Two-atom-thin topological crystalline insulators lacking out of plane inversion symmetry

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Abstract
A two-dimensional topological crystalline insulator (TCI) with a single unit cell (u.c.) thickness is demonstrated here. To that end, one first shows that tetragonal ($C_4$ in-plane) symmetry is not a necessary condition for the creation of zero-energy metallic surface states on TCI slabs of finite-thicknesses, because zero-energy states persist even as all the in-plane rotational symmetries—furnishing topological protection—are completely removed. In other words, zero-energy levels on the model are not due to (nor are they protected by) topology. Furthermore, effective two-fold energy degeneracies taking place at few discrete $k$-points away from zero energy in the bulk Hamiltonian—that are topologically protected—persist at the u.c. thickness limit. The chiral nature of the bulk TCI Hamiltonian permits creating a $2 \times 2$ square Hamiltonian, whose topological properties remarkably hold invariant at both the bulk and at the single u.c. thickness limits. The identical topological characterization for bulk and u.c.-thick phases is further guaranteed by a calculation involving Pfaffians. This way, a two-atom-thick TCI is deployed hereby, in a demonstration of a topological phase that holds both in the bulk, and in two dimensions.

Supplementary material for this article is available online

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(Some figures may appear in colour only in the online journal)

1. Introduction
Studies of the topology of the electronic band structure started by describing the quantum Hall effect according to the Chern number [1], and continued with the prediction of a quantum spin Hall (QSH) effect [2, 3]. Several QSH insulators were experimentally observed in mercury telluride quantum wells [4, 5], Bi$_{1-x}$Sb$_x$ alloys [6], and bismuth chalcogenides [7], and these materials are now commonly known as strong topological insulators (TIs) [8–10]. The topology of the electronic band structure of strong TIs guarantees the existence of robust surface conducting states in what is known as the bulk-boundary correspondence [8–12], and time-reversal symmetry (TRS) produces a Kramers degeneracy on strong TIs. TRS is antiunitary, and customarily written as the product of a unitary operator and the complex conjugation operator [13, 14]. While 2D materials such as stanene are predicted to be 2D TIs [15]...
and Bi2Se3 [8] is a 3D TI, it is rather unusual to find identical topological phases on the same model or material platform at both the 3D and 2D limits.

A TCI is a different kind of (weak) topological insulating phase that is protected by crystalline symmetries [16]; a spinless electronic system in which the product of a crystal symmetry and time reversal symmetry produces an effective Kramers degeneracy, and whose characterization [16] relies on integrals carried out over open paths in momentum space. The TCI phase has been observed on the (high-temperature) cubic phase of SnTe [17], which becomes a ferroelectric (not-TCI) phase at the unit cell thickness limit [18, 19]. Not relying on spin but rather on crystalline symmetries, TCIs present robust edge states against magnetic impurities [20]. A TCI phase has been predicted on inversion symmetric SnTe films [21–23] with a thickness larger than a single unit cell. The mirror reflection symmetry of Pb1−xSnxSe TCI was broken by strain [24], imparting mass to the otherwise massless Dirac fermions. In some TCIs, pressure induces a closing and subsequent reopening of the band gap [25]. Recent first-principles calculations and symmetry analyses predicted that atomically-thin transition metal dichalcogenides with a large band gap [26] and Slater–Koster integrals [27]—which only published last year, and which does contain the dispersion of a finite-size slab—to verify our assertion. This opportunity came to our attention while studying the properties of 1-unit-cell-thick, ferroelectric SnTe [18, 19, 31–33].

Here, we analyze the original TCI model, for which no dedicated studies at the unit cell thickness limit have been carried out yet. Indeed, Fu’s original publication introducing TCIs deals with a semi-infinite slab as the band structure of one of the two surfaces is not visible on the slab electronic dispersion (compare figure 2(b) on [16] to figure 3(b) on [30]—which only published last year, and which does contain the dispersion of a finite-size slab—to verify our assertion). This realization leaves a door for the study of topological properties of two-dimensional analogs of [16] wide-open; something that has not been done yet despite of that original’s work longevity. This opportunity came to our attention while studying the properties of 1-unit-cell-thick, ferroelectric SnTe [18, 19, 31–33].

We determine that the topological invariant characterizing the square of the original TCI Hamiltonian in [16]—one lacking inversion symmetry with respect to the x–y plane—does not change its form in going from the bulk (3D) onto a u.c.-thick slab that still maintains the crucial C4 in-plane rotational symmetry. Furthermore, if one accepts a definition of a topological invariant as (−1)p[π] at the 2D limit (because the Z and A points in momentum space lack meaning in 2D), such invariant remains negative and hence the TCI retains its topological properties when it is one u.c. thick. These unexpected results establish that the weak topology of the original TCI phase stands unaffected (i.e. that it persists) across spatial dimensions.

The following program is developed to that end: The original TCI Hamiltonian, and an analysis of hopping in terms of Slater–Koster integrals [34] are provided in section 2. The effective Kramers degeneracy, and the chiral nature of the Hamiltonian (which gives rise to a block diagonal square Hamiltonian), are discussed in section 3. An analysis of the electronic band structure of finite-size slabs is presented in section 4. The two-fold energy degeneracy of pairs of bands at the 1 and M points is emphasized, as well as the nodal nature of zero-energy surface states. Then, a Hamiltonian without in-plane symmetry is developed in section 5 to show how effective energy degeneracies underpinning the TCI are lifted by the reduction of symmetry. Two-fold degenerate states at the 1 and M points persist down to the u.c. thickness limit due to the block-diagonal nature of the Hamiltonian at these high-symmetry points, in which the pα and py states become decoupled. It is explicitly shown here that the bulk-boundary correspondence does not hold for the TCI: zero-energy surface states on exposed surfaces of sufficiently thick slabs persist even as the crystal symmetry is reduced and the effective degeneracy underpinning the TCI phase does not exist any longer. From this point on, the focus returns to the C4 in-plane symmetric TCI phase. We determine in section 6 that the TCI Hamiltonian [16] is a chiral square-root representation of another 2 × 2 Hamiltonian [35], whose vector field (an indicator of topology [12]) does not change its form in the bulk and the single u.c. thickness limits. The work ends with an explicit calculation of Pfaffians at the bulk and 2D limits in section 7, where an identical topological characterization—already obtained from a squared Hamiltonian—holds in 3D and 2D, for a double verification of the main claim made on this manuscript. Conclusions are provided in section 8.

2. Original TCI Hamiltonian and Slater–Koster analysis of its hopping terms

Consider the four-by-four two-atomic-site bulk Hamiltonian containing one px and one py orbital per atomic site on the tetragonal lattice defined in [16], on a u.c. whose volume a2c is depicted on figure 1(a), and in which hopping terms were indicated, too:

\[ H(\mathbf{k}) = \begin{pmatrix} H^A(k_x, k_y) & H^{AB}(k_x, k_y) \\ H^{BA}(k_x, k_y) & H^B(k_x, k_y) \end{pmatrix}, \]

with \( \mathbf{k} = (k_x, k_y, k_z) \),

\[ H^A(k_x, k_y) = 2\mu \begin{pmatrix} \cos(k_xa) & 0 \\ 0 & \cos(k_ya) \end{pmatrix} \]
\[ + 2\mu^2 \begin{pmatrix} \cos(k_xa) \cos(k_ya) & \sin(k_xa) \sin(k_ya) \\ \sin(k_xa) \sin(k_ya) & \cos(k_xa) \cos(k_ya) \end{pmatrix}, \]

(2)

(\( \alpha = A, \text{or} B \))

\[ H^{AB}(k_x, k_y) = [t'_1 + 2t'_2 (\cos(k_xa) + \cos(k_ya)) + t'_4 e^{ik_zc}] \sigma_0 \]
\[ = [f(k_x, k_y) + t'_4 e^{ik_zc}] \sigma_0 = f_b(\mathbf{k}) \sigma_0, \]

(3)

where \( b \) stands for bulk. \( H^A(\mathbf{k}) \) is a real matrix which only depends on \( k_x \) and \( k_y \) in [16], and such dependency was made explicit in equation (2).
In equations (1)–(3), σ₀ is the 2×2 identity matrix, \( t^A \) indicates the Hermitian adjoint operator, \( t^A = -t^B = 1, t^I = -t^2 = 0.5, t^I' = 2.5, t^I = 0.5, \) and \( t^I' = 2 \) (eV units are adopted for the hopping terms for definiteness here). Figure 1(b) shows the k-point path used to plot the bulk band structure on figures 1(c) and 1(d) shows the density of states (DOS) sampled over the tetragonal first Brillouin zone.

We link the tight-binding Hamiltonian in equation (1) to expressions provided by Slater and Koster involving \( p_x \) and \( p_y \) orbitals [34], to guide the design of low-symmetry Hamiltonians in section 5. Leaving the details to the supplementary material, \( H^{(2)}_{pp} = t^I', H^{(2)}_{pp} = 0, H^{(2)}_{pp} = t^I, \) and \( H^{(2)}_{pp} = 0, \) where the upper index \( j \) stands for first (11) or second (22) nearest neighbor.

As for out-of-plane second nearest neighbors, the \( p_x^A p_y^B \) entry is equal to zero, while the \( p_x^A p_y^B \) and \( p_y^A p_x^B \) entries become:

\[
2 \left[ \frac{a^2}{a^2 + c'^2} H^{(3)}_{pp} + \frac{c'^2}{a^2 + c'^2} H^{(3)}_{pp} \right] \cos(k_x a) + 2 H^{(2)}_{pp} \cos(k_x a), \quad \text{and} \]

\[
2 H^{(2)}_{pp} \cos(k_x a) + 2 \left[ \frac{a^2}{a^2 + c'^2} H^{(3)}_{pp} + \frac{c'^2}{a^2 + c'^2} H^{(3)}_{pp} \right] \cos(k_x a),
\]

respectively (\( a \) and \( c' \) were defined on figure 1(a)). According to equation (3), the term \( t^I' \) multiplying \( \cos(k_x a) + \cos(k_y a) \) is identical for \( p_x^A p_y^B \) and for \( p_y^A p_x^B \), which sets a constraint \( H^{(3)}_{pp} = H^{(3)}_{pp} = t^I' \) on equation (4). The present analysis will be employed to design a Hamiltonian on a monoclinic unit cell later on.

3. Effective Kramers degeneracy and chiral symmetry of the original TCI Hamiltonian

The \( C_4 \) symmetry within the \( xy \)-plane is represented by the \( e^{i\pi/2} \) operator, and the rotation matrix for the bulk Hamiltonian is built out of two copies of it, one per atom:

\[
\mathbb{U} = \begin{pmatrix} 0 & 1 & 0 & 0 \\ -1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & -1 & 0 \end{pmatrix},
\]

which has double degenerate ±i eigenvalues.

Imaginary contributions enter \( H(k) \) through \( t^I' e^{ik} \sigma_0 \) in equation (3). Nevertheless, this phase is real in the \( k_z = 0 \) and \( k_z = \pi \) planes (where Pfaffians will be computed [16], making the Hamiltonian real there as well. \( H(k_z, k_y, 0) \) and \( H(k_z, k_y, \pi/c) \) thus satisfy an effective Kramers degeneracy:

\[
H(k) = \Xi H(k) \Xi, \quad \Xi \left| \psi_{m}(k) \right\rangle = - \left| \psi_{m}(k) \right\rangle
\]

at these two planes, with \( \Xi = \mathbb{U}^2 T, m = 1, 2, \) and \( T \) being the antiunitary time reversal operator (a mere complex conjugation for the spinless fermions being considered here).

The choice \( t^I' = -t^I' \) and \( t^I' = -t^I' \) (i.e. \( H^{(b)}(k_z, k_y) = -H^{(b)}(k_z, k_y) \)) creates a chiral symmetry that makes the dispersion on equation (1) electron–hole symmetric:

\[
H(k) = \begin{pmatrix} H^A(k_z, k_y) & H^{AB}(k) \\ H^{AB}(k) & -H^A(k_z, k_y) \end{pmatrix}.
\]

The chirality of the bulk Hamiltonian, equation (7), implies that for any eigenvector with energy \( E : \left| \Psi_{E}(k) \right\rangle = (\psi^A_A, \psi^A_B, \psi^B_A, \psi^B_B)^T \), there exists an eigenvector with energy \( -E : \left| \Psi_{-E}(k) \right\rangle = (\psi^A_A, \psi^A_B, -\psi^B_A, -\psi^B_B)^T \) [36]. The electron–hole symmetry of \( H(k) \) is evident on figures 1(c) and (d), and it will lead to a block-diagonal square Hamiltonian in section 6.

When applied to the bulk Hamiltonian (equation (7)), equation (5) satisfies the symmetry requirements stated as equation (4) in [16]. In addition,

\[
[\mathbb{U}, H(k)] = 0,
\]

at \( \Gamma, M, Z, \) and \( A \). Equation (8) is crucial to ensure two-fold degenerate eigenvalues at these high-symmetry points. The energy degeneracies at these high-symmetry points are the crucial ingredient for the non-trivial topology of the original TCI model.

4. Electronic structure of slab Hamiltonians from the original TCI model

As indicated at the introduction, [16] emphasizes surface states on a semi-infinite slab configuration. Published only last year, [30] shows the electronic structure of a finite slab with surface
complex conjugation was used—as all matrices involved are shown on figure three, (e) two, and (f), (g) one u.c.s.
dispersion for TCI slabs with (a) twenty, (b) ten, (c) four, (d) slabs with sublattice thickness. Figure slabs with one u.c. thickness, and figures terminations on opposite sublattices for the first time. Here, we x (d) A-sublattice and (e) B-sublattice slabs. Slabs are periodic on the for selected slabs: (a) two u.c.s. (b) ‘short’ u.c. (c) ‘long’ u.c. Figure 2. Atomistic structure and schematics of hopping terms for selected slabs: (a) two u.c.s. (b) ‘short’ u.c. (c) ‘long’ u.c. (d) A-sublattice and (e) B-sublattice slabs. Slabs are periodic on the x- and y-directions.

terminations on opposite sublattices for the first time. Here, we aim to explore the topology of the original TCI model right at the 2D limit, and study slabs with varying thicknesses next with that goal in mind. To that end, figure 2(a) exemplifies a slab with two u.c. thickness, while figures 2(b) and (c) show slabs with one u.c. thickness, and figures 2(d) and (e) represent slabs with sublattice thickness. Figure 3 shows the electronic dispersion for TCI slabs with (a) twenty, (b) ten, (c) four, (d) three, (e) two, and (f), (g) one u.c.s.

To be explicit, all Hamiltonians arising from the structures shown on figure 2 are:

\[ H_{2uc}(k_x, k_y) = \begin{pmatrix}
H^A(k_x, k_y) & f_s(k_x, k_y) \sigma_0 \\
\tau^T f_s(k_x, k_y) \sigma_0 & -H^B(k_x, k_y)
\end{pmatrix} \]  \hspace{1cm} (9)

for the two-u.c. structure shown as figure 2(a), where no complex conjugation was used—as all matrices involved are real—and \( \tau \) is a 2 x 2 matrix full of zeroes. The eigenvalues of \( H_{2uc}(k_x, k_y) \) are displayed on figure 3(e).

As illustrated on figures 2(b) and (c), there are two different ways to build a slab with a single u.c. thickness. On the first choice (labeled short), the A and B atoms are separated by a distance \( c' \) (see figures 1(a) and 2(b)). In the second choice (dubbed long) the A atom is placed above the B atom at a height \( c - c' \) (figure 2(c)). The two Hamiltonians for these slabs are:

\[ H_l(k_x, k_y) = \begin{pmatrix}
H^A(k_x, k_y) & f_s(k_x, k_y) \sigma_0 \\
\tau^T f_s(k_x, k_y) \sigma_0 & -H^B(k_x, k_y)
\end{pmatrix} \]  \hspace{1cm} (10)

for the short u.c. slab (figure 2(b)), and

\[ H_l(k_x, k_y) = \begin{pmatrix}
H^A(k_x, k_y) & f_s(k_x, k_y) \sigma_0 \\
\tau^T f_s(k_x, k_y) \sigma_0 & -H^B(k_x, k_y)
\end{pmatrix} \]  \hspace{1cm} (11)

for the long u.c. slab (figure 2(c)). These slabs with one u.c. thickness are semiconductors with \( E_g = 2.122 \) eV or 3.956 eV energy band gaps, respectively (see figures 3(f) and 3(g)). No investigations of topological properties of those u.c.-thick slabs have ever been reported in the literature.

Figures 3(h) and (i) depict the electronic dispersions of slabs having a sublattice thickness, whose Hamiltonians are either \( H^A(k_x, k_y) \), or \( -H^B(k_x, k_y) \), respectively (see figures 2(d) and (e) for schematics), and which will be useful to fit electronic dispersions around the \( \bar{M} \) point. Building slabs with increased thickness is relatively straightforward. Defining

\[ \tau = \begin{pmatrix}
\sigma_0 \\
\tau^T \sigma_0
\end{pmatrix}, \]  \hspace{1cm} (12)

(which remains a real matrix), equation (9) can be rewritten as:

\[ H_{2uc}(k_x, k_y) = \begin{pmatrix}
H_l(k_x, k_y) & \tau \\
\tau^T & \sigma_0
\end{pmatrix} \hspace{1cm} (13)

where \( \tau^T \) is the transpose of \( \tau \). The plots shown on figures 3(a) through 3(d) are the eigenvalues of Hamiltonians built in a way similar to equation (13). For instance, figure 3(c) shows the eigenvalues of

\[ H_{4uc}(k_x, k_y) = \begin{pmatrix}
H_l(k_x, k_y) & \tau \\
\tau^T & \sigma_0
\end{pmatrix} \hspace{1cm} (14)

where \( \sigma_0 \) is a 4 x 4 matrix full of zeroes.

Horizontally scaled DOS plots confirm the existence of the electronic bandgaps \( E_g \) as observed in the band structure plots. (Explicit code for calculating band structures and DOS for slabs containing ten and two unit cells is facilitated as supporting material. We also provide a study of the difference among hybridized states near zero energy, and the block-diagonal nature of slab Hamiltonians at high-symmetry points there.) With the exception of figures 3(h) and (i) (which are slabs made out of only one sublattice, and whose electronic occupation differs at the \( \Gamma \) and \( \bar{M} \) points), all slabs leading to the band structures on figure 3 have opposite sublattice atoms at
Figure 3. Electronic dispersion of slabs containing \( n \) u.c.s: (a) \( n = 20 \), (b) \( n = 10 \), (c) \( n = 4 \), (d) \( n = 3 \), (e) \( n = 2 \), (f) \( n = 1 \), short u.c., and (g) \( n = 1 \), long u.c. As illustrated on figure 2, these slabs have atoms belonging to opposite sublattices facing vacuum. (h) Electronic dispersion for a slab with an \( A \)-sublattice thickness, and (i) with an \( B \)-sublattice thickness only. Orthogonality of states cannot be guaranteed away from the \( \bar{\Gamma} \) and \( \bar{M} \) points, leading to energy gaps \( E_g \) at zero energy that are confirmed by DOS calculations. Lower subplots are zoom-ins showing overlap with the dispersion of the \( A \)-sublattice (similar behavior exists for the dispersion around \( M \) at 1 eV). Two-fold degenerate energy crossings never split at the \( \bar{\Gamma} \) and \( \bar{M} \) points, regardless of slab thickness.

Figure 4. (a) Schematic depiction of a 40 u.c.-thick slab. (b) Nodal structure (blue curves) of the Fermi energy for this slab. (c) Zero energy states at the crossing marked by a rotated square in panel (b), located at \( 0.673 \bar{M} \).

There is always a region in \( k \)-space centered about \( \bar{M} \) in which the surface dispersion is given analytically by either \( H^A(k_x, k_y) \) (lower exposed, \( A \)-sublattice surface), or by \( -H^A(k_x, k_y) \) (upper exposed, \( B \)-sublattice surface), so that the dispersion of the surface bands can be known exactly; such discussion can be found as supporting material.

The electronic dispersion at zero energy will be discussed in section 5, making it necessary to identify the points in reciprocal space leading to zero energy states on sufficiently thick slabs. Figure 4(b) contains a depiction of zero-energy electronic dispersion contours for the slab with a thickness of 40 u.c.s, showcased on figure 4(a). The electronic bandgaps to be shown on figure 6 were obtained along the \( \bar{\Gamma} - M \) line (or the \( \bar{\Gamma} - C \) line for an oblique lattice). Figure 4(c) verifies that states at zero energy are surface states, as their amplitude increases toward the exposed surfaces.

5. 3D and 2D Hamiltonians arising from the original TCI model but with reduced symmetries

As indicated in [37], ‘the absence of gapless modes at boundaries that break the spatial symmetries does not indicate trivial bulk topology and therefore cannot be used to infer the topology of TCIs.’ In other words, the bulk-boundary correspondence (which applies to strong TIs [8–10]) does not hold for (weak) TCIs, and an explicit proof—which we have not found in the literature—is provided in what follows.

The proof requires showing that zero-energy states—that develop on sufficiently thick slabs—persist even when \( C_4 \) symmetry is removed, so that topological protection does not exist anymore. In pursuing the proof, one also sees the effective degeneracy being lifted at four high-symmetry points on the bulk first Brillouin zone (and, accordingly, on two \( k \)-points on the 2D Brillouin zone) when \( C_4 \) symmetry no longer holds. The link among \( C_4 \) symmetry and the effective degeneracy seen at the \( \bar{\Gamma} \) and \( M \) points in the bulk persists at the \( \bar{\Gamma} \) and \( \bar{M} \) points in 2D.

For this purpose, figure 5(a) depicts a monoclinic unit cell created by lattice vectors \( \mathbf{a} = a(1 + \epsilon, 0, 0), \mathbf{b} = a(\sin \delta, \cos \delta, 0), \) and \( \mathbf{c} = c(0, 0, 1) \), where the two parameters \( \epsilon \) and \( \delta \) take values from 0 to 1. The link among \( C_4 \) symmetry and the effective degeneracy seen at the \( \bar{\Gamma} \) and \( M \) points in the bulk persists at the \( \bar{\Gamma} \) and \( \bar{M} \) points in 2D.
showcasing an symmetry (\(\epsilon\) with \(R\) electron–hole symmetric). Two-fold energy degeneracies at symmetries). (b) The angle for second-nearest-neighbor hopping (a) Monoclinic lattice (having no in-plane rotational symmetries). (b) The angle for second-nearest-neighbor hopping terms changes from 45° to either \(\beta\) or \(\gamma\); one diagonal becomes larger (\(L_1\), while another (\(L_2\) shortens. (c) High-symmetry points and path in reciprocal space. (d) Band structure of the bulk Hamiltonian for occupied states (the band structure remains electron–hole symmetric). Two-fold energy degeneracies at \(\Gamma\), \(S\), \(Z\), and \(R\) high-symmetry points become lifted as the \(C_4\) in-plane symmetry (\(\epsilon = 0.0\) and \(\delta = 0.0\); upper subplot) is removed altogether (\(\epsilon = 0.1\) and \(\delta = 2.5\); lower subplot). (e) DOS showcasing an \(E_f\) = 0.56 eV bandgap for the monoclinic structure with \(\epsilon = 0.1\) and \(\delta = 2.5\). and \(\delta\) bring the unit cell away from a tetragonal structure onto the monoclinic one. As seen on figure 5(b), \(\epsilon > 0\) represents an elongation of one in-plane lattice vector (a), while \(\delta > 0\) indicates a tilt of the second in-plane lattice vector b onto a.

Non-zero values for \(\epsilon\) and \(\delta\) alter Slater–Koster integrals involved in hopping. For small values of \(\epsilon\), we introduce a reduction of \(t_{1}^H\) and of \(t_{2}^H\) along the \(x\)-direction by \(1 - \epsilon\). In-plane second nearest neighbor distances (whose original magnitude is \(\sqrt{2a}\)) get modified under the monoclinic symmetry as well (see figure 5(b)):

\[
L_1(\epsilon, \delta) = \sqrt{2a} \sqrt{(1 + \epsilon)(1 + \sin \delta) + \epsilon^2/2}, \quad \text{and}
\]

\[
L_2(\epsilon, \delta) = \sqrt{2a} \sqrt{(1 + \epsilon)(1 - \sin \delta) + \epsilon^2/2},
\]

leading to additional renormalization of hopping integrals. Angles among in-plane second nearest neighbors change from 45° on the tetragonal lattice onto:

\[
\cos \beta(\epsilon, \delta) = \sqrt{1 - a^2 \cos^2 \delta \over (L_1(\epsilon, \delta))^2}, \quad \text{and}
\]

\[
\cos \gamma(\epsilon, \delta) = \sqrt{1 - a^2 \cos^2 \delta \over (L_2(\epsilon, \delta))^2},
\]

and induce additional renormalization of hopping integrals. Figure 5(c) depicts the first Brillouin zone for the monoclinic lattice, whose high symmetry points were labeled following [38], and the monoclinic bulk Hamiltonian

\[
H_m(k) = \begin{pmatrix}
H_A^m(k_x, k_y) & H_B^m(k) \\
H_B^m(k) & -H_A^m(k_x, k_y)
\end{pmatrix},
\]

(17)

takes the following explicit form:

\[
H_A^m(k_x, k_y) = 2t_1^H \left( (1 - \epsilon) \cos (ka(1 + \epsilon)) + \sin^2 \delta \cos (a(\sin \delta k_x + \cos \delta k_y)) \right) + \left( \begin{array}{cc}
H_{m,p,p}^A & H_{m,p,p}^A \\
H_{m,p,p}^A & H_{m,p,p}^A
\end{array} \right)
\]

(18)

where

\[
H_{m,p,p}^A = 2t_1^H \sqrt{2a} \cos^2 \beta \cos [a(1 + \epsilon + \sin \delta)k_x + a \cos \delta k_y]
\]

\[
+ 2t_1^H \sqrt{2a} \cos \gamma \cos [a(1 + \epsilon - \sin \delta)k_x - a \cos \delta k_y],
\]

\[
H_{m,p,p}^A = 2t_1^H \sqrt{2a} \cos \beta \sin \beta \cos [a(1 + \epsilon + \sin \delta)k_x + a \cos \delta k_y]
\]

\[
- 2t_1^H \sqrt{2a} \cos \gamma \sin \gamma \cos [a(1 + \epsilon - \sin \delta)k_x - a \cos \delta k_y],
\]

The \((1 - \epsilon)\) prefactor seen on one term in equation (18) represents the decrease on the coupling strength along the \(x\)-direction of the original hopping term \(t_1^H\) due to the elongation of lattice vector a. The modification of the hopping
Table 1. Breakdown of the effective energy degeneracy of occupied states at high symmetry points on the (3D) monoclinic lattice, equation (17) (in eV). High symmetry points $H_2$ and $C$ turn into $M$, while $k$-points $M_2$ and $E$ turn into $A$ as the unit cell becomes tetragonal ($\epsilon = 0.0$ and $\delta = 0.0^\circ$).

|          | $\epsilon = 0.0$ and $\delta = 0.0^\circ$ | $\epsilon = 0.1$ and $\delta = 2.5^\circ$ | $\epsilon = 0.2$ and $\delta = 5.0^\circ$ |
|----------|------------------------------------------|-------------------------------------------|------------------------------------------|
| $\Gamma$ | $-7.1589, -7.1589$                      | $-7.0364, -6.9849$                      | $-6.9209, -6.8221$                      |
| $H_2$    | $-2.6926, -2.6926$                      | $-2.8618, -2.7235$                      | $-3.0457, -2.8160$                      |
| $C$      | $-2.6926, -2.6926$                      | $-2.8495, -2.7055$                      | $-3.0014, -2.7535$                      |
| $Z$      | $-3.9051, -3.9051$                      | $-3.7829, -3.6863$                      | $-3.6740, -3.4843$                      |
| $M_2$    | $-1.8028, -1.8028$                      | $-1.8050, -1.5765$                      | $-1.7912, -1.3644$                      |
| $E$      | $-1.8028, -1.8028$                      | $-1.8221, -1.5873$                      | $-1.8461, -1.4077$                      |

Table 2. Breakdown of the effective energy degeneracy of occupied states at high symmetry points on the oblique 2D lattice—equation (21)—for the short u.c. slab (in eV). High-symmetry $k$-points $H_2$ and $C$ turn into $M$ as the unit cell becomes square ($\epsilon = 0.0$ and $\delta = 0.0^\circ$).

|          | $\epsilon = 0.0$ and $\delta = 0.0^\circ$ | $\epsilon = 0.1$ and $\delta = 2.5^\circ$ | $\epsilon = 0.2$ and $\delta = 5.0^\circ$ |
|----------|------------------------------------------|-------------------------------------------|------------------------------------------|
| $\bar{\Gamma}$ | $-5.4083, -5.4083$                      | $-5.2830, -5.2143$                      | $-5.1671, -5.0339$                      |
| $H_2$    | $-1.1180, -1.1180$                      | $-1.3130, -0.9753$                      | $-1.4975, -0.9464$                      |
| $\bar{C}$ | $-1.1180, -1.1180$                      | $-1.3115, -0.9590$                      | $-1.4860, -0.8842$                      |

Table 3. Breakdown of the effective energy degeneracy of occupied states at high symmetry points on the oblique lattice for the long u.c. slab—equation (22)—in eV.

|          | $\epsilon = 0.0$ and $\delta = 0.0^\circ$ | $\epsilon = 0.1$ and $\delta = 2.5^\circ$ | $\epsilon = 0.2$ and $\delta = 5.0^\circ$ |
|----------|------------------------------------------|-------------------------------------------|------------------------------------------|
| $\bar{\Gamma}$ | $-3.6056, -3.6056$                      | $-3.5427, -3.4393$                      | $-3.4941, -3.2940$                      |
| $H_2$    | $-2.2361, -2.2361$                      | $-2.3117, -2.1380$                      | $-2.3806, -2.0785$                      |
| $\bar{C}$ | $-2.2361, -2.2361$                      | $-2.3152, -2.1353$                      | $-2.3913, -2.0717$                      |

terms $t'_2$ on the second matrix on equation (18)—representing the interactions among in-plane second nearest neighbors—is captured by the ratio $\frac{\sqrt{2\epsilon}}{\epsilon_{2,C}(\sigma_{\epsilon})}$ or $\frac{\sqrt{2\epsilon}}{\epsilon_{2,D}(\sigma_{\epsilon})}$, which are slightly larger or smaller from unity, respectively. A detailed derivation of the in-plane components of the monoclinic Hamiltonian is presented in the supplementary material.

The matrix coupling electrons among the $A$ and $B$ sublattices in the monoclinic lattice looks as follows:

$$H_m^{AB}(k) = \left[ t'_1 + 2t'_2(1 - \epsilon) \cos(k_{x\sigma}(1 + \epsilon)) \right] \sigma_0 + \left[ 2t'_2 \cos(k_{y\sigma}\sin\delta + \epsilon_{k\sigma}\cos\delta) + t'_2g_{k\epsilon} \right] \sigma_0,$$  

(19)
as, once again, only the bond with a non-zero projection onto the $x$-axis became elongated (leading to a single modified hopping $t'_2 \rightarrow -t'_2(1 - \epsilon)$ along the $x$-direction). The band structure for the bulk is shown on figure 5(d), and the DOS is depicted on figure 5(e) when $\epsilon = 0.1$ and $\delta = 2.5^\circ$. The breakdown of the energy degeneracy at high symmetry points is listed in table 1.

Defining

$$f_{s,m}(k_x, k_y) = t'_1 + 2t'_2(1 - \epsilon) \cos(k_{x\sigma}(1 + \epsilon)) + 2t'_2 \cos(k_{y\sigma}\sin\delta + \epsilon_{k\sigma}\cos\delta),$$  

(20)

(which remains real), and noticing that the vertical hopping $t'_2$ remains unaffected on the lowering of symmetry onto a monoclinic unit cell (such that $\tau$ in equation (12) remains unchanged), we find the effect of the lowering of symmetry on slabs with 1 u.c. thickness:

$$H_{s,m}(k_x, k_y) = \begin{pmatrix} H_{s,m}(k_x, k_y) & f_{s,m}(k_x, k_y) \sigma_0 & -f_{s,m}(k_x, k_y) \sigma_0 \\ f_{s,m}(k_x, k_y) \sigma_0 & -H_{s,m}(k_x, k_y) & \sigma_0 \\ -f_{s,m}(k_x, k_y) \sigma_0 & \sigma_0 & -H_{s,m}(k_x, k_y) \end{pmatrix},$$  

(21)

for the small u.c., or

$$H_{l,m}(k_x, k_y) = \begin{pmatrix} H_{l,m}(k_x, k_y) & t'_2 \sigma_0 & -H_{l,m}(k_x, k_y) \\ t'_2 \sigma_0 & -H_{l,m}(k_x, k_y) & \sigma_0 \\ -H_{l,m}(k_x, k_y) & \sigma_0 & H_{l,m}(k_x, k_y) \end{pmatrix},$$  

(22)

for the long u.c. A monoclinic 3D lattice becomes oblique in two-dimensions, and the breakdown of the effective degeneracy at the 2D limit for the oblique unit cell is demonstrated in tables 2 and 3 for both short and long u.c.s, respectively.

An observation from tables 1–3 is that the degenerate states protected by $C_2$ symmetry always are located at energies away from the Fermi energy; i.e. away from zero eV.

It is possible to build thicker oblique slabs following prescriptions similar to those leading to equation (14). For example, the oblique slab with 5 u.c. thickness displayed on figure 6(b) is given by:

$$H_{suc,m}(k_x, k_y) = \begin{pmatrix} H_{suc,m}(k_x, k_y) \tau & \circ & \circ & \circ \\ \circ & H_{suc,m}(k_x, k_y) \tau & \circ & \circ \\ \circ & \circ & H_{suc,m}(k_x, k_y) \tau & \circ \\ \circ & \circ & \circ & H_{suc,m}(k_x, k_y) \tau \end{pmatrix},$$  

(23)

with $\epsilon = 0.1$ and $\delta = 2.5^\circ$.

Oblique slabs with finite thicknesses prove that the emergence of zero-energy states is not due to topological
Figure 6. (a) Electronic structure of $n = 5$ u.c. thick slabs with topological protection (TCI: $\epsilon = 0$ and $C_3$ in-plane symmetry) or (b) without ($\epsilon = 0.1$ and $\delta = 2.5^\circ$ in equation (23); no in-plane symmetry). The energy gap $E_g$ is highlighted by dashed horizontal lines, and DOS plots are added to emphasize the bandgaps. (c) and (d) Emergence of zero-energy states on a thicker slab with (subplot (c)) or without topological protection (subplot (d)). (e) $E_g$ decays exponentially with the number $n$ of u.c.s, regardless of the presence of in-plane $C_s$ symmetry (in which topological protection exists) or the lack of it (where topological protection is absent). The single square for $n = 1$ is $E_g$ for the long u.c.

6. Square-root nature of original TCI Hamiltonian and its identical topology in 3D and 2D

The midgap states in the entanglement spectrum or in the entanglement Hamiltonian are customarily employed to distinguish between topologically trivial and non-trivial phases of TCIs [41–44]. Here, on the other hand, we will rely on the square Hamiltonian of the TCI, to demonstrate an identical topological characterization at the bulk and single u.c. limits of the TCI Hamiltonian, which serves as the first indication of the non-trivial topology of the TCI at the 2D limit.

Indeed, Arkinstall and coworkers proposed the creation of new topological phases by inducing a square root operation to

...
topological tight-binding Hamiltonians [35]. The square root operation provides a chiral symmetric arrangement of energy bands at positive and negative energies, inducing spectral symmetries at the expense of broken crystal symmetries. In this case, the broken symmetry is a lack of reflection with respect to the xy-plane by two atoms vertically stacked within the u.c. Arkinstall et al also indicate that the square root Hamiltonian of a topological phase retains topological features. There is intense and ongoing work on square-root descriptions of topological phases [45, 46].

From equations (2) and (3), \[ H^d(k_x,k_y) \] is shown on figure 2. \[ H^d(k_x,k_y) \] is proportional to \( \sigma_0 \), making the square Hamiltonian of the (bulk) TCI block-diagonal:

\[
(H(k))^2 = \begin{pmatrix} H^d_0(k) & 0 \\ 0 & H^b_0(k) \end{pmatrix},
\]

where \( H^d_0(k) \), defined as:

\[
H^d_0(k) = \begin{pmatrix} H^d_1(k) & H^d_{1b}(k) \\ H^d_{1b}(k) & H^d_2(k) \end{pmatrix},
\]

is a renormalized 2x2 Hamiltonian in which one of the two sublattices has been effectively removed [47], and \( b \) stands for bulk. Explicit calculations yield:

\[
d_0(k_x,k_y) = d_0(k_x,k_y) = \begin{pmatrix} d_0(k_x,k_y) \\ 0 \end{pmatrix},
\]

and:

\[
d_0(k_x,k_y) = 2h_0(k_x,k_y)h(k_x,k_y),
\]

where

\[
h_0(k_x,k_y) = t_0^i(k_x + k_y) + 2t_1^0 \cos k_y \cos k_x,
\]

\[
h(k_x,k_y) = (2t_3^0 \sin k_y) \cos k_x,
\]

which in turn originate from reparametrizing \( H^d(k_x,k_y) \) as \( h_0(k_x,k_y) \sigma_0 + h(k_x,k_y) \sigma \) (and whose vector field \( h(k_x,k_y) \) is shown on figure 7(a)). Equation (27) states that \( d_0(k_x,k_y) = d_0(k_x,k_y) \), so that the vector field does not depend on \( k_z \) in the bulk, giving rise to the two-dimensional plot shown on figure 7(b). (Strong TIs have circulating paths enclosing the origin [12], which is not the case on figure 7.)

The topological nature of \( H^d_0(k) \), encoded in \( d_0(k_x,k_y) \cdot \sigma \) [12], is obtained next. For definiteness, an expression for \( d_0(k_x,k_y) \) for small values of \( k_x \) and \( k_y \) was considered; \( |k_x| \ll \pi/a \) and \( |k_y| \ll \pi/a \). In that limit:

\[
h_0(k_x,k_y) \approx t_0^i \left( 2 - \frac{k_x^2}{2} \right) + 2t_1^0 \left( 1 - \frac{k_y^2}{2} \right),
\]

and:

\[
h(k_x,k_y) \approx 2t_3^0 \sin k_y \cos k_x.
\]

Crystal momenta \( k_x \) and \( k_y \) are now recast as \( k_0 \cos \theta \) and \( k_0 \sin \theta \), to determine the evolution of \( H^d_0(k) \) under an adiabatic rotation on k-space by \( 2\pi \), as encoded on \( d_0(k_x,k_y) \cdot \sigma \). To leading order, one gets around \( \Gamma \):

\[
d_0(k_0,\theta) \cdot \sigma \rightarrow 4k_0^4 \begin{pmatrix} t_1^0 \cos 2\theta - t_1^0 \sin 2\theta \\ t_1^0 \sin 2\theta \end{pmatrix},
\]

so that \( d_0(k_0,\theta) \cdot \sigma \) changes by \( 4\pi \) when \( \theta \) completes a cycle, for a winding number of 2.

The square roots of the eigenvalues from \( H^d_0(k) \) yield the dispersion of the Hamiltonian (equation (7)) shown on figure 1(c):

\[
E_{\pm}(k) = \pm \sqrt{\left| h_0(k_x,k_y) \right|^2 + \left| f_0(k_x,k_y) \right|^2}.
\]

The double \( \pm \) symbols label each of the four bulk bands, and the outer \( \pm \) signs explicitly indicate the electron–hole symmetry of the chiral bulk Hamiltonian.

An intriguing result from equations (26)–(28) is that the vector field \( d_0(k_x,k_y) \) (originally found for the bulk TCI Hamiltonian, and which turned out to be independent of \( k_z \)) remains unchanged even when the sample is one u.c. thick.

Indeed, the square of equation (10) is:

\[
(H_0(k_x,k_y))^2 = \begin{pmatrix} H^d_0(k_x,k_y) & 0 \\ 0 & H^b_0(k_x,k_y) \end{pmatrix},
\]

with

\[
H^d_0(k_x,k_y) = \left| H^d(k_x,k_y) \right|^2 + \left| f_0(k_x,k_y) \right|^2 \sigma_0 = d_{0z}(k_x,k_y) \sigma_0 + d_{0x}(k_x,k_y) \cdot \sigma,
\]

Explicit calculations yield:

\[
d_{0z}(k_x,k_y) = \left( 2h_0(k_x,k_y)h(k_x,k_y) \right),
\]

and:

\[
d_{0x}(k_x,k_y) = 2h_0(k_x,k_y)h(k_x,k_y),
\]
where $h_0(k_x, k_y)$ and $\mathbf{h}(k_x, k_y)$, as given by equation (28), suffer no single modification. This way, one concludes that:

$$d_i(k_x, k_y) = \mathbf{d}_i(k_x, k_y).$$

(35)

Given that the topology of a given electronic phase is encoded by its vector field, the topology of the square bulk Hamiltonian must be identical to the topology of the squared 2D Hamiltonian given by equation (10), which has never been identified with TCI behavior itself as far as we know.

In fact, it does not even matter what choice of u.c. one makes. Taking equation (11) now, one sees that

$$(H_i(k_x, k_y))^2 = \left( H_i^2(k_x, k_y) \right),$$

(36)

with

$$H_i^2(k_x, k_y) = \left( \mathbf{H}^A_i(k_x, k_y) \right)^2 + |f_i(k_x, k_y)|^2 \sigma_0 + d_i(k_x, k_y) \cdot \sigma.$$  

(37)

But

$$d_{0,i} = h_0(k_x, k_y)^2 + h_i(k_x, k_y)^2 + h_i(k_x, k_y)^2 + |f_i(k_x, k_y)|^2,$$

(38)

with $h_0(k_x, k_y)$, $h_i(k_x, k_y)$, and $h_i(k_x, k_y)$ still as given by equation (28), and

$$\mathbf{d}_i(k_x, k_y) = 2h_0(k_x, k_y)\mathbf{h}(k_x, k_y),$$

(39)

where $h_0(k_x, k_y)$ and $\mathbf{h}(k_x, k_y)$ remain as written down in equation (28) still, so that

$$\mathbf{d}_i(k_x, k_y) = \mathbf{d}_i(k_x, k_y) = \mathbf{d}_i(k_x, k_y).$$

(40)

In other words, the square of the TCI phase (equation (1)), and the squares of the two-dimensional Hamiltonians given by equations (10) and (11) encode the very same topological features, so that the topological characterization of the (bulk) $H_i^2(k)$ holds on identical at the single u.c. thickness limit.

We end this work by computing Pfaffians next. That last calculation will help re-emphasize the existence of a TCI phase in 2D.

### 7. Topological invariants from Pfaffians of the original TCI Hamiltonian in the bulk and 2D limits

As indicated before, midgap states in the entanglement spectrum can be employed to distinguish between topologically trivial and non-trivial phases of TCI [41–44]. On the other hand, Pfaffians are bona-fide tools for the determination of topology as well, and those will be employed in what follows.

The bulk Hamiltonian (equation (7)) takes the following generic form at high-symmetry points:

$$H_b = \begin{pmatrix} \Lambda & 0 & \lambda_b & 0 \\ 0 & \Lambda & 0 & \lambda_b \\ \lambda_b & 0 & -\Lambda & 0 \\ 0 & \lambda_b & 0 & -\Lambda \end{pmatrix},$$

(41)

Table 4. Parameters $\Lambda$, $\lambda_b$, and two-fold degenerate eigenvalues $E_{\pm} = \pm \sqrt{\Lambda^2 + \lambda^2_b}$ at $\Gamma$, $M$, $Z$ and $A$ k-points for the bulk Hamiltonian. The values of $\Lambda$ and $\lambda_b$ can be directly replaced into equations (49) and (50) at these high-symmetry $k$-points. All values are given in eV.

| $\Gamma$ | $M$ | $Z$ | $A$ |
|---|---|---|---|
| $\Lambda$ | $3$ | $-1$ | $3$ | $-1$ |
| $\lambda_b$ | $13/2$ | $5/2$ | $5/2$ | $-3/2$ |
| $E_{\pm}$ | $\pm 7.1589$ | $\pm 2.6926$ | $\pm 3.9051$ | $\pm 1.8028$ |

with real $\Lambda$ and $\lambda_b$ listed in table 4. The operator $H_b$ turns out to be the Hamiltonian of two non-interacting identical dimers. To see this, we use the operator $\mathcal{P}$

$$\mathcal{P} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}$$

(42)

to obtain:

$$H_b' = \mathcal{P} H_b \mathcal{P}^T = \begin{pmatrix} \Lambda & \lambda_b & 0 & 0 \\ \lambda_b & -\Lambda & 0 & 0 \\ 0 & 0 & \Lambda & \lambda_b \\ 0 & 0 & \lambda_b & -\Lambda \end{pmatrix}$$

(43)

which are the two subblocks argued for previously; the degeneracy at high-symmetry points is due to the $C_4$ in-plane symmetry of the model. The eigenvalues of $H_b'$ are doubly degenerate, and take on the following two values:

$$E_{\pm, b} = \pm \sqrt{\Lambda^2 + \lambda^2_b},$$

(44)

Slab Hamiltonians containing n u.c.s can also be permuted at the $\Gamma$ and $M$ points by a generalization of $\mathcal{P}$, leading to $n$ two-fold degenerate crossings at these high-symmetry points. (Such phenomenology can be most clearly observed on figures 3(c)–(g), in which the number of bands is small; see supporting materials.)

The two-dimensional Hamiltonian of a single $A$-sublattice slab gives the metallic eigenvalue spectrum shown in figure 3(h), in which quadratic dispersions with energy degeneracy at the $M$ point at $-1$ eV can still be seen with clarity. Its counterpart, the energy dispersion of the two-dimensional $B$-sublattice shown in figure 3(i), is the negative of the dispersion for $A$-sublattice atoms, due to the relative minus sign in its hopping terms with respect to the former sublattice (see definitions for hopping terms after equation (5)). At the sublattice limit, $H'$ still remains diagonal at the $\Gamma$ and $M$ points, and it yields two-fold degenerate eigenvalues:

$$H^A = \begin{pmatrix} \Lambda & 0 \\ 0 & \Lambda \end{pmatrix}$$

(45)

at sublattice $A$, and

$$H^B = \begin{pmatrix} -\Lambda & 0 \\ 0 & -\Lambda \end{pmatrix}$$

(46)
at sublattice B, with Λ still given in table 4. Such degeneracies, and the magnitudes of the degenerate eigenvalues are confirmed on figures 3(h) and (i). This is why these doubly-degenerate high-symmetry points never split in energy, despite of slab thickness, thus underscoring the topological protection.

The structure of the bulk Hamiltonian, equation (7), at the Γ, M, Z and Λ k-points maintains its form at the Λ and M points in the single u.c. limit. In the two possible 2D cases (short or long u.c.), one replaces Λ by either λ or Λ, whose magnitudes are listed in table 5.

Setting the label η—which can either be b, s, or l—to describe the bulk Hamiltonian or the short or long unit cell Hamiltonians at high symmetry points, one writes down the general expression:

\[ H_{\eta} = \begin{pmatrix} \Lambda & 0 & \lambda_{\eta} & 0 \\ 0 & \Lambda & 0 & \lambda_{\eta} \\ \lambda_{\eta} & 0 & -\Lambda & 0 \\ 0 & \lambda_{\eta} & 0 & -\Lambda \end{pmatrix}, \] (47)

with real Λ and λη listed in tables 4 and 5. The eigenvalues of \( H_{\eta} \) are doubly degenerate, and take on the following values:

\[ E_{\pm,\eta} = \pm \sqrt{\Lambda^2 + \lambda_{\eta}^2}, \] (48)

Only the two occupied (and degenerate) eigenvectors are needed to determine the topology of the Hamiltonian [48–51], and these have the following real entries:

\[ |v_1\rangle = \frac{1}{\sqrt{(\Lambda - E_{-\eta})^2 + \lambda_{\eta}^2}} \begin{pmatrix} -\lambda_{\eta} \\ 0 \\ \Lambda - E_{-\eta} \\ 0 \end{pmatrix}, \] (49)

\[ |v_2\rangle = \frac{1}{\sqrt{(\Lambda - E_{-\eta})^2 + \lambda_{\eta}^2}} \begin{pmatrix} 0 \\ -\lambda_{\eta} \\ 0 \\ \Lambda - E_{-\eta} \end{pmatrix}, \] (50)

whose general structure remains invariant in the bulk and u.c. limits. This realization will simplify the calculation of Pfaffians momentarily.

### 7.1. Pfaffians in 3D and 2D

Pfaffians are next explicitly computed to provide further credence to the u.c.-thick TCI.

First:

\[ A = i(\langle u_m | \nabla_k | u_m \rangle = i(\Xi_{u_m} | \nabla_k \Xi_{u_m}) \]

\[ = i(\langle u_m | \nabla_\perp \Xi_{u_m} | u_m \rangle = i(\langle u_m | \nabla_\perp \Xi_{u_m} | u_m \rangle. \] (51)

This way, \( A = -i(\langle u_m | \nabla_k | u_m \rangle = -A \), so \( A = 0 \) at these planes. In the 2D limit, \( k_z = 0 \) as well, making \( A = 0 \).

The reason for writing down the eigenvectors explicitly is that the sole replacement of the given magnitude of Λ, λη, and \( E_{\pm,\eta} \) guarantees that no extra phases are inserted into these vectors at the start of the open paths (one from Γ to M, and another from Z to A in the bulk case, and one from Λ to M in the single u.c. case). While one has freedom to choose the eigenvector’s phase at one high-symmetry point in the Brillouin zone, one must follow the state continuously along the open path to another high-symmetry point in order not to add undue discontinuities on the vector’s amplitudes [10]. Furthermore, a non-arbitrary sequencing of the valence band eigenvectors at the Λ, M, Z, and Λ (Γ and M) points in the bulk (1 u.c. thick slab) case becomes necessary so that no undesired permutation of degenerate states produces additional negative signs when computing the Pfaffian. To assign the proper ordering of states at high-symmetry points, the evolution of the Hamiltonian at points in k-space that are located infinitesimally away from the energy-degenerate high-symmetry points was first determined. This process is explicitly delineated to break degeneracies at the Γ and M points in the bulk TCI now.

To leading order in \( \delta \) at k-point \( (\delta, \delta, 0) \):

\[ \delta H_{\delta}(\Gamma) = \delta^2 \begin{pmatrix} -2 & 1 & -1 & 0 \\ 1 & -2 & 0 & -1 \\ -1 & 0 & 2 & -1 \\ 0 & -1 & -1 & 2 \end{pmatrix}. \] (52)

Similarly, at \( (\pi/a - \delta, \pi/a - \delta, 0) \) one gets:

\[ \delta H_{\delta}(M) = \delta^2 \begin{pmatrix} 0 & 1 & 1 & 0 \\ 1 & 0 & 0 & 1 \\ 1 & 0 & 0 & -1 \\ 0 & 1 & -1 & 0 \end{pmatrix}. \] (53)

Similar expressions can be found for \( \delta H_{\delta}(Z) \) and \( \delta H_{\delta}(A) \).

We solved for the eigenvalues and eigenvectors of \( H_{\delta}(\Gamma) + \delta H_{\delta}(\Gamma) \), \( H_{\delta}(M) + \delta H_{\delta}(M) \), \( H_{\delta}(Z) + \delta H_{\delta}(Z) \), and \( H_{\delta}(A) + \delta H_{\delta}(A) \) numerically, and the ordered sequence of eigenvalues and eigenvectors is shown in table 6. In addition, amplitudes of the eigenvectors that ensure the continuous evolution of the wavefunctions’ amplitude across the Brillouin zone are shown in figure 8(a) (Γ – M path) and figure 8(b) (Z – M path).

As indicated earlier on, the two-fold degeneracy persists at the 2D limit, requiring a process to order eigenstates as well. For the Hamiltonian for the short u.c. \( (\eta = s) \) at k-point \( (\delta, \delta) \), \( \delta H_{s}(\Gamma) \) is identical to the one given on equation (52), and similarly, \( \delta H_{s}(M) \) is identical to equation (53) in that 2D limit.
Table 6. Bulk eigenvalues at locations in reciprocal space slightly away from high-symmetry points, and eigenvectors with lifted degeneracies as $\delta \to 0$. Ordered vectors (according to the lifted degeneracies) are the ones employed to compute Pfaffians with a well defined sequence.

| $\Gamma + (\delta,\delta,0)$ | $M - (\delta,\delta,0)$ | $Z + (\delta,\delta,0)$ | $A - (\delta,\delta,0)$ |
|-----------------------------|-----------------------------|-----------------------------|-----------------------------|
| $-\sqrt{205-12487}/2$, $|u_1| = \frac{|u_1| + |u_2|}{\sqrt{2}}$ | $-\sqrt{292+2837}/2$, $|u_1| = \frac{|u_1| - |u_2|}{\sqrt{2}}$ | $-\sqrt{61+4437}/2$, $|u_1| = \frac{|u_1| - |u_2|}{\sqrt{2}}$ | $-\sqrt{13+445}/2$, $|u_1| = \frac{|u_1| + |u_2|}{\sqrt{2}}$ |
| $-\sqrt{205-12487}/2$, $|u_2| = \frac{|u_1| - |u_2|}{\sqrt{2}}$ | $-\sqrt{292+12487}/2$, $|u_2| = \frac{|u_1| + |u_2|}{\sqrt{2}}$ | $-\sqrt{61+9237}/2$, $|u_2| = \frac{|u_1| - |u_2|}{\sqrt{2}}$ | $-\sqrt{13-2837}/2$, $|u_2| = \frac{-1}{\sqrt{2}}$ |

Figure 8. Eigenvectors along the (a) $\Gamma - M$ and (b) $Z - A$ paths for the bulk TCI. (c) and (d) are eigenvectors along the $\bar{\Gamma} - M$ paths for the short and long unit cell slabs, respectively. Despite of small differences on amplitudes, the ordering of occupied states along the bulk $\Gamma - M$ path remains preserved along the $\bar{\Gamma}$ to $M$ path in the single u.c. limit.

Table 7. 1 u.c. eigenvalues (in eV) at locations in reciprocal space slightly away from high-symmetry points ($\delta = 0.01 \frac{\pi}{\sqrt{2}}$), and eigenvectors with lifted degeneracies as $\delta \to 0$. Vectors with lifted degeneracies are the ones employed to compute Pfaffians. Note that the ordered eigenvectors have an identical form to those listed for $k$-points $\Gamma$ and $M$ in table 6.

| $\bar{\Gamma} + (\delta,\delta); \eta = s$ | $M - (\delta,\delta); \eta = s$ | $\bar{\Gamma} + (\delta,\delta); \eta = l$ | $M - (\delta,\delta); \eta = l$ |
|-------------------------------|-------------------------------|-------------------------------|-------------------------------|
| $-5.40819$, $|u_1| = \frac{|u_1| + |u_2|}{\sqrt{2}}$ | $-1.11817$, $|u_1| = \frac{|u_1| - |u_2|}{\sqrt{2}}$ | $-3.60547$, $|u_1| = \frac{|u_1| - |u_2|}{\sqrt{2}}$ | $-2.23611$, $|u_1| = \frac{|u_1| - |u_2|}{\sqrt{2}}$ |
| $-5.40808$, $|u_2| = \frac{|u_1| - |u_2|}{\sqrt{2}}$ | $-1.11799$, $|u_2| = \frac{|u_1| + |u_2|}{\sqrt{2}}$ | $-3.60530$, $|u_2| = \frac{|u_1| - |u_2|}{\sqrt{2}}$ | $-2.23602$, $|u_2| = \frac{|u_1| - |u_2|}{\sqrt{2}}$ |

As for the long u.c. ($\eta = l$), one gets:

$$ \delta H_f(T) = \delta^2 \begin{pmatrix} -2 & 1 & 0 & 0 \\ 1 & -2 & 0 & 0 \\ 0 & 0 & 2 & -1 \\ 0 & 0 & -1 & 2 \end{pmatrix}, $$ \hspace{1cm} (54)

and

$$ \delta H_f(M) = \delta^2 \begin{pmatrix} 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 \\ 0 & 0 & -1 & 0 \end{pmatrix}, $$ \hspace{1cm} (55)

respectively.

We now endeavor to construct the matrix $w(k)$ whose entries are given by $w_j(k) = \langle u_j(k)|U|u_j(k)\rangle = \langle u_j(k)|U|u_j(k)\rangle$. The topological invariant is a property of the occupied bands, so $w(k)$ is a $2 \times 2$ matrix. We use the states listed in table 6 for $|u_i(k)\rangle$ for the bulk TCI. The following explicit matrices thus result,

$$ w(\Gamma) = \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}, \quad w(M) = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}, $$ \hspace{1cm} (56)

and

$$ w(Z) = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}, \quad w(A) = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}. $$ \hspace{1cm} (57)

The Pfaffian is equal to $w_{12}(k)$, yielding $(-1)^{\nu_{\bar{T}}} = Pf[w(\Gamma)]/Pf[w(M)] = -1$ and $(-1)^{\nu_{\bar{Z}}} = Pf[w(Z)]/Pf[w(A)] = 1$. The topological index is given by their product:

$$ (-1)^{\nu_{\bar{Z}}} = (-1)^{\nu_{\bar{T}}}(-1)^{\nu_{\bar{Z}}} = -1, $$ \hspace{1cm} (58)
and is nontrivial. Crucially for the argument that follows, 

\[ (-1)^{\nu s} = -1, \quad \text{while} \quad (-1)^{\nu M} = +1. \]

Table 7 and figures 8(c) and (d) contain the ordered eigenvalues at the \( \Gamma \) and \( M \) points for the short \( (\eta = s) \) and long \( (\eta = l) \) u.c. thick Hamiltonians; the eigenvector’s structure and ordering is identical to that seen on table 6 for the \( \Gamma \) and \( M \) points and on figure 8(a), which leads to \((-1)^{\nu M} = -1\). \( Z \) and \( A \) are not defined at the 2D limit, so that

\[ (-1)^{\nu s} = (-1)^{\nu M} = -1. \] (59)

Along with the discussion around figure 7 concerning the identical topology of \( H^2 \) in the 3D and 2D limit, equation (59) constitutes one of the main findings of the present work, in which we have thus proven that single u.c.-thick slabs are TCIs on their own merit.

8. Conclusion

We considered finite-thickness slabs of a TCI Hamiltonian. We showed by explicit slab calculations—in which all in-plane symmetries were removed—that the bulk-boundary correspondence does not hold on the TCI phase. Furthermore, we relied on the chiral nature of the model to create square Hamiltonians whose topological characterization in the bulk persisted in unit cell thick slabs lacking in-plane inversion symmetry due to their bipartite nature. The identical topological characterization is further confirmed by a calculation of topological invariants using the Pfaffians at the bulk and two-dimensional limits. The approaches followed here, as well as the discovery of the 2D TCI phase on the original model Hamiltonian, are novel results as far as we can tell.

Data availability statement

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

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