This is the accepted manuscript made available via CHORUS. The article has been published as:

Importance of electron correlations in understanding photoelectron spectroscopy and Weyl character of MoTe_2

Niraj Aryal and Efstratios Manousakis

Phys. Rev. B **99**, 035123 — Published 11 January 2019

DOI: [10.1103/PhysRevB.99.035123](https://doi.org/10.1103/PhysRevB.99.035123)

Importance of electron correlations in understanding the photo-electron spectroscopy and the Weyl character of MoTe₂

Niraj Aryal⁽¹⁾ and Efstratios Manousakis^(1,2)

⁽¹⁾ *Department of Physics and National High Magnetic Field Laboratory,
Florida State University, Tallahassee, FL 32306-4350, USA*

⁽²⁾ *Department of Physics, University of Athens, Panepistimiopolis, Zografos, 157 84 Athens, Greece*
(Dated: November 1, 2018)

We study the role of electron correlations in the presumed type II Weyl semimetallic candidate γ -MoTe₂ by employing density functional theory (DFT) where the on-site Coulomb repulsion (Hubbard U) for the Mo 4d states is included within the DFT+U scheme. We show that pure DFT calculations fail to describe important features of the light-polarization dependence of the angular resolved photoemission intensity which can be accounted for by including the role of the Hubbard U. At the same time while pure DFT calculations cannot explain the angular dependence of the Fermi surface as revealed by quantum oscillation experiments (a fact which had raised doubt about the presence of the Weyl physics in γ -MoTe₂) inclusion of such on-site Coulomb repulsion can. We find that while the number of Weyl points (WPs) and their position in the Brillouin Zone change as a function of U, a pair of such WPs very close to the Fermi level survive the inclusion of these important corrections. Our calculations suggest that the Fermi surface of γ -MoTe₂ is in the vicinity of a correlations-induced Lifshitz transition which can be probed experimentally and its interplay with the Weyl physics might be intriguing.

I. INTRODUCTION

Transition metal dichalcogenides (TMDs) have continued to surprise physicists and chemists for decades by providing an avalanche of materials with intriguing chemical, mechanical, electronic and optical properties, of both fundamental and technological implications¹⁻³. Among several other properties of fundamental importance in physics, more recently, DFT calculations⁴⁻⁶ predict that the lesser known inversion-symmetry-breaking TMDs γ -WTe₂ and γ -MoTe₂ host Lorentz invariance violating type-II Weyl Fermions. In a similar fashion to their type-I counterparts (namely, (Ta,Nb)(As,P)), these materials are predicted to host Weyl nodes (WN) at the boundaries of the electron and hole pockets as well as topological Fermi arcs which connect Weyl nodes of opposite chiralities⁷⁻¹⁰. Different anomalies in the transport experiments, such as extremely high carrier mobility^{11,12}, and chiral anomaly induced negative longitudinal magnetoresistance¹³ are considered indirect evidence of Weyl Fermions.

Angular resolved photoemission spectroscopy (ARPES) is undoubtedly a direct and widely accepted probe of the above mentioned features of the electronic structure and it has successfully identified type-I Weyl candidates by directly imaging the WNs and Fermi arc states^{7,8}. However, in materials which are candidates for realizing type-II Weyl Fermions, ARPES experiments are not as convincing and unambiguous as in the type-I case because of the coexistence of the bulk electron and hole pockets with surface arc-states and the presence of both trivial and non-trivial Fermi arcs^{14,15}. Nevertheless, in the candidate material γ -MoTe₂ several ARPES studies have claimed to observe Weyl points (WPs) and non-trivial Fermi arcs in agreement with the DFT calculations¹⁶⁻¹⁹. On the other hand, the pre-

dicted electron and hole pockets from DFT calculations, which give rise to the WPs and non-trivial topology, fail to explain the experimentally observed quantum oscillation (QO) frequencies²⁰ in MoTe₂. This failure of the DFT calculations to explain the QO frequencies has raised a certain degree of doubt about the existence of these WPs, given the fact that they were the predictions of such calculations. Also, the high sensitivity of the WPs (of their number, their location and even of their presence) to the slight change in the lattice parameters^{4,21}, the fact that even the presence of Fermi arcs does not necessarily imply the existence of the WPs and non-trivial topology^{14,22} and the approximate way of incorporating the exchange-correlation effects in the DFT, upon which all the predictions about Weyl materials are based, has made this field both exciting and controversial. A significant amount of effort and resources have been devoted by the scientific community to understand and possibly use these Weyl and other topological materials in novel electronics, therefore, it is very important to make sure that their characterization, especially with regard to these fundamental properties is accurate.

In Ref. 20, the authors have empirically shown that in order to make any sensible comparison between the experimentally observed QO frequencies and the DFT calculated ones for the Weyl candidate MoTe₂, the relative band energies of the electron and hole pockets have to be shifted in opposite directions by a constant momentum-independent amount of energy (~ 50 meV). These authors also showed that such a shift was enough to explain the ARPES observed band structure. This adjustment of the band structure, however, sheds doubt about the existence of the Weyl points whose presence depends strongly on small amounts of energy displacement of the hole and electron bands.

In this paper, we resolve the above described discrepancy by studying the electronic structure of MoTe₂ by means of the DFT+U method. Our results show that the “ad hoc” shifting of the bands attempted in Ref. 20 is not only empirical but also justified. The electron and hole bands, predicted to form WPs, are the result of hybridization of Mo-*d* and Te-*p* orbitals (see Fig. 7 in Appendix A. Hence, as expected introduction of a Hubbard U term on the Mo *d*-orbital to take into account the effects of Coulomb correlations arising from the more localized nature of this orbital would lead to different contributions to the energy of these bands. A reasonable value of Hubbard U ($\approx 3\text{eV}$) provides a better agreement with the QO and ARPES data. In addition, we demonstrate that in order to explain the polarization dependence of the ARPES results we need to introduce a non-zero U. More importantly, while many other pairs of Weyl points vanish by increasing the value of U an interesting Weyl pair very near the Fermi level is found to survive. Also, our calculations indicate that the system is in close vicinity to a Lifshitz transition and the WPs are much closer to the Fermi level than predicted by DFT calculations and these predictions can be explored experimentally.

II. COMPUTATIONAL DETAILS

The ab-initio calculations for MoTe₂ are carried out using the Quantum Espresso²³ implementation of the density functional theory (DFT) in the GGA framework including spin-orbit coupling (SOC). The Perdew-Burke-Ernzerhof (PBE) exchange correlation functional²⁴ was used with fully relativistic norm conserving pseudopotentials generated using the optimized norm-conserving Vanderbilt pseudopotentials as described in Ref. 25. All GGA+U calculations presented in this paper are done within the rotationally invariant scheme of Liechtenstein *et al.*²⁶. We used a *k*-mesh of $15 \times 9 \times 5$ to sample the BZ for the self-consistent calculations and also an energy cutoff of 650 eV was used after carrying out careful convergence tests. The Fermi surface sheets were visualized using the XCrysden software²⁷. The angular dependence of the quantum oscillation frequencies was calculated using the skeaf code²⁸. Wannier90²⁹ software was used to construct a tight binding Hamiltonian consisting of Mo 4*d* and Te 5*p* states without maximal localization procedure. The Wannierisation method enabled us to calculate the Fermi surface, band structure and Berry curvature using a very dense *k*-mesh. The constructed tight binding Hamiltonian was fed into WannierTools software³⁰ to calculate the slab band structure, slab Fermi surface and to identify the Weyl points.

III. RESULTS AND DISCUSSION

A. Band Structure and Fermi surface with Hubbard U on Mo-*d* orbitals

In Fig. 1 and 2, we present the band structure and Fermi surface of MoTe₂ obtained for different values of a Hubbard U on the Mo-*d* orbital. The electron bands around the *Y* high symmetry point feel the biggest effect as they have the highest proportion of Mo-*d* orbital. This is also seen in the FS diagram (Fig.2 (b) and (c)) where the smaller hole pockets vanish when $U \neq 0$. Furthermore, as these electron bands around the *Y* point are pushed above E_F by the application of U, the electrons in these states occupy the lower energy hole bands. Hence, the hole pockets around the Γ point appear to shrink (i.e., the hole-bands are pushed down relative to the E_F) while they have negligible Mo-*d* contributions. Interestingly, the direction of movement of the electron and hole bands with respect to the E_F is very similar to the prediction of Ref. 20 which was done in order to match the experimental data. Note that the magnitude of the shift of the electron bands is small compared to the value of U because of the low filling factor of these bands.

Fig. 2 (b), (c) and (d), illustrates the evolution of the Fermi surface (FS) for $U=0, 3$ and 5 eV respectively. Both the electron and hole bands are affected, mostly along the k_y direction (i.e., the *Y* high symmetry direction in the band structure shown in Fig. 1(b)); the small electron pocket along this direction disappears and the hole pocket shrinks. Our FS for non-zero U value is consistent with the HSE calculation of Ref. 31. However, the shape of the FS, in particular that of the hole pocket which change as a function of U, is different from ours. As a general trend, the “head” of the “star” hole pocket at Γ shrinks by increasing U, whereas the “arms” stay more or less the same. For $U \sim 4.5$ eV (see Appendix), we find a Lifshitz transition of the hole pocket where its central region disconnects from the “kidney”-like structures as seen in Fig. 2 (d).

B. Comparison with ARPES

In Fig. 3, we compare the ARPES¹⁸ results and the calculated projected density of states for different values of U as obtained from a slab calculation. We can see clearly that DFT overestimates the size of both the hole and electron pockets. In particular, notice the significant difference between the ARPES and DFT hole pocket around the Γ point. The size of the hole pocket decreases significantly, especially around the Γ point when a non-zero U is used. A good agreement with the ARPES experiment is found for $U \sim 3$ eV as seen in Fig. 3(c). As discussed in the previous paragraph, for $U \sim 4.5$ eV a Lifshitz transition occurs, however, beyond that value of U (Fig. 3(d)) the central hole pocket is very small as

FIG. 1. Comparison between the band structure of MoTe₂ with $U = 0$ eV (dotted line) and (a) $U = 3$ eV (b) $U = 5$ eV (solid lines). The inset is a zoomed version of the bands around Γ -point.

FIG. 2. Comparison between bulk Fermi surface of MoTe₂ for (a) $U = 0$, (b) 3 and (c) 5 eV respectively.

compared to that seen in the ARPES. We note that in the ARPES measurement of Fig. 3(a) the central hole pocket is of negligible intensity appearing as white color. However, different regions of the Fermi surface are visible under different polarizations of light as shown in Ref. 22. Next, by calculating the ARPES light-polarization dependence we show that while it cannot be captured by the DFT calculation, the DFT+ U calculation yields a good qualitative account.

The bands that cross the Fermi surface have different distribution of the p and d orbitals which are visible under different directions of polarization of light. We show below by simple symmetry arguments that the matrix elements of the operator $\vec{\epsilon} \cdot \nabla$ (where $\vec{\epsilon}$ is the direction of the light polarization) which enters in ARPES intensity follows the different rules for the different orbital character of the components of each band.

We start by recalling the very general expression for the ARPES intensity^{32–35} in the one-step model, i.e.,

$$I = \frac{e\hbar}{mc} \sum_i B_{ii} |\vec{A} \cdot \langle f | \vec{p} | i \rangle|^2, \quad (1)$$

where B_{ii} are the corresponding matrix elements of the

imaginary part of the retarded single-electron Green's function G_1^+ (i.e., the spectral function) in the basis formed by all the initial states $|i\rangle$ of the electronic system in the solid, i.e.,

$$B_{ii} = -\frac{1}{\pi} \langle i | \text{Im} G_1^+ | i \rangle. \quad (2)$$

Here the electromagnetic vector potential is a vector parallel to the direction $\vec{\epsilon}$ of the polarization of the incident photon.

Here we wish to identify for a given direction of the light polarization which orbital characters of each band does not contribute to the ARPES intensity.

We will consider the usual geometry of scattering in ARPES where the detected photo-ejected electron has momentum parallel to the surface³², i.e.,

$$|f\rangle = |\mathbf{k}_{||}\rangle, \quad (3)$$

$$\langle \vec{r} | \mathbf{k}_{||} \rangle \sim e^{i(k_x x + k_y y)}, \quad (4)$$

where the x and y directions are the two directions parallel to the slab surface in our calculation. The initial

FIG. 3. Comparison between ARPES and DFT calculation. (a) ARPES measured Fermi surface taken from Ref. 18. (b,c,d) Calculated projected local density of states at (0,0,1) surface for the slab evaluated at $E = E_F$ for $U=0, 3$ and 5 eV respectively. The surface Fermi-arcs are shown in red and they are in similar positions as found by ARPES.

FIG. 4. Comparison between ARPES and the results of the DFT calculation of the polarization dependence. Figs. (a,d) and (c,f) are the calculated bulk FS for $U=0$ and $U=3$ eV calculations respectively whereas (b) and (e) are ARPES measured FS taken from Ref. 22 for the vertical and horizontal polarization of light respectively. In Figs. (a) and (c) the contribution from only the Te- (p_x, p_y) and Mo- $(d_{x^2-y^2}, d_{xy}, d_z^2)$ orbitals is included whereas in Figs. (d) and (f), only the contribution for the Te- (p_z) and Mo- (d_{xz}, d_{yz}) orbitals is included. In Fig. b we have added the dotted-line enclosure of the “kidney”-like features of the ARPES to guide the eye since for the case of vertical polarization they are very faint as in our calculations (Fig. c).

state is usually expanded within the unit cell as³²

$$\langle \vec{r} | i \rangle = \sum_{l,m,\alpha} i^l C_{l,m}^\alpha R_l^\alpha(r) Y_{l,m}(\theta, \omega), \quad (5)$$

where α denotes different basis states and $R_l^\beta(r)$ is the radial part of the Bloch wavefunction. In our case the states under consideration contributing to the ARPES intensity are mixtures of p and d character only. Therefore, we write

$$\langle \vec{r} | i \rangle = \sum_{l,\nu\alpha} i^l D_{l,\nu}^\alpha \mathcal{R}_l^\alpha(r) Q_{l\nu}(x, y, z), \quad (6)$$

where $Q_{l\nu}$ are the familiar linear combinations of the $Y_{l,m}$ for a given fixed value of l and for $l = 1$, they are

$$Q_{11}(x, y, z) = p_x, \quad (7)$$

$$Q_{12}(x, y, z) = p_y, \quad (8)$$

$$Q_{13}(x, y, z) = p_z. \quad (9)$$

and for $l = 2$

$$Q_{21}(x, y, z) = d_{z^2}, \quad (10)$$

$$Q_{22}(x, y, z) = d_{x^2-y^2}, \quad (11)$$

$$Q_{23}(x, y, z) = d_{xy}, \quad (12)$$

$$Q_{24}(x, y, z) = d_{xz}, \quad (13)$$

$$Q_{25}(x, y, z) = d_{yz}. \quad (14)$$

Notice that because $\mathcal{R}_l(r)$ is an even function of x , y and z , and because the final state given by Eq. 4 is only a function of x and y , using the form given in Eq. 6 as initial states, it is straightforward to show that the matrix elements of the operator $\vec{\epsilon} \cdot \nabla$ which enters in Eq. 1 follows the following rules for the different orbital character of the components of each band. (a) When $\vec{\epsilon} \parallel \hat{z}$, from the p -orbitals only the p_z character contributes to the intensity. From the d -orbitals, only the d_{xz} and d_{yz} contribute. (b) When $\vec{\epsilon} \perp \hat{z}$, from the p -orbitals only the p_x and p_y character contribute to the intensity. From the d -orbitals, only the d_{xy} and $d_{x^2-y^2}$ and d_{z^2} contribute.

In Fig. 4 we present the results of our calculation where we only include the non-zero contributions for a given direction of light polarization. We also compare these results obtained with $U=0$ (DFT) and $U=3$ eV to the ARPES measurements²². Figs. 4(a),4(d) and Figs. 4(c),4(f) are the calculated bulk FS for $U=0$ and $U=3$ eV calculations respectively, whereas Figs. 4(b),4(e) are the ARPES measured FS for the vertical and horizontal polarization of light respectively. The main feature of the results is that the part of the hole pocket which encircles the Γ point has strong polarization dependence which is captured by our calculation for $U=3$ eV, while the DFT calculation fails to do so. In addition, the “kidney”-like parts of the hole pocket are faint in the case of vertical polarization and more intense in the case of horizontal polarization, a feature which is also present in the $U=3$ eV calculation and is absent in the DFT calculation. We consider these features which can be captured only by the DFT+ U calculation a “smoking gun” that the DFT+ U calculation gives a much better picture of what happens near the FS of γ -MoTe₂.

C. Comparison with Quantum Oscillation Experiment

Direct comparison of the calculated angular dependence with the experimentally measured QO data did not turn out to be straightforward because we found that the calculation shows significant difference between the spin-orbit (SO) split bands. This is in contrary to the experimentally observed QO FFT spectra (see Fig. 4 (b) of Ref. 20) where the twin peaks with similar angular dependence, identified as SO partners, have much smaller SO splittings. Overestimation of SOC strengths in DFT calculations, thereby giving inaccurate values of experimental observables, is not a new finding by itself and has been discussed in previous works³⁶. Hence, we averaged

over the angular dependence of the SO partners which is justified because of their similar angular dependence (see Fig. 10 in the Appendix C for comparison without averaging).

In Ref. 20, it was shown that the DFT calculated angular dependence of the Fermi surface does not agree with the experimentally measured QO frequencies. In particular, a larger than 1000 T QO frequency along the c -axis is absent and this is also true for the QO frequencies reported in Ref. 31. In Fig. 5 the angular dependence of the orbits measured by QO experiments is compared to our results using $U=0, 3$ and 5 eV. In Fig. 5(a) and Fig. 5(b), both the spin-orbit partners are averaged whereas in Fig. 5(c), only the electron partners are averaged, as the smaller of the hole pockets further disintegrates into smaller pieces and hence, they have different angular dependence. Though the size of both the electron and hole pockets decrease as a function of U , the hole pockets are the ones most affected. When the U value is increased in order to obtain the Lifshitz transition, the large hole pocket of size ≈ 1500 T collapses to two small sized pockets of approximate sizes 500 T and 100 T (see also Fig. 8(b) in Appendix B for other values of U). This effect is dramatic and can provide a plausible answer for the absence of large sized hole pockets in the QO experiments. However, as can be inferred from Fig. 4, this is contrary to the observation of the ARPES experiments. On the other hand, if we adopt a moderate value of $U=3$ eV, it can account for most of the Quantum Oscillation frequencies (including the hole orbit along the a -axis) and at the same time can explain the ARPES experiments including the polarization dependence of the intensity. The large hole orbit of size greater than 1000 T not seen in QO experiments along the c -axis can not be accounted with this value of U ; however, it is not unusual in a QO experiment to miss an orbit along a certain direction, especially when the mass of the carrier is very anisotropic^{37,38}. The fact, however, that γ -MoTe₂ is near the Lifshitz transition, which in practical terms means just 25 meV (we find that this corresponds to less than ~ 2 % per chemical formula electron doping) away from the transition, it is expected to have a significant effect in transport phenomena. The reason is that the size of the large hole pocket near Γ changes rapidly by external causes.

D. Fate of the Weyl point on application of U

As noted by other authors^{4,14,21,39}, the exact position and even the presence of WPs is very sensitive to the slightest change in the lattice parameters and also in the different implementations of DFT. Therefore, first we identify the position of the WPs for MoTe₂ by looking at the Berry curvature vector plot. (See Fig. 11 in Appendix D which illustrates the location of the source and sink of Berry curvature.) In total, there are 4 pairs of WPs, each on one quadrant, all of which are located

FIG. 5. Comparison between the experimental angular dependence and the calculated angular dependence for (a) DFT (b) $U=3$ eV (c) $U=5$ eV. In Figs. (b) and (c), the calculated orbits that do not correspond to the experimental ones are drawn with dashed lines.

FIG. 6. Band structure along the Weyl points W_1 and W_2 for (a) $U = 0$ eV, (b) $U = 3$ eV and in (c) for $U = 3$ eV along a direction perpendicular to the W_1W_2 line which passes through the W_2 point and lies on the XY plane.

on the $k_z = 0$ plane and are related by the crystal symmetry. The band structure along these WPs is shown in Fig. 6 (a) which is similar to the one shown in Ref.21.

We also examined how the WPs evolve as a function of U . We found that the number of the WPs and their position in the BZ change with increasing U in a very non-trivial way. For example, for $U=0.75$ eV, we find 12 pairs of WPs out of which only 4 pairs are located on the $k_z = 0$ plane. For $U=2$ eV and $U=4$ eV, there are respectively 8 and 6 pairs of WPs, none of which are located on the $k_z = 0$ plane. Also, we found that the source and sinks of the WPs move farther from one-another as U increases. In Fig.6, we present the band structure along the WPs for $U=3$ eV along the direction W_1W_2 (Fig. 6(b)) and along the perpendicular direction (Fig. 6(c)) where it is shown that one pair of surviving WPs is very close to the E_F . Notice that the WP, which is very close to the FS, seems to be a type-I along the W_1W_2 direction; however, when we calculated the band dispersion along the direc-

tion perpendicular to W_1W_2 (Fig. 6(c)), it becomes clear that its character is of type-II. In Appendix E, we introduce a simple model for an inversion symmetry breaking quasi-2D system and demonstrate how the WPs can be destroyed or created by the presence of the Hubbard U . Using this model we find that the WPs move in k -space with the application of U until they annihilate.

IV. CONCLUSION AND OUTLOOK

In summary, we found that a modest value of U (close to 3 eV) gives good qualitative agreement with both the QO experiments and the ARPES measured Fermi surface. The “smoking gun” that our calculation provides a much more accurate interpretation of the FS of γ -MoTe₂ is that it can explain the polarization dependence of ARPES, while the DFT calculation fails. We find a Lifshitz transition for $U \sim 4.5$ eV and, because of the more accurate account of the experimental results by using a not too different value of U , this suggests that γ -MoTe₂ could be in the close vicinity to this transition produced by the effects of Coulomb correlations. Experimentally, the Lifshitz transition can be probed by a small amount of doping which can be introduced either chemically or by making a device with a back-gate potential which can effectively introduce charge. We find that the number of WPs and their position in the BZ change non-linearly with U . Interestingly, the WPs closest to the Fermi level survive for the U value which accurately describes both ARPES and QO experiments and this is a very positive result of the present work regarding a controversial issue.

Note added. After this paper was submitted for peer review and it was uploaded on the arxiv⁴⁰, another paper⁴¹ was uploaded on the arxiv later discussing the importance of correlations to describe the ARPES measured Fermi surface and the onset of a simple Lifshitz transition different than the one found in the our work. The Lifshitz transition (LT) discussed in this reference (which is now published⁴²) happens at around $U=2$ eV where the small pockets close to the Brillouin zone edge disappear. We have also discussed this occurrence for non-zero U value in the present paper but we have not highlighted it, because the Fermi-surface sheets which are affected by this LT are located far away from the location of the Weyl points inside the Brillouin zone. In our present paper we have focused on a more dramatic Lifshitz transition happening at a higher value of U (>4 eV) which involves the bands forming the Weyl points. Furthermore, one important claim made in the above mentioned publication⁴² is that there is a hybrid Weyl point for a non-zero U value where a type-I and a type-II Weyl point co-exist and it was claimed that mathematically such a state can be considered as an analogue to the black hole horizon. The authors drew this conclusion by looking at the band structure along certain Brillouin-zone directions only. We would like to point out that, as discussed in the present paper (see our Figs. 6(b) and (c)),

both Weyl points are of type-II. In order to prove that a Weyl cone is of type-I, one has to verify that it appears to be of type-I character when plotted along all directions, not just along specific Brillouin-zone directions⁵.

V. ACKNOWLEDGMENTS

This work was supported in part by the U.S. National High Magnetic Field Laboratory, which is funded by NSF DMR-1157490 and the State of Florida.

Appendix A: Orbital projected band structure from DFT

In Figs. 7(a) and 7(b), we present the projected band structure of MoTe₂ onto the Mo-*d* and Te-*p* orbitals respectively. The contribution from other orbitals close to the Fermi level is negligible, hence we focused on only these two kind of orbitals. It is seen that the electron and hole bands have hybridization of the Mo-*d* and Te-*p* orbitals.

Appendix B: Angular dependence of hole pocket as a function of *U*

Since the biggest disagreement with the quantum oscillation (QO) experiments arises from the absence of the large-sized orbits (≥ 1500 T) which correspond to the hole pockets in the DFT calculations, we focus only on these hole pockets. In Fig. 8(a) we illustrate how the biggest of the hole pockets evolve as a function of *U* for the values of *U* ranging from 0 (i.e. DFT) to 5eV. In Fig. 8(b), we show the calculated angular dependence for different values of *U*. The size of the hole pockets decrease as a function of *U*. Notice how the frequency at *c*-axis collapses from 1500 T to 500 T when varying *U* from 4 to 5 eV because of a Lifshitz transition. This is seen in Fig. 8(a) (iv) where the central part of the hole pocket separates from the “kidneys”.

As a verification that the transition occurs near 4.5 eV, we performed calculations for *U*=4.3 eV where we find that this value of *U* is below *U_c* and for *U*=4.7 eV where we find that this value of *U* is above *U_c* as seen in Figs. 9 (a) and (b) respectively.

Appendix C: Comparison of Fermi surface with experimental angular dependence

In the main text, we presented the comparison between the calculated and the angular dependence of the Fermi surface as obtained by quantum oscillation (QO) experiments by averaging the spin-orbit partners in our calculations. This is because the spin-orbit partners seen in the experiments had much less splitting than that seen

FIG. 7. Band structure of MoTe₂ obtained from GGA calculation along $R - \Gamma - X - Y - \Gamma$ high symmetry directions showing the projection onto (a) Mo-*d* orbitals and (b) Te-*p* orbitals. The size of the dots is proportional to the percentage of the corresponding orbitals. The contributions from other orbitals are negligible, hence not shown here for clarity.

in our calculations. In this section, we show the comparison without averaging the spin-orbit partners. Fig. 10 shows the angular dependence of the electron and hole pockets for both of the spin-orbit partners obtained from the DFT calculation. As can be concluded by examining this figure, because of the large size of the effect of the SOC on the orbits not seen in the QO experiments, prevents us from making any meaningful comparison. In the QO experiments there are two very close to each other frequencies for every orbit, which have been interpreted in Ref. 20 as the SO partners. When averaging both frequencies in each of these pairs in the experimental data and in the DFT calculations the identification of the orbits becomes clearer.

Appendix D: Fate of the Weyl points

First, we identify the position of each Weyl point for our MoTe₂ crystal without the application of *U* by looking at the Berry curvature vector plot for $k_z = 0$ in

FIG. 8. (a) Evolution of the biggest hole pocket for different values of U (i) $U=0$ (ii) $U=2$ and (iii) $U=4$ eV (iv) $U=5$ eV. (b) Comparison of the angular dependence of the hole pocket shown in (a) for different values of U .

Fig. 11(a). Our calculation shows Weyl points at $W_1 = (0.102\frac{2\pi}{a}, 0.058\frac{2\pi}{b}, 0.0)$ and $W_2 = (0.104\frac{2\pi}{a}, 0.017\frac{2\pi}{b}, 0.0)$. The Figure clearly shows the source and the sink of the Berry curvature as marked by W_2 and W_1 respectively. In total, there are 4 pairs of Weyl points, each on one quadrant, all of which are located at the $k_z = 0$ plane and are related by the crystal symmetry. The Berry curvature vector plot for $U = 0.75$ eV is shown in Fig. 11(b) which shows that the position of the Weyl point has changed.

Our calculation finds that though the number and position of Weyl points change as a function of U , some pairs of Weyl points survive the introduction of U . The band structure along one such pair of Weyl points is shown in Fig. 12 for different values of U .

Appendix E: A Simple Model of Weyl Semimetal

We present a simple 4×4 lattice model of a Weyl semimetal in order to demonstrate that the Weyl points can be destroyed or created by the action of a Hubbard term and we treat the electronic correlation term within mean-field theory. This corresponds to the GGA+ U approach where the on-site Coulomb repulsion term is also factorized as in the MFT. Though our goal in this paper

FIG. 9. Bulk band structure of MoTe₂ for (a) $U = 4.3$ eV which is before the onset of the LT and (b) $U = 4.7$ eV which is after the LT as indicated by the submerging of the valley of the hole pocket close to the Γ point inside the Fermi sea.

is to understand the effect of U in MoTe₂ specifically, our model is general enough to explain the role of U in the other type-II Weyl candidate, WTe₂, which is of the same crystal family as MoTe₂.

Since the bands forming Weyl points in MoTe₂ are comprised of hybridised Mo- d and Te- p orbitals, we can write an effective 4-band model using these two hybridised orbitals along with the spin degree of freedom as our basis. However, as seen in Fig. 7, they are not of pure d or p character; hence the effect of the Hubbard U term on the Mo d -orbital on each of the bands will be proportional to the product of the U value and the percentage of d orbital in each of the bands. We will call these orbitals A and B, where orbital A refers to the orbital having higher proportion of Mo- d (i.e. d -like) and B refers to the orbital having higher proportion of Te- p orbital (i.e. p -like). Hence, our basis consists of the following four states: $|A \uparrow\rangle, |B \uparrow\rangle, |A \downarrow\rangle, |B \downarrow\rangle$.

FIG. 10. Comparison between the experimental and the DFT calculated angular dependence without averaging the spin-orbit partners where e1 and e2 correspond to electron pockets and h1 and h2 correspond to hole pockets.

1. A 2D Weyl model

First, we write a continuum version of the 2D Dirac model which are centered around (k_x^0, k_y^0) . These Dirac points will be converted to Weyl points after the addition of spin-orbit coupling (SOC) term. In the following subsection, we will discuss a 3D model obtained from this 2D model by stacking these 2D planes on top of each other. Our Hamiltonian for the 2D-Dirac point can be written as:

$$H(\vec{k}) = \begin{bmatrix} 0 & k'^- & 0 & 0 \\ k'^+ & 0 & 0 & 0 \\ 0 & 0 & 0 & k'^- \\ 0 & 0 & k'^+ & 0 \end{bmatrix}, \quad (\text{E1})$$

where $k^\pm = k_x \pm ik_y$ and $k'_{x(y)} = k_{x(y)} - k_{x(y)}^0$.

With the inclusion of a Rashba type spin-orbit interaction of the form $2\lambda(\vec{\sigma} \times \vec{k}) \cdot \hat{z}$ that introduces spin-hopping from A sublattice to B and vice-versa, our Hamiltonian becomes:

$$H(\vec{k}) = \begin{bmatrix} 0 & k'^- & 0 & 2\lambda i k^- \\ k'^+ & 0 & 2\lambda i k^- & 0 \\ 0 & -2\lambda i k^+ & 0 & k'^- \\ -2\lambda i k^+ & 0 & k'^+ & 0 \end{bmatrix}. \quad (\text{E2})$$

This model gives eigenvalues $E = \pm\sqrt{X}$ where, $X = |k'^2| + 4\lambda^2|k^2| \pm 4\lambda k'_x|k|$. The expression for X can be rewritten as $X = k_y'^2 + (k'_x \pm 2\lambda|k|)^2$. Thus, $X = 0$ when $k'_y = 0$ and $k_x = k_x^0 \pm 2\lambda|k|$. The condition for k_x gives two values of k_x where zero energy occurs, i.e., at $\kappa_x^\pm = k_x^0 \pm \sqrt{k_x^{02} - (1 - 4\lambda^2)(k_x^{02} - 4\lambda^2 k_y^{02})}$. $\kappa_x^\pm$ is real if $\frac{(1 - 4\lambda^2)(k_x^{02} - 4\lambda^2 k_y^{02})}{k_x^{02}} \leq 1$. In particular, for $k_x^0 = k_y^0$, we find that we have two zero energy solutions if $\lambda^2 \leq$

FIG. 11. Fig. (a) and Fig. (b) show the vector plot of the x and y component of the Berry curvature on the $k_z = 0$ plane around the Weyl points for $U = 0$ and $U = 0.75$ eV respectively. The relative size of the length of the vector is proportional to the magnitude of the Berry curvature.

0.5. In general, the existence of Weyl points depends on the value of λ, k_x^0 and k_y^0 . In Figs. 13(b) and 13(c), we show the dispersion for values of the Rashba coupling parameter 0 and 0.2 respectively. The former one gives a Dirac cone whereas the latter one gives two Weyl nodes separated along the k_x direction.

2. A 3D Weyl model

Having understood the effect of a Rashba term in this 2D Dirac model, we wish to address the problem which is more relevant to our system of γ -MoTe₂. In γ -MoTe₂, since the d and p orbitals contributing to the Weyl points arise from two different atoms, it is more realistic to consider different onsite energies, say E_A and E_B for the two orbitals. Hence, our Hamiltonian for the 2D-Weyl point take the following form:

$$H_{2D}(\mathbf{k}) = \epsilon \hat{I} + \begin{bmatrix} \delta E & k'^- & 0 & 2\lambda i k^- \\ k'^+ & -\delta E & 2\lambda i k^- & 0 \\ 0 & -2\lambda i k^+ & \delta E & k'^- \\ -2\lambda i k^+ & 0 & k'^+ & -\delta E \end{bmatrix} \quad (\text{E3})$$

where, $\epsilon = \frac{(E_A + E_B)}{2}$, $\delta E = \frac{(E_A - E_B)}{2}$, and $\hat{I}$ is the 4×4 identity matrix.

FIG. 12. Band structure along the Weyl points W_1 and W_2 for (a) $U = 0.75$ eV, (b) $U = 2$ eV and (c) $U = 4$ eV. Notice the character transformation of the W_2 Weyl point from type-II to type-I when changing the value of U from $U=0$ to $U=2$ and beyond along this direction. However, as discussed in the main text, plotting bands along other directions reveals that the character of this Weyl point is of type-II. Also notice that the W_1 Weyl point moves towards E_F significantly for non-zero U .

Since the identity term in Eqn. E3 is just a momentum-independent overall shift of the bands, we will neglect it in the following discussion. The eigenvalues of Eqn. E3 are given by $E = \pm \sqrt{(\delta E)^2 + X}$ which yield gapped Weyl points when $E_A \neq E_B$.

Now, we will stack these gapped 2D Weyl planes along the z direction to form a quasi-2D crystal which can de-

scribe the weak interlayer coupling in these exfoliable materials. Such an approach of constructing higher dimensional topological systems from lower dimensional topological systems have been applied before in Refs. 43 and 44. In doing so, we will consider the effective hopping from A to A orbital to be different from the hopping from B to B orbital. This is realistic for materials like MoTe_2 and WTe_2 because the crystal structure, shown in Fig. 13(a), consists of buckled planes of Mo and Te atoms. In this case the out-of-plane Te-Te atomic distance is smaller than that of the Mo-Mo case. In fact, for these materials, the Mo-Mo distance is even larger than Te-Mo distance, which allows us to even neglect the hopping from Mo-Mo orbitals; however, our qualitative results remain unaffected by either of these two approaches. By using different onsite energies on A and B orbitals and/or different effective hoppings between the same orbitals, we break the inversion symmetry in our model.

Now the Hamiltonian for a 3D lattice becomes:

$$H_{3D}(\mathbf{k}) = -2t_{AB} \cos(k_z) \hat{I} + H'_{2D}(\mathbf{k}), \quad (\text{E4})$$

where, $H'_{2D}(\mathbf{k})$ is similar to $H_{2D}(\mathbf{k})$ except the diagonal δE term is replaced by $Y = \delta E - 2\delta t_z \cos(k_z c)$ such that $\delta t_z = \frac{(t_A - t_B)}{2}$ and $t_{AB} = \frac{(t_A + t_B)}{2}$. The introduction of the “buckling” along the z -direction in our Hamiltonian gives back the Weyl physics which was gapped in the 2D case because $E = \pm \sqrt{Y^2 + X}$ and $Y = 0$ if $\delta E = 2\delta t_z \cos(k_z c)$. Hence, the zero energy solution occurs for $(\kappa_x^\pm, k_y^0, \kappa_z)$ where, $\kappa_z c = \cos^{-1}(\frac{\delta E}{2\delta t_z})$ and the solution exists if $2|\delta t_z| \geq |\delta E|$. Note that the k_z dependent identity term makes the Weyl cone always of type-II nature with a tilt along the k_z direction because $2t_{AB} \geq |\delta t_z|$. Let us analyze the following two conditions:

1. For the equality condition, we obtain a zero energy solution for $k_z c = \pi$. This gives linear dispersion along the k_x and k_y directions but quadratic dispersion along the k_z direction. Hence, the zero energy solution exists for $(\kappa_{x\pm}, k_y^0, \pi)$.
2. For $\delta t > \delta E$, there are four values of $\vec{k}$ which yield a zero energy solution, the following $(\kappa_x^\pm, k_y^0, \kappa_{z\pm})$ where $\kappa_z = \cos^{-1}(\frac{\delta E}{2\delta t_z})$ ($\cos(k_z)$ crosses the $\frac{\delta E}{2\delta t_z}$ line twice when k_z varies from 0 to 2π). Expanding Y around κ_z , we find that $Y(k_z) = 2\delta t_z \sin(\kappa_z) k_z$. Since $\sin(\kappa_z)$ is a constant, the dispersion is linear along k_z as well (it is linear along the k_x and k_y directions before adding the Rashba term which simply shifts them for an appropriate value of the λ parameter.). Next, we will analyze how the inclusion of the Hubbard U term changes the Weyl physics.

FIG. 13. (a) Crystal structure of MoTe₂. The black solid line gives the boundary of the unit cell. Notice the “stacking” of Mo and Te planes along the c axis. (b) and (c) shows the birth of Weyl points out of the Dirac point due to the Rashba term for the 2-D model. In (b) the Dirac cone is four-fold degenerate and centered at (0.1,0.1) whereas in (c) the Dirac cone forms two Weyl points at (0.0544, 0.1) and (0.1835, 0.1) for a value of the Rashba coupling constant $\lambda = 0.2$.

3. The effect of the Hubbard U

In order to analyze the role of Coulomb correlations in the Weyl physics, we add a Hubbard U term U_A on the A orbital and a U_B on the B orbital, where $U_x = f_x U$, $x \in A, B$ such that f_x denotes the percentage of the Mo- d orbital in the x orbital and $f_A \neq f_B$. For the physics we are interested in, using two different Hubbard U terms is qualitatively similar to using only one U term and setting the other to zero. Hence, we will set $U_B = 0$ and write $U_A = g(U)$ where $g(U)$ is some function of U proportional to the percentage of the d orbital. These considerations yield the following Hamiltonian:

$$\hat{H} = \sum_k c_k^\dagger H_{3D}(\mathbf{k}) c_k + g(U) \sum_{i \in A} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} \quad (\text{E5})$$

where, $H_{3D}(\mathbf{k})$ is defined by Eqn. E4 and $c_k^\dagger = (c_{k\uparrow}^{A\dagger}, c_{k\uparrow}^{B\dagger}, c_{k\downarrow}^{A\dagger}, c_{k\downarrow}^{B\dagger})$. Writing $\hat{n}_{i\sigma} = \langle n_{i\sigma} \rangle + (\hat{n}_{i\sigma} - \langle n_{i\sigma} \rangle)$, where the second term is the fluctuation about the average term, we obtain the following mean-field Hubbard Hamiltonian:

$$\begin{aligned} \hat{H}_{MF} = & \sum_k c_k^\dagger H_{3D}(\mathbf{k}) c_k + g(U) \sum_{i \in A, \sigma} \hat{n}_{i\sigma} \langle n_{i\bar{\sigma}} \rangle \\ & + g(U) \sum_{i \in A} \langle \hat{n}_{i\uparrow} \rangle \langle \hat{n}_{i\downarrow} \rangle \hat{I}, \end{aligned} \quad (\text{E6})$$

where σ and $\bar{\sigma}$ denote spins of opposite flavors.

Since MoTe₂ is nonmagnetic, $\langle \hat{n}_{i\uparrow} \rangle = \langle \hat{n}_{i\downarrow} \rangle = \frac{1}{2}$. Then we obtain:

$$\hat{H} = \sum_k c_k^\dagger H_0(k) c_k + \frac{g(U)}{2} \sum_{k, \sigma} \hat{n}_{k\sigma}^A + \frac{g(U)N}{4}, \quad (\text{E7})$$

where, N is the total number of lattice sites. Eqn. E7 reduces to a form similar to Eqn. E4 except of the addition of $\frac{g(U)}{4}$ term in the identity and the diagonal term is replaced by $Y = [\delta E - 2\delta t_z \cos(k_z c) + \frac{g(U)}{4}]$. $Y = 0$ as long as $\cos(k_z c) = \frac{(\delta E + \frac{g(U)}{4})}{2\delta t_z}$. Hence, the Weyl point shifts as a function of U and finally gaps out for large enough U such that $|\delta E + \frac{g(U)}{4}| > 2|\delta t_z|$.

¹ Q. H. Wang, K. Kalantar-Zadeh, A. Kis, J. N. Coleman, and M. S. Strano, Nat. Nanotechnol. **7**, 699 (2012).
² J. Wilson and A. Yoffe, Adv. Phys. **18**, 193 (1969).
³ L. F. Mattheiss, Phys. Rev. B **8**, 3719 (1973).
⁴ Z. Wang, D. Gresch, A. A. Soluyanov, W. Xie, S. Kushwaha, X. Dai, M. Troyer, R. J. Cava, and B. A. Bernevig, Phys. Rev. Lett. **117**, 056805 (2016).
⁵ A. A. Soluyanov, D. Gresch, Z. Wang, Q. Wu, M. Troyer, X. Dai, and B. A. Bernevig, Nature **527**, 495 (2015).

⁶ T.-R. Chang, S.-Y. Xu, G. Chang, C.-C. Lee, S.-M. Huang, et al., Nat. Commun. **7**, 10639 (2016).
⁷ S.-Y. Xu, I. Belopolski, N. Alidoust, M. Neupane, G. Bian, et al., Science **349**, 613 (2015).
⁸ S.-M. Huang, S.-Y. Xu, I. Belopolski, C.-C. Lee, G. Chang, et al., Nat. Commun. **6**, 7373 (2015).
⁹ S.-Y. Xu, N. Alidoust, I. Belopolski, Z. Yuan, G. Bian, Chang, et al., Nat. Phys. **11**, 748 (2015).
¹⁰ N. P. Armitage, E. J. Mele, and A. Vishwanath,

- Rev. Mod. Phys. **90**, 015001 (2018).
- ¹¹ Z. Wang, Y. Zheng, Z. Shen, Y. Lu, H. Fang, F. Sheng, Y. Zhou, X. Yang, Y. Li, C. Feng, and Z.-A. Xu, Phys. Rev. B **93**, 121112 (2016).
 - ¹² C. Shekhar, A. K. Nayak, Y. Sun, M. Schmidt, M. Nicklas, I. Leermakers, Zeitler, *et al.*, Nat. Phys. **11**, 645 (2015).
 - ¹³ X. Huang, L. Zhao, Y. Long, P. Wang, D. Chen, Z. Yang, H. Liang, *et al.*, Phys. Rev. X **5**, 031023 (2015).
 - ¹⁴ F. Y. Bruno, A. Tamai, Q. S. Wu, I. Cucchi, C. Barreteau, A. de la Torre, S. McKeown Walker, S. Riccò, *et al.*, Phys. Rev. B **94**, 121112 (2016).
 - ¹⁵ T. M. McCormick, I. Kimchi, and N. Trivedi, Phys. Rev. B **95**, 075133 (2017).
 - ¹⁶ L. Huang, T. M. McCormick, M. Ochi, Z. Zhao, M.-T. Suzuki, R. Arita, Wu, *et al.*, Nat. Mater. **15**, 1155 (2016).
 - ¹⁷ K. Deng, G. Wan, P. Deng, K. Zhang, S. Ding, E. Wang, M. Yan, Huang, *et al.*, Nat. Phys. **12**, 1105 (2016).
 - ¹⁸ J. Jiang, Z. Liu, Y. Sun, H. Yang, C. Rajamathi, Y. Qi, L. Yang, C. Chen, H. Peng, *et al.*, Nat. Commun. **8**, 13973 (2017).
 - ¹⁹ A. Tamai, Q. S. Wu, I. Cucchi, F. Y. Bruno, S. Riccò, T. K. Kim, M. Hoesch, C. Barreteau, *et al.*, Phys. Rev. X **6**, 031021 (2016).
 - ²⁰ D. Rhodes, R. Schönemann, N. Aryal, Q. Zhou, Q. R. Zhang, *et al.*, Phys. Rev. B **96**, 165134 (2017).
 - ²¹ Y. Sun, S.-C. Wu, M. N. Ali, C. Felser, and B. Yan, Phys. Rev. B **92**, 161107 (2015).
 - ²² A. Crepaldi, G. Autès, A. Sterzi, G. Manzoni, M. Zaccagna, F. Cilento, *et al.*, Phys. Rev. B **95**, 041408 (2017).
 - ²³ P. Giannozzi, S. Baroni, and N. B. *et al.*, Journal of Physics: Condensed Matter **21**, 395502 (2009).
 - ²⁴ J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).
 - ²⁵ D. R. Hamann, Phys. Rev. B **88**, 085117 (2013).
 - ²⁶ A. I. Liechtenstein, V. I. Anisimov, and J. Zaanen, Phys. Rev. B **52**, R5467 (1995).
 - ²⁷ A. Kokalj, J. Mol. Graph. Model. **17**, 176 (1999).
 - ²⁸ P. Rourke and S. Julian, Computer Physics Communications **183**, 324 (2012).
 - ²⁹ A. A. Mostofi, J. R. Yates, G. Pizzi, Y.-S. Lee, I. Souza, D. Vanderbilt, and N. Marzari, Computer Physics Communications **185**, 2309 (2014).
 - ³⁰ Q. Wu, S. Zhang, H.-F. Song, M. Troyer, and A. A. Soluyanov, Computer Physics Communications **224**, 405 (2018).
 - ³¹ Y. Qi, P. G. Naumov, M. N. Ali, C. R. Rajamathi, W. Schnelle, *et al.*, Nat. Commun. **7**, 11038 (2016).
 - ³² M. Lindroos, S. Sahrakorpi, and A. Bansil, Phys. Rev. B **65**, 054514 (2002).
 - ³³ J. Pendry, Surface Science **57**, 679 (1976).
 - ³⁴ C. Caroli, D. Lederer-Rozenblatt, B. Roulet, and D. Saint-James, Phys. Rev. B **8**, 4552 (1973).
 - ³⁵ Z.-H. Zhu, C. N. Veenstra, G. Levy, A. Ubaldini, P. Syers, N. P. Butch, J. Paglione, M. W. Haverkort, I. S. Elfimov, and A. Damascelli, Phys. Rev. Lett. **110**, 216401 (2013).
 - ³⁶ A. Lusakowski, P. Bogusławski, and T. Radzyński, Phys. Rev. B **83**, 115206 (2011).
 - ³⁷ R. Schönemann, N. Aryal, Q. Zhou, Y.-C. Chiu, K.-W. Chen, T. J. Martin, *et al.*, Phys. Rev. B **96**, 121108 (2017).
 - ³⁸ K.-W. Chen, X. Lian, Y. Lai, N. Aryal, Y.-C. Chiu, *et al.*, Phys. Rev. Lett. **120**, 206401 (2018).
 - ³⁹ D. Di Sante, P. K. Das, C. Bigi, Z. Ergönenc, N. Gürtler, Krieger, *et al.*, Phys. Rev. Lett. **119**, 026403 (2017).
 - ⁴⁰ N. Aryal and E. Manousakis, arXiv preprint arXiv:1808.06445 (2018).
 - ⁴¹ N. Xu, Z. Wang, A. Magrez, P. Bugnon, H. Berger, C. Matt, V. Strocov, N. Plumb, M. Radovic, E. Pomjakushina, *et al.*, arXiv preprint arXiv:1808.08816 (2018).
 - ⁴² N. Xu, Z. W. Wang, A. Magrez, P. Bugnon, H. Berger, C. E. Matt, V. N. Strocov, N. C. Plumb, M. Radovic, E. Pomjakushina, K. Conder, J. H. Dil, J. Mesot, R. Yu, H. Ding, and M. Shi, Phys. Rev. Lett. **121**, 136401 (2018).
 - ⁴³ A. A. Burkov and L. Balents, Phys. Rev. Lett. **107**, 127205 (2011).
 - ⁴⁴ S. Ganeshan and S. Das Sarma, Phys. Rev. B **91**, 125438 (2015).