MoTe$_{2}$: A Type-II Weyl Topological Metal
Zhijun Wang, Dominik Gresch, Alexey A. Soluyanov, Weiwei Xie, S. Kushwaha, Xi Dai, Matthias Troyer, Robert J. Cava, and B. Andrei Bernevig
DOI: 10.1103/PhysRevLett.117.056805
MoTe$_2$: A Type-II Weyl Topological Metal

Zhijun Wang$^1$, Dominik Gresch$^2$, Alexey A. Soluyanov$^2$, Weiwei Xie$^3$, S. Kushwaha$^3$, Xi Dai$^4$, Matthias Troyer$^2$, Robert J. Cava$^3$, and B. Andrei Bernevig$^1$

$^1$Department of Physics, Princeton University, Princeton, NJ 08544, USA
$^2$Theoretical Physics and Station Q Zurich, ETH Zurich, 8093 Zurich, Switzerland
$^3$Department of Chemistry, Princeton University, Princeton, NJ 08540, USA and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

(Dated: June 30, 2016)

Based on the ab initio calculations, we show that MoTe$_2$, in its low-temperature orthorhombic structure characterized by an X-ray diffraction study at 100 K, realizes 4 type-II Weyl points between the $N$-th and $N$+1-th bands, where $N$ is the total number of valence electrons per unit cell. Other WPs and nodal lines between different other bands also appear close to the Fermi level due to a complex topological band structure. We predict a series of strain-driven topological phase transitions in this compound, opening a wide range of possible experimental realizations of different topological semimetal phases. Crucially, with no strain, the number of observable surface Fermi arcs in this material is 2 – the smallest number of arcs consistent with time-reversal symmetry.

PACS numbers: 73.20.At, 71.20.Be

The ability of gapless band structures to host topological features was first discussed in the context of liquid He [1, 2]. It recently became relevant to crystalline materials with the experimental discovery [3, 4] of the theoretically predicted [5, 6] Weyl semimetals (WSM) in the TaAs family of compounds. In WSMs a topologically protected band crossing of two bands occurs in the close vicinity of the Fermi level forming a gapless node [7–9]. The low energy Hamiltonian for such semimetals is that of a Weyl fermion [10], which exhibits interesting spectroscopic and transport phenomena such as Fermi arcs [9, 11] and the chiral anomaly [7, 12–15].

It was recently shown [16] that in materials Weyl fermions come in two flavors: while the type-I Weyl point (WP) (the condensed matter counterpart of the high-energy theory Weyl fermion) is associated with a closed point-like Fermi surface, its newly proposed type-II cousin [16] appears at the boundary of electron and hole pockets, and has transport properties that are very different from those of the usual, type-I WSM. Another kind of topological metal – a nodal line metal [17–25] – occurs when bands cross along a line in the Brillouin zone (BZ), giving rise to surface states shaped like the surface of a drum [25]. The existence of such a nodal line requires the presence of a symmetry, such as mirror symmetry (or the combination of time reversal and inversion in the absence of spin-orbit coupling (SOC) [26]), in the material.

A WP is associated with a topological charge, since it represents a sink or source of Berry curvature. A nodal line is associated with a Berry phase of $\pi$ along any mirror-symmetric closed trajectory linking with the line. According to the fermion doubling theorem [27, 28] the number of sinks in a crystal has to be equal to the number of sources, meaning that WPs can only appear and annihilate in pairs of opposite topological charge. In non-magnetic materials the presence of time-reversal symmetry dictates the minimal number of WPs to be four, giving rise to two Fermi arcs on the surface of the material. The WSMs experimentally discovered [3, 4, 29] and theoretically predicted [5, 6, 30–32] to date all have more than the minimal number of WPs, as well as a multitude of Fermi arcs, which prevent clean spectroscopy. Of type-I WSMs the TaAs family [5, 6], hosts 24 WPs and the recently predicted type-II WSM WTe$_2$ hosts 8 of them between the $N$-th and $N$ + 1-th bands, where $N$ is the total number of valence electrons per unit cell. Below we refer to the bands below band $N$ inclusive as valence, and the ones above as conduction.

In search for other type-II WSMs, it is natural to look at compounds chemically similar to WTe$_2$. One such compound, MoTe$_2$ in a previously unreported orthorhombic phase was argued to be a strong candidate for another realization of type-II WSM [16, 33]. A very recent interesting work [33] reported that orthorhombic MoTe$_2$ also hosts 8 type-2 WPs in the $k_z = 0$ plane between bands $N$ and $N$ + 1 (as WTe$_2$), for the MoTe$_2$ crystal structure measured at 120 K. By further analyzing the MoTe$_2$ structure reported in Ref. [33], we found that in addition there are 16 WPs out of the $k_z = 0$ plane also formed by bands $N$ and $N$ + 1, located in the immediate vicinity of the Fermi level [34].

In this paper we present the experimental structure of MoTe$_2$ at 100 K and use it to perform first principles and tight-binding calculations of the band structure topology around the Fermi level. Our calculation suggests a different topological physics around the Fermi level than that reported in Ref. [33]: we find only 4 type-II WPs (which we call $W$) between bands $N$ and $N$ + 1.55 meV above the Fermi level. These WPs give rise to only 2 clean visible Fermi arcs on the surface of this material. In this sense MoTe$_2$ represents a “hydrogen atom” of time-reversal ins-
variant WSMs, having the minimal possible number of WPs consistent with time-reversal symmetry. We provide arguments that the difference with the 8 WPs in the $k_z = 0$ plane reported in Ref. [33] comes from the high sensitivity of the band structure of MoTe$_2$ to even small changes in lattice parameters – MoTe$_2$ lies on a cusp between a transition from 4 to 8 nodes in between valence and conduction bands in the $k_z = 0$ plane. Indeed, there are small differences in the crystallographic data of Ref. [33] at 120 K and the one reported here at 100 K resulting in the unit cell volume decrease of about 1%, suggesting the possibility of a temperature-driven topological phase transition.

Another recent work [35] predicts the existence of WPs in Mo doped WTe$_2$. That prediction is obtained by interpolating between the tight-binding models of WTe$_2$ and a theoretically relaxed orthorhombic MoTe$_2$. Such interpolation represents a very strong approximation for the band structure of a doped compound, which, together with the above discussed sensitivity of the WPs to even small differences in the experimental crystal structure, makes the predictions of the work [35] unreliable. More valuable discussions about strain-driven topological phase transition are presented in the Supplemental Material (SM) [36].

Moreover, in metals it is important to look at other topological features near the Fermi level, not only those formed between valence and conduction bands, since the occupation becomes a function of crystal momentum $k$. For example, the topological features near the Fermi level, not only those close in energy to the $W$ points (see details in SM [36]), are located in the $k_z = 0$ plane (more details in SM [36]).

We find also two nodal lines close to the Fermi level, formed by the valence bands $N-1$ and $N$, protected by mirror symmetry. Unlike $W$ points that are formed at the boundary of electron and hole pockets, the additional topological features arise at the touching points of two pockets of the same carriers. Thus, despite a complex topological band structure, the surface Fermi arcs arising due to the four type-II $W$ points are rather clean and should be easy to see in spectroscopic experiments, while the surface states associated with the additional topological crossings overlap with surface projections of the bulk states.

We grew samples by slow cooling and performed diffraction measurements at 100 K. Our results establish that MoTe$_2$ has a new low temperature orthorhombic $1T'$-phase (as was previously reported in [33]), which we designate as the $\gamma$-phase (More details in SM [36] for the full structure characterization). Using this crystallographic data, we perform ab initio calculations based on the density functional theory (DFT) [38, 39] and the generalized gradient approximation (GGA) for the exchange-correlation potential [40]. We first compute the band structure of MoTe$_2$ without spin-orbit coupling (SOC), as illustrated in Fig. 1(b). We find 2 mirror-protected nodal lines in the $k_y=0$ plane (more details in SM [36]) and 12 WPs formed by valence and conduction bands, 4 of which are located in the $k_z = 0$ plane ($W_1$ points) and 8 are out of that plane ($W_2$ points), as shown in Tab. I.

The strong SOC of Mo 4d- and Te 5p-states which dominate the physics around $E_F$ significantly changes the band structure as shown in Fig. 1(c). We first elucidate the topological crossings between valence and conduction bands. The two nodal loops present without SOC become fully gapped. The structure of WPs also changes significantly: the WPs at $k_z \neq 0$ disappear, while only four WPs are found in the $k_z = 0$ plane, still allowed by the $C_{2v}$ symmetry [16, 26] (see symmetries in SM [36]). The coordinates of these points ($W$) are given in Tab. I. Their location and Chern numbers (see SM [36] for the details of Chern number calculation) are illustrated in Fig. 2. The separation between the nearest points with opposite Chern numbers in the unstrained MoTe$_2$ is $\approx 10\%$ of the reciprocal lattice constants $|G_{\gamma}|$ meaning that the...
TABLE I. WPs of MoTe$_2$. The positions (in reduced coordinates $k_x$, $k_y$, $k_z$), Chern numbers, and the energy relative to the $E_F$ are given. W1 and W2 are the WPs formed by bands $N/2$ and $N/2 + 1$ in the absence of SOC, while $W$ are the WPs formed by bands $N$ and $N + 1$ with full SOC. The coordinates of the other points are related to the ones listed by the reflections $M_{x,y}$. 

<table>
<thead>
<tr>
<th>Weyl points</th>
<th>coordinates $(k_x \frac{2\pi}{a}, k_y \frac{2\pi}{b}, k_z \frac{2\pi}{c})$</th>
<th>Chern number $E - E_F$ (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>W1</td>
<td>$(0.1819, 0.1721, 0)$</td>
<td>+1   -38</td>
</tr>
<tr>
<td>W2</td>
<td>$(0.1300, 0.0793, \pm 0.298)$</td>
<td>-1   -18</td>
</tr>
<tr>
<td>W</td>
<td>$(0.1011, 0.0503, 0)$</td>
<td>+1   +55</td>
</tr>
</tbody>
</table>

Topological Fermi arcs should be easily observable in this material. While, as shown below, other topological gapless features show up very close to the $E_F$ in between bands other than $N$ and $N + 1$, it is the $W$ WPs that are most important as they give rise to the only Fermi arcs not superimposed on bulk states upon surface projection.

We computed topological invariants to establish the existence of the WPs, and to prove that no additional WPs are present in between the valence and conduction bands. The first invariant is the Chern number, associated with each of the WPs, which was computed both from the Wannier-based tight-binding model [41, 42], and directly from first-principles calculations [43] (see SM [36]). The result of this calculation is illustrated in Tab. 1 and Fig. 2, where the WPs and their Chern numbers are shown in the BZ.

Further insight into the topology of the Bloch bands of MoTe$_2$ is obtained by computing the $\mathbb{Z}_2$ invariant of the lowest $N$ bands on the time-reversal symmetric planes $k_z = 0, \pi$. The $k_z = 0$ plane contains the WPs and is gapless; hence no such invariant can be defined on this plane. The other five planes, however, are gapped between bands $N$ and $N + 1$. Of these planes only the $k_y = 0$ one has a non-trivial $\mathbb{Z}_2$ invariant. This means that the $k_y = 0$ cut of the BZ is analogous to the 2D BZ of a quantum spin Hall insulator that carries an odd number of Kramers pairs of edge states. The lack of a nontrivial $\mathbb{Z}_2$ invariant on all the other planes implies the existence of disconnected Fermi surfaces. Notice that a connected Fermi sea of the surface states, be it strong or weak topological or trivial insulator, does not lead to only one nontrivial $\mathbb{Z}_2$ index on a high symmetry plane. Fig. 3 shows the surface spectral function for the (001)-surface of MoTe$_2$, and topological Fermi arcs crossing the $k_y = 0$ plane are clearly visible.

![FIG. 2. Weyl points in the Brillouin zone of MoTe$_2$. Four Weyl points formed by bands $N$ and $N + 1$ in the Brillouin zone are shown. The blue and red colors indicate Chern numbers +1 and -1 respectively.](image1)

![FIG. 3. Spectral function of the (001)-surface of the orthorhombic MoTe$_2$ at 20 meV below the Fermi level. A projection of a big hole-like pocket is in the center, with the projections of the electron-pockets to the left and to the right from it. In the gap of these two kinds of pockets, topological Fermi arcs are marked with arrows. The WPs’ projections are denoted by x, with blue and black colors indicating Chern numbers +1 and -1 respectively.](image2)

In type-I WSMs tuning the Fermi level to the energy of the WP results in the surface Fermi arcs connecting projections of the WPs on the particular surface. Type-II WPs appear at the boundary between the pockets, when $E_F$ is tuned to the WP energy; hence projections of bulk carrier pockets necessarily appear in the surface electronic density of states irrespective of the Fermi energy in relation to the WP. The Fermi arcs can in this case be hidden within the projection of the bulk pockets on the surface, but they can still be revealed by tuning the chemical potential (see SM [36]). This is illustrated for MoTe$_2$ in Fig. 3, where we chose a spectroscopically reachable value for the chemical potential of $-20$ meV below $E_F$. The clean Fermi arcs have been revealed in the angle-resolved photoemission spectroscopy measurement [44]. A projection of a big hole-like bulk pocket is seen in the center of the surface BZ, with the projections of the electron-pockets to the left and to the right from it. Unlike the case of type-I WSMs, where a Fermi arc connects surface projections of WPs of opposite chirality, the Fermi arcs illustrated in Fig. 3 are of different nature.

The projections of the $W$ WPs are within the pro-
and $N + 1$ is designated with a $\times$ sign, while the Weyl point between bands $N + 1$ and $N + 2$ is shown as a $+$ sign. The circles mark the crossings of the plane with a nodal line formed by bands $N + 1$ and $N$. Pockets formed by bands $N = 1$, $N$, $N + 1$ and $N + 2$ are shown in green, red, blue and cyan correspondingly.

A cross section of the bulk Fermi surface in the $k_z = 0$ plane around the $k_x = 0$ plane related by mirror $M_y$. A further check of the topological nature of this ring is obtained by computing the Berry phase of a loop trajectory that links with the nodal line. We find this to be equal to $\pi$, as expected for a nodal line.

In conclusion, based on the DFT calculations, we have studied in detail the topological properties of a new orthorhombic $\gamma$-phase of MoTe$_2$, which has been experimentally characterized. The $ab$ initio calculations suggest that unlike the $\alpha$ and $\beta$ phases, the $\gamma$-phase hosts a multitude of topological features around the Fermi level including type-II WPs and nodal lines. The WPs found between the bands $N$ and $N + 1$ band come in a single quadruplet, the smallest number allowed by time-reversal symmetry. This allows for a particularly clean Fermi arc structure on the surface of MoTe$_2$, which should be readily observable in spectroscopic measurements [44]. Other WPs and nodal lines between different other bands also appear in MoTe$_2$, but their spectroscopic signatures on the surface overlap with those of the projected bulk Fermi surfaces.

Acknowledgments We thank Binghai Yan for helpful discussions. This work was supported by NSF CAREER DMR-095242, ONR - N00014-14-1-0330, ARO MURI W911NF-12-1-0461, NSF-MRSEC DMR-0819860, Packard Foundation and Keck Grant. D.G., A.A.S. and M.T. were supported by Microsoft Research, the European Research Council through ERC Advanced Grant SIMCOFE, the Swiss National Science Foundation through the National Competence Centers in Research MARVEL and QSCIT. Z.W. and X.D. were supported by the National Natural Science Foundation of China (No. 11504117), the 973 program of China (No. 2013CB921700), and the “strategic Priority Research Program (B)” of the Chinese Academy of Sciences (No. XDB07020100).

FIG. 4. Contour plot of the bulk Fermi surface at $k_z = 0$ for different values of $E_F$. The Weyl point $W$ between bands $N$ and $N + 1$ is designated with a $\times$ sign, while the Weyl point between bands $N + 1$ and $N + 2$ is shown as a $+$ sign. The circles mark the crossings of the plane with a nodal line formed by bands $N + 1$ and $N$. Pockets formed by bands $N = 1$, $N$, $N + 1$ and $N + 2$ are shown in green, red, blue and cyan correspondingly.
Zhijun Wang and Dominik Gresch contributed equally to this work.

[34] Two of these points are at $k = (0.12661133, 0.0996582, 0.21142578)$ (20 meV above $E_F$) and $k = (0.11034922, 0.07301836, 0.14727305)$ (36 meV above $E_F$). Another 6 points are related to these ones by the mirror reflections $M_x$ and $M_y$, and 8 more points are located symmetrically to these ones about the $k_z = 0$ plane.
[36] See Supplemental Material [url], which includes Refs. [45–68].
(1999).


