Fermi arc electronic structure and Chern numbers in the type-II Weyl semimetal candidate Mo\textsubscript{x}W\textsubscript{1−x}Te\textsubscript{2}

Ilya Belopolski,\textsuperscript{1,2} Su-Yang Xu,\textsuperscript{1} Yukiai Ishida,\textsuperscript{2} Xingchen Pan,\textsuperscript{3} Peng Yu,\textsuperscript{4} Daniel S. Sanchez,\textsuperscript{1} Hao Zheng,\textsuperscript{1} Madhab Neupane,\textsuperscript{5} Nasser Alidoust,\textsuperscript{1} Guoqing Chang,\textsuperscript{6,7} Tay-Rong Chang,\textsuperscript{8} Yun Wu,\textsuperscript{4} Guang Bian,\textsuperscript{1} Shin-Ming Huang,\textsuperscript{6,7,10} Chi-Cheng Lee,\textsuperscript{5,7} Daixiang Mou,\textsuperscript{9} Lunan Huang,\textsuperscript{8} You Song,\textsuperscript{11} Baigeng Wang,\textsuperscript{3} Guanghou Wang,\textsuperscript{3} Yao-Wen Yeh,\textsuperscript{12} Nan Yao,\textsuperscript{12} Julien E. Rault,\textsuperscript{13} Patrick Le Févre,\textsuperscript{13} François Bertran,\textsuperscript{13} Horng-Tay Jeng,\textsuperscript{8,14} Takeshi Kondo,\textsuperscript{2} Adam Kaminski,\textsuperscript{9} Hsin Lin,\textsuperscript{6,7} Zheng Liu,\textsuperscript{4,15,16} Fengqi Song,\textsuperscript{3,1} Shik Shin,\textsuperscript{2} and M. Zahid Hasan\textsuperscript{1,12,§}

\textsuperscript{1}Laboratory for Topological Quantum Matter and Spectroscopy (B7), Department of Physics, Princeton University, Princeton, New Jersey 08544, USA
\textsuperscript{2}The Institute for Solid State Physics (ISSP), University of Tokyo, Kashiwa-no-ha, Kashiwa, Chiba 277-8581, Japan
\textsuperscript{3}National Laboratory of Solid State Microstructures, Collaborative Innovation Center of Advanced Microstructures, and Department of Physics, Nanjing University, Nanjing, 210093, People’s Republic of China
\textsuperscript{4}Centre for Programmable Materials, School of Materials Science and Engineering, Nanyang Technological University, 639798, Singapore
\textsuperscript{5}Department of Physics, University of Central Florida, Orlando, Florida 32816, USA
\textsuperscript{6}Centre for Advanced 2D Materials and Graphene Research Centre, National University of Singapore, 6 Science Drive 2, 117546, Singapore
\textsuperscript{7}Department of Physics, National University of Singapore, 2 Science Drive 3, 117546, Singapore
\textsuperscript{8}Department of Physics, National Tsing Hua University, Hsinchu 30013, Taiwan
\textsuperscript{9}Ames Laboratory, U.S. DOE and Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011, USA
\textsuperscript{10}Department of Physics, National Sun Yat-sen University, Kaohsiung 80424, Taiwan
\textsuperscript{11}State Key Laboratory of Coordination Chemistry, School of Chemistry and Chemical Engineering, Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing, 210093, People’s Republic of China
\textsuperscript{12}Princeton Institute for Science and Technology of Materials, Princeton University, Princeton, New Jersey, 08544, USA
\textsuperscript{13}Synchrotron SOLEIL, L’Orme des Merisiers, Saint-Aubin-BP 48, 91192 Gif-sur-Yvette, France
\textsuperscript{14}Institute of Physics, Academia Sinica, Taipei 11529, Taiwan
\textsuperscript{15}NOVITAS, Nanoelectronics Centre of Excellence, School of Electrical and Electronic Engineering, Nanyang Technological University, 639798, Singapore
\textsuperscript{16}CINTRA CNRS/NTU/THALES, UMI 3288, Research Techno Plaza, 50 Nanyang Drive, Border X Block, Level 6, 637553, Singapore

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It has recently been proposed that electronic band structures in crystals can give rise to a previously overlooked type of Weyl fermion, which violates Lorentz invariance and, consequently, is forbidden in particle physics. It was further predicted that Mo\textsubscript{x}W\textsubscript{1−x}Te\textsubscript{2} may realize such a type-II Weyl fermion. Here, we first show theoretically that it is crucial to access the band structure above the Fermi level \( \varepsilon_F \) to show a Weyl semimetal in Mo\textsubscript{x}W\textsubscript{1−x}Te\textsubscript{2}. Then, we study Mo\textsubscript{x}W\textsubscript{1−x}Te\textsubscript{2} by pump-probe ARPES and we directly access the band structure \( \varepsilon > 0.2 \text{ eV} \) above \( \varepsilon_F \) in experiment. By comparing our results with \textit{ab initio} calculations, we conclude that we directly observe the surface state containing the topological Fermi arc. We propose that a future study of Mo\textsubscript{x}W\textsubscript{1−x}Te\textsubscript{2} by pump-probe ARPES may directly pinpoint the Fermi arc. Our work sets the stage for the experimental discovery of the first type-II Weyl semimetal in Mo\textsubscript{x}W\textsubscript{1−x}Te\textsubscript{2}.

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I. INTRODUCTION

Weyl fermions have been known since the early twentieth century as chiral particles associated with solutions to the Dirac equation at zero mass \([1,2]\). In particle physics, imposing Lorentz invariance uniquely fixes the dispersion for a Weyl fermion. However, effective field theories in condensed matter physics are not required to obey Lorentz invariance, leaving a freedom in the Weyl fermion dispersion. Recently, it was discovered that this freedom allows a new type of Weyl fermion to arise in a crystalline band structure, distinct from the Weyl fermion relevant to particle physics \([3–9]\). This type-II Weyl fermion strongly violates Lorentz invariance and has a dispersion characterized by a Weyl cone, which is tilted over on its side. It was further predicted that a type-II Weyl semimetal arises in WTe\textsubscript{2} \([3]\). Concurrently, MoTe\textsubscript{2} and Mo\textsubscript{x}W\textsubscript{1−x}Te\textsubscript{2} were predicted to be Weyl semimetals \([10–12]\) and, more recently, several additional type-II Weyl semimetal candidates have been proposed \([13–15]\). All theoretical studies found that all Weyl points in the Mo\textsubscript{x}W\textsubscript{1−x}Te\textsubscript{2} series are above the Fermi level \( \varepsilon_F \). While angle-resolved photoemission spectroscopy (ARPES) would be the technique of choice to directly demonstrate a type-II Weyl semimetal in Mo\textsubscript{x}W\textsubscript{1−x}Te\textsubscript{2}, conventional ARPES can only study occupied electron states, below \( \varepsilon_F \), making it challenging to access the Weyl semimetal state in Mo\textsubscript{x}W\textsubscript{1−x}Te\textsubscript{2}. Nonetheless, several ARPES works attempt to access the Weyl semimetal state in MoTe\textsubscript{2} and WTe\textsubscript{2} by studying the band structure above \( \varepsilon_F \) in the tail of the Fermi-Dirac distribution \([16–18]\), while other works have tried to demonstrate a type-II Weyl semimetal.
in MoTe$_2$ and WTe$_2$ in ARPES by studying only the band structure below $\varepsilon_F$ [19–23]. We note also a recent study of type-II Weyl fermions in an unrelated compound [24,25].

Here, we first argue that it’s crucial to study the band structure above $\varepsilon_F$ to show a Weyl semimetal in Mo$_x$W$_{1-x}$Te$_2$, even if the Fermi arcs fall partly below the Fermi level. Next, we experimentally demonstrate that we can access states sufficiently far above $\varepsilon_F$ using a state-of-the-art photoemission technique known as pump-probe ARPES. We find excellent agreement between our pump-probe ARPES data and ab initio calculations, suggesting that Mo$_0$W$_{1-x}$Te$_2$ is a type-II Weyl semimetal. We propose that a future pump-probe ARPES study may directly pinpoint the topological Fermi arc. In this way, our results set the theoretical and experimental groundwork for demonstrating the first type-II Weyl semimetal in Mo$_x$W$_{1-x}$Te$_2$. Our work also opens the way to studying the unoccupied band structure and time-relaxation dynamics of transition metal dichalcogenides by pump-probe ARPES.

II. THEORY FOR TYPE-II WEYL POINTS

Can we show that a material is a Weyl semimetal if the Weyl points are above the Fermi level, $\varepsilon_W > \varepsilon_F$? For the simple case of a well-separated type-I Weyl point of chiral charge $\pm 1$, it is easy to see that this is true. Specifically, although we cannot see the Weyl point itself, the Fermi arc extends below the Fermi level, see Fig. 1(a). Therefore, we can consider a closed loop in the surface Brillouin zone which encloses the Fermi point. By counting the number of surface state crossings on this loop we can demonstrate a nonzero Chern number [26].

In our example, we expect one crossing along the loop. By contrast, this approach fails for a well-separated type-II Weyl point above $\varepsilon_F$. In particular, recall that when counting Chern numbers, the loop we choose must stay always in the bulk band gap. As a result, for the type-II case we cannot choose the same loop as in the type-I case since the loop would run into the bulk hole pocket. We might instead choose a loop which is slanted in energy, see Fig. 1(b), but such a loop would necessarily extend above $\varepsilon_F$. Alternatively, we can consider different constant energy cuts of the type-II Weyl cone. In Figs. 1(c)–1(e), we show constant-energy cuts of the type-II Weyl cone and Fermi arc. We see that we cannot choose a closed loop around the Weyl point by looking only at one energy because the loop runs into a bulk pocket. However, we can build up a closed loop from segments at energies above and below the Weyl point, as in Figs. 1(c) and 1(e). But again, we must necessarily include a segment on a cut at $\varepsilon > \varepsilon_W$. We find that for a type-II Weyl semimetal, if the Weyl points are above the Fermi level, we must study the unoccupied band structure.

Next, we argue that in the specific case of Mo$_x$W$_{1-x}$Te$_2$ we must access the unoccupied band structure to show a Weyl semimetal. In the Supplemental Material, we present a detailed discussion of the band structure of Mo$_x$W$_{1-x}$Te$_2$ [27]. Here we only note a key result, consistent among all ab initio calculations of Mo$_x$W$_{1-x}$Te$_2$, that all Weyl points are type II and are above the Fermi level [3,10–12]. These facts are essentially sufficient to require that we access the unoccupied band structure. However, it is useful to provide a few more details. Suppose that the Fermi level of Mo$_x$W$_{1-x}$Te$_2$ roughly corresponds to the case of Fig. 1(c). To count a Chern number using only the band structure below the Fermi level, we need to find a path enclosing a nonzero chiral charge while avoiding the bulk hole and electron pockets. We can try to trace a path around the entire hole pocket. However, as we will see in Fig. 2, the Weyl point projections all fall in one large hole pocket at $\varepsilon_F$. As a result, tracing around the entire hole pocket encloses zero chiral charge, see also an excellent related discussion in Ref. [28]. Therefore, demonstrating a Weyl semimetal in Mo$_x$W$_{1-x}$Te$_2$ requires accessing the unoccupied band structure.

III. ELECTRONIC STRUCTURE OF MO$_x$W$_{1-x}$Te$_2$

We briefly introduce the occupied band structure of Mo$_x$W$_{1-x}$Te$_2$. We present ARPES spectra of Mo$_{0.45}$W$_{0.55}$Te$_2$ below the Fermi level, see Figs. 2(a)–2(k). We observe a
FIG. 2. Mo$_{0.45}$W$_{0.55}$Te$_2$ below the Fermi level. (a)–(g) Conventional ARPES spectra of the constant-energy contour. We observe a palmier-shaped hole pocket and an almond-shaped electron pocket. The two pockets approach each other and we directly observe a beautiful avoided crossing near $\epsilon_F$ where they hybridize, in (a). This hybridization is expected to give rise to Weyl points above $\epsilon_F$. (h)–(k) ARPES measured $E_B$-$k_y$ dispersion maps along the cuts shown in (f). We expect Weyl points or Fermi arcs above the Fermi level at certain $k_y$ where the pockets approach. (l)–(n) Constant-energy contours for Mo$_{0.4}$W$_{0.6}$Te$_2$ from ab initio calculations. The black and white dots indicate the Weyl points, above $\epsilon_F$. Note the excellent overall agreement with the ARPES spectra. The offset on the $k_y$ scale on the ARPES spectra is set by comparison with calculation. (o) Cartoon of the palmier and almond at the Fermi level. Based on calculation, we expect Weyl points above $\epsilon_F$ where the pockets intersect.

palmier-shaped hole pocket and an almond-shaped electron pocket, which chase each other as we scan in binding energy. We find excellent agreement between our ARPES results and ab initio calculation, see Figs. 2(l)–2(n). Based on calculation, at two energies above $\epsilon_F$, the pockets catch up to each other and intersect, forming two sets of Weyl points $W_1$ and $W_2$, see Fig. 2(o) and also the Supplemental Material [27].

Next, we show that we can directly access the relevant unoccupied states in Mo$_x$W$_{1-x}$Te$_2$ with pump-probe ARPES. In our experiment, we use a 1.48 eV pump laser to excite electrons into low-lying states above the Fermi level and a 5.92 eV probe laser to perform photoemission [29]. We first study Mo$_{0.45}$W$_{0.55}$Te$_2$ along $\Gamma$-$\bar{\Gamma}$ at fixed $k_z = 0$ Å$^{-1}$, see Figs. 3(a) and 3(b). The sample responds beautifully to the pump laser, and we observe a dramatic evolution of the bands up to energies $> 0.2$ eV above $\epsilon_F$. We find similarly that we can directly access the unoccupied band structure on a cut at fixed $k_y \sim k_W$, see Figs. 3(c) and 3(d). Further, by plotting constant-energy cuts we can directly observe that the almond pocket continues to grow above $\epsilon_F$, while the palmier pocket recedes, consistent with calculation, see Figs. 3(e)–3(j). We note that all available calculations of Mo$_x$W$_{1-x}$Te$_2$ place the Weyl points $< 0.1$ eV above the Fermi level. In addition, the Weyl point projections are all predicted to lie within 0.25 Å$^{-1}$ of the $\bar{\Gamma}$ point [3,10–12]. Our pump-probe measurement easily accesses the relevant region of reciprocal space to show a Weyl semimetal in Mo$_x$W$_{1-x}$Te$_2$ for all $x$.

IV. SIGNATURES OF A WEYL SEMIMETAL

We next present evidence for a Weyl semimetal in Mo$_{0.45}$W$_{0.55}$Te$_2$. The agreement between our pump-probe ARPES spectra and ab initio calculation strongly suggests that Mo$_{0.45}$W$_{0.55}$Te$_2$ is a Weyl semimetal. We consider the spectrum at fixed $k_z \sim k_W$, see Figs. 4(a)–4(d). We find an upper electron pocket (1) with a short additional surface state (2), a lower electron pocket (3), and the approach between hole and electron bands (4), all in excellent agreement with the calculation. We also note the excellent agreement in
the constant energy contours both above and below \( \varepsilon_F \), as discussed above. In addition to this overall agreement between experiment and theory, we might ask if there is any direct signature of a Weyl semimetal that we can pinpoint from the experimental data alone [26]. First, we consider a measurement of the Chern number, following the prescription discussed above for type-II Weyl cones. Formally, the Fermi arc will contribute one crossing, while the trivial surface state will generically meet at \( \varepsilon_F \) with net contribution zero. However, in our experiment, the finite resolution prevents us from carrying out this counting. In particular, the linewidth of the surface state is comparable to its energy dispersion, so we cannot determine this counting. In particular, a full\( k_y \) dependence may further show a systematic evolution of the Fermi arc.

Next, we note that since the chiral charges of all Weyl point projections are \( \pm 1 \), we expect a disjoint arc connecting pairs of Weyl points, in one of two possible configurations, Figs. 4(e) and 4(f). From our calculation, we expect case Fig. 4(e). However, we observe no such disjoint arc in (3) in our spectrum. From calculation, we see that this is perhaps reasonable because the Fermi arc is adjacent to trivial surface states, see the dotted lines in Figs. 4(c) and 4(d). Indeed, we can understand the Fermi arcs in Mo\(_{0.45}\)W\(_{0.55}\)Te\(_2\) as arising from the large electronlike surface state (3) of Figs. 4(c) and 4(d), which we can imagine as being present whether or not there are Weyl points. Then, we can tune the system through a topological phase transition. Before the transition, the surface state is entirely trivial. After the transition, the Weyl point projections sit on (3) and “snip out” a topological Fermi arc from the large surface state. Formally, the Fermi arc terminates strictly on the Weyl points, while the remaining trivial surface states merge into the bulk in some generic way near the Weyl points. However, within any reasonable resolution, the topological and trivial surface states appear to connect at the Weyl points. As a result, we see no disjoint Fermi arc.

We might then ask if we can observe a kink, since the Fermi arc and the trivial surface state will generically meet at some angle. We note that we observe a kink in calculation, but not in the ARPES spectra presented here. We propose that a more complete pump-probe ARPES study of Mo\(_{0.45}\)W\(_{1-\varepsilon}\)Te\(_2\) may show such a kink. In particular, a full \( k_y \) dependence may catch a kink or “ripple” in the surface state, signaling a topological Fermi arc. If there is a difference in how well localized the Fermi arc is on the surface of the sample, compared to the trivial surface state, then a difference in the photoemission cross section may make one or the other feature brighter, allowing us to directly detect the arc. A composition dependence may further show a systematic evolution of the kink, which would also prove an arc. Such an analysis is beyond the scope of this work. Here, we have shown theoretically that accessing the unoccupied band structure is necessary to show a Weyl semimetal in Mo\(_{0.45}\)W\(_{1-\varepsilon}\)Te\(_2\) and, in addition, we have directly accessed the unoccupied band structure in experiment and observed the surface state containing the topological Fermi arc. These results set the
stage for directly demonstrating that Mo$_x$W$_{1-x}$Te$_2$ is a type-II Weyl semimetal.

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