Second-order topology in Ta$_2$M$_3$Te$_5$ ($M = \text{Ni, Pd}$) monolayers

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Based on first-principles calculations, we propose that Ta$_2$M$_3$Te$_5$ ($M = \text{Pd, Ni}$) monolayers have second-order topology due to double-band inversion. Since inversion ($P$) and time-reversal ($T$) symmetries are preserved in the systems, the 2D Hamiltonian is real and its topology is classified by Stiefel-Whitney (SW) numbers ($w_1$, $w_2$). Using the Wilson loop method, we compute $w_1 = 0$ and $w_2 = 1$ for Ta$_2$Ni$_3$Te$_5$, indicating a second SW insulator. We conclude that Ta$_2$Ni$_3$Te$_5$ monolayer is a 2D second-order topological insulator (SOTI) with gapped edge states and localized corner states. By analyzing atomic band representations, we find that the monolayer is an unconventional insulator with an essential BR at an empty site, i.e., $A_3\oplus 4e$, which is consistent with the second-order topology of the second SW insulator. We demonstrate that its unconventional nature is due to the remarkable double-band inversion on $\Gamma$–$Y$. Although the double-band inversion always happens in the monolayers of $A_2M_{1,3}X_5$ family, the band gap varies form Ta$_2$Ni$_3$Te$_5$, to Ta$_2$Pd$_3$Te$_5$, to Nb$_2$Pd$_3$Te$_5$ monolayers, giving rise to exotic states, such as 2D SOTI, quantum spin Hall effect, excitonic insulator, and superconductivity. These layered compounds of $A_2M_{1,3}X_5$ family provide a good platform for realizing the 2D SOTI and exploring the interplay between topological order and interactions.

Introduction. In higher-order topological insulators (HOTI)\textsuperscript{1–12}, the in-gap states can be found in $(d - n)$-dimensional edges $(n > 1)$, such as the corner states of two-dimensional (2D) systems or the hinge states of three-dimensional systems. Different from topological insulators with $(d - 1)$-dimensional edge states, the Chern numbers or $Z_2$ numbers in HOTIs are zero. The higher-order topology can be captured by topological quantum chemistry\textsuperscript{13–15}, nested Wilson loop method\textsuperscript{1,8} and second Stiefel-Whitney class\textsuperscript{16–22}. Using topological quantum chemistry\textsuperscript{13}, the HOTI can be diagnosed by the decomposition of atomic band representations (aBRs) as an unconventional insulator with mismatching of electronic charge centers and atomic positions\textsuperscript{14,15,23}. Similar to dipoles (Berry phase) for topological insulators, the HOTI can be understood by multipole moments\textsuperscript{1}. In a 2D system, the second-order topology corresponds to the quadrupole moment, which can be diagnosed by the nested Wilson loop method\textsuperscript{1,8,20}. When the system contains space-time inversion symmetries, such as $PT$ and $C_2zT$, where the $P$ and $T$ represent inversion and time-reversal symmetries, the second-order topology can be described by the second SW number ($w_2$)\textsuperscript{20}. The second SW number $w_2$ is a well-defined 2D topological invariant of an insulator only when the first SW number $w_1 = 0$. Usually, a 2D SOTI with $w_1 = 0$, $w_2 = 1$ has gapped edge states and degenerate localized corner states, which are pinned at zero energy (being topological) in the presence of chiral symmetry. When the degenerate corner states are in the energy gap of bulk and edge states, the fractional corner charge can be maintained due to filling anomaly\textsuperscript{24}.

So far, various 2D systems are proposed to be SOTI or SW insulators, such as monolayer graphdiyne\textsuperscript{25,26}, liganded Xenes\textsuperscript{22,27}, β-Sb monolayer\textsuperscript{15,28} and Bi/ EuO\textsuperscript{29}. However, no transition-metal compound has been proposed to be a 2D SOTI. After considering many-body interactions, superconductivity, excitonic insulator and Luttinger liquid can be expected in the transition-metal SOTI. In recent years, van der Waals (vdW) layered materials of $A_2M_{1,3}X_5$ ($A=\text{Ta, Nb}$; $M = \text{Pd, Ni}$; $X = \text{Se, Te}$) family have attracted attentions because of their special properties, such as quantum spin Hall effect in Ta$_2$Pd$_3$Te$_5$ monolayer\textsuperscript{30,31}, excitons in Ta$_2$NiSe$_5$\textsuperscript{32–34}, and superconductivity in Nb$_2$Pd$_3$Te$_5$ and

![FIG. 1. (a) The crystal structure, Wyckoff positions and Brillouin zone (BZ) of Ta$_2$Ni$_3$Te$_5$ monolayer. (b) Band structure and irreps at Y and $\Gamma$ of Ta$_2$Ni$_3$Te$_5$ monolayer. (c) The 1D $k_x$-direct Wilson bands as a function of $k_x$ calculated in the DFT code. (d) Close-up of the green region in (c), with one crossing of Wilson bands at $\theta = \pi$ indicating the second SW class $w_2 = 1$.](cond-mat.soft/12_May_22/2205.05839v1)
doped Ta$_2$Pd$_3$Te$_5$\textsuperscript{35}. In particular, the monolayers of $A_2M_{1,3}X_5$ family can be exfoliated easily, serving as a good platform for studying topology and interactions in lower dimensions.

In this work, we predict that based on first-principles calculations, Ta$_2$Ni$_3$Te$_5$ monolayer is a 2D SOTI. Using the Wilson-loop method, we show that its SW numbers are $w_1 = 0$ and $w_2 = 1$, corresponding to a 2D SOTI. We also solve the aBR decomposition for Ta$_2$Ni$_3$Te$_5$ monolayer, and find that it is unconventional with an essential BR at an empty Wyckoff position (WKP), $A_g@4c$, which originates from the remarkable double-band inversion on $\Gamma$–Y line. To verify the SOTI phase, we compute the energy spectrum of Ta$_2$Ni$_3$Te$_5$ monolayer with open boundary conditions in both $x$ and $y$ directions and obtain four degenerate corner states. Additionally, the double-band-inversion picture widely happens in the band structures of Ta$_2$NiSe$_5$ and $A_2$Pd$_3$Te$_5$ monolayers. Ta$_2$Pd$_3$Te$_5$ monolayer being at phase boundary can be tuned to a SOTI with uniaxial strain or exciton condensation. In summary, the Ta$_2$M$_3$Te$_5$ monolayers are new 2D SOTI candidates for experimental realization.

**Band structures.** The space group of relaxed Ta$_2$Ni$_3$Te$_5$ monolayer is $Pmmn$ (No. 59). The symmetry operators are inversion, $C_{2x} = \{C_{2x}\{1/2, 0, 0\}$, $C_{2y} = \{C_{2y}\{0, 1/2, 0\}$ and $C_{2z} = \{C_{2z}\{1/2, 1/2, 0\}$. Although $P$ is weakly broken in the exfoliated monolayer from the bulk, it will be regained after relaxation. The change of the band structure is minute, as presented in Fig. S1(c-d) in Appendix A.

The band structure of Ta$_2$Ni$_3$Te$_5$ monolayer in Fig. 1(b) suggests that it is an insulator with a band gap of 65 meV. We have checked that spin-orbit coupling has a little effect. As shown in the orbital-resolved band structures of Figs. 3(a-c), although low-energy bands near the Fermi level ($E_F$) are mainly contributed by Ta-$d_{z^2}$ orbitals (two conduction bands) and Ni$_4$-$d_{xz}$ orbitals (two valence bands), the inverted bands of $\{Y2;GM1-,GM3-\}$ come from Te-$p_x$ orbitals. The irreducible representations (irreps) at $Y$ and $\Gamma$ are denoted for the inverted bands in Fig. 1(b). We notice that the double-band inversion between $\{Y2;GM1-,GM3-\}$ bands and $\{Y1;GM1+,GM3+\}$ bands is remarkable, about 1 eV, which will be discussed in detail below.

**Atomic band representations.** In a unit cell of Ta$_2$Ni$_3$Te$_5$ monolayer in Fig. 1(a), four Ta atoms, four Ni$_B$ atoms and eight Te atoms are located at 4c WKPs. The rest two Te atoms and two Ni$_A$ atoms are differently located at 2a and 2b WKPs respectively. They form 1D Ta$_2$Te$_5$ double chains. Unlike only $A$-type voids filled in Ta$_2$NiSe$_5$, both $A$- and $B$-types of tetrahedral voids are filled by Ni$_A$ and Ni$_B$ respectively in Ta$_2$Ni$_3$Te$_5$\textsuperscript{30} (Fig. 1(a)).

To analyze the band topology, the decomposition of aBR is performed. The aBRs are obtained from the crystal structure by pos2aBR, and irreps of occupied states are calculated by IRVSP at high-symmetry $k$-points. Then, the aBR decomposition is solved online –

\textbf{http://tm.iphy.ac.cn/UnconvMat.html}. The results are listed in Table S2 of Appendix B. Instead of being a sum of aBRs, we find that the aBR decomposition of the occupied bands has to include an essential BR at an empty WKP, i.e., $A_g@4c$. As illustrated in Fig. 1(a), the charge centers of the essential BR are located at the middle of Ni$_B$-Ni$_B$ bonds (i.e., the 4e WKP), indicating that the Ta$_2$Ni$_3$Te$_5$ monolayer is a 2D unconventional insulator with second-order topology.

**Double-band inversion.** In an ideal atomic limit, Te-$p$ orbitals and Ni-$d$ orbitals are occupied, while Ta-$d$ orbitals are fully unoccupied. Thus, all the occupied bands are supposed to be the aBRs of Te-$p$ and Ni-$d$ orbitals, as shown in the left panel of Fig. 2(a). However, in the monolayers of $A_2M_{1,3}X_5$ family (see their band structures in Appendix A), a double-band inversion happens between the occupied aBR $A''@4c$ (Te-$p_x$) and unoccupied aBR $A'@4c$ (Ta-$d_{z^2}$), as shown in the right two panels of Fig. 2(a). When the double-band inversion happens between $\{Y2;GM1-,GM3-\}$ and $\{Y1;GM1+,GM3+\}$ in Y–$\Gamma$ line, it results in a semimetal for Ta$_2$NiSe$_5$ monolayer (215-type; Fig. 2(b)). When it happens between $\{Y2;GM1-,GM3-\}$ and $\{Y1;GM1+,GM3+\}$ in Y–$\Gamma$ line, the system becomes a 2D SOTI for Ta$_2$Ni$_3$Te$_5$ monolayer (235-type), resulting in the essential BR of $A_g@4c$.

**Second Stiefel-Whitney class $w_2 = 1$.** To identify the second-order topology of the monolayers, we compute the second SW number by the Wilson-loop method. The first SW class ($w_1$) is,

$$w_1 \bigg| _C = \frac{1}{\pi} \int _C d\mathbf{k} \cdot \text{Tr} \mathbf{A} (\mathbf{k}) \quad (1)$$

![FIG. 2. (a) The diagram of double-band inversion along Y–$\Gamma$ line. The schematic band structures of (b) 215-type semimetal and (c) 235-type insulator. The double-band inversion happens in both cases. The crossing points on the $\Gamma$–X line are part of the $Z_2$ monopole nodal line.](image-url)
where $\mathcal{A}_{mn}(k) = \langle u_m(k) | \nabla_k | u_n(k) \rangle$\textsuperscript{19}. The second SW class ($w_2$) can be computed by the nested Wilson-loop method, or simply by $m$ module 2, where $m$ is the number of crossings of Wilson bands at $\theta = \pi$. It should be noted that $w_1 = 0$, $w_2$ can be unchanged when choosing the unit cell shifting a half lattice constant. The 1D Wilson-loops are computed along $k_y$. The computed phases of the eigenvalues of Wilson-loop matrices $W_y(k_y)$ (Wilson bands) are shown in Fig. 1(c) as a function of $k_y$. The results show that the first SW class is $w_1 = 0$. In addition, there is one crossing of Wilson bands at $\theta = \pi$ [Fig. 1(d)], indicating the second SW class $w_2 = 1$. It is also computed by the nested Wilson-loop method in Appendix C. Therefore, the Ta$_2$Ni$_3$Te$_5$ monolayer is a SOTI with a nontrivial second SW number.

**Edge spectrum and corner states.** From the orbital-resolved band structures (Fig. 3), the MLWFs of Ta-$d_{xz}$, Ni$_A$-$d_{xz}$ and Te-$p_x$ orbitals are extracted, to construct a 2D tight-binding (TB) model of Ta$_2$Ni$_3$Te$_5$ monolayer. As shown in Fig. 3(d), the obtained TB model fits the DFT band structure well. First, we compute the (01)-edge spectrum with open boundary condition along $y$. Instead of gapless edge states for a 2D Z$_2$-nontrivial insulator, gapped edge states are obtained for the 2D SOTI [Fig. 3(e)]. Then, we explore corner states as the hallmark of the 2D SOTI. We compute the energy spectrum for a nanodisk. For concreteness, we take a rectangular-shaped nanodisk with 50 $\times$ 10 unit cells, preserving both $M_x$ and $M_y$ symmetries in the 0D geometry. The obtained discrete spectrum for this nanodisk is plotted in the inset of Fig. 3(f). Remarkably, one observes four degenerate states near $E_F$. The spatial distribution of these four-fold modes can be visualized from their charge distribution, as shown in Fig. 3(f). Clearly, they are well localized at the four corners, corresponding to isolated corner states.

**Minimum model for the 2D SOTI.** As shown in Fig. 2, the minimum model for the 2D SOTI should be consisted of two BRs of $A'$@4e and $A''@4e$. Based on the situation of Ta$_2$Ni$_3$Te$_5$ monolayer in Fig. 2(c), the minimum effective model is derived as below,

$$
H_{TB}(k) = 
\begin{pmatrix}
H_{Ta}(k) & H_{hyb}(k) \\
H_{hyb}(k)^\dagger & H_{Ni}(k)
\end{pmatrix}
$$

(2)

The terms of $H_{Ta}(k)$, $H_{Ni}(k)$ and $H_{int}(k)$ are 4 $\times$ 4 matrices, which read

$$
H_{Ta}(k) = [\varepsilon_s + 2t_{s1}\cos(k_x)]\sigma_0\tau_0 + t_{s2}\gamma_1(k) + t_{s3}\sigma_0\tau_x, \\
H_{Ni}(k) = [\varepsilon_p + 2t_{p1}\cos(k_x)]\sigma_0\tau_0 + t_{p2}\gamma_2(k) + t_{p3}\sigma_0\tau_x, \\
H_{hyb}(k) = 2t_{sp}\sin(k_x)\gamma_3(k).
$$

(3)

The $\gamma_{1,2,3}(k)$ matrices are given explicitly in Appendix D. We find that $t_{s1} < 0$ and $t_{p1}, t_{p2} > 0$ for the $A_2M_{1,3}X_5$ family ($t_{s3}$ and $t_{p3}$ are small). When $\varepsilon_s + 2t_{s1} - 2|t_{s2}| < \varepsilon_p + 2t_{p1} + 2t_{p2}$, the double-band inversion happens in the monolayers of this family. By fitting the DFT bands, we obtain $t_{s2} > 0$ for the 215-type, while $t_{s2} < 0$ for the 235-type (Table 1).
Analytic solution of (01)-edge states As the remnants of the SOTI phase, the localized edge states can be solved analytically. For the (01)-edge, one can treat the full $H_{\text{TB}}(k)$ as two parts, $H_0(k)$ and $H'(k)$,

$$H_0(k) = \begin{pmatrix} t_{s2} \gamma_1(k) + t_{s3} \sigma_0 \tau_x & 0 \\ 0 & t_{p2} \gamma_2(k) + t_{s3} \sigma_0 \tau_x \end{pmatrix}$$

$$H'(k) = \begin{pmatrix} [\varepsilon_s + 2t_{s1} \cos(k_x)] \sigma_0 \tau_0 & \sigma_0 \gamma_b(k) \\ \sigma_0 \gamma_b(k)^\dagger & [\varepsilon_p + 2t_{p1} \cos(k_x)] \sigma_0 \tau_0 \end{pmatrix}$$

(4)

Without loss of generality, we take $t_{p2} = -t_{s2}$ and $t_{p3} = -t_{s3}$. Note that there is a pair of Dirac points $(\pm k^D_x, 0)$, with $k^D_x = \arccos \left( \frac{1}{2} \left( \frac{t_{33}}{t_{s2}} \right)^2 - 1 \right)$. Since $k_x$ is still a good quantum number on the (01)-edge, expanding $k_y$ to the second order, the zero-mode equation

$$H_0(k_x, -i\partial_y) \Psi(k_x, y) = 0$$

can be solved for $y \in [0, +\infty)$. Taking the trial solution of $\Psi(k_x, y) = \psi(k_x)e^{\lambda y}$, we obtain the secular equation and the solution of $\lambda = \pm \lambda_\pm$, where

$$\lambda_\pm = 1 \pm \sqrt{\frac{t_{s2}^2}{(1 + \cos(k_x))t_{s2}^2} - 1}$$

(5)

With the boundary conditions $\Psi(k_x, 0) = \Psi(k_x, +\infty) = 0$, only $-\lambda_+$ are permitted.

In the $k_x$ regime of $[-k^D_x, k^D_x]$, the edge zero-mode states are $\Psi(k_x, y) = [C_1(k_x)\phi_1(k_x) + C_2(k_x)\phi_2(k_x)]$.

$$\phi_1(k_x) = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 \\ -1 + e^{-ik_x} & t_{s2} & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}^T$$

$$\phi_2(k_x) = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 \end{pmatrix}^T$$

(6)

The edge zero states are Fermi arcs that linking the pair of projected Dirac points $(\pm k^D_x, 0)$. Once $H'(k)$ included, the effective edge Hamiltonian is,

$$H_{\text{edge}}^{\text{eff}} = \langle \Phi | H(k) | \Phi \rangle$$

$$= \begin{pmatrix} \varepsilon_s + 2t_{s1} \cos(k_x) & 0 \\ 0 & \varepsilon_p + 2t_{p1} \cos(k_x) \end{pmatrix}$$

(7)

where $|\Phi\rangle \equiv |\phi_1(k_x), \phi_2(k_x)\rangle$. Two edge spectra are obtained as the remnants of the 2D SOTI.

Discussion. In Ta$_2$NiSe$_5$ monolayer, the double-band inversion has also happened between Ta-$d_{z^2}$ and Te-$p_x$ states, about 0.4 eV, resulting in a semimetal with a pair of $Z_2$-monopole nodal lines in the 215-type$^{30}$. The highest valence bands (HVB) on Y-T are from the inverted Ta-$d_{z^2}$ states. However, in Ta$_2$Ni$_5$Te$_5$ monolayer, the double-band inversion strength becomes remarkable, $\sim$ 1 eV, which is ascribed to the filled $B$-type voids and more extended Te-$p_z$ states. On the other hand, the HVB becomes Ni$_{1-d_{z^2}}$ states (slightly hybridized with Te-$p_x$ states). It is insulating with a small gap of 65 meV. When it comes to A$_2$Pd$_3$Te$_5$ monolayer, the remarkable inversion strength is similar to that of Ta$_2$Ni$_5$Te$_5$. But the HVB of Pd$_{1-d_{z^2}}$ states go upwards further due to more expansion of the Se orbitals and the energy gap becomes almost zero in Ta$_2$Pd$_3$Te$_5$. In short, the double-band inversion always happens in these monolayers, while the band gap of the 235-type changes from positive (Ta$_2$Ni$_5$Te$_5$), to nearly zero (Ta$_2$Pd$_3$Te$_5$) with a tiny band overlap, to negative (Nb$_2$Pd$_3$Te$_5$), as shown in Fig. S1. Since their bulk materials are vdW layered compounds, the bulk topology and properties strongly rely on the band structures of the monolayers in the A$_2$M$_{1,3}X_5$ family.

As we find in Ref. $^{30}$, the band topology of Ta$_2$Pd$_3$Te$_5$ monolayer is lattice sensitive, although the intrinsic monolayer is predicted to be a quantum spin Hall insulator upon including spin-orbit coupling (SOC). By applying $> 1\%$ uniaxial compressive strain along $b$, it becomes a $Z_2$-trivial insulator, being a 2D SOTI. One the other hand, due to the quasi-1D crystal structure, the screening effect of carriers is relatively weak and the electron-hole Coulomb interaction is substantial for exciton condensation$^{37,38}$, resulting in an exciton insulator. Since the second-order topology is due to the remarkable double-band inversion and protected by the second SW number, this excitonic insulating phase could still share the second-order topology, as long as $C_{2,T}$ or $PT$ symmetry is still preserved, possessing gapped edge states. With the large double-band inversion (HOTI topology) and small band energy gap/overlap ($Z_2$ topology$^{30}$ with SOC), these vdW layered materials provide a good platform to study the interplay between the topology and interactions (Fig. 4), which can be easily manipulated by external magnetic and electric fields. The coexistence of topological and excitonic order could generate topological excitons in the systems$^{39}$. Additionally, it is easy to

| TABLE I. Parameters of the TB model for the 2D SOTI. |
|---|---|---|---|---|---|---|---|
| $\varepsilon_s$ | $t_{s1}$ | $t_{s2}$ | $t_{s3}$ | $t_{p1}$ | $t_{p2}$ | $t_{p3}$ |
| 1.05 | -0.6 | -0.11 | -0.02 | 0.3 | -2.8 | 0.8 | 1.0 | 0.2 |
mechanically exfoliate into large flakes with long straight edges along the 1D chain due to the strong anisotropy of bonds. The in-gap edge states as remnants of the SOTI phase are responsible for the observed Luttinger-liquid behavior. According to the above discussion, we summarize the diverse properties of $A_2M_{1,3}X_5$ family in Fig. 4.

In conclusion, we predict that $\text{Ta}_2M_3\text{Te}_5$ monolayers can be SOTIs by solving aBR decomposition and computing SW numbers. Through aBR analysis, we conclude that the higher-order topology comes from an essential BR at an empty site ($A_2^g@A_2^c$), and it originates from the remarkable double-band inversion. The double-band inversion also happens in the band structure of the $\text{Ta}_3\text{NiSe}_5$ monolayer. The second SW number of $\text{Ta}_2\text{Ni}_3\text{Te}_5$ monolayer is $w_2 = 1$, corresponding to a second-order topology in 2D. Therefore, we obtain edge states and corner states of the monolayer. Due to the weak screening effect of the quasi-1D crystals, these vdW layered compounds of $A_2M_{1,3}X_5$ family provide a good platform for exploring the interplay between topological order and interactions in the future.

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APPENDIX

A. The band structures of the monolayers of A$_2$M$_{1.3}$X$_5$ family

Our first-principles calculations were performed within the framework of the density functional theory (DFT) using the projector augmented wave (PAW) method$^{11,12}$, as implemented in Vienna $ab$-initio simulation package (VASP)$^{13,14}$. The Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation exchange-correlations functional$^{15}$ was used. Spin-orbit coupling is neglected in the calculations. In the self-consistent process, 16 $\times$ 4 $\times$ 1 $k$-point sampling grids were used, and the cut-off energy for plane wave expansion was 500 eV. The irreducible representations (irreps) were obtained by the program IRVSP$^{46}$. The maximally localized Wannier functions (ML-WFs) were constructed by using the Wannier90 package$^{17}$. The edge spectra are calculated using surface Green’s function of semi-infinite system$^{18,49}$.

There are two phases of bulk Ta$_2$NiTe$_5$, which are C2/c (No. 15, low-temperature phase) and Cmcm (No. 63, high-temperature phase). As shown in Fig. S1(a-b), both of the exfoliated Ta$_2$NiTe$_5$ monolayers are semimetal, and compatibility relationship are consistent with band inversion process in Fig. 2(a). The space group of bulk Ta$_2$M$_3$Te$_5$($M$ = Ni, Pd) is Pnma (No. 62), and space group of the corresponding exfoliated monolayer is Pmn2$_1$ (No. 31). However, the relaxed Ta$_2$Ni$_3$Te$_5$ monolayer structure has inversion symmetry, and the corresponding space group becomes Pnma (No. 59). Thus, the inversion was imposed into Ta$_2$M$_3$Te$_5$ monolayers with tiny atom positions movements, and the corresponding band structures are also calculated, as shown in Fig. S1(c-h). In addition, with inversion symmetry, the irreps can be distinguished by parity and the aBR decomposition can be done easily.

![Image of band structures](image)

**FIG. S1.** The comparisons of band structures of (a-b) Ta$_2$NiSe$_5$, (c-d) Ta$_2$Ni$_3$Te$_5$, (e-f) Ta$_2$Pd$_3$Te$_5$, (g-h) Nb$_2$Pd$_3$Te$_5$ monolayers. The lower panels share the same space group #59. The 235-type band gap ($E_g = E_{GM4-} - E_{GM4+}$) changes for 0.068 eV for Ta$_2$Ni$_3$Te$_5$, to -0.023 eV for Ta$_2$Pd$_3$Te$_5$, to -0.124 eV for Nb$_2$Pd$_3$Te$_5$ monolayers.

B. The aBR decomposition of Ta$_2$Ni$_3$Te$_5$

As shown in Table. S1, we find that $A'^@4c + A''@4c = A g@4c + A u@4c$ in aBRs for Pmnn space group. The WKPs, aBRs and the aBR decomposition of Ta$_2$Ni$_3$Te$_5$ monolayer are given in Table. S2.

**TABLE S1.** The aBRs for Pmnn space group at 4e and 4c WKPs, we can find $A'^@4c + A''@4c = A g@4c + A u@4c$.

| | $A'^@4c$ | $A''@4c$ | $A g@4c$ | $A u@4c$ |
|---|---|---|---|---|
| $\Gamma$ | $\Gamma_1 \oplus \Gamma_2 \oplus \Gamma_4^+ \oplus \Gamma_4^-$ | $\Gamma_1^- \oplus \Gamma_2^- \oplus \Gamma_3^- \oplus \Gamma_4^+$ | $\Gamma_1^+ \oplus \Gamma_2^+ \oplus \Gamma_3^+ \oplus \Gamma_4^+$ | $\Gamma_1^- \oplus \Gamma_2^- \oplus \Gamma_3^- \oplus \Gamma_4^-$ |
| $S$ | $S_1 \oplus S_2$ | $S_1 \oplus S_2$ | $S_1 \oplus S_2$ | $S_1 \oplus S_2$ |
| $X$ | $X_1 \oplus X_2$ | $X_1 \oplus X_2$ | $X_1 \oplus X_2$ | $X_1 \oplus X_2$ |
| $Y$ | $2Y_1$ | $2Y_2$ | $Y_1 \oplus Y_2$ | $Y_1 \oplus Y_2$ |
with classified using the SW class instead of the Chern class. And the second SW class can be determined by a $Z_w$ systems with $C_k$. Diagonalizing thus the final Berry phase of nested Wilson loop is, 

$$W_{k_x} = \prod_{j=0}^{N-1} M_{y}(k_x, k_y^{j+1}).$$

Diagonalizing $W_{y}(k_x)$, we can get the eigenvalues $W_{y}(k_x) = e^{i\theta_l(k_x)} (l = 1, 2, ..., n_{occ})$ and the corresponding eigenvectors $|w_{y}(k_x)\rangle$. The phase is also called the Wannier charge center (WCC). In systems with $PT$ symmetry, the Hamiltonian can be transformed to be real. In this condition, the system can be classified using the SW class instead of the Chern class. And the second SW class can be determined by a $Z_2$ number $w_2$, which is corresponding to the number of cross points at $\theta = \pi$ modulo 2. When calculating nested Wilson loop, we define $M^{mn}_{xy}(k_x, k_x^{j+1}) = \langle w_{my}(k_x^{j})|w_{ny}(k_x^{j+1})\rangle (k_x^{j} = 2\pi j/N; m, n = 1, 2, ..., n')$. Similar to the Wilson-loop method,

$$W_{xy} = \prod_{j=0}^{N-1} M_{xy}(k_x^{j}, k_x^{j+1}).$$

Thus the final Berry phase of nested Wilson loop is,

$$\Phi = -\text{Im} \ln |\det(W_{xy})|.$$
D. The Effective model for a 2D SOTI

With unit cell defined in Fig. S2(a), basis chosen as
\[
(|k, \text{Ta}_1, d_{z^2}) , |k, \text{Ta}_2, d_{z^2}) , |k, \text{Ta}_3, d_{z^2}) , |k, \text{Ta}_4, d_{z^2}) ,
|k, \text{Ni}_1, d_{xz}) , |k, \text{Ni}_2, d_{xz}) , |k, \text{Ni}_3, d_{xz}) , |k, \text{Ni}_4, d_{xz}) ,
\]
\[(13)\]
hoppings considered listed in Fig. S2(b-h), and being Fourier transformed with atomic position \(\tau\) excluded \(\text{i.e., lattice gauge; denoted as } H(k)\), the minimum effective eight-band tight-binding Hamiltonian of \(\text{Ta}_2\text{Ni}_3\text{Te}_5\) are given as Eq. (3) in the main text, with \(\gamma\)-matrices defined as
\[
\begin{align*}
\gamma_1(k) &= \begin{pmatrix}
0 & 0 & 0 & e^{-ik_y} (1 + e^{-ik_x}) \\
0 & 0 & 1 + e^{ik_x} & 0 \\
e^{ik_y} (1 + e^{ik_x}) & 0 & 0 & 0
\end{pmatrix} \\
\gamma_2(k) &= \begin{pmatrix}
0 & 0 & 0 & e^{ik_y} \\
0 & 0 & e^{-ik_x} & 0 \\
0 & e^{-ik_y} (1 + e^{ik_x}) & 0 & 0 \\
1 + e^{ik_x} & 0 & 0 & 0
\end{pmatrix} \\
\gamma_3(k) &= \begin{pmatrix}
0 & e^{-ik_y} & 0 & 0 \\
1 & 0 & 0 & 0 \\
0 & 0 & e^{ik_y} & 0 \\
0 & 0 & 0 & 1
\end{pmatrix}
\end{align*}
\[(14a)\] \[(14b)\] \[(14c)\]

Therefore, the full tight-binding (TB) Hamiltonian is
\[
H_{\text{TB}}(k) = \begin{pmatrix} H_{\text{Ta}}(k) & H_{\text{hyb}}(k) \\ H_{\text{hyb}}(k)^\dagger & H_{\text{Ni}}(k) \end{pmatrix}
\]
\[
(15)
\]

FIG. S2. (a) Unit cell of minimum effective model of \(\text{Ta}_2\text{Ni}_3\text{Te}_5\). (b)-(h) Hoppings considered in effective model. The hoppings are represented by bonds between Ta and Ni atoms. And the plus and minus represent the positive and negative values of hoppings.