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<td>Belopolski, Ilya; Sanchez, Daniel S.; Ishida, Yukiaki; Pan, Xingchen; Yu, Peng; Xu, Su-Yang; Chang, Guoqing; Chang, Tay-Rong; Zheng, Hao; Alidoust, Nasser; Bian, Guang; Neupane, Madhab; Huang, Shin-Ming; Lee, Chi-Cheng; Song, You; Bu, Haijun; Wang, Guanghou; Li, Shisheng; Eda, Goki; Jeng, Horng-Tay; Kondo, Takeshi; Lin, Hsin; Liu, Zheng; Song, Fengqi; Shin, Shik; Hasan, M. Zahid</td>
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Discovery of a new type of topological Weyl fermion semimetal state in Mo$_x$W$_{1-x}$Te$_2$

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The recent discovery of a Weyl semimetal in TaAs offers the first Weyl fermion observed in nature and dramatically broadens the classification of topological phases. However, in TaAs it has proven challenging to study the rich transport phenomena arising from emergent Weyl fermions. The series Mo$_x$W$_{1-x}$Te$_2$ are inversion-breaking, layered, tunable semimetals already under study as a promising platform for new electronics and recently proposed to host Type II, or strongly Lorentz-violating, Weyl fermions. Here we report the discovery of a Weyl semimetal in Mo$_x$W$_{1-x}$Te$_2$ at $x = 25\%$. We use pump-probe angle-resolved photoemission spectroscopy (pump-probe ARPES) to directly observe a topological Fermi arc above the Fermi level, demonstrating a Weyl semimetal. The excellent agreement with calculation suggests that Mo$_x$W$_{1-x}$Te$_2$ is a Type II Weyl semimetal. We also find that certain Weyl points are at the Fermi level, making Mo$_x$W$_{1-x}$Te$_2$ a promising platform for transport and optics experiments on Weyl semimetals.
The recent discovery of the first Weyl semimetal in TaAs has opened a new direction of research in condensed matter physics. Weyl semimetals are fascinating because they give rise to Weyl fermions as emergent electronic quasiparticles, which can give rise to Weyl fermions as emergent electronic quasiparticles, have an unusual topological classification closely related to the integer quantum Hall effect, and host topological Fermi arc surface states. These properties give rise to many unusual transport phenomena, including negative longitudinal magnetoresistance from the chiral anomaly, an anomalous Hall effect, the chiral magnetic effect, non-local transport and novel quantum oscillations. Although many recent works have studied transport properties in TaAs, transport experiments are challenging because TaAs and its isoelectronic cousins have a three-dimensional crystal structure with irrelevant metallic bands and many Weyl points. As a result, there is a need to discover new Weyl semimetals better suited for transport and optics experiments and eventual device applications.

Recently, the Mo\(_{1-x}\)W\(_x\)Te\(_2\) series has been proposed as a new Weyl semimetal. Unlike TaAs, Mo\(_{1-x}\)W\(_x\)Te\(_2\) has a layered crystal structure and is rather widely available as large, high-quality single crystals. Indeed, MoTe\(_2\) provides the possibility to realize a tunable Weyl semimetal, which may be important for transport measurements and applications. Recently, it was also discovered theoretically that WTe\(_2\) hosts a novel type of strongly Lorentz-violating Weyl fermion, or Type II Weyl fermion, long ignored in quantum field theory. This offers a fascinating opportunity to realize in a crystal an emergent particle forbidden as a fundamental particle in particle physics. There are, moreover, unique transport signatures associated with strongly Lorentz-violating Weyl fermions. For all these reasons, there is considerable interest in demonstrating that Mo\(_{1-x}\)W\(_x\)Te\(_2\) is a Weyl semimetal. At the same time, it is important to note that ab initio calculations predict that the Weyl points in Mo\(_{1-x}\)W\(_x\)Te\(_2\) are above the Fermi level. This makes it challenging to access the Weyl semimetal state with conventional angle-resolved photoemission spectroscopy (ARPES). Recently, we have demonstrated that we can access the unoccupied band structure of Mo\(_{1-x}\)W\(_x\)Te\(_2\) by pump-probe ARPES to the energy range necessary to study the Weyl points and Fermi arcs. As a further consideration, despite the promise of Mo\(_{1-x}\)W\(_x\)Te\(_2\) for transport, if the Weyl points are far from the Fermi level, then the novel phenomena associated with the emergent Weyl fermions and violation of Lorentz invariance will not be relevant to the material’s transport properties.

Here we report the discovery of a Weyl semimetal in Mo\(_{1-x}\)W\(_x\)Te\(_2\) at doping \(x=25\%\). We use pump-probe ARPES to study the band structure above the Fermi level and we directly observe two kinks in a surface state band. We interpret the kinks as corresponding to the end points of a topological Fermi arc surface state. We apply the bulk-boundary correspondence and argue that since the surface state band structure includes a topological Fermi arc, Mo\(_{1-x}\)W\(_x\)Te\(_2\) is a Weyl semimetal. The end points of the Fermi arc also allow us to fix the energy and momentum locations of the Weyl points. We find excellent agreement with our ab initio calculation. However, crucially, we find that certain Weyl points have lower binding energy than expected from calculation and, in fact, are located very close to the Fermi level. This unexpected result suggests that our Mo\(_{0.75}\)W\(_{0.25}\)Te\(_2\) samples may be useful to study the unusual transport phenomena of Weyl semimetals and, in particular, those particularly exotic phenomena arising from strongly Lorentz-violating Weyl fermions. Our work also sets the stage for the first tunable Weyl semimetal. Our discovery of a Weyl semimetal in Mo\(_{1-x}\)W\(_x\)Te\(_2\) provides the first Weyl semimetal outside the TaAs family, as well as a Weyl semimetal which may be tunable and easily accessible in transport studies. Taken altogether with calculation, our experimental results further show that we have realized the first Weyl semimetal with Type II, or strongly Lorentz-violating, emergent Weyl fermions.

Results

Overview of the crystal and electronic structure. We first provide a brief background of Mo\(_{1-x}\)W\(_x\)Te\(_2\) and study the band structure below the Fermi level. WTe\(_2\) crystallizes in an orthorhombic Bravais lattice, space group \(Pmn\alpha\) (\#31), lattice constants \(a=6.282\ \text{Å},\ b=3.496\ \text{Å},\ c=14.07\ \text{Å}\), as shown in Fig. 1a (ref. 43). Crucially, the crystal has no inversion symmetry, a requirement for a Weyl semimetal. The crystals we study are flat, shiny, layered and beautiful, see Fig. 1b. The natural cleaving plane is (001), with surface and bulk Brillouin zones, as shown in Fig. 1c. We first consider the overall band structure of WTe\(_2\). There are two bands, one electron and one hole pocket, near the Fermi level, both very near the \(\Gamma\) point of the bulk Brillouin zone, along the \(\Gamma\)–\(Y\) line. Although the bands approach each other and Weyl points might be expected to arise where the bands cross, it is now understood that WTe\(_2\) is in fact very close to a phase transition between a Weyl semimetal phase and a trivial phase, so that the electronic structure of WTe\(_2\) is too fragile to make it a compelling candidate for a Weyl semimetal. Next, we interpolate between \(ab\ initio\) Wannier function-based tight-binding models for WTe\(_2\) and MoTe\(_2\) to study Mo\(_{1-x}\)W\(_x\)Te\(_2\) at arbitrary \(x\) (ref. 22). For a wide range of \(x\), we find a robust Weyl semimetal phase. In Fig. 1e,f, we show where the Weyl points sit in the Brillouin zone. They are all located close to \(\Gamma\) in the \(k_x=0\) momentum plane. There are two sets of Weyl points, \(W_1\) at binding energies \(E_0=0.045\ E_F\) and \(W_2\) at \(E_0=-0.066\ E_F\), all above the Fermi level \(E_F\). In addition, the Weyl points are almost aligned at the same \(k_y=\pm k_W\), although this positioning is not known to be in any way symmetry-protected. We also note that the Weyl cones are all tilted over, corresponding to strongly Lorentz-violating or Type II Weyl fermions, see Fig. 1g (ref. 23). Next, we study a Fermi surface of Mo\(_{1-x}\)W\(_x\)Te\(_2\) for \(x=45\%\) using incident light with photon energy \(h\nu=6.36\ E_F\), shown in Fig. 1h. We observe two pockets, a palmier-shaped pocket closer to the \(\Gamma\) point of the surface Brillouin zone and an almond-shaped pocket sitting next to the palmier pocket, further from \(\Gamma\). The palmier pocket is a hole pocket, while the almond pocket is an electron pocket. We note that we see an excellent agreement between our results and an \(ab\ initio\) calculation of Mo\(_{1-x}\)W\(_x\)Te\(_2\) for \(x=40\%\), shown in Fig. 1i. At the same time, we point out that the electron pocket of the Weyl points is nearly absent in this ARPES spectrum, possibly due to low photoemission cross section at the photon energy used. However, as we will see below, we do observe this electron pocket clearly in our pump-probe ARPES measurements, carried out at a slightly different photon energy, \(h\nu=5.92\ E_F\). On the basis of our calculations and preliminary ARPES results, we expect that the Weyl points sit above the Fermi level, where the palmier and almond pockets approach each other. We also present an \(E_F-k_y\) spectrum in Fig. 1j, where we see how the palmier and almond pockets nest into each other. We expect the two pockets to chase each other as they disperse above \(E_F\), giving rise to Weyl points, see Fig. 1k.

Unoccupied band structure of Mo\(_{1-x}\)W\(_x\)Te\(_2\). Next, we show that pump-probe ARPES at probe photon energy \(h\nu=5.92\ E_F\) gives us access to the bulk and surface bands participating in the Weyl semimetal state in Mo\(_{0.25}\)W\(_{0.75}\)Te\(_2\), both below and above \(E_F\). In Fig. 2a-c, we present three successive ARPES spectra of Mo\(_{0.25}\)W\(_{0.75}\)Te\(_2\) at fixed \(k_y\) near the predicted position of the Weyl
points. We observe a beautiful, sharp band near \(E_F\), whose sharp character suggests that it is a surface band, and broad continua above and below the Fermi level, whose broad character suggests that they are bulk valence and conduction bands. In Fig. 2d–f, we show the same cuts, with guides to the eye to mark the bulk valence and conduction band continua. We also find that we can track the evolution of the bulk valence and conduction bands clearly in our data with \(k_y\). Specifically, we see that both the bulk valence and conduction bands disperse toward negative binding energies as we sweep \(k_y\) closer to \(\Gamma\). At the same time, we note that the bulk valence band near \(\Gamma\) is only visible near \(k_x \sim 0\) and drops sharply in photoemission cross-section away from \(k_x \sim 0\). In Fig. 2g,h we present a comparison of our ARPES data with an \textit{ab initio} calculation of Mo\(_{0.25}\)W\(_{0.75}\)Te\(_2\) (ref. 22). We also mark the location of the three successive spectra on a Fermi surface in Fig. 2i. We include as well the approximate locations of the Weyl points, as expected from calculation. We find excellent correspondence between both bulk and surface states. We add that we directly observe an additional surface state detaching from the bulk conduction band well above the Fermi level and that we can also match this additional surface state well between our ARPES spectra and calculation. Our pump-probe ARPES results clearly show both the bulk and surface band structure of Mo\(_{0.25}\)W\(_{0.75}\)Te\(_2\), both below and above \(E_F\), and with an excellent correspondence with calculation.

**Observation of a topological Fermi arc above the Fermi level.**

Now we show that we observe signatures of a Fermi arc in Mo\(_{0.25}\)W\(_{0.75}\)Te\(_2\). We consider the cut shown in Fig. 3a, repeated from Fig. 2b, and we study the surface state. We observe two kinks in each branch, at \(E_B \sim -0.005\,\text{eV}\) and \(E_B \sim 0.05\,\text{eV}\). This kink is a smoking-gun signature of a Weyl point\(^2\). We claim that each kink corresponds to a Weyl point and that the surface state passing through them includes a topological Fermi arc. To show these kinks more clearly, in Fig. 3b, we show a second derivative plot of the spectrum in Fig. 3a. In Fig. 3c we also present a cartoon of the kink in our data, with the positions of the \(W_1\) and \(W_2\) Weyl points marked. Again, note that although the \(W_1\) and \(W_2\) are not located strictly at the same \(k_y\), we expect the \(k_y\) separation to be on the order of \(10^{-4}\,\text{Å}^{-1}\) from calculation, so that we can consider them to lie at the same \(k_y\) within experimental resolution. We emphasize that from our pump-probe ARPES spectrum, we can directly read off that the energy separation of the Weyl points is \(\sim 0.05\,\text{eV}\) and that the \(W_1\) are located at \(\sim -0.005\,\text{eV}\). We also present a quantitative analysis of our data, showing a kink. To do this, we fit the surface state momentum distribution curves (MDCs) to a Lorentzian distribution and we plot the train of peaks corresponding to the surface state band. We note that we simultaneously fit the topological surface state, the bulk valence and conduction states, and the trivial surface state above the conduction band. In Fig. 3d we plot
the resulting band dispersions in white and observe an excellent fit to our spectrum. Next, we define a kink as a failure of the train of Lorentzian maxima to fit to a quadratic band. In particular, over a small energy and momentum window, any band should be well-characterized by a quadratic fit, so the failure of such a fit in a narrow energy window implies a kink. After fitting the topological surface state to a quadratic polynomial we find two mismatched regions, marked in Fig. 3e, demonstrating two kinks.

For comparison, we plot the energy positions of the $W_1$ and $W_2$ as read off directly from Fig. 3a. We find an excellent agreement between the qualitative and quantitative analysis, although we note that the fit claims that the $W_2$ kink is lower in energy. To illustrate the success of the Lorentzian fit, in Fig. 3f,g we present two representative MDCs at energies indicated by the green arrows. We see that the Lorentzian distributions provide a good fit and take into account all bands observed in our spectra. The raw data, the second derivative plots and the Lorentzian fitting all show two kinks, providing a strong signature of Fermi arcs.

To show that we have observed a topological Fermi arc, we compare our experimental observation of two surface state kinks with a numerical calculation of Mo$_{0.25}$W$_{0.75}$Te$_2$. In Fig. 4a,b, we mark the energies of the Weyl points as well as the band minimum of the surface state in our ARPES spectrum and in calculation. We see that the energy difference between the Weyl
points is $\sim 0.02$ eV in calculation but $\sim 0.05$ eV in experiment. Moreover, the band minimum $E_{\text{min}}$ is at $\sim E_F$ in calculation, but at $E_F \sim 0.06$ eV in experiment. The difference in $E_{\text{min}}$ suggests either that our sample is electron-doped or that the $k_y$ position of the Weyl points differs in experiment and theory. Next, crucially, we observe that, in disagreement with calculation, the $W_1$ are located only $\sim 0.005$ eV above $E_F$. This suggests that the Weyl points and Fermi arcs in our Mo$_{0.25}$W$_{0.75}$Te$_2$ samples may be accessible in transport. This result is particularly relevant because MoTe$_2$, WTe$_2$ and other transition metal dichalcogenides are already under study as platforms for novel electronics$^{26–30}$. Since the Weyl points of Mo$_x$W$_{1-x}$Te$_2$ may be at the Fermi level, it is

Figure 3 | Direct experimental observation of Fermi arcs in Mo$_{0.25}$W$_{0.75}$Te$_2$. (a) To establish Fermi arcs in Mo$_{0.25}$W$_{0.75}$Te$_2$ we focus on the spectrum shown in Fig. 2b, with $k_y \sim k_W$. We observe two kinks in the surface state, at $E_B \sim -0.005$ eV and $E_B \sim -0.05$ eV. (b) The kinks are easier to see in a second-derivative plot of panel a. (c) Same as panel a, but with a guide to the eye showing the kinks. The Weyl point projections are at the locations of the kinks. The surface state with the kinks is a topological Fermi arc. (d) To further confirm a kink, we fit Lorentzian distributions to our data. We capture all four bands in the vicinity of the kinks: the bulk conduction and valence states, the topological surface state and an additional trivial surface state merging into the conduction band at more negative $E_B$. We define a kink as a failure of a quadratic fit to a band. We argue that for a small energy and momentum window, any band should be well-characterized by a quadratic fit and that the failure of such a fit shows a kink. (e) By matching the train of Lorentzian peaks of the topological surface state (red) to a quadratic fit (blue) we find two mismatched regions (shaded in yellow), showing two kinks. The purple arrows show the location of the Weyl points, taken from panel c, and are consistent with the kinks we observe by fitting. (f,g) Two characteristic MDCs at energies indicated by the green arrows in panel e. We see that the Lorentzian distributions provide a good fit and capture all bands observed in our spectra.
possible that transport measurements may detect a signature of the strongly Lorentz-violating Weyl fermions or other unusual transport phenomena associated with Weyl semimetals in Mo\(_{0.25}\)W\(_{0.75}\)Te\(_2\). We summarize our results in Fig. 4c. We directly observe, above the Fermi level, a surface state with two kinks (shown in red). By comparing our results with ab initio calculation, we confirm that the kinks correspond to Weyl points. Furthermore, the excellent agreement of our experimental results with calculation shows that we have realized the first Type II Weyl semimetal.

**Limits on directly observing Type II Weyl cones.** So far we have studied the surface states of Mo\(_{0.25}\)W\(_{0.75}\)Te\(_2\) and we have argued that Mo\(_{0.25}\)W\(_{0.75}\)Te\(_2\) is a Weyl semimetal because we observe a topological Fermi arc surface state. However, topological Fermi arcs cannot strictly distinguish between bulk Weyl cones that are of Type I or Type II. While the excellent agreement with calculation suggests that Mo\(_{0.25}\)W\(_{0.75}\)Te\(_2\) is a Type II Weyl semimetal, we might ask if we can directly observe a Type II Weyl cone in Mo\(_{0.25}\)W\(_{0.75}\)Te\(_2\) by ARPES. This corresponds to observing the two branches of the bulk Weyl cone, as indicated by the purple dotted
circles in Fig. 4c. We reiterate that one crucial obstacle in observing a Type II Weyl cone is that all the recent calculations on WTe₂, Mo₇/W₁₋ₓTe₂ and MoTe₂ predict that all Weyl points are above the Fermi level²²–²⁵. As we have seen, using pump-probe ARPES, we are able to measure the unoccupied band structure and show a Fermi arc. However, in our pump-probe ARPES measurements, we find that the photoemission cross-section of the bulk bands is too weak near the Weyl points. At the same time, our calculations suggest that for a reasonable quasiparticle lifetime and spectral linewidth, it may be difficult to resolve the two branches of the Weyl cone. We conclude that it is challenging to directly access the Type II Weyl cones in MoₓW₁₋ₓTe₂.

Considerations regarding trivial surface states. One obvious concern in the interpretation of our experimental result is that we observe two kinks in the surface state, but we expect a disjoint segment based on topological theory. In particular, all calculations show that all Weyl points in MoₓW₁₋ₓTe₂ have chiral charge 1 ± 1 (refs 22–25). However, our observation of a kink suggests that there are two Fermi arcs connecting to the same Weyl point, which requires a chiral charge of 1 ± 2. To resolve this contradiction, we study the calculation of the surface state near the Weyl points, shown in Fig. 4g. We observe, as expected, a Fermi arc (red arrow) connecting the Weyl points. However, at the same time, we see that trivial surface states (yellow arrows) from above and below the band crossing merge with the bulk bands in the vicinity of the Weyl points. As a result, there is no disjoint arc but rather a large, broadband surface state with a ripple arising from the Weyl points. We can imagine that this broadband surface state exists even in the trivial phase. Then, when the bulk bands cross and give rise to Weyl points, a Fermi arc is pulled out from this broadband surface state. At the same time, the remainder of the broadband surface state survives as a trivial surface state. In this way, the Fermi arc is not disjoint but shows up as a ripple. We observe precisely this ripple in our ARPES spectra of MoₓW₁₋ₓTe₂.

As a further check of our analysis, we perform a Lorentzian fit of an ARPES spectrum at kₓ shifted away from the Weyl points, shown in Fig. 4d, the same cut as Fig. 2c. We show the Lorentzian fit in Fig. 4e and a quadratic fit to the train of peaks in Fig. 2f. In sharp contrast to the result for kₓ ≈ kₓ, there is no ripple in the spectrum and the quadratic provides an excellent fit. This result is again consistent with our expectation that we should observe a ripple only at kₓ near the Weyl points. Our results also set the stage for the realization of the first tunable Weyl semimetal in MoₓW₁₋ₓTe₂. As we vary the composition, we expect to tune the relative separation of the Weyl points and their position in energy relative to E_F. In Fig. 4h–k, we present a series of calculations of MoₓW₁₋ₓTe₂ for x = 10, 25, 40 and 100%. We see that the separation of the Weyl points increases with x and that the W₁ approach E_F for larger x. We propose that a systematic composition dependence can demonstrate the first tunable Weyl semimetal in MoₓW₁₋ₓTe₂.

Discussion

We have demonstrated a Weyl semimetal in MoₓW₁₋ₓTe₂ by directly observing kinks and a Fermi arc in the surface state band structure. Taken together with calculation, our experimental data show that we have realized the first Type II Weyl semimetal, with strongly Lorentz-violating Weyl fermions. We point out that in contrast to concurrent works on the Weyl semimetal state in MoTe₂ (refs 44–52), we directly access the unoccupied band structure of MoₓW₁₋ₓTe₂ and directly observe a Weyl semimetal with minimal reliance on calculation. In particular, our observation of a surface state kink at a generic point in the surface Brillouin zone requires that the system be a Weyl semimetal. The excellent agreement with calculation serves as an additional, independent check of our experimental results. We also reiterate that unlike MoTe₂, MoₓW₁₋ₓTe₂ opens the way to the realization of the first tunable Weyl semimetal. Lastly, we note that MoTe₂ is complicated because it is near a critical point for a topological phase transition. Indeed, one recent theoretical work²³ shows that MoTe₂ has four Weyl points, while another²⁵ finds eight Weyl points. This is, moreover, similar to the case of WTe₂, which is near the critical point for a transition between eight Weyl points and zero Weyl points. By contrast, MoₓW₁₋ₓTe₂ sits well within the eight Weyl point phase for most x, as confirmed explicitly here and by calculation in ref. 22. The stability of the topological phase of MoₓW₁₋ₓTe₂ simplifies the interpretation of our data. By directly demonstrating a Weyl semimetal in MoₓW₁₋ₓTe₂, we provide not only the first Weyl semimetal beyond the TaAs family, but the first Type II Weyl semimetal, as well as a Weyl semimetal which may be tunable and which may be more accessible for transport and optics studies of the fascinating phenomena arising from emergent Weyl fermions in a crystal.

Methods

Pump-probe ARPES. Pump-probe ARPES measurements were carried out using a hemispherical Scienta R4000 analyser and a mode-locked Ti:Sapphire laser system that delivered 1.48 eV pump and 5.92 eV probe pulses at a repetition rate of 250 kHz (ref. 53). The time and energy resolution were 300 fs and 15 meV, respectively. The spot diameters of the pump and probe lasers at the sample were 250 and 85 μm, respectively. Measurements were carried out at pressures <5 × 10⁻¹¹ Torr and temperatures ~8 K.

Sample growth. Single crystals of MoₓW₁₋ₓTe₂ were grown using a chemical vapor transport technique with iodine as the transport agent. Stoichiometric Mo, W and Te powders were ground together and loaded into a quartz tube with a small amount of I. The tube was sealed under vacuum and placed in a two-zone furnace. The hot zone was maintained at 1,050 °C for 2 weeks and the cold zone was maintained at 950 °C. The dopant distribution is not uniform particularly near the crystal surface. The composition of the selected sample was determined by an energy dispersive spectroscopy measurement with a scanning electron microscope.

Ab initio calculations. The ab initio calculations were based on the generalized gradient approximation⁴⁴ using the full-potential projected augmented wave method⁶⁶,⁶⁷ as implemented in the VASP package³⁵. Experimental lattice constants were used for both WTe₂ (ref. 58) and MoTe₂. A 15 × 11 × 7 Monkhorst-Pack k-point mesh was used in the calculations. The spin-orbit coupling effects were included in calculations. To calculate the bulk and surface electronic structures, we constructed first-principles tight-binding model Hamilton by projecting onto the Wannier orbitals²⁸–⁴⁴, which use the VASP-WANNIER90 interface⁶². We used W d orbitals, Mo d orbitals, and Te p orbitals to construct Wannier functions and without perform the procedure for maximizing localization. The electronic structure of the MoₓW₁₋ₓTe₂ samples with finite doping was calculated by a linear interpolation of tight-binding model matrix elements of WTe₂ and MoTe₂. The surface states were calculated from the surface Green’s function of the semi-infinite system⁶³,⁶⁴.

Data availability. The data relevant to the findings of this study are available from the corresponding authors on reasonable request.

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