A Charge-Density-Wave Weyl Semimetal

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Employing both theoretical and experimental techniques, we discover that the paradigmatic quasi-one-dimensional material (TaSe₄)₂I, which at room temperature respects the body-centered tetragonal, chiral space group (SG) 97 (I422), is actually a Weyl semimetal whose Weyl points become gapped by a charge-density wave (CDW). Using first-principles calculations, we determine that the Fermi surface of (TaSe₄)₂I is dominated by topological contributions from bulk Weyl points, which appear near the kₚ = ±π/c planes and lie within a narrow energy range of the Fermi level (E_F) −10 meV < (E − E_F) < 15 meV. We find that the low-energy electronic structure of (TaSe₄)₂I is characterized by one set of 16 symmetry-related C = ±1 Weyl points below E_F, and four additional sets of 8 C = ±1 Weyl points above E_F. (TaSe₄)₂I has previously been highlighted for exhibiting a characteristic CDW. To elucidate the origin of the CDW, we use first principles to calculate the electronic susceptibility, whose real part diverges at several high-density spots that correspond to Fermi-surface “nesting” vectors that link Weyl points with opposite chiral charges. We then use X-ray diffraction (XRD) and ARPES probes on (TaSe₄)₂I samples to experimentally confirm the CDW phase. In our XRD experiments, we observe the onset of satellite reflections at T_C = 248 K, which is in close agreement with the value of T_C = 260 K measured in previous works. The satellite reflections appear at q = [mη(π/a), nη(π/a), oδ(π/c)], where η = 0.027 ± 0.001, δ = 0.012 ± 0.001, and m + n + o ∈ 2Z, implying that the CDW-modulated structure, like the unmodulated parent structure of (TaSe₄)₂I, is also body-centered tetragonal. We find that the experimentally observed satellite reflections exhibit close agreement with the high-density spots (Weyl-point nesting vectors) in the electronic susceptibility obtained from first principles. Calculating the surface states through surface Green’s functions, we observe pairs of well-isolated topological Fermi arcs on both the TaSe₄-chain and I-atom terminations of the conventional-cell (110)-surface. We then calculate the quasiparticle interference patterns of the Fermi arcs. Because crystals in SG 97 are structurally chiral (i.e., they lack improper rotation symmetries, such as inversion or mirror), then (TaSe₄)₂I in its high-temperature phase is also a strong candidate for the experimental observation of the quantized circular photogalvanic and chiral magnetic effects. Furthermore because its CDW couples momentum-separated Weyl points with opposite chiral charges, then (TaSe₄)₂I in its low-temperature phase provides a venue for the examination of topological insulating phases with strong interactions, such as axion insulators.

INTRODUCTION

The discovery of graphene [1] and the recognition of its band structure’s relationship with topology [2], has ignited intense interest in searching for additional topological semimetals. In just a short time, this search has yielded a tremendous diversity of topological and crystalline semimetallic phenomena [3–13]. Among these, topological semimetals, whose nodal features range from conventional band-inversion Weyl fermions [14–24] to more exotic double-Weyl [25, 26], unconventional [12, 27–30], and higher-order [31–33] variants, have shown particular promise for the observation of novel topological physics in gapless systems. In conventional Weyl semimetals, such as TaAs [16–18] and the tilted type-II variants WTe₂ [21] and MoTe₂ [19, 20, 22], bands at the Fermi energy meet at generic points in the Brillouin zone (BZ) in twofold, linear degeneracies that are monopoles of Berry curvature with quantized Chern numbers |C| = 1, and therefore represent condensed-matter realizations of Weyl fermions [14]. In the presence of additional crystal symmetries, this degeneracy and dispersion can be enhanced, resulting in topological chiral fermions with
larger Chern numbers, such as the double-Weyl fermions in SrSi$_2$ [26] and the modified spin-3/2 and double-spin-1 unconventional chiral fermions in the RhSi family [12, 28–30]. All of these topologically chiral semimetals share a common bulk-boundary correspondence: the surface projections of opposite chiral fermions are connected by topological surface Fermi arcs [14]. These surface Fermi arcs have emerged as the primary experimental means of confirming the topology of bulk chiral fermions in semimetallic compounds, and their signatures have been observed in angle-resolved photoemission spectroscopy (ARPES) experiments [15–22, 34–38] and scanning tunneling microscopy probes of quasiparticle interference (QPI) [39–41].

Particularly great interest has been devoted to the theoretical prediction and experimental discovery of bulk signatures of gapless topological phases. Such proposed signatures include negative magnetoresistance [18, 42], the intrinsic spin Hall effect [43], and the anomalous Hall effect [25, 44–46]. Recently, additional signatures have also been proposed in the subset of topologically chiral semimetals for which chiral fermions with opposite Chern numbers lie at different energies; these effects include the chiral magnetic effect (CME) [47–52] and the quantized circular photoanalogic effect (CPGE) [53, 54]. For nonmagnetic compounds, namely those with time-reversal (T) symmetry, this energy offset can only be realized in the absence of bulk improper rotation symmetries, i.e., in structurally chiral crystals [28]. Exploiting this result, the CPGE has been predicted in the structurally chiral Ag$_2$Se, TiTeO$_6$, and RhSi families [28, 29, 54, 55], and was recently observed in RhSi [56]. Finally, signatures of the long-sought “axion insulator” [57] as well as other, correlated topological phases have been observed in chiral semimetals for which pairs of chiral fermions have become coupled by (typically incommensurate) charge- or spin-density waves (CDWs and SDWs, respectively) [58–61].

In this work, we employ both first-principles calculations and experimental probes to predict that quasi-one-dimensional (1D) crystals in the chain structure of (TaSe$_4$)$_2$I [62–64] are in fact Weyl semimetals whose Weyl points (WPs) become coupled by the onset of a CDW. Though (TaSe$_4$)$_2$I has previously been highlighted for exhibiting linear crossings near the Fermi energy ($E_F$) [65] and Kramers-Weyl fermions far below $E_F$ [28], our work represents the first recognition that (TaSe$_4$)$_2$I hosts topological chiral fermions (i.e. WPs) at $E_F$, and the first reported link between the bulk WPs and the CDW wavevector. Furthermore, because (TaSe$_4$)$_2$I crystallizes in chiral space group (SG) 97 (I422), it hosts WPs of opposite topological chirality at different energies [28, 29, 53], and therefore provides a promising platform for the observation of the CME and the CPGE. Specifically, in (TaSe$_4$)$_2$I, the entire Fermi surface is formed from the cones of 48 WPs, which we designate as the “Fermi-surface WPs” (FSWPs). The 48 FSWPs lie within 15 meV of $E_F$: 16 $C = +1$ FSWPs lie $\sim 9$ meV below $E_F$, and the remaining 32 lie above. The net +16 chiral charge of the WPs below $E_F$ is by far the largest value predicted to date in a real material. Additionally, previous experiments have shown that (TaSe$_4$)$_2$I transitions into an incommensurate CDW phase when cooled just below room temperature [66, 67]. Using first-principles calculations, we compute the high-density electronic susceptibility $\chi$ vectors, which we determine to largely coincide with the nesting vectors between FSWPs with both the same and opposite chiral charges [see Section H of the Supplementary Material (SM H)]. We then perform X-ray diffraction (XRD) and angle-resolved photoemission spectroscopy (ARPES) experiments on (TaSe$_4$)$_2$I samples to determine the gap and modulation vectors of the CDW phase (see SM I), which we find to be in good agreement with the electronic susceptibility peaks originating from nesting vectors between FSWPs with opposite chiral charges. Taken together, our results strongly suggest that (TaSe$_4$)$_2$I is the first known material candidate for a Weyl-semimetal-derived axion insulator.

RESULTS AND DISCUSSION

Structure. (TaSe$_4$)$_2$I (Inorganic Crystal Structure Database [70] No. 35190, further details available at https://topologicalquantumchemistry.org/#/detail/35190 [13, 71–73]) crystallizes in a quasi-1D, body-centered tetragonal chiral structure in SG 97 (I422) with the reciprocal lattice vectors:

$$\vec{k}_1 = \left( \frac{2\pi}{a}, \frac{2\pi}{c}, 0 \right), \ \vec{k}_2 = \left( \frac{2\pi}{a}, 0, \frac{2\pi}{c} \right), \ \vec{k}_3 = \left( \frac{2\pi}{a}, \frac{2\pi}{c}, 0 \right),$$

where $a = 9.59$ Å and $c = 12.64$ Å are the conventional-cell lattice constants [Ref. [62] and Fig. 1(a,b)]. The conventional cell of (TaSe$_4$)$_2$I contains two TaSe$_4$ chains aligned along the $c$-axis and four iodine atoms separating the chains [Fig. 1(a,b)]. Each chain is formed of four alternating layers of Ta atoms and rectangles with four Se atoms on each corner, for a total of 4 Ta atoms and 16 Se atoms per chain. Within the conventional cell, there are two chains, implying a total chemical formula of (TaSe$_4$)$_2$I per conventional cell [Fig. 1(a,b)]. Because the volume of the primitive (unit) cell in body-centered SG 97 (I422) is half of that of the conventional cell, then the primitive cell of (TaSe$_4$)$_2$I only contains half of the atoms in the conventional cell shown in Fig. 1(a,b) [i.e. the total chemical formula is (TaSe$_4$)$_2$I$_2$ per primitive cell]. Within each TaSe$_4$ chain, the Se rectangles and Ta atoms deviate only slightly from a regular eightfold screw arrangement along the $c$-axis in which consecutive Se rectangles (and Ta
panel of (d), moving along Γ−Z in SM B. Further below E and chiral charges of all 48 FSWPs are provided in Table I, and characteristic band dispersions for each FSWP are comprised the entire Fermi surface, we designate them the “Fermi-surface WPs” (FSWPs). The momentum-space coordinates −4 because T−E at the Fermi energy (E<0). Because (TaSe4)2I crystallizes in the body-centered tetragonal space group 97 (I422) [62], then its primitive cell contains half as many atoms as the conventional cell shown in (a,b) [SM J]. (c) The bulk BZ and its projections onto the conventional-cell (001)- and (110)-surfaces. Throughout this work, all high-symmetry points (HSPs) in the bulk BZ are expressed in terms of the conventional-cell reciprocal lattice vectors (a,a,c) [alternatively, the primitive-cell reciprocal lattice vectors are given in Eq. (1)]. In the units of (kx,ky,kz), the HSPs in (c) and their coordinates are respectively given by: Γ (0,0,0), Z (0,0,1), P (1/2,1/2,1/2), N (1/2,0,1/2), and X (1/2,1/2,0). (d) Electronic band structure of (TaSe4)2I with spin-orbit coupling. Bands along ΓZ cross at the Fermi energy (E_F) near the halfway points k_z = ±π/c – a remnant of a filling-enforced nodal plane [68, 69] that is present in the band structure of a crystal of decoupled TaSe₄ chains (SM J). Away from the high-symmetry BZ lines, there are 48 Weyl points (WPs) lying within the energy range −10 meV < (E−E_F) < 15 meV; because the cones of these 48 WPs comprise the entire Fermi surface, we designate them the “Fermi-surface WPs” (FSWPs). The momentum-space coordinates and chiral charges of all 48 FSWPs are provided in Table I, and characteristic band dispersions for each FSWP are provided in SM B. Further below E_F, there are also eight C₄ᵥ-symmetric chiral fermions (WPs) along ΓZ. As shown in the left inset panel of (d), moving along ΓZ in increasing k_z, four of the eight C₄ᵥ-symmetric WPs exhibit the compensating chiral charges −2, +1, −1, and +2 [their time-reversal (T) partners along ΓZ with negative values of k_z also exhibit the same charges, because T does not invert the chiral charge of a WP [14, 28]]. In the right inset panel, we show that bands along PN are gapped at E_F. (e) Band dispersion along two FSWPs, located respectively at K₁ = (−0.283180, 0.469691, 0.510336) (W⁺₁) and K₂ = (0.306427, 0.000000, 0.560108) (W⁻₁). Bands from the two FSWPs in (e) are separated by a large nontrivial energy window of 0.123 eV, or on the order of 1400 K.

FIG. 1. (Color online) The crystal structure, 3D bulk and 2D surface Brillouin zones (BZs), and electronic structure of (TaSe₄)₂I. The crystal structure is shown from both (a) top [(001)] and (b) tilted side perspectives, where throughout this work, surface terminations are labeled using the conventional-cell lattice vectors (a,a,c). Because (TaSe₄)₂I crystallizes in the body-centered tetragonal space group 97 (I422) [62], then its primitive cell contains half as many atoms as the conventional cell shown in (a,b) [SM J]. (c) The bulk BZ and its projections onto the conventional-cell (001)- and (110)-surfaces. Throughout this work, all high-symmetry points (HSPs) in the bulk BZ are expressed in terms of the conventional-cell reciprocal lattice vectors (a,a,c) [alternatively, the primitive-cell reciprocal lattice vectors are given in Eq. (1)]. In the units of (kx,ky,kz), the HSPs in (c) and their coordinates are respectively given by: Γ (0,0,0), Z (0,0,1), P (1/2,1/2,1/2), N (1/2,0,1/2), and X (1/2,1/2,0). (d) Electronic band structure of (TaSe₄)₂I with spin-orbit coupling. Bands along ΓZ cross at the Fermi energy (E_F) near the halfway points k_z = ±π/c – a remnant of a filling-enforced nodal plane [68, 69] that is present in the band structure of a crystal of decoupled TaSe₄ chains (SM J). Away from the high-symmetry BZ lines, there are 48 Weyl points (WPs) lying within the energy range −10 meV < (E−E_F) < 15 meV; because the cones of these 48 WPs comprise the entire Fermi surface, we designate them the “Fermi-surface WPs” (FSWPs). The momentum-space coordinates and chiral charges of all 48 FSWPs are provided in Table I, and characteristic band dispersions for each FSWP are provided in SM B. Further below E_F, there are also eight C₄ᵥ-symmetric chiral fermions (WPs) along ΓZ. As shown in the left inset panel of (d), moving along ΓZ in increasing k_z, four of the eight C₄ᵥ-symmetric WPs exhibit the compensating chiral charges −2, +1, −1, and +2 [their time-reversal (T) partners along ΓZ with negative values of k_z also exhibit the same charges, because T does not invert the chiral charge of a WP [14, 28]]. In the right inset panel, we show that bands along PN are gapped at E_F. (e) Band dispersion along two FSWPs, located respectively at K₁ = (−0.283180, 0.469691, 0.510336) (W⁺₁) and K₂ = (0.306427, 0.000000, 0.560108) (W⁻₁). Bands from the two FSWPs in (e) are separated by a large nontrivial energy window of 0.123 eV, or on the order of 1400 K.

atoms) are related by 45° turns and c/4 translation in the z direction. [Fig. 1(b)]. The nearly-preserved eightfold screw symmetry reflects that an isolated TaSe₄ chain is invariant under the non-crystallographic chiral rod group (p3122)RG [Refs. [74–77] and SM J]. Because the rectangle of Se atoms within each layer lies far from the limit of a perfect square, then the nearly-preserved eightfold screw symmetry generates a sixteen-fold pattern of Se atoms when each chain is viewed along the c axis, as shown in Fig. 1(a) [if the Se rectangles were deformed to squares, then the Se atoms would only form an eightfold pattern]. SG 97 is generated by body-centered tetragonal lattice translations and the proper rotation symmetries C₄ᵥ and C₂x, where C₄ᵥ is a rotation by 360°/n about the i-axis. Because SG 97 does not contain improper rotation symmetries, it characterizes crystals with structural handedness [28]. As the monopole chiral charges (Chern numbers) of chiral fermions are left invariant under proper rotations and T [23], WPs with opposite charges are free to lie at different energies in (TaSe₄)₂I. Though this effect has been predicted in Kramers-Weyl [27, 28]
correspondingly observe weak dispersion along PN high-symmetry lines in the first BZ [Fig. 1(c)]. We show the band structure of (TaSe$_4$)$_2$I in Fig. 1(d), which represents the “enforced” semimetallic crossings along ΓZ predicted from the Topological Quantum Chemistry and band connectivity [13, 79–82] analyses of the electronic structure of (TaSe$_4$)$_2$I performed in [71, 83, 84]. However, because the C$_{4z}$-symmetric chiral fermions below $E_F$ represent the “enforced” semimetallic crossings along ΓZ, we observe 48 WPs within 15 meV of $E_F$, which we designate the “Fermi-surface WPs” (FSWPs), as well eight C$_4$-symmetric chiral fermions (two pairs of conventional WPs and two pairs of double-Weyl points [25, 26]) along ΓZ lying between 16 – 20 meV below $E_F$ [shown in the left inset panel of Fig. 1(d)]. The C$_{4z}$-symmetric chiral fermions below $E_F$ are only weakly separated [Δ$k_z ∼ 0.002(2π/c)$] and carry a net-zero chiral charge within each narrow grouping, then they are not likely to contribute experimentally detectable Fermi-arc surface states. Additionally, because all eight enforced chiral fermions are fully occupied and carry compensating chiral charges, then they do not contribute to topological response probes, such as the CPGE [53, 54]. Therefore, we will neglect the C$_{4z}$-enforced WPs below $E_F$ in further discussions of the chiral fermions (WPs) in (TaSe$_4$)$_2$I.

In Table I, we list the coordinates and multiplicities of the 48 FSWPs. Under the SG 97 (I422) symmetries C$_{4z}$, C$_{2x(2y)}$, C$_{2z+}(x−y)$, and T, a WP at a generic crystal momentum $(k_x, k_y, k_z)$ is related to 15 additional WPs with the same Chern number, for a total of 16 WPs located at $(±k_x, ±k_y, ±k_z)$ and $(±k_y, ±k_z, ±k_z)$ in the reciprocal coordinates of the conventional-cell reciprocal lattice vectors. Using the notation employed by the KVEC tool on the Bilbao Crystallographic Server [72, 73], this can be summarized with the statement that WPs at generic crystal momenta occupy the 160 reciprocal-space Wyckoff position (i.e., lie within a multiplicity-16 “star”) in SG 97. In the remainder of this section, we will use crystal symmetries to derive the momentum-space coordinates and multiplicities of the symmetry-related sets of FSWPs, which we will then label with the reciprocal-space Wyckoff positions listed

| WPs Multiplicity | Coordinates | $E - E_F$ (meV) | Chern Number |
|-------------------|-------------|----------------|--------------|
| $W_1^+$           | 16          | 0.28318, 0.46969, 0.51034 | -8.928 | +1 |
| $W_2^+$           | 8           | 0.13178, 0.13178, 0.57513 | 6.195  | +1 |
| $W_3^+$           | 8           | 0.32795, 0.00000, 0.56275 | 9.497  | −1 |
| $W_4^+$           | 8           | 0.30643, 0.00000, 0.56011 | 12.666 | −1 |
| $W_5^+$           | 8           | 0.41074, 0.41074, 0.51572 | 12.906 | −1 |
in KVEC (see the Methods section for further details).

First, in the most general reciprocal-space Wyckoff position in SG 97 (16o), crystal symmetries generate eight of the sites, and \( T \) symmetry generates the other eight sites. In (TeSe\(_2\))\(_2\)I, 16 symmetry-related FSWPs with the same Chern number \((C = +1)\) lie below the Fermi energy and occupy 16o; we denote them as \( W^+_I \) in Table I. When placed at a higher-symmetry location in the BZ, a WP becomes related by symmetry to fewer WPs than one placed at a generic \( k \) point. In (TeSe\(_2\))\(_2\)I, we additionally identify four groupings of 8 symmetry-related WPs at less general \( k \) points than 16o, all of which lie above \( E_F \) (see Table I). A WP at \((k_x, 0, k_z)\) is left invariant under the combined symmetry \( C_{2y} \times T \) [85], whereas under the other symmetries of SG 97, it is related to 7 other WPs, resulting in 8 total WPs at \((\pm k_x, 0, \pm k_z)\) and \((0, \pm k_x, \pm k_z)\). We observe two sets of WPs with these reciprocal-space coordinates (the 8n position), which we denote as \( W^+_I \) and \( W^-_I \), respectively. Similarly, a WP at \((k_x, k_y, k_z)\) is left invariant under \( C_{2z-y} \times T \), whereas under the other symmetries of SG 97, it is related to 7 other WPs, resulting in 8 total WPs at \((\pm k_x, \pm k_y, \pm k_z)\). We also observe two sets of WPs with these coordinates (8m), which we denote as \( W^+_I \) and \( W^-_I \), respectively. We provide further analyses of the locations and surface projections of the 48 FSWPs in (TeSe\(_2\))\(_2\)I in SM A. In SM J, we also show that the WPs in (TeSe\(_2\))\(_2\)I are theoretically capable of being pairwise annihilated through a series of (large) band-inversion transitions, and therefore, that (TeSe\(_2\))\(_2\)I is not a filling-enforced semimetal [68, 69] (though we also show in SM J that an isolated TaSe\(_4\) chain conversely is a filling-enforced 1D semimetal). Furthermore, because crystals in SG 97 are structurally chiral, then WPs in (TeSe\(_2\))\(_2\)I with opposite Chern numbers generically lie at different energies [28]; specifically, of the 48 FSWPs in Table I, only the 16 \( C = +1 \) WPs \( W^+_I \) lie below \( E_F \). While examples of filling-enforced semimetals with energetically separated, oppositely charged chiral fermions have been identified in previous works [29, 30, 54], (TeSe\(_2\))\(_2\)I represents an extremely rare example of a band-inversion Weyl semimetal for which all of the symmetry-related occupied chiral fermions nearest the Fermi energy exhibit the same Chern number. The net \( C = +16 \) chiral charge of the occupied WPs in (TeSe\(_2\))\(_2\)I is by far the largest value yet predicted. (TeSe\(_2\))\(_2\)I samples should therefore exhibit a strongly amplified response in quantized bulk topological chiral probes, such as the CME and the CPGE.

Weyl-Point Coupling in First-Principles Electronic Susceptibility Calculations and in X-Ray Diffraction and ARPES Experiments. Having theoretically established that the high-temperature phase of (TeSe\(_2\))\(_2\)I is a Weyl semimetal, we will now demonstrate a relationship between the bulk Fermi-surface WPs (FSWPs) and the modulation vectors of the CDW phase. First, to characterize the electronic contribution to the CDW phase, we will calculate the electronic susceptibility from first principles (SM H). Next, we will compare the resulting scattering \((q)\) vectors to “nesting” vectors between the FSWPs. Finally, we have performed experimental investigations of (TeSe\(_2\))\(_2\)I samples using XRD and ARPES probes (SM I). As we will discuss below, we find close agreement between the CDW modulation vectors obtained in XRD experiments, the nesting vectors between the FSWPs, and the high-density spots in the electronic susceptibility. Taken together, this implies that the CDW in (TeSe\(_2\))\(_2\)I couples the bulk FSWPs, including, as we will see, FSWPs with opposite chiral charges.

We begin by performing first-principles calculations to quantitatively characterize the electronic contribution of the Fermi surface of the high-temperature phase of (TeSe\(_2\))\(_2\)I to the CDW that emerges in its low-temperature phase [86–88]. Rather than calculating the simple Fermi-surface nesting vectors, which have been shown to provide an incomplete characterization of CDW phases [89], we instead calculate the real part of the bare electronic susceptibility in the constant-matrix approximation \( \chi_q = \sum_k [n_F(\epsilon_k) - n_F(\epsilon_{k+q})]/(\epsilon_k - \epsilon_{k+q}) \), where \( n_F(\epsilon) = 1/[\exp(\epsilon/k_B T) + 1] \) denotes the Fermi-Dirac distribution function (SM H). We find that in \( \chi_q \), all of the peaks (high-density spots) are concentrated in the vicinity of the \( q_z = 0, \pm 0.024(2\pi/c) \), and \( \pm 0.090(2\pi/c) \) planes [Fig. 2(a–c), respectively]. To understand the origin of the peaks in \( \chi_q \), we compare the \( q \) vectors of the strong peaks to Fermi surface nesting vectors. Remarkably, we find that almost all of the peaks in \( \chi_q \) are located at \( q \) vectors that match the nesting vectors between FSWPs, including those with opposite chiral charges [Fig. 2(a)]. In Table II, we enumerate (up to symmetry-equivalent \( q \) vectors) the peaks in \( \chi_q \) that link FSWPs; the remaining peaks are analyzed in SM H.

To further characterize the CDW phase, we also carried out XRD experiments on single-crystal (TeSe\(_2\))\(_2\)I samples. Specifically, when a crystal with the lattice constants \( a, b, c \) is periodically modulated, as occurs in a CDW phase, then satellite Bragg reflections begin to appear in XRD probes at the momentum-space locations \( Q = G + q \), where \( G = ha^* + kb^* + lc^* \) are the larger reciprocal lattice vectors of the smaller unit cell of the unmodulated (high-temperature) structure, and \( q = m\eta_1 + n\eta_2 + o\delta \) are the smaller modulation vectors of the (typically incommensurate) CDW-modulated structure. Examining the results of our XRD probes of (TeSe\(_2\))\(_2\)I, we observe the appearance of satellite reflections in the vicinities of the \( G = (110), (420), (620) \), and (554) main reflections after samples were cooled below \( T_C \approx 248 \) K (see Fig. S11 in SM I.1), representing clear evidence of a CDW transition, albeit at a slightly lower temperature than the value of \( T_C \approx 260 \) K previously measured in (TeSe\(_2\))\(_2\)I [66, 67]. Probing the XRD data in the vicinities of the main reflections \( G \), we identify the CDW
This implies that satellite spots in the CDW phase appear in a body-centered tetragonal arrangement governed by \([mn\pi/a, mn\pi/c, o\delta(\pi/c)]\), where \(m+n+o=\mu, \mu \in \mathbb{Z}\) (further details provided in SM I.1). As shown in Fig. 2 and Table II, the electronic susceptibility peaks and FSWP nesting vectors calculated from first-principles closely match integer multiples of the experimentally-obtained CDW modulation basis vectors \([m\pi/a, n\pi/c, o\pi/c]\) (SM I.1), and the value of \(\text{Re}(\chi_q)\) in relative units. In particular, \(q_1\), \(q_2\), \(q_3\), \(q_4\), and \(q_5\) nest FSWPs with opposite chiral charges.

We further confirmed that the CDW in \((\text{TaSe}_4)_2\text{I}\) opens an insulating gap by performing ARPES probes of samples at 100 K and 270 K, which are respectively well below and above the CDW transition temperatures observed in our XRD probes (\(\approx 248\) K, see SM I.1 for more details) and in previous works (260 K) [66, 67]. In the low-temperature phase, we observe a gap of roughly 0.12 eV, which shrinks to less than 0.04 eV when samples are warmed to 270 K (see SM I.2 for more details). We attribute this change in gap size to a transition from a low-temperature phase with a CDW-induced band gap into the high-temperature Weyl-semimetal phase predicted in this work.

**Surface States.** Weyl semimetals most notably exhibit characteristic topological Fermi-arc surface states. To confirm the presence of topological surface Fermi arcs in \((\text{TaSe}_4)_2\text{I}\), we calculate the surface states with semi-infinite open boundary conditions using surface Green’s functions [90, 91]. \((\text{TaSe}_4)_2\text{I}\) is known to experimentally cleave on the conventional-cell
The conventional-cell (110)-surface projections of the bulk Fermi surface of (TaSe$_4$)$_2$I form four time-reversal pairs of separated islands in each surface BZ. In Fig. 3(a,b), we enclose the islands with dashed blue lines, and label the islands (and their time-reversal) partners $\alpha$ ($\alpha'$), $\beta$ ($\beta'$), $\gamma_1$ ($\gamma_1'$), and $\gamma_2$ ($\gamma_2'$) [surface TRIM points are labeled with $\times$ symbols in Fig. 3(a,b), see SM C for more details]. Each island is formed from the projected bulk Fermi pockets of the FSWPs, and can thus carry a total chiral charge. To determine the total chiral charge of each island, we sum the chiral charges of the bulk FSWPs that project to it; because two symmetry-related bulk WPs can project to the same momentum in the surface BZ, we introduce arrows in Fig. 3(a,b) to distinguish the projections of single FSWPs.
narrow the surface Fermi arcs are largely localized within a
within each surface BZ. Like the projected bulk states,
eight topological Fermi-arc surface states are present
in Fig. 3(a,b) at the (110)-surface BZ [horizontal and vertical cuts in
topology of the surface Fermi arcs, we calculate the
terminations of the (110)-surface of (TaSe
of the FSWPs, we infer that
with the same chiral charge. From the surface projections
from the projections of two symmetry-related FSWPs
exhibits a $C = -4$ topological spectrum (four Fermi-arc
states with negative velocity cross the Fermi energy), and
the vertical cut [Fig. 3(d,f), respectively] displays a trivial
spectrum. This can be understood by recognizing that the horizontal cut in Fig. 3(a,b) along $k_z = 0.54(2\pi/c)$
is equivalent to a loop around $\beta$, which we previously
determined to carry a net chiral charge of $C = -4$ from
the FSWPs that project to it. Conversely, the vertical cut in Fig. 3(a,b), which lies along the projection of a
$T$-invariant bulk plane, is required to exhibit a net-zero
(trivial) Chern number. Consequently, on both possible
terminations, no topological surface states cross $E_F$
along the vertical line at $k_\parallel = 0.50(2\pi/\sqrt{2}a)$ [Fig. 3(d,f)].
Interestingly, on the I-atom termination [Fig. 3(b)],
the four surface Fermi arcs at $k_\parallel < 0$ exhibit a different
connectivity than in (a), and the four surface Fermi arcs
at $k_\parallel > 0$ all intersect at a single (TRIM) point. Because
the bulk projections and Fermi level in Fig. 3(a,b) are
the same, then we attribute the difference in Fermi-arc

![FIG. 4. (Color online) Quasiparticle interference (QPI) patterns of the surface Fermi arcs of the TaSe$_4$-chain termination of the conventional-cell (110)-surface of (TaSe$_4$)$_2$I at $E_F$. (a) The (110)-surface states, as well as the projections of bulk states, in the full surface BZ, which is spanned by the surface-BZ reciprocal lattice vectors $a^*_1$ and $a^*_2$ (see SM C for more details). There are 16 topological Fermi arcs in (a); the 8 Fermi arcs in the black box at $k_z = \pi/c$ [i.e., in the region of the surface BZ shown in
Fig. 3(a)] are related to the 8 Fermi arcs outside of the box at $k_z = -\pi/c$ by linear combinations of $a^*_1, 2$. (b) To emphasize the
topological contributions to the QPI from the surface Fermi arcs, we filter out the projections of the bulk states and focus on
the Fermi arcs in the top half of the conventional surface BZ [black box in (a)]. We then enumerate all of the spin-conserving
(blue) and nonconserving (green) scattering vectors between the Fermi arcs for a scalar impurity based on the Fermi-arc spin
polarization as calculated from first principles (see SM D). Because the Fermi pockets at $k_z = \pm \pi/c$ are related by surface
reciprocal lattice vectors $[a^*_1, 2$ in (a)], we only label in (b) the independent scattering processes within a single surface BZ;
larger scattering vectors between surface states at $k_z = \pm \pi/c$ are also permitted, but are related to the vectors in (b) by linear
combinations of $a^*_1, 2$. Next, to numerically characterize scattering between the surface Fermi arcs, we calculate and compare two
approximations of the QPI: the joint density of states (JDOS) and the spin-dependent scattering probability (see Ref. [39]
and SM E for more details), the results of which are shown in (c,d) respectively, and are labeled using the scattering vectors
identified in (b). Because the Fermi arcs in (b) appear in pairs (e.g. 1 and 2) with largely opposite spin polarization directions
[see Fig. S4(a) in SM D], the JDOS exhibits characteristic pairs of scattering states that alternate between spin-conserving (e.g. $Q_{11,i}$) and nonconserving (e.g. $Q_{11,i}^*$) scattering processes. In the SSP (d), the spin-nonconserving scattering processes within
each pair are visibly suppressed (though we still label them with green arrows to emphasize their relative absence in the SSP).
Near $q = \vec{0}$, all of the pairs of surface Fermi arcs in (b) also contribute to a central set of three features in the JDOS (c) and
one feature in the SSP (d), as highlighted in [39]. The centralmost set of scattering states originates from scattering within
the same Fermi arc (e.g. $Q_{11,i}$) and is spin-conserving; therefore it is visible in both the JDOS and the SSP. Conversely, the two
features to the left and the right of it (e.g. $Q_{12,i}$ and its time-reverse) are spin-nonconserving, and are largely absent in the SSP.
To infer the temperature dependence of the QPI pattern, we also calculate the surface Fermi-arc states at increasing effective
temperatures, the results of which are shown in Fig. S7 in SM G.
from the projections of two symmetry-related FSWPs
with the same chiral charge. From the surface projections
of the FSWPs, we infer that $\alpha, \beta, \gamma_1,$ and $\gamma_2$ respectively
carry the net chiral charges $-4, -4, +4,$ and $+4$ (SM C).
Because the chiral charge of a WP does not change sign
under $T$ [14], then $\alpha^*, \beta^*, \gamma_1^*$, and $\gamma_2^*$ also exhibit the same
net charges of $-4, -4, +4,$ and $+4$, respectively.

On both the TaSe$_4$-chain [Fig. 3(a)] and I-atom
[Fig. 3(b)] terminations of the (110)-surface of (TaSe$_4$)$_2$I,
eight topological Fermi-arc surface states are present
within each surface BZ. When the projected bulk states,
the surface Fermi arcs are largely localized within a
narrow $k_z$ range near $k_z = \pm \pi/c$. To diagnose the
topology of the surface Fermi arcs, we calculate the
surface-state energy dispersion on closed loops traversing
the (110)-surface BZ [horizontal and vertical cuts in
Fig. 3(a,b) at $k_z = 0.54(2\pi/c)$ and $k_\parallel = 0.50(2\pi/\sqrt{2}a)$, respectively]. On both the TaSe$_4$-chain and I-atom
terminations, the horizontal cut [Fig. 3(c,e), respectively]
connectivity between (a,b) to surface Lifshitz transitions driven by the additional layer of (110)-surface iodine atoms that is present in (b).

Finally, to further understand symmetry-allowed scattering processes in (TaSe$_4$)$_2$I, we calculate the QPI patterns of the Fermi-arc surface states in Fig. 3(a,b). Following the prescription in [39], we calculate and compare two different autocorrelators for each possible (110)-surface termination:

$$\text{JDOS}(q, E) = \int \frac{d^3 k}{(2\pi)^3} \sum_{\sigma} A_{\sigma\sigma}(k, E) \sum_{\sigma'} A_{\sigma'\sigma}(k + q, E)$$

$$\text{SSP}(q, E) = \int \frac{d^3 k}{(2\pi)^3} \sum_{\sigma\sigma'} \left[ A_{\sigma\sigma}(k, E) A_{\sigma'\sigma}(k + q, E) \right]$$

(3)

where the JDOS($q, E$) and the SSP($q, E$) respectively represent the joint density of states and the spin-dependent scattering probability (see SM E for more details). For the TaSe$_4$-chain termination at $E - E_F = 0$, the surface Fermi surface consists of eight topological Fermi-arc states within each surface BZ [e.g., the eight Fermi arcs within the black rectangle in Fig. 4(a), which is the same region of the surface BZ shown in Fig. 3(a)]. The eight Fermi arcs can be divided into four pairs (i.e., two pairs and their time-reversal partners) with largely reversed in-plane spin textures (e.g., Fermi arcs 3 and 4 have nearly opposite spin textures, as do their time-reversal partners, 3’ and 4’) [SM D]. Because of the action of T symmetry, T-related arcs (e.g., 1 and 1’) also exhibit opposite spin polarization directions [see Fig. S4(a) in SM D for more details]. Because QPI is measured with scanning tunneling microscopy (STM), which is surface-sensitive, we filter out the projections of the bulk Fermi surface to emphasize the topological contributions of the surface Fermi arcs [Fig. 4(b)]. Calculating the QPI pattern of the surface Fermi arcs in Fig. 4(b) for a scalar impurity [Fig. 4(c)], we observe pairs of arc-like scattering states in the JDOS (e.g., $Q_{12'}$ and $Q_{11'}$) formed from one set of spin-conserving (blue, e.g., $Q_{12'}$) and one set of spin-nonconserving (green, e.g., $Q_{11'}$) scattering processes. In the SSP [Fig. 4(d)], the spin nonconserving processes are visibly suppressed (though we still label them with green arrows to emphasize their relative absence). In SM E, we analyze the QPI pattern of the iodine-atom termination, which is more complicated than the QPI pattern of the TaSe$_4$-chain termination, because the additional surface iodine atoms draw the four Fermi arcs at $k_z > 0$ together into a surface Lifshitz critical point near $E_F$ [Fig. 3(b)].

Summary. Using first-principles calculations and experimental probes, we have demonstrated that (TaSe$_4$)$_2$I is a structurally chiral conventional Weyl semimetal whose Fermi surface is entirely composed of the cones of conventional ($|C| = 1$) WPs, which, when a (TaSe$_4$)$_2$I crystal is cooled below $T_c \sim 248$ K, become linked and gapped by a CDW. We specifically determined that the Fermi surface derives from 24 time-reversal pairs of WPs (FSWPs) near the $k_z = \pm \pi/c$ planes that vary in energy between 10 meV below $E_F$ to 15 meV above $E_F$. We observe that some of the WPs with opposite chiral charges are well-separated by energy windows as large as 0.123 eV, allowing their Fermi pockets to be treated separately. Crucially, we find close agreement between the FSWP nesting vectors, the electronic susceptibility peaks obtained from first-principles, and the CDW modulation vectors obtained from XRD experiments, indicating that the CDW in (TaSe$_4$)$_2$I couples bulk WPs, including those with opposite chiral charges. Because it hosts occupied WPs with a large net chiral charge, and features an experimentally accessible CDW phase that links momentum-separated Weyl fermions, (TaSe$_4$)$_2$I provides a unique and promising platform for the experimental observation of topological chiral response effects and exotic interacting topological phases.

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METHODS

We performed ab-initio calculations based on density functional theory (DFT) as implemented in the FPLO package [93], and used the full-potential local-orbital basis within the generalized gradient approximation (GGA) [94], fully incorporating the effects of spin-orbit coupling (SOC). The projected atomic Wannier functions (PAWFs) were constructed using the Ta, d, Se, p, and I p orbitals to reproduce the band structures obtained from the ab-initio calculations. The surface states were calculated with a semi-infinite open boundary structure by using a tight-binding model constructed from the above PAWFs. All calculations were performed employing the experimental lattice parameters [62].

In our first principles calculations of (TaSe$_4$I)$_2$ we observe Weyl points (WPs) both at and below $E_F$. In particular, the WPs closest to $E_F$ (the Fermi-surface Weyl points (FSWPs)) subdivide into sets of WPs related by the (chiral) crystal symmetries of SG 97 (I422), where all of the WPs within each set lie at the same energy and exhibit the same chiral charge (Ref. [28] and Table I of the main text). In order to classify the symmetry-related sets of FSWPs, we compare them to the output of the KVEC tool on the Bilbao Crystallographic Server [72, 73]. For each SG, KVEC lists the momentum-space Wyckoff positions (i.e. the “momentum stars”) generated by the unitary crystal symmetries of the type-I magnetic subgroup of the SG [78]; specifically, KVEC does not incorporate the action of time-reversal ($T$) symmetry, which is present in real materials, such as (TaSe$_4$I)$_2$, and must also be included to correctly determine the locations of the symmetry-related FSWPs. However, we can circumvent the absence of $T$ symmetry in KVEC in the particular case of SG 97 by recognizing that inversion symmetry ($I$) and $T$ symmetry have the same action on crystal momenta:

$$T\vec{k} = I\vec{k} = -\vec{k},$$

(4)

even though they act differently on Bloch wavefunctions [95]. SG 97 is generated by:

$$\{T|000\}, \{C_4|000\}, \{C_{2z}|000\},$$

(5)

as well as body-centered tetragonal lattice translations [78]. We can add $I$ symmetry to SG 97 to generate its index-2 supergroup SG 139 (I4/mmm):

$$I4/mmm \equiv E(I422) \cup \{I|000\}(I422),$$

(6)

where $E$ is the identity element. Crucially, Eqs. (4) through (6) imply that the reciprocal-space Wyckoff positions of the type-I magnetic subgroup of SG 139 have the same multiplicities and momentum-space coordinates [though not the same little (co)groups] as those in $T$-symmetric SG 97. We are therefore able to use the output of KVEC for SG 139 to check our analysis of the symmetry-related FSWPs in (TaSe$_4$I)$_2$. In the Results and Discussion section of the main text, we use the output of KVEC for SG 139 to identify the reciprocal-space Wyckoff positions of the FSWPs listed in Table I of the main text, and to confirm their multiplicities and expected locations. We note that because KVEC lists the reciprocal-space Wyckoff positions with respect to the conventional BZ, which is twice as large as the primitive BZ of body-centered SGs 97 and 139, the reciprocal-space Wyckoff positions and multiplicities referenced in this work contain half as many sites as those listed in KVEC. For example, when the effects of $T$ symmetry are incorporated, a (Kramers- [28]) WP lying at $\Gamma$ in SG 97 would be labeled as occupying the 2a reciprocal-space Wyckoff position in KVEC, but would be labeled in this work as occupying the 1a position, because there is only one $\Gamma$ point in the first BZ of SG 97.

We also performed XRD and ARPES experiments on (TaSe$_4$I)$_2$ samples to study the CDW wavevector and electronic band structure, respectively. The XRD experiments were carried out on beamline 25B of the European Synchrotron Radiation Facility in Grenoble, France using a six-circle diffractometer and a wavelength of $\lambda=0.71$ Å. The whisker-shaped (TaSe$_4$I)$_2$ sample was mounted on a copper sample holder oriented with its c-axis perpendicular to the incoming beam and cooled to a minimum temperature of 88 K using a flow of liquid nitrogen. We then measured the intensities of the diffracted X-rays near several main Bragg reflections, employing a two-dimensional (2D) pixel detector to collect the data from both 1D line scans and 2D reciprocal-space maps (RSMs). ARPES measurements were performed at the high-resolution branch of beamline I05, Diamond Light Source (DLS) with a Scienta R4000 analyzer. The photon-energy range for the DLS was 30-220 eV. The angles of the emitted photoelectrons were measured with a resolution of 0.2°, and their energies were measured at an overall resolution of < 15 meV. After samples were glued to the sample holder, they were then cleaved in situ to expose the (110)-surface, which is the favored cleavage plane of (TaSe$_4$I)$_2$ [65]. Throughout our ARPES experiments, samples were kept at a pressure of < 1.5 x 10$^{-10}$ Torr, and measurements of the low- and high-temperature phases of (TaSe$_4$I)$_2$ were performed at 100 K and 270 K, respectively.

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A. Distribution of the Bulk Weyl Points in (TaSe$_4$)$_2$I

In (TaSe$_4$)$_2$I, we identify 24 pairs of WPs in the bulk within 15 meV of the Fermi energy ($E_F$), which we designate as the “Fermi-surface” WPs (FSWPs). As shown in Table I of the main text, which lists the coordinates of all 48 FSWPs with respect the conventional reciprocal cell, the bulk FSWPs divide into five sets of symmetry-related WPs. Within each set, the FSWPs are related to each other by the symmetry elements of SG 97 (I$4_22$): $C_4$, $C_2(x_2y_2)$, $C_2(x+y,(2x-y))$, and $T$. Because all of these operations are proper rotations or time-reversal, which do not change the sign of the chiral charge of a WP [28], then all of the WPs within each set exhibit the same chiral charge.

**FIG. S1.** (Color online) The distribution of Fermi-surface WPs (FSWPs) in the first bulk Brillouin zone (BZ). To highlight the 3D distribution of the FSWPs, we present both (001)-surface (top) (a) and (110)-surface (side) (b) views of their locations in the first bulk BZ. The black lines in (a,b) represent the boundaries of 2D slices of the bulk BZ [Fig. 1(c) of the main text] taken at different bulk momenta parallel to the projection direction. Specifically, the larger octagon in (a) represents the boundary of the bulk $k_z = 0$ plane [containing $\Gamma$, $\Sigma$, $Y$, and $X$ in Fig. 1(c) of the main text], and the smaller square represents the boundary of the $k_z = 2\pi/c$ plane (containing $Z$, $\Sigma_1$, and $Y_1$). In (b), the larger octagon represents the boundary of the $k_x + k_y = 0$ plane (containing $\Gamma$, $X$, $P$, $Y_1$, and $Z$), and the smaller diamond represents the boundary of the $k_x + k_y = \frac{2\pi}{2\sqrt{2}}$ plane (containing $Y$ and different, symmetry-related $X$ and $P$ points). Additionally, in (b), $a_{1,2}^*$ indicate the (110)-surface BZ primitive reciprocal lattice vectors [Fig 4(a) of the main text], and the dashed blue box indicates the boundary of the second (conventional) surface BZ, which we will later examine in detail in SM C. The five sets of WPs listed in Table I of the main text are respectively indicated with boxes ($W_1^+$), circles ($W_2^+$), upward-pointing triangles ($W_3^-$), downward-pointing triangles ($W_4^-$), and diamonds ($W_5^-$). In (a), because of $C_2$ and $C_2y$ symmetry, each of the depicted WPs actually represents a pair of WPs with the same chiral charge lying at the opposite momenta $\pm k_z$. All of the symbols in (a) consequently represent FSWP projections with chiral charge $|C| = 2$. In (b), the relationship between the bulk WPs and symmetries is more complicated. We therefore introduce arrows to indicate which symbols in (b) correspond to the surface projections of only a single bulk FSWP; in (b), like in (a), the symbols without arrows indicate the projections of two bulk FSWPs with the same chiral charge. Each of the symbols with arrows in (b) consequently represents a surface projection with chiral charge $|C| = 1$, whereas each of the symbols without arrows represents a surface projection with chiral charge $|C| = 2$. The momentum-space coordinates of the $W_3^-$ and $W_5^-$ FSWPs are very close to each other (but not exactly the same, see Table I of the main text for the exact coordinates), and so the upward- and downward-pointing triangles appear at nearly the same locations in (a) and (b).

In Fig. S1(a,b), we show (001)-surface (top) and (110)-surface (side) views of the bulk FSWPs in the first BZ, respectively. The solid lines indicate the projected boundary of the first bulk BZ, and the differently shaped symbols each denote one set of symmetry-related WPs; a key matching each symbol to each set of WPs is given in the top panel.
of Fig. S1. Each of the depicted FSWPs in Fig. S1 lies within the first bulk BZ; later, in SM C and F respectively, we will determine the locations of the projections of the bulk FSWPs in the (110) and (001) surface BZs. Because (TaSe$_4$)$_2$I crystallizes in a body-centered tetragonal structure [space group (SG) 97 (I422)], then bulk BZ slices taken at different values of a perpendicular momentum exhibit a different shape (Fig. 1(c) of the main text). Specifically, the larger octagon in Fig. S1(a) represents the boundary of the bulk $k_z = 0$ plane [containing $\Gamma$, $\Sigma$, $Y$, and $X$ in Fig. 1(c) of the main text], and the smaller square represents the boundary of the $k_z = 2\pi/c$ plane (containing $Z$, $\Sigma_1$, and $Y_1$). In Fig. S1(b), the larger octagon represents the boundary of the $k_x + k_y = 0$ plane (containing $\Gamma$, $X$, $P$, $Y_1$, and $Z$), and the smaller diamond represents the boundary of the $k_x + k_y = \frac{\pi}{a\sqrt{2}}$ plane (containing $Y$ and different, symmetry-related $X$ and $P$ points). Note that while the FSWPs are distributed over a wide range in $k_{x,y}$ [Fig. S1(a)], they all lie within a close vicinity of the $k_z = \pm \pi/c$ planes [Fig. S1(b)]. As explicitly shown in SM J, this distribution of nodal points reflects that a crystal of decoupled TaSe$_4$ chains and iodine atoms is a filling-enforced semimetal [68, 69] with 4$_2$-screw- and $T$- symmetry enforced nodal surfaces lying near $k_z = \pi/c$.

B. Band Dispersions of the Bulk Weyl Points

To diagnose each bulk FSWP in (TaSe$_4$)$_2$I as type-I (untitled) or type-II (tilted) [21], we perform detailed three-dimensional band structure calculations. Specifically, by calculating the Fermi surface at different energy contours, we determine that $W_3^-$ and $W_4^-$ (upward- and downward-pointing triangles in Fig. S1, respectively) are type-I WPs, whereas $W_1^+$, $W_2^+$, and $W_5^-$ (boxes, circles, and diamonds in Fig. S1, respectively) are type-II WPs. In Fig. S2, we show typifying band dispersions of one FSWP within each of the five symmetry-related sets listed in Table I of the main text.

![Typifying band dispersions of WPs](image)

FIG. S2. (Color online) Typifying band dispersions of WPs within each of the five symmetry-related groupings of Fermi-surface WPs in (TaSe$_4$)$_2$I (Table I of the main text).

C. Bulk Fermi Surface Projections in the Conventional-Cell (110)-Surface BZ

In order to understand the distribution of the topological Fermi arcs in (TaSe$_4$)$_2$I, we determine the net chiral charges of the projections of the bulk Fermi surface onto the surface BZs. Because (TaSe$_4$)$_2$I experimentally favors cleaving along the conventional-cell (110)-plane [65], we focus in this section on the conventional-cell (110)-surface projections of the bulk Fermi surface, where all momentum-space vectors are given with respect to the conventional-cell reciprocal lattice vectors ($k_x \frac{2\pi}{a}$, $k_y \frac{2\pi}{c}$, $k_z \frac{2\pi}{c}$). Later, in SM G, we similarly analyze the projections of the bulk WPs onto the (001)- and (100)-surfaces.

To project the bulk Fermi surface onto the (110)-surface, we must carefully treat the bulk body-centered tetragonal position- and reciprocal-space lattice vectors of (TaSe$_4$)$_2$I. In the units of the conventional-cell lattice translations ($a$, $a$, $c$), the position-space lattice vectors of the (110)-surface are $a_1 = (-a/2, a/2, -c/2)$ and $a_2 = (-a/2, a/2, c/2)$. Thus, the (110)-surface primitive reciprocal lattice vectors are $a_1^* = (-\pi/a, \pi/a, -2\pi/c)$ and $a_2^* = (-\pi/a, \pi/a, 2\pi/c)$. To construct the (110)-surface BZ [Fig. S3(a)], we first define $k_\parallel$ in the (110) direction of the conventional cell [i.e., $k_\parallel = -k_x + k_y$], and then form the linear combinations:

$$k_{1,2} = k_\parallel \pm k_z,$$

which lie parallel to $a_{1,2}^*$. For simplicity, however, in this work, even though the (110)-surface BZ is defined as the square indexed by the primitive-surface-cell momenta $k_{1,2}$, we will still use the more natural conventional-cell
monenta $k_1 = -k_2 + k_y$ and $k_z$ to index momentum-space coordinates along the (110)-surface. $k_\parallel$ and $k_\perp$ are periodic with respect to the conventional (110)-surface BZ [dashed square in Fig. S3(a)], which is twice as large as the primitive surface BZ [black squares in Fig. S3(a)], because it contains both the first and second primitive surface BZs. Restricting to the dashed blue square in Fig. S3(a), we project the bulk Fermi pockets of the 48 FSWPs discussed in SM A and listed in Table I of the main text. Because all of the projected pieces of the bulk Fermi surface lie in the vicinity of the $k_z = \pm \pi/c$ lines, which are related by linear combinations of the surface reciprocal lattice vectors $a_1^*, \alpha$, then we are free to focus on the projected Fermi pockets near $k_z = \pi/c$ [solid black rectangle in Fig. S3(a)], which is shown in a larger, rescaled view in Fig. S3(b). As discussed in Fig. 3 of the main text, the projected bulk Fermi pockets split into four pairs of islands related by the action of $\mathcal{T}$ symmetry. In Fig. S3, we enclose the islands of projected bulk states with dashed blue lines, and denote the surface TRIM points $-a_1^*/2$ and $a_1^*/2$ with $\times$ symbols. Labeling islands (and their time-reversal partners) with unprimed (primed) symbols, we respectively denote the four islands (and their time-reversal partners) as $\alpha$, $\beta$, $\gamma_1$, and $\gamma_2$.

To obtain the net chiral charge of each projected island, we project the bulk FSWPs from SM A onto the (110)-surface BZ in Fig. S3(b) (employing the labeling scheme of the WPs in Fig. S1). In Fig. S3(c), we show an even closer view of the projected Fermi surface in the vicinity of $k_\parallel = 0, k_z = \pi/c$. As discussed in SM A, the arrows in Fig. S3(c) point to FSWP projections with net chiral charges $|C| = 1$; the symbols in Fig. S3(c) without arrows indicate two bulk FSWPs of the same chiral charge $C = \pm1$ that lie at the same values of $k_\parallel$ and $k_z$, and thus project to the same point in the (110)-surface BZ. In Fig. S3, we enclose the islands of projected bulk states with dashed blue lines, and denote the surface TRIM points $-a_1^*/2$ and $a_1^*/2$ with $\times$ symbols. To calculate the topological charge of each island, we simply sum the projected Chern numbers of the WPs that project to the same point in the (110)-surface BZ. Therefore, the symbols without arrows in Fig. S3(c) indicate bulk projections with net chiral charges $|C| = 2$. As shown in Fig. S2, $W_{-2}^+$ and $W_{1}^-$ (upward- and downward-pointing triangles, respectively) are type-I WPs, whereas $W_{1}^+$ (boxes), $W_{2}^-$ (circles), and $W_{5}^-$ (diamonds) are type-II WPs. To calculate the topological charge of each island, we simply sum the projected Chern numbers of the WPs that project to that island. From this counting, we determine the net chiral charges of $\alpha$, $\beta$, $\gamma_1$, and $\gamma_2$ to be $C = -4$, $-4$, $+4$, and $+4$, respectively. Because $\mathcal{T}$ symmetry does not affect the chiral charge of a WP [14], then the time-reversal partners of each island carry the same respective chiral charges.

FIG. S3. (Color online) Bulk Fermi surface projected onto the conventional-cell (110)-surface BZ. (a) The (110)-surface BZs (black squares), whose reciprocal lattice vectors are $a_1^*, a_2^*$, and the projections of the bulk Fermi surface; the blue dashed square in (a) indicates the conventional surface BZ indexed by $k_\parallel$ and $k_z$, which is twice as large as the primitive surface BZ (black squares). (b) An enlarged and rescaled view of the projected Fermi pockets near $k_z = \pi/c$ in the solid black rectangle in (a); the projected bulk states in (b) are the same as those shown [along with the (110)-surface states] in Fig. 3(a) of the main text. In (b), we denote the (110)-surface time-reversal-invariant momenta (TRIM points) at $-a_1^*/2$ and $a_1^*/2$ with $\times$ symbols. The projections of the bulk Fermi pockets form four time-reversal pairs of islands, each of which is shown enclosed in a dashed blue line in (b) and (c), and carries a net topological chiral charge. The four islands (and their time-reversal partners) are respectively denoted as $\alpha$, $\beta$, $\gamma_1$, and $\gamma_2$. (c) A closer view of the projected bulk Fermi surface in the vicinity of $k_1 = 0, k_z = \pi/c$. In both (b) and (c), we employ the labeling scheme of Fig. S1, in which the projections of the bulk WPs $W_{1}^+, W_{2}^+, W_{1}^-, W_{5}^-$ are respectively indicated by boxes, circles, upward-pointing triangles, downward-pointing triangles, and diamonds. As previously in Fig. S1, we use arrows to indicate FSWP projections with total chiral charges of $|C| = 1$; the symbols without arrows, which indicate the superposed projections of two bulk FSWPs with the same chiral charge, carry net charges of $|C| = 2$. 


D. Spin Texture of the (110)-Surface Fermi Arcs

In Fig. S4(a-d), we show the spin texture of the (110)-surface states of (TaSe$_4$)$_2$I calculated on a slab Wannier-based tight-binding Hamiltonian from first-principles. Specifically, for each state within the (110)-surface Fermi arcs [Fig. 3(a,b) of the main text], we calculate the spin expectation value vector $\langle \vec{S} \rangle = (\langle S_\parallel \rangle, \langle S_z \rangle, \langle S_\perp \rangle)$, where $S_\parallel, S_\perp = -S_x \pm S_y$. Because we find that $\langle S_\perp \rangle \ll \langle S_\parallel \rangle, \langle S_z \rangle$ at all surface $k$ points, then we only show the in-plane components $(\langle S_\parallel \rangle, \langle S_z \rangle)$ in Fig. S4. In Fig. S4(a), we show the spin texture of the TaSe$_4$-chain termination at $E = E_F$. (b-d) Spin textures of the I-atom termination at $E - E_F = -0.004$, 0, and 0.004 eV, respectively. On both terminations, the surface Fermi arcs appear in pairs (e.g. 3 and 4) with largely reversed in-plane spin textures. Each Fermi arc (e.g. 3) is additionally related by $T$ to a partner (e.g. 3') with an opposite in-plane spin texture (surface TRIM points are indicated with $\times$ symbols in Fig. S4). At the Fermi energy ($E - E_F = 0$), four of the Fermi arcs on the iodine-atom termination (1, 2, 1', and 2') intersect at a junction that is representative of a surface Lifshitz critical point [Fig. S4(c)] between the Fermi-arc connectivity in Fig. S4(b) and the connectivity in Fig. S4(d). Specifically, the projected bulk states and their respective chiral charges are the same in Fig. S4(a,c), and the system Fermi level is independent of the surface termination; therefore, the Lifshitz critical point that appears in (c) and not in (a) is a consequence of the different surface atoms in the TaSe$_4$ and I terminations, as opposed to a shift of the Fermi energy. A similar surface-dependent Lifshitz transition was recently realized in experiment in [96]. Additionally, as shown in Fig. 3(a,b) of the main text,
on the iodine-atom termination, Fermi arcs 3 and 4 and their time-reversal partners 3' and 4' exhibit a different connectivity in Fig. S4(b-d) than they do on the TaSe$_4$-chain termination (a); this is also the result of a surface Lifshitz transition, because the chiral charges of the projected bulk states are the same in (a-d).

E. Quasiparticle Interference on the Iodine-Atom Termination of the Conventional-Cell (110)-Surface

In both the main text [Fig. 4(c,d)] and in this section, we calculate the quasiparticle interference (QPI) patterns of the Fermi-arc surface states in Fig. 3(a,b) of the main text using the prescription developed in [39]. First, because scanning tunneling spectroscopy (STM) measurements are surface sensitive, and to emphasize topological contributions to the QPI, we set the surface Green’s function to zero at each of the $k$ points in which it only contains contributions from the projected bulk states in Fig. S3; the Green’s function in the remaining $k$ points only contains contributions from surface-localized topological Fermi-arc states. Next, we calculate and compare two different autocorrelators for each possible conventional-cell (110)-surface termination:

\[
\text{JDOS}(q, E) = \int \frac{d^3 k}{(2\pi)^3} \sum_\sigma A_{\alpha \sigma}(k, E) \sum_{\sigma'} A_{\sigma' \sigma}(k + q, E),
\]

\[
\text{SSP}(q, E) = \int \frac{d^3 k}{(2\pi)^3} \sum_{\sigma \sigma'} [A_{\sigma \sigma'}(k, E) A_{\sigma' \sigma}(k + q, E)],
\]

where the JDOS$(q, E)$ and the SSP$(q, E)$ respectively represent the joint density of states and the spin-dependent scattering probability. First, we calculate the imaginary part of the surface Green’s function $A_{ij \sigma \sigma'}(k, E)$ at each (110)-surface $k$ point. We then trace out the orbital components indexed by $i,j$, leaving $A_{\sigma \sigma'}(k, E)$, which we take to approximate the spin-resolved Fourier-transformed density of states [39]. Finally, we calculate the two autocorrelators in Eq. (8), which differ by whether we first matrix multiply $A_{\sigma \sigma'}(k, E)$ and $A_{\sigma' \sigma}(k + q, E)$ and then trace over the result (SSP), or whether we first individually trace over $A_{\sigma \sigma'}(k, E)$ and $A_{\sigma' \sigma}(k + q, E)$ and then multiply the scalar results (JDOS).

In Fig. 4(c,d) of the main text, we show and analyze the QPI of the TaSe$_4$-chain termination of the (110)-surface states of (TaSe$_4$)$_2$I at $E - E_F = 0$, calculated using the methodology detailed in this section. We then additionally analyze the QPI of the alternative, iodine-atom termination (110)-surface states. In Fig. S5(a-c), we show the JDOS and SSP [Eq. (8)] of only the I-atom-termination (110)-surface topological Fermi arcs at $E - E_F = -0.004, 0, 0.004$ eV. As with the TaSe$_4$-chain termination, the I-atom-termination surface Fermi arcs appear in pairs (e.g. 1 and 2) with opposite spin textures [Fig. S4(b-d)]. Therefore at all energies, scattering states appear in spin-conserving (blue, e.g. $\vec{Q}_{12}^i$) and nonconserving (green, e.g. $\vec{Q}_{11}^i$) pairs (Fig. S5). The spin-conserving scattering processes are present in both the JDOS and the SSP, whereas the spin-nonconserving processes are largely suppressed in the SSP (though we still label them with green arrows to emphasize their relative absence). Additionally, at energies above and below $E_F$ near $\vec{q} = 0$, all of the pairs of surface Fermi arcs contribute to a central set of three features in the JDOS (c) and one feature in the SSP (d), as highlighted in [39] and discussed in Fig. 4 of the main text. Unlike with the TaSe$_4$-chain termination, at the Fermi energy, two of the I-atom surface Fermi arcs and their time-reversal partners (1, 2, 1’, and 2’) merge in a surface Lifshitz transition [Fig. S5(b) and SM D] that represents the critical point separating the Fermi-arc connectivity in Fig. S5(a) from the connectivity in Fig. S5(c). At $E = E_F$, short-range scattering processes from the four arcs at the Lifshitz point overwhelm both the JDOS and SSP at small $\vec{q}$ [Fig. S5(e,h), respectively].
FIG. S5. (Color online) Quasiparticle interference (QPI) for the topological Fermi arcs on the iodine-atom termination of the conventional-cell (110)-surface of (TaSe$_4$)$_2$I at $E - E_F = -0.004$, 0, 0.004 eV. In all panels, we label spin-conserving (nonconserving) scattering processes with blue (green) arrows. (a,b,c) The (110)-surface Fermi arcs at $E - E_F = -0.004$, 0, 0.004 eV, respectively. In both this figure and in Fig. 4(c,d) of the main text, the QPI is only calculated for the surface Fermi-arc states; specifically, because STM measurements are surface sensitive, we have set the surface Green’s function to zero at the $k$ points in which its only contributions come from bulk states. (a,c) Above and below $E_F$, the surface Fermi arcs in appear in pairs (e.g. 1 and 2) with largely opposite spin polarization directions [Fig. S4(b)]. Therefore, the JDOS (d,f) exhibits characteristic pairs of scattering states that alternate between spin-conserving (e.g. $\vec{Q}_{12}$) and nonconserving (e.g. $\vec{Q}_{11'}$) scattering processes. Conversely, in the SSP (g,i), the spin-nonconserving scattering processes within each pair are visibly suppressed (though we still label them with green arrows to emphasize their relative absence in the SSP). Near $\vec{q} = 0$, all of the pairs of surface Fermi arcs in (a,c) also contribute to a central set of three features in the JDOS (d,f) and one feature in the SSP (g,i), as highlighted in [39] and in Fig. 4 of the main text. Unlike in the TaSe$_4$-chain termination [Fig. 3(a) of the main text], at $E - E_F = 0$, two of the surface Fermi arcs and their time-reversal partners (1, 2, 1′, and 2′) intersect in (b) at a surface Lifshitz critical point (SM D) between the Fermi-arc connectivity in (a) and the connectivity in (c). Short-range scattering processes from surface states in the momentum-space vicinity of the Lifshitz point combine to produce a large signal near $\vec{q} = 0$ in the JDOS (e) and the SSP (h) at $E - E_F = 0$. In addition to all of the scattering processes depicted in this figure and in Fig. 4 of the main text, scattering is also allowed at larger $q$ vectors between the surface Fermi-arc states at $k_z = \pm \pi/c$ [Fig. S3(a)]. However, because the eight surface Fermi arcs at $k_z = -\pi/c$ are related to the arcs at $k_z = \pi/c$ by surface reciprocal lattice vectors [$a_1^*, a_2^*$ in Fig. S3(a)], then all of the longer scattering processes between $k_z = \pm \pi/c$ can be expressed as the sum of the short-range processes shown here and a linear combination of $a_1^*, a_2^*$. 
F. Surface States on the Conventional-Cell (001)- and (100)-Surfaces

Though it is experimentally favorable to cleave (TaSe$_4$)$_2$I along the conventional-cell (110)-surface [65], in this section, we also calculate the surface states of the conventional-cell (001)- and (100)-surfaces at the Fermi energy, which are shown in Fig. S6(a,b), respectively. Like the (110)-surface [Fig. 3(a,b) of the main text], both the (001)- and (100)-surfaces also exhibit topological Fermi-arc surface states.

G. Conventional-Cell (110)-Surface States at Different Temperatures

To show the temperature dependence of the topological surface Fermi arcs in (TaSe$_4$)$_2$I, we additionally calculated the conventional-cell (110)-surface states at increasing effective temperatures by increasing the state-smearing parameter $\epsilon$ in the surface Green’s function $A_{ij\sigma\sigma'}(k, E) = 1/[E - H_{ij\sigma\sigma'}(k) + i\epsilon]$, where $\epsilon = k_BT$. In Fig. S7(a-d), we respectively plot the (110)-surface states at $\epsilon = 0.0001978$, 0.0086, 0.02236, and 0.043, which respectively correspond to effective temperatures of $T = 2.3$ K, 100 K, 260 K, and 500 K. Above $T_C$ for the CDW phase, which lies between 248 K and 260 K (Refs. [66, 67] and SM I), the surface Fermi arcs become increasingly difficult to resolve [Fig. S7(c,d)]. However, even at high temperatures, the presence of surface Fermi arcs can still be inferred by observing that the surface Fermi surface is continuous, whereas the surface projections of the bulk Fermi surface form disconnected islands (SM C).

H. Electronic Susceptibility

In some cases, a charge-density wave (CDW) cannot be explained using the simple picture of Fermi-surface nesting vectors [89, 97–99]. Therefore, still focusing on the electronic contribution to the CDW instability (as opposed to the phonon contribution), we perform the more accurate calculation of electronic susceptibility. Specifically, when the real part of the electronic susceptibility $\chi_q$ diverges, there is a strong electronic contribution towards a CDW.
FIG. S7. (Color online) (a-d) The conventional-cell (110)-surface states of (TaSe$_4$)$_2$I at $E = E_F$ calculated at the effective temperatures $T = 2.3$ K, 100 K, 260 K, and 500 K, respectively. (a,b) While the surface Fermi arcs at $T = 2.3$ K and $T = 100$ K are clear and distinct, they were calculated at effective temperatures below $T_C$ for the CDW phase, which is known to occur between 248 K and 260 K (Refs. [66, 67] and SM I). (c,d) However, even above $T_C$, the surface Fermi arcs are still visible as pieces of a continuous surface Fermi surface, though they quickly become difficult to resolve at room temperature and above.

transition [89]. In the constant-matrix approximation:

$$\chi_\mathbf{q} = \sum_\mathbf{k} \left[ n_F(\epsilon_\mathbf{k}) - n_F(\epsilon_{\mathbf{k+q}}) \right] / (\epsilon_\mathbf{k} - \epsilon_{\mathbf{k+q}}),$$

(9)

where $n_F(\epsilon) = 1/[\exp(\epsilon/k_B T) + 1]$ is the Fermi-Dirac distribution function. Using a Wannier-based tight-binding Hamiltonian, we compute $\chi_\mathbf{q}$ in the full 3D BZ of (TaSe$_4$)$_2$I, which we plot in Fig. 2(b-d) of the main text. We find that nearly all of the observed peaks in the electronic susceptibility can be attributed to nesting vectors between the bulk FSWPs. As shown in Fig. S8, the remaining two peaks:

$$\tilde{\mathbf{q}}_1 = \left[ 1.00000 \left( \frac{2\pi}{a} \right), 0.00000 \left( \frac{2\pi}{a} \right), 0.00000 \left( \frac{2\pi}{c} \right) \right], \quad \tilde{\mathbf{q}}_2 = \left[ 0.06062 \left( \frac{2\pi}{a} \right), 0.06062 \left( \frac{2\pi}{a} \right), 0.02400 \left( \frac{2\pi}{c} \right) \right],$$

(10)

originate from nesting vectors between bulk Fermi pockets away from the nodal centers of the FSWPs [though because (TaSe$_4$)$_2$I in SG 97 (I422) is a symmorphic chiral crystal, then all of its Fermi pockets originate from the (possibly narrowly split) topological bands of bulk chiral fermions [28]]. Remarkably, we observe that for the peaks in $\chi_\mathbf{q}$ that do correspond to FSWP nesting vectors (Table II of the main text), all of the $q$ vectors closely match integer multiples of the experimentally-obtained CDW modulation vectors (SM I.1), implying that the CDW in (TaSe$_4$)$_2$I links bulk WPs, including those with opposite chiral charges. In Table II of the main text, we list all of the peaks in the electronic susceptibility whose $q$ vectors match nesting vectors between FSWPs, and express their $q$ vectors in terms of the experimentally-observed CDW modulation vectors (SM I.1).
FIG. S8. (Color online) The origin of the electronic susceptibility peaks not listed in Table II of the main text. In (TaSe$_4$I)$_2$, almost all of the peaks in the electronic susceptibility ($\chi_q$) calculated from first-principles [Fig. 2(b-d) of the main text] match nesting vectors between FSWPs; (e,f) the remaining peaks [\(q_{1,2}\) in Eq. (10)] instead match nesting vectors between bulk Fermi pockets away from the nodal centers of the FSWPs. (a) The bulk Fermi surface of (TaSe$_4$I)$_2$ projected in the z [(001)] direction, where the red (blue) colors indicate electron (hole) pockets. In Fig. 2(b) of the main text, there is a strong (red) peak at \(q_1\) [reproduced in (e)]. A large number of both electron- and hole-like Fermi pockets are nested by \(q_1\) in (a). This occurs because \(q_1\) is a reciprocal lattice vector in a primitive tetragonal crystal of decoupled TaSe$_4$ chains (SM J), and because interchain coupling is weak in (TaSe$_4$I)$_2$ [62]. (b) Superposition of the Fermi pockets at \(k_z = 0.012(2\pi/c)\) (black teardrops) and \(k_z = -0.012(2\pi/c)\) (blue teardrops). (c) ([d]) A closer view of the Fermi pockets in the dashed [solid] square in (b), and the result of calculating their isolated contributions to \(\chi_q\) by restricting the sum in Eq. (9) to \(k\) points within each pair of Fermi pockets. In both (c,d), there are strong contributions to the electronic susceptibility originating from \(q_6\), which we show in the main text to correspond to nesting vectors between two W$^+$ FSWPs [Table II of the main text]. However, in the electronic susceptibility contribution shown in (c,d), there is also a weak contribution from the diagonal nesting vector \(q_2\). (f) Crucially, even though \(q_6\) is stronger in (c,d) than \(q_2\), because \(\chi_q\) contains the summed contributions of all pairs of Fermi pockets [Eq. (9) with the sum taken over all \(k\) points in the first BZ], then there is still a strong peak in \(\chi_q\) corresponding to \(q_2\). Specifically, the strong vectors shown in (c,d) are both labeled with \(q_6\) because they are related by symmetry; however, because the strong vectors in (c,d) are not parallel, then they only contribute individually at different locations in \(\chi_q\) in (f). Conversely, because the weak vector \(q_2\) is present at exactly the same angle in (c,d), then its contribution to \(\chi_q\) adds constructively, leading to a large peak at \(q_2\) in (f).
I. Experimental Data

We also performed X-ray diffraction (XRD) and angle-resolved photoemission spectroscopy (ARPES) experiments to study the CDW wavevector and electronic band structure of \((\text{TaSe}_4)_2\text{I}\), respectively. In SM I.1 and SM I.2, we respectively provide details of our XRD and ARPES investigations.

I.1 X-Ray Diffraction Experiments

In order to determine the modulation vectors and confirm the CDW transition temperature \(T_C\) of \((\text{TaSe}_4)_2\text{I}\), we performed X-ray diffraction (XRD) experiments. We specifically carried out our XRD experiments on a whisker-shaped \((\text{TaSe}_4)_2\text{I}\) sample at beamline 25B of the European Synchrotron Radiation Facility in Grenoble, France using a six-circle diffractometer and a wavelength of \(\lambda=0.71\ \text{Å}\). The sample was mounted on a copper holder, oriented with its c-axis perpendicular to the incoming beam, and cooled using a flow of liquid nitrogen to reach a minimum temperature of 88 K. Throughout this section, distances in reciprocal space are given with respect to the previously established conventional-cell lattice constants \(a = b = 9.59\ \text{Å}\) and \(c = 12.64\ \text{Å}\) of the high-temperature phase of \((\text{TaSe}_4)_2\text{I}\) [62].

As the temperature of the sample was lowered, we searched for satellite reflections as evidence of the onset of a CDW phase. Specifically, when a crystal with the lattice constants \(a, b, c\) is periodically modulated, as occurs in a CDW phase, then XRD probes begin to exhibit satellite Bragg reflections at the momentum-space locations \(\mathbf{Q} = \mathbf{G} + \mathbf{q}\), where \(\mathbf{G} = h\mathbf{a}^* + k\mathbf{b}^* + l\mathbf{c}^*\) are the larger reciprocal lattice vectors of the smaller unit cell of the unmodulated (high-temperature) structure, and \(\mathbf{q} = m\mathbf{n}_1 + m\mathbf{n}_2 + o\mathbf{d}\) are the smaller modulation vectors of the (typically incommensurate) CDW-modulated structure. In \(\mathbf{G}\) \((\mathbf{q})\), the conventional reciprocal-space lattice (modulation) vectors along \(k_x\) \((q_x)\), \(k_y\) \((q_y)\), and \(k_z\) \((q_z)\) are respectively given by \(\mathbf{a}^*\) \((\eta_1)\), \(\mathbf{b}^*\) \((\eta_2)\), and \(\mathbf{c}^*\) \((\delta)\). The main purpose of our XRD investigations was to experimentally obtain the values of \(\eta_1/\eta_2\) and \(\delta\); we additionally used the appearance of satellite reflections to obtain an estimate of \(T_C\) in our \((\text{TaSe}_4)_2\text{I}\) sample, as detailed below.

We identified the locations of the satellite peaks by performing systematic XRD scans along several directions in \(k\) space, employing a two-dimensional (2D) pixel detector to collect the data from both 1D line scans and 2D reciprocal-space intensity maps (RSMs) near the \(\mathbf{G} = (110), (420), (620),\) and \((554)\) Bragg reflections. We selected these reflections, because the \((110), (420),\) and \((620)\) reflections were the simplest to measure in our c-axis directed whisker-shaped sample, and because satellite reflections near the \((554)\) reflection were previously studied in [88]. All scans for satellite peaks were performed at a temperature range of roughly 88 K to 100 K, which is well below the reported CDW transition temperature of 260 K [66, 67]. In Fig. S9, we show the results of an XRD intensity line scan along the \(k_{x+y}\) direction at \(k_z = 0\) near the \(\mathbf{G} = (110)\) main Bragg reflection. The intense peak at \(k_{x+y} = \sqrt{2}(\pi/a)\) \((\mathbf{Q} = \mathbf{G})\) reflects the average structure, which coincides with the unmodulated (high-temperature) crystal structure of \((\text{TaSe}_4)_2\text{I}\). We observe two weak satellite peaks at \(\mathbf{q} = \pm(\pi/2a), \pm(\pi/2c), 0\), where \(\eta = 0.027 \pm 0.001\). Because, as we will show, across all measured main reflections, we did not observe any satellite peaks closer to their main reflections than \(\mathbf{q}\) is to \(\mathbf{G} = (110)\), then we label the two \(\mathbf{q}\) vectors in Fig. S9 with the full indices \(\mathbf{Q} = (hkl, mno) = (110,110)\) and \((110,110)\), respectively. We will see that this implies that the CDW-modulated structure, like the unmodulated parent structure, of \((\text{TaSe}_4)_2\text{I}\), is body-centered tetragonal, and that its satellite reflections appear at momenta that are periodic with respect to the reciprocal basis vectors:

\[
\tilde{\mathbf{q}}_1 = \left[0, \eta\left(\frac{2\pi}{a}\right), 0, \delta\left(\frac{2\pi}{c}\right)\right], \quad \tilde{\mathbf{q}}_2 = \left[\eta\left(\frac{2\pi}{a}\right), 0, \delta\left(\frac{2\pi}{c}\right)\right], \quad \tilde{\mathbf{q}}_3 = \left[\eta\left(\frac{2\pi}{a}\right), \eta\left(\frac{2\pi}{a}\right), 0\right],
\]

such that satellite spots only appear at \(\mathbf{q}\) vectors satisfying \(m + n + o = \mu, \mu \in 2\mathbb{Z}\). This is consistent with the recognition, discussed by Korekawa et al. [100, 103], that CDW-modulated structures frequently preserve the point group symmetries of their unmodulated parent structures, and that their satellite spots frequently follow the same reciprocal Bravais lattice (but not with the same lattice spacing) as the high-temperature crystal structure. This effect has been observed in both natural crystals, such as feldspars, and in ultra-thin films [102]. In Fig. S9, we more generally only observe satellite spots for which \(m + n = \mu, \mu \in 2\mathbb{Z}\), which is consistent with Eq. (11) [we have not yet introduced satellite spots with nonzero \(o\), though in a body-centered CDW phase, they will satisfy the more general relation \(m + n + o = \mu, \mu \in 2\mathbb{Z}\)], i.e., Eq. (11)]. We note that the first-order satellites in Fig. S9, \(\mathbf{q} = (110)\) and \((110)\), are extremely faint; they appear at an intensity that is about four orders of magnitude weaker than the main reflection at \(\mathbf{G} = (110)\). To explain this, we note that it has been well established in previous works that, while the locations of satellites spots are broadly governed by the reciprocal modulation vectors [here Eq. (11)], their intensities can vary in
FIG. S9. (Color online) XRD scan along the \( k_{x+y} \) direction across the \( G = (110) \) main reflection of our (TaSe\(_4\))\(_2\)I sample, plotted on a logarithmic intensity scale and collected within a temperature range of roughly 88 K to 100 K, which is well below the reported CDW transition temperature of 260 K [66, 67]. We use arrows to indicate the locations of the \( q \) plotted on a logarithmic intensity scale and collected within a temperature range of roughly 88 K to 100 K, which is well below the main reflection at \( G = (110) \). In previous investigations of the CDW phase in (TaSe\(_4\))\(_2\)I, the \( q = (110) \) and \( (1\bar{1}0) \) satellite reflections were not reported at all [87, 88], most likely due to the limited resolution and laboratory X-ray beam intensities available at the time. Because it is quite difficult to account for all of the factors that determine the relative intensities of satellite spots in a CDW phase [88, 100–102], we will leave a detailed analysis of the faintness of the first-order satellites for future works. However, in other works [88, 100–102], the relative intensities of CDW satellite reflections were well captured by a Bessel-function harmonic modulation ansatz in which the intensity of each satellite spot is approximated to be proportional to \( J_\eta(2Q \cdot U) \), where \( J_\eta \) is the \( \eta \)th-order Bessel function, \( Q \) is the satellite position, and \( U \) is a position-space vector characterizing the magnitude and direction of the CDW modulation [88, 100–102].

In a set of satellite reflections characterized by the harmonic modulation ansatz \( J_\eta(2Q \cdot U) \), satellite reflections generally exhibit lower intensities at larger values of \( Q \), following a pattern that is well described by just the envelope function of the Bessel function \( J_\eta \). However, it is also possible for some of the satellite reflections, such as the first-order spots \( q = (110) \) and \( (1\bar{1}0) \), to lie close to the zeroes of the Bessel function, and therefore exhibit disproportionately weak intensities compared to their relatively small \( q \) vectors. In the inset panel, we show a schematic depicting the relative locations in the \( k_{x+y} \)-plane of the main and satellite Bragg reflections [where distances between the main \( G = (110) \) reflection (central red circle) and its satellites (blue squares) are not drawn to scale],

a complicated manner as a function of the indices of \( Q = (hkl, mno) \) [88, 100–102]. This effect is frequently modeled using a harmonic modulation ansatz in which the intensity of each satellite spot is approximated to be proportional to \( J_\eta(Q \cdot U) \), where \( J_\eta(x) \) is the \( \eta \)th-order Bessel function, \( Q \) is the satellite position, and \( U \) is a position-space vector characterizing the magnitude and direction of the CDW modulation [88, 100–102]. In a set of satellite reflections characterized by the harmonic modulation ansatz \( J_\eta(Q \cdot U) \), satellite reflections generally exhibit lower intensities at larger values of \( Q \), following a pattern that is well described by just the envelope function of the Bessel function \( J_\eta \). However, it is also possible for some of the satellite reflections, such as the first-order spots \( q = (110) \) and \( (1\bar{1}0) \) in Fig. S9, to lie close to the zeroes of the Bessel function [i.e., at values of \( Q \) for which \( J_\eta(Q \cdot U) = 0 \)], such that they exhibit weak (or vanishing) intensities. This can cause satellite reflections with small \( q \) vectors [like the first-order spots \( q = (110) \) and \( (1\bar{1}0) \) in Fig. S9], which lie at values of \( Q \) for which the envelope function of \( J_\eta \) is large, to nevertheless appear with weak intensities in XRD probes. In practice, it still remains quite difficult to perform a detailed quantitative analysis of the \( Q \)-dependence of satellite spot intensities; therefore we leave such an analysis for future works.

The body-centered arrangement of satellite positions in Fig. S9 also provides new perspective on previous XRD probes of the CDW phase of (TaSe\(_4\))\(_2\)I. In earlier works [87, 88], satellite reflections were observed at \( q_{xy} = [\pm \eta(\frac{2\pi}{a}), \pm \eta(\frac{2\pi}{a})] \), where \( \eta \approx 0.05 \) and \( q = q_{xy} + q_z \delta \); specifically, the authors of those works did not observe the faint \((mn) = (11)\) and \((1\bar{1}1)\) satellite spots that we observe in Fig. S9. We therefore now recognize that the satellite reflections with the smallest in-plane \( q \) vectors previously observed in [87, 88] were in fact higher-order reflections with \( q_{xy} = (mn) = 2(22) \) and \( 2(\bar{2}2) \), and that the earlier experiments in those works most likely did not observe the
where η appear near all main reflections 4 phase of our (TaSe 2) sample is body-centered tetragonal [100, 102, 103] which we will later confirm by comparing the scans presented in this figure and in Fig. S9 to the 2D reciprocal-space maps (RSMs) shown in Fig. S11, we interpret the satellite peaks observed in this scan as lying at a separation of 2Δ [see the inset panel for a schematic depicting the relative locations (not drawn to scale) and indices of the main (red circle) and satellite (blue squares) reflections]. Along with the scan presented in Fig. S9, this implies that, if the CDW-modulated phase is body-centered tetragonal, then the satellite peaks in the CDW phase of our (TaSe 2)I sample should exhibit a spacing governed by the reciprocal basis vectors in Eq. (11), such that they appear near all main reflections G at q = (mno) = [mη(±π/m), nη(±π/n), oδ(±π/2)], where η = 0.027 ± 0.001, δ = 0.012 ± 0.001, and m + n + o = μ, μ ∈ 2Z.

Next, we determine the satellite spacing along the k 3 direction by performing a line scan with varying q 3 at a fixed position in the k xy plane Q xy = (hk, mn) = (11, 11), where Q xy = G xy + q xy and G = G xy + q 3 c 3. In this notation, when q 3 = 0, our scan passes through the previous satellite reflection Q = (hk, mno) = (110, 110) in Fig. S9. The results of our scan along q 3 near the G = (110) main reflection are plotted in Fig. S10 on a linear intensity scale. We observe that the satellite spots in Fig. S10 exhibit a nearly regular q 3 separation of [0.0247 ± 0.0020](2π/a). As discussed previously in the text surrounding Eq. (11), anticipating that the CDW phase in (TaSe 2)I exhibits a body-centered (tetragonal) modulated structure, which implies that only satellite spots satisfying m + n + o = μ, μ ∈ 2Z are visible in XRD probes [100, 102, 103], then we interpret the satellite peaks in Fig. S10 as lying at a separation of 2Δ. This implies that the first visible satellite spot in Fig. S10, which lies at q = [0.027(±π/a), 0.027(±π/a), 0.0247(±π/a)], is actually the q = (112) reflection, i.e. a reflection located q 3 = 2Δ above the k 3 = 0 plane. Along with the in-plane satellite positions q xy = (mn) previously determined in Fig. S9, this implies that, if the CDW-phase modulation basis vectors of our (TaSe 2)I sample are body-centered tetragonal [Eq. (11)], then the locations of the satellite reflections are given by q = (mno) = [mη(±π/m), nη(±π/n), oδ(±π/2)], where η = 0.027 ± 0.001, δ = 0.012 ± 0.001, and m + n + o = μ, μ ∈ 2Z.

In order to complete our determination that the CDW-modulated structure is body-centered tetragonal, and to confirm the values of η and δ obtained from the line scans in Figs. S9 and S10 respectively, we additionally recorded detailed 2D RSMs near the G = (420) and (620) main reflections. In Fig. S11(a) [Fig. S11(b)], we show an RSM collected in the k x,y plane (k x + y = constant) in the vicinity of the G = (420) [(620)] main reflection, plotted on a logarithmic intensity scale. As in Figs. S9 and S10, data for the RSMs shown in Fig. S11 was collected within a temperature range of 88 K to 100 K, which is well below the reported CDW transition temperature of 260 K [66, 67]. The positions of the satellite reflections labeled in Fig. S11 agree with the modulation vectors q = (mno) = [mη(±π/m), nη(±π/n), oδ(±π/2)], where η = 0.027 ± 0.001 and δ = 0.012 ± 0.001, obtained from the line-scan data shown in Figs. S9 and S10, as well as

FIG. S10. (Color online) XRD scan along the k 3 direction through the Q = (hkl, mno) = (110, 110) satellite reflection in Fig. S9, plotted on a linear intensity scale and collected within a temperature range of roughly 88 K to 100 K, which is well below the reported CDW transition temperature of 260 K [66, 67]. The satellite peaks along this scan exhibit a nearly regular spacing of [0.0247 ± 0.0020](2π/a) in the q 3 direction. Anticipating that the CDW-modulated structure, like the unmodulated parent structure of (TaSe 2)I, is body-centered tetragonal [100, 102, 103] [which we will later confirm by comparing the scans presented in this figure and in Fig. S9 to the 2D reciprocal-space maps (RSMs) shown in Fig. S11], we interpret the satellite peaks observed in this scan as lying at a separation of 2Δ [see the inset panel for a schematic depicting the relative locations (not drawn to scale) and indices of the main (red circle) and satellite (blue squares) reflections]. Along with the scan presented in Fig. S9, this implies that, if the CDW-modulated phase is body-centered tetragonal, then the satellite peaks in the CDW phase of our (TaSe 2)I sample should exhibit a spacing governed by the reciprocal basis vectors in Eq. (11), such that they appear near all main reflections G at q = (mno) = [mη(±π/m), nη(±π/n), oδ(±π/2)], where η = 0.027 ± 0.001, δ = 0.012 ± 0.001, and m + n + o = μ, μ ∈ 2Z.
FIG. S11. (Color online) 2D reciprocal space maps (RSMs) recorded near the $G = (420)$ (a) and (620) (b) main reflections in our $(TaSe_4)_2I$ sample, collected in the $k_{x,y}$- and $k_{x+y,z}$-planes, respectively. In both (a,b), satellite intensities are plotted on a logarithmic color scale, as indicated by the color bar below (b). As in Figs. S9 and S10, data for the RSMs shown in Fig. S11 was collected within a temperature range of 88 K to 100 K, which is well below the reported CDW transition temperature of 260 K [66, 67]. In both (a,b) we label the indices $q = (mno)$ of the visible satellite spots closest to the main reflections $G$, whose positions agree with the modulation vectors $q = q_{xy} + q_z \delta = (mno) = [m\eta(2\pi a), n\eta(2\pi a), o\delta(2\pi c)]$, where $\eta = 0.027 \pm 0.001$ and $\delta = 0.012 \pm 0.001$, obtained from the line scans shown in Figs. S9 and S10. In (a), data for our RSM was collected at $q_z = 0.024(2\pi/c)$, i.e. $2\delta$ above the main $G = (420)$ reflection, which is itself only residually visible in (a); therefore, the indicated yellow spots in (a) at $q_x = q_y = \pm \eta(2\pi a) \ |q_{xy}| = \eta\sqrt{2}(2\pi a) = 0.039(2\pi a)$] correspond to the $q = (112)$ and $q = (112)$ satellite reflections.

To summarize, we have both performed XRD line scans (Figs. S9 and S10) and collected XRD RSMs (Fig. S11) along the $k_{x,y}$, $k_{x+y}$, and $k_z$ directions, revealing a pattern of satellite reflections that implies that the CDW-modulated structure in $(TaSe_4)_2I$ is body-centered tetragonal, and thus that it respects the same point group symmetries and exhibits satellite reflections with the same reciprocal Bravais lattice geometry (but not with the same lattice spacing) as its high-temperature chiral crystal structure in SG 97 ($I422$).

Finally, to obtain an estimate for the critical temperature $T_C$ of the transition in $(TaSe_4)_2I$ from the low-temperature CDW phase to the high-temperature Weyl-semimetal phase, we measured the temperature dependence of the satellite reflection intensities. In Fig. S12, we plot the summed intensities of all satellite reflections in the vicinity of the $G = (110)$ main reflection as a function of temperature. We observe that all satellite spots simultaneously disappear at a transition temperature of $T_C = 248$ K, which is representative of a transition away from a CDW phase. This value of $T_C$ is slightly lower than, but still in close agreement with, the value of $T_C = 260$ K measured in previous works [66, 67, 87, 104].

I.2 ARPES Experiments

ARPES measurements were performed at the high-resolution branch of beamline I05 of the Diamond Light Source (DLS) with a Scienta R4000 analyzer. The photon energy range for the DLS was 30-220 eV. During measurements, $(TaSe_4)_2I$ samples were kept at a pressure of $< 1.5 \times 10^{-10}$ Torr. The angles of the emitted photoelectrons were
FIG. S12. (Color online) Total measured satellite intensity in our \((\text{TaSe}_4)_2\text{I}\) sample in the vicinity of the \(G = (110)\) main reflection as a function of temperature. The blue squares represent collected data and the dashed line is a smooth, close-fit interpolation to guide the eye. We observe that all satellites simultaneously disappear at \(T_C = 248\) K, representing a signature of a transition away from a CDW phase.

measured with a resolution of 0.2°, and their energies were measured at an overall resolution of < 15 meV. After samples were glued to the sample holder, they were then cleaved in situ to expose the (110)-surface, which is the favored cleavage plane of \((\text{TaSe}_4)_2\text{I}\) [65].

In Fig. S13, we plot the high-resolution band dispersion along \(\Gamma Z\) measured using an incident photon energy of 72 eV. Measurements of the low-temperature and high-temperature phases of \((\text{TaSe}_4)_2\text{I}\) were performed at 100 K and 270 K, respectively, where previous literature has determined that a CDW transition occurs in \((\text{TaSe}_4)_2\text{I}\) at \(T_C \approx 260\) K [66, 67] [though our XRD data in SM I.1 (Fig. S12) shows the emergence of a CDW at the slightly lower temperature of \(T_C = 248\) K]. We were unable to perform measurements below 100 K due to a significant charging effect, which we attribute to a rapid increase in resistance. Specifically, in insulating materials, such as our \((\text{TaSe}_4)_2\text{I}\) sample in its CDW phase, the emission of photoelectrons depletes the total sample charge. This results in a finite surface voltage that reduces the kinetic energies of subsequent outgoing photoelectrons, which we observe in our ARPES measurements. Conversely, in the high-temperature data [Fig. S13(b)], the spectrum does not exhibit a charging effect, but is instead visibly affected by thermal broadening. To set the Fermi energy, we placed our \((\text{TaSe}_4)_2\text{I}\) sample in contact with polycrystalline Au, whose photoemission spectrum is well known. In agreement with previous works [65, 105], both the high- and low-temperature ARPES spectra of our \((\text{TaSe}_4)_2\text{I}\) sample showed signatures of strong polaronic effects that renormalize the electronic band structure to higher binding energies by \(E_p \approx 0.22\) eV.

In Fig. S13, we show the band dispersion of our \((\text{TaSe}_4)_2\text{I}\) sample measured along \(\Gamma Z\) through ARPES. In both the low- and high-temperature phases (upper and lower panels, respectively, in Fig. S13), the valence bands show two components with a separation of about 0.1 eV. We observe that the energy gap between the top of the valence band and the bottom of the conduction band is roughly 0.12 eV in the low-temperature phase [Fig. S13 (upper panels)], but is significantly smaller (< 0.04 eV) in the high-temperature phase [Fig. S13 (lower panels)]. We attribute this change in gap size to a transition from a low-temperature phase with a CDW-induced band gap into the high-temperature Weyl-semimetal phase predicted in this work. At both of the sample temperatures used in our ARPES probes (100 K and 270 K), the energy bands display strong dispersion in the \(k_z\) direction (see Fig. S13).
FIG. S13. (Color online) Band dispersion of (TaSe$_4$)$_2$I as measured in angle-resolved photoemission (ARPES) experiments. In the upper (lower) panels, we respectively plot the raw ARPES data, the second derivative of the data along ΓZ, and the stacking energy distribution curves (EDCs) at 100 K (270 K). The valence and conduction bands are respectively indicated by dashed magenta and green lines. In the raw data, we mark the polaron gap, the CDW-induced band gap (in the low-temperature phase), and the valence band splitting. The blue curves in the EDC plots (rightmost panels) mark the value of $k_z$ at which the gap between the top of the valence band and the bottom of the conduction band is at a minimum.
J. Understanding the Electronic Structure of (TaSe₄)₂I from the Perspective of Filling-Enforced Gaplessness

As shown in Figs. 1(d) and 4(a) of the main text, in the high-temperature Weyl semimetal phase of (TaSe₄)₂I in the body-centered tetragonal SG 97 (I422), the electronic structure is quasi-1D, and the entire Fermi surface lies in the vicinity of the \( k_z = \pm \pi/c \) planes. However, in SG 97, generic points in the \( k_z = \pm \pi/c \) planes are not fixed by a symmetry (e.g. \( C_{2z} \times T \)), as they would be in a primitive tetragonal structure with a periodicity of \( c \) in the \( z \) direction [though the high-symmetry and TRIM (i.e. non-generic) points \( P \) and \( N \) conversely do lie in the \( k_z = \pm \pi/c \) planes in SG 97, as shown in Fig. 1(c) of the main text] [72, 73, 78]. Therefore, it is natural to ask, in a real material such as (TaSe₄)₂I, whose electronic structure is not fine-tuned like that of a toy model, why the entire Fermi surface remains concentrated near \( k_z = \pm \pi/c \) in the absence of a pinning symmetry. In this section, we will show how the shape and localization of the Fermi surface of (TaSe₄)₂I derive from the electronic structure of decoupled TaSe₄ chains, which do exhibit nonsymmorphic-symmetry- and electronic-filling-enforced band crossings near \( k_z = \pi/c \).

We begin by considering a single, isolated TaSe₄ chain that is infinite in the \( z \) (c-axis) direction. Compared to the more familiar space group (SG) symmetry elements [78], the symmetry generators for an isolated TaSe₄ chain are relatively unusual. In addition to \( T \) symmetry, twofold rotations about the \( x \) axis, and \( z \)-direction lattice translation, a TaSe₄ chain is generated by the combination of 45° rotation about the \( z \) axis and \( c/4 \) translation in the \( z \) direction:

\[
T = \{ T|0 \}, \quad C_{2x} = \{ C_{2x}|0 \}, \quad T_z = \{ E|c \}, \quad s_{8z} = \{ C_{8z} | \frac{c}{4} \},
\]

(12)

where \( E \) is the identity element. An isolated TaSe₄ chain is therefore invariant under the non-crystallographic chiral rod group \((p8\overline{2}22)_{RG}\) [74, 75], given in the notation of [31]. Specifically, because the eightfold screw symmetry \( s_{8z} \) is not an element of any of the 230 3D SGs [78], then \((p8\overline{2}22)_{RG}\) does not have a space supergroup, unlike the 75 crystallographic rod groups [76, 77]. We will later show in this section that the process of forming a 3D crystal by coupling an array of TaSe₄ chains in the \( xy \)-plane, along with the introduction of iodine atoms, breaks the non-crystallographic \( s_{8z} \) symmetry while preserving its crystallographic square:

\[
s_{4z} = (s_{8z})^2 = \{ C_{4z} | \frac{c}{2} \},
\]

(13)

which we will see to be a symmetry of the conventional cell of (TaSe₄)₂I when translations in the \( xy \)-plane are taken into account.

As discussed in [68, 69], in nonsymmetric symmetry groups, the glide and screw symmetries place constraints on the electronic fillings at which a symmetry-preserving [single-particle (band) or interacting] gap can ever be present. Specifically, in a \( T \)-symmetric, nonsymmorphic 1D crystal with a \( c \) (z-) directed screw axis of the form:

\[
s_{nB} = \left\{ C_n | \frac{B}{n} \right\},
\]

(14)

where \( n, B \in \mathbb{Z}^+ \) and \( n > B \), insulating gaps are only permitted (absent non-minimally connected bands [13, 79, 106, 107]) at the electronic fillings [68, 69]:

\[
\nu \in 2 \left( \frac{n}{B} \right) \mathbb{Z}.
\]

(15)

This is because the \((B/n)c\) translation in \( s_{nB} \) divides each unit cell into \( n/B \) symmetry-related segments in which states in position-space (atomic orbitals in the single-particle limit) are twofold degenerate due to Kramers’ theorem (\( T \) symmetry). Consequently, in the terminology of [68, 69], \( 2(n/B)\mathbb{Z} \) is the “minimal-insulating filling.” Absent interactions, Eq. (15) implies that spinful bands in a 1D screw-symmetric crystal appear with a minimal connectivity of \( 2(n/B) \) [68, 69, 108], and that they exhibit filling-enforced (nodal) degeneracies [11, 69] (i.e., filling-enforced gaplessness) at other (frequently even) values of the system filling \( \nu \).

Using Eq. (15), we determine that an isolated TaSe₄ chain in rod group \((p8\overline{2}22)_{RG}\), for which \( n = 8 \) and \( B = 2 \) [Eq. (12)], exhibits a minimal-insulating filling of:

\[
\nu_{\text{chain}} \in 8\mathbb{Z}.
\]

(16)

We next compare \( \nu_{\text{chain}} \) to the number of electrons in a single TaSe₄ chain at both charge neutrality and at its oxidation state when paired with iodine atoms in 3D (TaSe₄)₂I crystals. To begin, the electronic configurations of Ta and Se are respectively given by [109]:

\[
\text{Ta} \equiv [\text{Xe}]4f^{14}5d^36s^2, \quad \text{Se} \equiv [\text{Ar}]3d^{10}4s^24p^4,
\]

(17)
such that Ta and Se respectively carry the core and valence electron numbers:

\[ N^v_{Ta} = 68, \ N^v_{Se} = 5, \ N^c_{Se} = 28, \ N^c_{Se} = 6. \]  \hspace{1cm} (18)

A single TaSe4 chain consists of four repeated units of Ta atoms and rectangles with Se atoms on each of the four corners [Fig. 1(a,b) of the main text]. Therefore, a single TaSe4 chain carries the total core and valence electron numbers:

\[ N^v_T = 4N^v_{Ta} + 16N^c_{Se} = 720, \ N^c_T = 4N^c_{Ta} + 16N^v_{Se} = 116. \]  \hspace{1cm} (19)

First, because \( N^v_T \mod 8 = 0 \), then a gap is permitted between bands induced from the core orbitals and those from the valence atomic orbitals. Therefore, for now, we will focus our analysis on the valence atomic orbitals and electrons. In a 3D crystal of (TaSe4)2I, there are 2 iodine atoms per each TaSe4 chain. Because iodine atoms prefer to realize the ionic oxidation state \( \Gamma^- \) by removing an electron from another atom, then, anticipating constructing a 3D crystal of TaSe4 chains and iodine atoms, we remove two valence electrons from the isolated TaSe4 chain that we are presently analyzing:

\[ \tilde{N}^v_T = N^v_T - 2 = 114, \]  \hspace{1cm} (20)

realizing an overall chemical formula of \( [(TaSe_4)_4]^{2+} \). Because \( \tilde{N}^v_T \mod 8 \neq 0 \), then we predict that an isolated \( [(TaSe_4)_4]^{2+} \) chain is a filling-enforced semimetal. Specifically, if all of the bands in an isolated \( [(TaSe_4)_4]^{2+} \) chain are minimally connected, then \( \tilde{N}^v_T \mod 8 = 2 \) implies the presence of a quarter-filled set of eight connected, spinful bands (four connected bands per spin in the limit of vanishing SOC) at the Fermi level. In Fig. S14(a), we show the band structure of a single, isolated TaSe4 chain calculated from first principles with the Fermi level set to that of \( [(TaSe_4)_4]^{2+} \). The band structure in Fig. S14(a) was specifically obtained by removing all of the atoms in the conventional cell of a 3D crystal of \( (TaSe_4)_2I \) except for a single TaSe4 chain, increasing the cell volume to fully decouple the remaining TaSe4 chains, manually restoring the weakly broken \( s_{82} \) screw symmetry of the decoupled chains [Fig. 1(a,b) of the main text and Eq. (12)], and calculating the electronic structure along the \( k_z \) direction with the Fermi level lowered by two electrons to that of \( [(TaSe_4)_4]^{2+} \). To highlight the role of filling-enforced gaplessness, the band structure in Fig. S14(a) was calculated without spin-orbit coupling (SOC); therefore, away from \( k_z = 0, \pi/c \), the bands in (a) are spin-degenerate. At \( k_z = \pi/c \) in Fig. S14(a), we observe a half-filled, fourfold nodal degeneracy (two degenerate states per spin), which is locally protected by the combination of spinless \( T \) and \( (s_{82})^2 = s_{42} \) screw symmetry [Eq. (13)]. This confirms that \( [(TaSe_4)_4]^{2+} \) is a filling-enforced semimetal with a quarter-filled eightfold band connectivity at \( E_F \) (four connected bands per spin without SOC), as discussed in the text following Eq. (20).

We pause to note that, as shown in [69, 110–113], if we were to introduce SOC, which is generically present and non-negligible in a real \( [(TaSe_4)_4]^{2+} \) chain, then the nodal degeneracy at \( k_z = \pi/c \) in Fig. S14(a) would split into a pair of twofold linear crossings at time-reversed values of \( k_z \), realizing an “hourglass”-like band structure. However, whether or not SOC is present, a symmetry-preserving gap still cannot be opened in \( (p8_2 2 2)_{RG} \) at \( \nu = \tilde{N}^v_T \) [Eqs. (16) and (20)]. Therefore, \( [(TaSe_4)_4]^{2+} \) would remain a filling-enforced (hourglass) semimetal if SOC were introduced. However, for simplicity, we will continue to artificially neglect SOC until the final stage of the discussion in this section, and will for now consider all of the bands in the electronic structure of a single \( [(TaSe_4)_4]^{2+} \) chain to be spin-degenerate.

While continuing to focus on a single \( [(TaSe_4)_4]^{2+} \) chain, we next restore charge neutrality by introducing two iodine (\( I^- \)) ions. Specifically, we add two \( I^- \) ions to each 1D unit cell of the previous isolated \( [(TaSe_4)_4]^{2+} \) chain – one at \( z = 0 \) and another at \( z = c/2 \). The resulting chain carries a total chemical formula of \( (TaSe_4)_4I_2 \) per unit cell. For simplicity, for now, we will not discuss the locations of the iodine atoms in the \( xy \)-plane, and will instead only consider their effects on the rod group symmetries and the electronic filling. Because there are only two iodine ions, then the ions do not respect the non-crystallographic \( s_{82} \) screw symmetry [Eq. (12)], though they do respect its square \( 4_2 \) [Eq. (13)]. Therefore, the addition of two iodine atoms has lowered the symmetry of the chain to that of the crystallographic rod group \( (p4_2 2 2)_{RG} \) [76, 77], which is generated by:

\[ \mathcal{T} = \left\{ \mathcal{T}\{0 \}, \ C_{2x} = \left\{ C_{2x}\{0 \}, \ T_z = \left\{ E\{c \}, \ s_{42} = \left\{ C_{4_2}\{c/2 \} \right\} \right\}. \right\} \]  \hspace{1cm} (21)

We can determine the minimal insulating filling of \( (p4_2 2 2)_{RG} \) by recognizing that:

\[ P4_2 2 2 = E(\{p4_2 2 2\}_{RG}) \cup T_z(\{p4_2 2 2\}_{RG}), \]  \hspace{1cm} (22)
We could have also equivalently derived \( \nu'_{\text{chain}} \) by recognizing that the 4\(_2\) screw symmetry in \((p4_222)_{RG}\) [Eq. (13)] divides each chain unit cell into 4\(_2\)-related halves, which, along with \( T \) symmetry, enforces a minimal band connectivity of 4 in the single-particle limit [13, 69, 79, 106, 107].

Next, we compare \( \nu'_{\text{chain}} \) to the electronic filling. Because all of the (formerly) valence atomic orbitals of the iodine ions are occupied by electrons, we take all of the iodine ion electrons to be (closed-shell) core electrons [109]. Therefore, from the electronic configuration of an iodine ion:

\[
I^- \equiv [Kr]4d^{10}5s^25p^6, \tag{24}
\]
we obtain:

\[ N_{T^+}^c = 54, \ N_{T^-}^c = 0. \]  

(25)

For a TaSe\(_4\) chain with two iodine atoms [which exhibits a total chemical formula of (TaSe\(_4\))\(_2\)I\(_2\) per 1D unit cell], this implies that:

\[ N_{T^+}^{c'} = N_{T^+}^c + 2N_{T^-}^c = 828, \ N_{T^-}^{c'} = \tilde{N}_{T^-}^{c'} = 114. \]  

(26)

Because \( N_{T^+}^{c'} \mod 4 = 0 \) [Eq. (23)], then we can again take the core electrons and atomic orbitals to be separated from the valence electrons and orbitals by an energy gap, and can restrict focus to the valence states, as we did previously in the text following Eq. (19). Crucially, for the valence electrons, \( N_{T^+}^{c'} \mod 4 \neq 0 \), implying that a TaSe\(_4\) chain, when it is doped with two iodine atoms, remains a filling-enforced semimetal. Specifically, if all of the bands in an isolated (TaSe\(_4\))\(_4\)I\(_2\) chain are minimally connected, then \( N_{T^+}^{c'} \mod 4 = 2 \) implies the presence of a half-filled set of four connected, spinful bands (two connected bands per spin in the limit of vanishing SOC) at the Fermi level of a single TaSe\(_4\) chain doped with two iodine atoms. Because we have previously shown in Fig. S14(a) that the filling-enforced gaplessness of a TaSe4 chain that is missing two electrons manifests as a \( 4 \frac{2}{2} \)\( 4 \) of a single TaSe\(_4\) chain doped with two iodine atoms. Because we have previously shown in Fig. S14(a) that the filling-enforced gaplessness of a single TaSe\(_4\) chain doped with two iodine atoms [i.e. (TaSe\(_4\))\(_4\)I\(_2\)] still respects \( 4 \) screw symmetry [Eq. (13)], then we conclude that the filling-enforced gaplessness of a single chain of (TaSe\(_4\))\(_4\)I\(_2\) also manifests as a filling-enforced nodal degeneracy near \( k_z = \pi/c \).

For our final step towards constructing the 3D unit cell of (TaSe\(_4\))\(_2\)I, we simply superpose (but do not yet couple) two copies of the previous (TaSe\(_4\))\(_4\)I\(_2\) chain. We perform this intermediate step because the conventional cell of (TaSe\(_4\))\(_2\)I consists of two TaSe\(_4\) chains and four iodine atoms, for a total chemical formula of (TaSe\(_4\))\(_8\)I\(_4\) [Fig. 1(a,b) of the main text]. This superposition does not change the symmetry, and therefore the rod group of the superposed chains remains \( (p422)_{RG} \), and the minimal insulating filling remains \( \nu^{c}_{\text{chain}} \in 4Z \) [Eq. (23)]. However, superposing two chains doubles the total number of electronic states, as well as the total number of electrons. We schematically depict the band structure of two decoupled (TaSe\(_4\))\(_4\)I\(_2\) chains in the absence of SOC in Fig. S14(b). Before introducing any coupling between the chains, in the limit of perturbatively weak SOC employed in this section, the bands in Fig. S14(b) away from \( k_z = 0, \pi/c \) are \textit{fourfold degenerate} (one band per spin per chain), and there is now an (unstable) \textit{eightfold} nodal degeneracy at \( k_z = \pi/c \). The instability of this eightfold nodal degeneracy can be understood from two perspectives. First, the symmetries of rod group \( (p422)_{RG} \) cannot stabilize an eight-dimensional corepresentation [11, 12, 78] or spinful eightfold band connectivity [13, 79]. Second, from the perspective of minimal insulating filling, the total number of core and valence electrons in the doubled chain is:

\[ N_{T^+}^{c'} = 2N_{T^+}^c = 1656, \ N_{T^-}^{c'} = 2N_{T^-}^c = 228, \]  

(27)

where \( N_{T^+}, N_{T^-} \) are defined in Eq. (26). Because \( N_{T^+}^{c'} \mod 4 = N_{T^-}^{c'} \mod 4 = 0 \), then an insulating gap is permitted at the Fermi energy in the doubled chain, and hence, the eightfold degeneracy at \( k_z = \pi/c \) in Fig. S14(b) can be gapped without lowering the system symmetry.

Finally, having established the electronic filling of two superposed and decoupled TaSe\(_4\) chains doped with two iodine atoms each, we can construct the 3D unit cell of (TaSe\(_4\))\(_2\)I, which contains the same atoms and exhibits the same (as well as additional) symmetries. We begin by placing an array of \( z \)-directed TaSe\(_4\) chains, separated by a distance \( a \) in the \( x \) and \( y \) directions, at \( (x, y) = (a/2, 0) \) in each unit cell. We then, as previously, remove two electrons from each chain, such that the chains now each have an overall chemical formula \([(TaSe_4)_4]^{2+}\) \textit{as described in the text surrounding Eq. (27)} while preserving \( C_{2z}, s_{4z}, \) and \( C_{4z} \) symmetries as respectively defined in Eqs. (21) and (28). The resulting crystal of \([(TaSe_4)_4]^{2+}\) chains and \( I^- \) ions is identical to the structure shown in Fig. 1(a,b) of the main text. While containing the same number of states and electrons [Eq. (27)] and respecting the same symmetries [Eq. (21)] as the two superposed (TaSe\(_4\))\(_4\)I\(_2\) chains doped with two iodine atoms, this structure also exhibits the filling-enforced gaplessness of a single TaSe\(_4\) chain that is missing two electrons, manifests as a \( 4 \frac{2}{2} \)\( 4 \) of a single TaSe\(_4\) chain doped with two iodine atoms, and therefore respects \( 4 \) screw symmetry [Eq. (13)], then we conclude that the filling-enforced gaplessness of a single chain of (TaSe\(_4\))\(_4\)I\(_2\) also manifests as a filling-enforced nodal degeneracy near \( k_z = \pi/c \).
chains discussed in the text surrounding Eq. (27), the 3D crystal also respects additional symmetries, which include $C_{4z}$ about $x = y = 0$ [Eq. (28)] and translations in the $x$ and $y$ directions:

$$T_x = \{ E|a00 \}, \quad T_y = \{ E|0a0 \}. \quad (30)$$

Furthermore, because the chains are not centered at $x = y = 0$, then the previous generating symmetries of rod group ($P_{422}$)$_{BC}$ [Eq. (21)] now additionally contain translations in the $x\overline{y}$-plane when enforced in the 3D crystal. Specifically, while the previous twofold rotation about the $x$-axis can be expressed in 3D without additional translations:

$$C_{2x} = \{ C_{2z}|000 \}, \quad (31)$$

the previous $4_2$ screw symmetry from Eq. (13) contains additional translations when enforced in 3D about $(x, y) = (0, a/2)$:

$$s_{4z} = \left\{ C_{4z}|a a c \right\}, \quad (32)$$

Crucially, by combining the (inverse of the) new $C_{4z}$ symmetry [Eq. (28)] and the previous $4_2$ screw symmetry [Eq. (32)], we realize a new, purely translational symmetry:

$$T_{BCT} = \left\{ E|a a c \right\}, \quad (33)$$

which we recognize as one of the primitive lattice vectors of a body-centered tetragonal SG [78]. Specifically, we recognize Eqs. (28), (31), and (33), as the generating elements of SG 97 ($I422$), the SG of (TaSe$_4$)$_2$I [62]. Using the MINSUP tool on the Bilbao Crystallographic Server [72, 73, 114], we confirm that SG 97 ($I422$) is indeed a $k$-type index-2 supergroup of SG 93 ($P_{422}$), the space supergroup of the rod group of decoupled TaSe$_4$ chains and iodine atoms [Eq. (22)], and is specifically generated by:

$$I422 \equiv E(P_{422}) \cup T_{BCT}(P_{422}), \quad (34)$$

where $T_{BCT}$ is defined in Eq. (33).

Though the body-centered structure generated by Eqs. (28), (31), and (33) contains all of the same symmetries as the 3D primitive tetragonal crystal [SG 93 ($P_{422}$)] that we previously constructed from two [(TaSe$_4$)$_2$]$^{2+}$ chains and four I$^-$ ions [text surrounding Eq. (28)], it contains half as many atoms (states) per unit cell as the previous structure in SG 93. Furthermore, because the generating translations in the body-centered structure [Eq. (33) and its $C_{2z}$ and $C_{4z}$ conjugates] are shorter than the previous lattice translation $T_z = \{ E|0 \}$ of isolated TaSe$_4$ chains [Eq. (21)], then the TRIM points in the larger BZ of the body-centered structure (SG 97) do not lie in the same locations that they did previously in the smaller BZ of the larger (conventional) cell (SG 93). This has the effect of unfolding the weak-SOC (unstable) eightfold degeneracy at $k_z = \pi/c$ in Fig. S14(b) into a pair of fourfold nodal planes at $k_z = \pm \pi/c$ in the BZ of the body-centered structure. When the effects of SOC and interchain coupling in the $x\overline{y}$-plane are incorporated, the nodal planes at $k_z = \pm \pi/c$ split into 48 Fermi-surface WPs away from $k_{x,y} = 0$ [Table I of the main text], as well as four time-reversal pairs of $C_{4z}$-symmetry-enforced chiral fermions (WPs) below the Fermi energy [Fig. 1(d) of the main text and Fig. S14(c)]. In agreement with our determination that the eightfold degeneracy at $k_z = \pi/c$ in Fig. S14(b) can be fully gapped without breaking a symmetry, the eight $C_{4z}$-symmetric chiral fermions along $k_z = k_y = 0$ in (TaSe$_4$)$_2$I could in principle be removed through a (very large) band inversion. Specifically, while (TaSe$_4$)$_2$I is an “enforced semimetal” in the nomenclature of [71] (further details available at https://topologicalquantumchemistry.org/#/detail/35190 [13, 71–73]), because all of the elementary band representations in SG 97 are twofold-connected [72, 73, 79], then the $C_{4z}$-symmetric WPs along $k_x = k_y = 0$ can be removed by (symmetry-preserving) pairwise annihilation. Crucially, while the momentum separation of the chiral fermions at $k_z = \pm \pi/c$ is large in the BZ of 3D (TaSe$_4$)$_2$I in body-centered SG 97 ($I422$), it is still small in the limit of decoupled chains, because $k_z = \pm \pi/c$ are related by $(2\pi/c)\hat{z}$, which is a reciprocal lattice vector in the primitive tetragonal structure of decoupled chains [SG 93 ($P_{422}$), generated by Eqs. (21) and (30)].

As discussed in the main text, though the $C_{4z}$-symmetric WPs along $k_x = k_y = 0$ in (TaSe$_4$)$_2$I lie below the Fermi level and sit in narrowly-separated groupings with compensating chiral charges [Fig. S14(c)], and are thus unlikely to contribute in experiment to transport or exhibit observable Fermi-arc surface states, we have shown that they still play a key role in understanding the symmetry-enforced band connectivity of (TaSe$_4$)$_2$I. Specifically, although
the Fermi pockets and $C_4$-enforced WPs are not pinned to $k_z = \pm \pi/c$ in SG 97, they still appear localized close to $k_z = \pm \pi/c$ in the electronic structure of (TaSe$_4$)$_2$I in its high temperature phase [Fig. 1(d) of the main text]. This occurs precisely because interchain coupling is weak in (TaSe$_4$)$_2$I [62], such that the Fermi pockets and WPs remain (perturbatively) close to their original, filling-enforced location(s) ($k_z = \pi/c$) in the electronic structure of decoupled TaSe$_4$ chains [Fig. S14(a)].

In summary, in this section, we have shown that the Fermi surface of the high-temperature (Weyl semimetal) phase of (TaSe$_4$)$_2$I is localized in the vicinity of $k_z = \pm \pi/c$ because (TaSe$_4$)$_2$I derives from weakly coupled TaSe$_4$ chains, which exhibit filling-enforced nodal degeneracies close to $k_z = \pi/c$ [Fig. S14(a)]. This provides additional insight on the CDW phase in (TaSe$_4$)$_2$I. Specifically, while we have shown in Table II of the main text and in SM I.1 that the CDW gap in (TaSe$_4$)$_2$I originates from coupling 3D WPs with opposite chiral charges, the symmetry and filling analysis performed in this section suggests that the CDW phase could in principle also be constructed by weakly coupling an array of interacting, filling-enforced semimetallic wires with nonsymmorphic rod group ($p4_222$)$_{RG}$. In this construction, each wire would then become gapped by electron-electron interactions, either through symmetry-lowering in the mean-field, or through another, more exotic mechanism that has not yet been elucidated [11, 68, 115]. To conclude, the presence of a CDW in (TaSe$_4$)$_2$I, which opens a symmetry- and filling-enforced gap in the limit of decoupled TaSe$_4$ chains, is also consistent with the recent recognition that filling-enforced semimetals; such as SrIrO$_3$ [69, 116, 117], CuBi$_2$O$_4$ [12, 115, 118], and organic stable radicals [119]; are often susceptible in experiment to interacting instabilities.