian is divided into surface and bulk parts. The spectral functions and Fermi surface are calculated with the surface Green’s functions of the semi-infinite system. Both spectral functions and the Fermi surface are projected to the upmost septuple layer.

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