Uniaxial strain-induced phase transition in the 2D topological semimetal IrTe$_2$

Strain is ubiquitous in solid-state materials, but despite its fundamental importance and technological relevance, leveraging externally applied strain to gain control over material properties is still in its infancy. In particular, strain control over the diverse phase transitions and topological states in two-dimensional transition metal dichalcogenides remains an open challenge. Here, we exploit uniaxial strain to stabilize the long-debated structural ground state of the 2D topological semimetal IrTe$_2$, which is hidden in unstrained samples. Combined angle-resolved photoemission spectroscopy and scanning tunneling microscopy data reveal the strain-stabilized phase has a 6 x 1 periodicity and undergoes a Lifshitz transition, granting unprecedented spectroscopic access to previously inaccessible type-II topological Dirac states that dominate the modified inter-layer hopping. Supported by density functional theory calculations, we show that strain induces an Ir to Te charge transfer resulting in strongly weakened inter-layer Te bonds and a reshaped energetic landscape favoring the 6x1 phase. Our results highlight the potential to exploit strain-engineered properties in layered materials, particularly in the context of tuning inter-layer behavior.
Using external stimuli to manipulate the diverse phenomena observed in quantum materials may allow for tunable control over technologically relevant material properties. Within this context uniaxial strain has recently emerged as a powerful approach to influence the properties of solids and offers a path to tailor both physical properties and device functionalities, particularly in the 2D transition metal dichalcogenides (TMDs). While efforts to control phase transition behavior with strain have focused predominantly on oxide materials, there also exist many opportunities within the 2D semimetals, which routinely host multiple nearly degenerate structural, electronic, and topological phases, thereby making them sensitive to external perturbation. In this regard, the family of layered tellurides are particularly promising, a prime example of which is $1T$-IrTe$_2$. This high-atomic number material is predicted to have a type-II bulk Dirac semimetal with a Dirac point slightly above the Fermi level and presents first-order bulk phase transitions to a $5 \times 1 \times 5$ structure at 280 K, and to an $8 \times 1 \times 8$ structure at 180 K. At the surface a complex staircase of nearly degenerate low-temperature phases with periodicity $3n+2$ (i.e., $8 \times 1, 11 \times 1, 17 \times 1$) coexist over scales of a few tens of nanometers. All of the broken-symmetry phases display characteristic quasi-1D modulations typically identified as Ir dimers, although the changes to the in-plane bonding suggest a multicenter bond as a more complete description (for brevity we will continue to use “dimers” throughout the text). The proposed ground state, a $6 \times 1$ phase, is typically observed only within nanoscale regions, making it all but inaccessible to most techniques. As a result, the electronic structure of the ground state, as well as any influence of the phase transitions on the bulk Dirac states, remains unclear, hindering efforts to elucidate the transition mechanism or exploit the topological properties. A number of phases can also be produced via doping: superconductivity is induced by partial substitution of Ir with Pt or Pd, or by temperature quenching, while partial substitution of Te with Se induces charge order. Further emphasizing the metastable nature of the material, the range of competing phases observed in IrTe$_2$ strongly implies that its macroscopic behavior may be tunable via strain, allowing individual phases to be selectively stabilized without the need for external doping.

In this work, by applying a modest uniaxial tensile strain ($\varepsilon \sim 0.1\%$) to IrTe$_2$ single crystals, we demonstrate the selective stabilization of a single structural phase transition with domain sizes four orders of magnitude larger than in unstrained samples. Complementary real and momentum space probes reveal this as a $6 \times 1$ charge ordered phase, a configuration that maximizes both the formation of Ir dimers and of Ir to Te charge transfer. We show that strain initiates this charge transfer already at room temperature, thereby removing the near degeneracy of the $3n+2$ ladder of phases and favoring the $6 \times 1$ phase at low temperatures. This energetic bias allows unprecedented spectroscopic access to the ground state of IrTe$_2$, including the previously unobserved bulk Dirac-like states, which undergo a Lifshitz transition due to the charge transfer. Concurrently, charge transfer results in a significant weakening of the majority of interlayer Te bonds in the unit cell, resulting in a tenfold reduction of interlayer hopping in the relevant states, and leaving the bulk Dirac states as the dominant interlayer transport channel. These results demonstrate the power of strain to influence phase transitions, bonding and topology in the layered tellurides, and more broadly in the 2D semimetals.

**Results and discussion**

**Strain-stabilized electronic structure.** Figure 1a shows the hexagonal crystal structure, typical of the layered TMDs, in the high-temperature $1 \times 1$ phase of $1T$-IrTe$_2$. In comparison, all low-temperature phases in IrTe$_2$, including the $6 \times 1$ (Fig. 1b), are characterized by the formation of Ir dimers stabilized by electronic energy gain. The density of these dimers increases in each of the successive charge ordered phases, reaching a maximum in the $6 \times 1$ phase, which is generally considered as the ground state of the system. The band dispersion along the high-symmetry $K\bar{L}L$ direction of the bulk Brillouin zone (Fig. 1c), as obtained by angle-resolved photoemission spectroscopy (ARPES), is displayed in Fig. 1e for the high-temperature phase in an unstrained sample. The corresponding Fermi surface (Fig. 1h) shows three-fold rotational symmetry consistent with the literature. Upon cooling the unstrained sample (Fig. 1f), subtle changes occur due to the phase transitions. The overall form of the electronic structure in the low-temperature phase strongly resembles that at high temperatures, but with broadened bands and a reduction of spectral weight. The lack of clear features results from the presence of multiple domains and phases (see Fig. 2). While the disappearance of the three-fold symmetry in the Fermi surface (Fig. 1f) implies a dominant domain orientation within the probed region (50 $\mu$m), the absence of a single-phase domain hinders analysis of the resulting electronic structure. In dramatic contrast, the strained sample (Fig. 1g) displays a rich spectrum of remarkably sharp bands over a wide energy range, implying a uniform signal over the probed region originating from a single phase. Strain was applied along the $a$-axis of the high-temperature phase using the home-built device pictured in Fig. 1d (see Methods). Of particular interest are sharp surface states and an apparent bulk-like hyperbolic dispersion close to $E_F$ (red arrows, Fig. 1g), discussed in more detail below, which are undiscernible in unstrained samples. The Fermi surface (Fig. 1j), reveals a clear directionality, breaking the rotational symmetry of the high-temperature phase and resulting in a mirror-plane along the $k_x = 0$ line. Cuts along the $k_x$ direction (Supplementary Fig. S2) showing repeated surface and bulk states reveal this phase has a $6 \times 1$ in-plane periodicity, which is difficult to discern in Fig. 1j due to the small size of the repeated features and the variation of spectral weight.

**Real-space structure and mapping of the strained phase.** The $6 \times 1$ periodicity of the strain-induced state is confirmed in Fig. 2, which demonstrates the effect of strain on the real-space surface structure as revealed by low-temperature scanning tunneling microscopy (STM) measurements. Unstrained samples display a mixture of differently oriented domains (Fig. 2a), which form due to the three-fold degeneracy of the high-temperature phase. Within these rotational domains, there exist multiple phases with different $3n+2$ periodicities (Fig. 2d). Again, in contrast, strained samples reveal a clear unidirectional domain (Fig. 2b), with a single $6 \times 1$ periodicity (Fig. 2f). The line cut in Fig. 2g shows that two nonidentical groups of three atoms comprise the $6 \times 1$ periodicity in agreement with previous work. Although the individual STM images are limited in size, the same $6 \times 1$ phase—always with the same orientation—is found in multiple images across the sample surface over hundreds of microns: more than half of the sample area (Supplementary Fig. S3). This macroscopic domain size is also seen in low energy electron diffraction (LEED), which averages over a region of similar dimensions (Fig. 2c and e). In the strained case, we indeed record a single domain orientation with $6 \times 1$ periodicity, contrasting the clear three-fold directionality of the unstrained sample. Further corroboration is obtained from micro-ARPES mapping (Fig. 2h), obtained by integrating the intensity of the sharp surface states characteristic of the $6 \times 1$ phase (Supplementary Fig. S4) across the sample surface. This
The experimental $k_x$ and $k_y$ directions are marked. c) Bulk Brillouin zone of the high-temperature phase showing the high-symmetry points. d) Photograph of the strain device. Samples are oriented to have strain along the $a$-direction of the high-temperature phase. e-g) ARPES measurements ($h\nu = 90.5$ eV) of (e) the high-temperature unstrained sample obtained along the LAL direction of the bulk Brillouin zone, (f) the low-temperature ($T = 30$ K) unstrained sample obtained along $HAH$ and (g) in the low-temperature strained sample. The cut along $k_y$ in (g) corresponds to along the Ir-dimer chain direction, i.e., the $x\bar{1}$ direction of the 6 × 1 unit cell ($b$-axis in real space). The intensity scale encodes the photoemission intensity measured in the experiment, which is proportional to the one-particle removal function, $A^- (k, \omega)$. h-j) corresponding Fermi surfaces for the three cases in (e)-(g). The corresponding Brillouin zones for the $1 \times 1$ and $6 \times 1$ phases are overlaid. Additional constant energy cuts are shown in Supplementary Fig. S1. Cuts along the $k_x$ direction (charge ordering direction, $q_{CO}$) revealing the 6 × 1 period are shown in Supplementary Fig. S2.

reveals that the 6 × 1 phase is found over a continuous region of dimensions $\sim 0.5 \times 0.4$ mm$^2$.

**Charge transfer and impact on interlayer bonding.** As is typical of phase transitions in the metal-chalcogenides, evidence of charge transfer is observed during the formation of the 6 × 1 phase. Due to the presence of polymeric bonds, the structure of IrTe$_2$ lies between that of pure 2D and 3D materials. This results in an Ir$^{3+}$ configuration and hence a partial charge of Te$_{1.5}$ on average in the high-temperature phase. In the low-temperature phase, a charge $\delta$ is transferred, which produces modified Ir$^{3+\delta}$ and Te$_{1.5-\delta/2}^{3-}$ species. The electronic energy gain from Ir-dimer formation competes with the lattice deformation energy, making a complete dimerization of the surface energetically unstable. As a result, both Ir$^{3+\delta}$ and Ir$^{3+\delta^+}$ species are present in the low-temperature phase. This can be readily observed in X-ray photoemission spectroscopy (XPS) where the two distinct peaks appear in the Ir 4$f$ spectra as shown in Fig. 3b. We note that the higher binding energy of the second peak implies reduced screening of the core potential, consistent with a reduced electronic density on the Ir atom (Ir$^{3+\delta}$). Further evidence for a charge transfer is discussed below (Fig. 4). The ratio of the peak areas tracks the relative dimer density and indicates a mixture of phases in unstrained samples. In contrast, for strained samples an increase of the Ir$^{3+\delta^+}$ peak produces a ratio that accords perfectly with the expectation for a single 6 × 1 phase (0.67). Crucially, XPS measurements in the high-temperature phase reveal a small population of the Ir$^{3+\delta^+}$ species is already evident above the transition temperature in strained samples, which is absent for unstrained samples. This implies strain actually induces a charge transfer from Ir to Te, and that this is central to understanding the phase stabilization. The analysis of the peak ratio in the high-temperature phase gives only 0.14, well below the ratio of 0.4 obtained in the 5 × 1 phase, which has the lowest dimer density of the ordered phases. An open question is whether the appearance of the second charge peak in the high-temperature phase implies the existence of dimers above the phase transition temperature, and indeed whether an ordered phase can be induced at room temperature by increasing the strain level. We note, however, that dimer formation depends on the competition between electronic and lattice energy, and it is therefore possible in the strained system that a charge transfer is induced without dimer formation. Finally, we remark that no evidence for a continuous phase transition is observed in temperature dependent ARPES at this strain level (Supplementary Fig. S5).
In order to gain more insight into this redistribution of charge in the strain-stabilized phase, in Fig. 3c, d we compare the calculated charge distributions in the $1 \times 1$ and $6 \times 1$ phases, respectively. Particularly notable is that there is a clear increase of charge density in the interlayer Te–Te region as a result of the phase transition, as charge is moved away from the Ir$^{3+\delta+}$ sites and onto the Te atoms. This implies the strain-induced charge transfer that produces the Ir$^{3+\delta+}$ signal.
in the high-temperature phase also redistributes charge into the interlayer region. The impact of this charge transfer on the out-of-plane Te–Te bonds\(^{25,32}\) is significant (the effect on the in-plane bonds has been addressed previously\(^{18}\)). A particularity of the tellurides in comparison to other TMDs is the presence of 3D polymeric bonding structures\(^{12}\), in place of the usual van der Waals gap. IrTe\(_2\) indeed contains a network of weak interlayer covalent bonding structures\(^{12}\), in place of the usual van der Waals gap.

The resulting interlayer bond weakening has been termed “depolymerization”\(^{12,25,26}\), although quantities relevant for bonding such as the out-of-plane bond strengths and hopping have not previously been addressed in detail for the low-temperature phase. We provide a direct experimental quantification of the effect that the bond weakening has on the electronic structure and electronic hopping in the out-of-plane direction.

We do so by comparing the out-of-plane \((k_z)\) dispersion (Fig. 4b) in the high-temperature and 6 × 1 phases, and reemphasize that it is only via the strain stabilization that we are able to access the electronic structure of the pure 6 × 1 phase. Between the low- and high-temperature phases, the majority of states maintain their small out-of-plane dispersion. However, a sizeable change in the warping of the Fermi contour is observed for bulk states on either side of the Brillouin zone boundary, highlighted by the blue lines in the two panels of Fig. 4b. In general, such warped Fermi surface contours are characteristic of a strong anisotropy in the electronic hopping parameters and are routinely observed in low-dimensional materials. A small warping corresponds to a low coupling between chains (1D) or planes (2D) along the relevant real-space direction\(^{34,35}\). In the case of the present out-of-plane dispersion, the narrowing of the warping in the \(k_z\) direction corresponds to a reduction of the interlayer hopping in the high-temperature phase. In contrast, significant dispersion is observed for these same states in the \((k_x, k_y)\) plane, highlighting the quasi-2D behavior of IrTe\(_2\) in the 6 × 1 phase.

In the low-temperature 6 × 1 phase, comparisons to other TMDs is the presence of 3D polymeric bonding structures\(^{12}\), in place of the usual van der Waals gap. IrTe\(_2\) indeed contains a network of weak interlayer covalent bonding structures\(^{12}\), in place of the usual van der Waals gap.

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These observations strongly implicate the interlayer bond weakening in the mechanism of the strain-stabilized phase transition\textsuperscript{25,26,32}. By inducing charge transfer, and hence interlayer bond weakening already in the high-temperature phase, strain reduces the amount of electronic energy that can be gained through dimerization of the Ir\textsuperscript{3+}\textsuperscript{4+} ions. This therefore destabilizes the nearly degenerate low-temperature phases, and pushes the system to favor the formation of the phase with the highest dimer density and hence highest possible gain in electronic energy i.e., the 6 × 1 phase. In this way, the less stable 3n + 2 phases are removed from the low-temperature phase diagram. The effect of strain is thus twofold: first, by defining a preferential direction, it breaks the degeneracy of the three-fold dimer orientation;\textsuperscript{36} second, it biases the energetic landscape of the system in favor of the 6 × 1 phase. Although the effects of strain are subtle in the high-temperature phase, as expected for the perturbative strain level applied, they pave the way for the stabilized phase transition, with dramatic results at low temperatures. We note that similar bonding behavior is realized in a number of di- and tri-telluride materials spanning the 2D and 3D regimes\textsuperscript{12}, suggesting strain or electrical gating as powerful methods to control structural behavior and dimensionality in this class of materials.

**Topological Lifshitz transition.** In contrast to the discussion above, a state dispersing in \(k_x\) appears around \(k_x = 0\) in the low-temperature phase (red arrow, Fig. 4b). Due to its strong out-of-plane dispersion (Supplementary Fig. S6), this state is only observed close to \(k_y = 5 \text{ Å}^{-1}\), i.e., the bulk A-point. This feature corresponds to the triangular block of states observed in Fig. 1g, and shown again at different \(k_y\) positions in Fig. 4d. The strong \(k_x\) dependence and “filled-in” nature of these states resulting from the projection of the bulk manifold reveals them as bulk states. Of note is that these bulk states have the cone-like hyperbolic dispersion of massless Dirac fermions (Supplementary Fig. S7), which occur ubiquitous in the group-10 TMDs and are predicted in IrTe\textsubscript{2}.\textsuperscript{13} However, the location of the type-II Dirac point at room temperature is above \(E_F\), hence inaccessible to ARPES, while at low temperature the mixture of phases typically hides their true nature. The observed shift of the Dirac point to 350 meV below \(E_F\) in the 6 × 1 phase occurs as the Dirac states are derived from the out-of-plane Te 5p\(_x\) orbitals and hence are strongly doped by the charge transfer into Te states as described above. As we have shown, it is only possible to access these states spectroscopically in strained samples. By moving the Dirac point and the electron-like portion of the Dirac cone into the occupied states, strain produces a Lifshitz transition similar to the temperature-driven transitions observed in WTe\textsubscript{2}\textsuperscript{37} and ZrTe\textsubscript{2}\textsuperscript{38}. Such a dramatic change in Fermi surface topology is likely to have a significant impact on the transport properties in this material. In particular, the topological nature of the states involved in the transition may explain the observed large, nonsaturating magnetoresistance\textsuperscript{39} similar to the behavior in other layered di-tellurides\textsuperscript{40–42}. The strain-stabilized order may even enhance such effects, paving the way to tunable magneto-resistive behavior. Given the out-of-plane character of the 5p\(_y\) orbitals involved in the Dirac states, it is plausible that particularly large changes in interlayer transport may be observed, and investigations of the resistivity anisotropy using e.g., focused ion beam methods\textsuperscript{43} are highly desirable in this regard. The potentially topological nature of this out-of-plane transport makes IrTe\textsubscript{2} layers especially interesting for tuning interlayer behavior in heterostructure architectures\textsuperscript{44,45}.

While a detailed discussion of the topological properties is beyond the scope of the current article, we nonetheless highlight a surprising observation regarding these Dirac-like states: the dispersion is not compatible with a single Dirac cone. This can be seen, for example, from the cut at \(k_y = -0.05 \text{ Å}^{-1}\) in Fig. 4d, which reveals two partially overlapping cone-like dispersions. Indeed, the in-plane dispersion of these Dirac states (Fig. 4c) reveals rich structures related to these bulk states, comprising the central hyperbola around \(k_y = 0 \text{ Å}^{-1}\), which has a bow tie-like Fermi contour, and additional hyperbolic cones centered at around \(k_y = \pm 0.15 \text{ Å}^{-1}\) (see also Supplementary Fig. S7), which form asymmetric arcs. The spacing of this latter behavior is compatible with the periodicity imposed by the 6 × 1 phase, but their unusual distributions and the origin of the additional central bow tie structure remain unclear. Further detailed investigations including theoretical work will be required to clarify the nature of these states.

**Conclusion.** In summary, we have selectively stabilized the 6 × 1 charged ground state of the layered topological semimetal IrTe\textsubscript{2} by employing uniaxial strain. The induced macroscopic domain sizes allow detailed insights into the electronic structure at the surface in both real and momentum space. Charge transfer in the strain-stabilized phase strongly reduces the out-of-plane Te bond strengths, electronically decoupling the layers and resulting in a Lifshitz transition, granting access to a previously inaccessible bulk-Dirac dispersion that acts as the main interlayer hopping channel. Complementary measurements of the transport properties of the 6 × 1 phase, as well as of monolayer IrTe\textsubscript{2}, are therefore highly desirable. We note that in contrast to the tensile strain utilized here, uniaxial compression may stabilize superconducting behavior\textsuperscript{22–24} which, concomitant with the topological states, opens the possibility of strain-tunable topological superconductivity in IrTe\textsubscript{2}.

**Methods**

**Sample growth and characterization.** Single crystals of IrTe\textsubscript{2} were grown using the self-flux method\textsuperscript{46–48}. Samples were characterized by magnetic susceptibility and resistivity measurements\textsuperscript{29}, confirming the bulk phase transition temperatures of \(T_N = 278 \text{ K}\) and \(T_S = 180 \text{ K}\) in unstrained samples. Samples to be prepared for straining were chosen to have large flat areas with minimal cracks or flakes at the surface as viewed under an optical microscope, in order to allow for more homogeneous strain application. Bulk samples were initially cleaved with a scapel to remove thicker layers, and then mounted onto the unstrained device and further thinned by Scotch tape cleaving.

**Strain device and characterization.** The strain device, shown in Fig. 1d, is a home-built design consisting of three parts: a molybdenum (Mo) base plate, a copper beryllium (CuBe) bridge, and a rounded aluminum (Al) block, which is placed under the bridge. The maximum height of the Al block is machined to be slightly larger than the distance from the underside of the top of the CuBe-bridge to the surface of the base plate. Thus, when the pieces are screwed together, the CuBe-bridge is forced to bend by the Al block. Samples were oriented with Laue diffraction such that the bending axis was perpendicular to one of the three-fold symmetric directions in the high-temperature phase i.e., along the \(a\)-axis of the high-temperature phase (or equivalently, the \(b\)-axis). This is along the in-plane bond direction half of the Ir-Ir dimers in the 6 × 1 unit cell. The sample was mounted on the CuBe-bridge of the strain device using a two-part epoxy (EPO-TEK E4110), which was cured and allowed to cool before strain was applied. Strain was applied manually by tightening the screws on the underside of the device, which connect the Mo base plate with the Cu-bridge. To ensure maximum directed strain, the screws were tightened in pairs. All four screws were tightened loosely, following which the two screws on one side of the bridge were fully tightened. The remaining two screws were then gradually tightened in an alternating fashion, in order to allow as even an application of strain across the device as possible. The strain magnitude was calibrated using commercial strain gauges (Omega Engineering) with nominal resistance 350 Ω and gauge factor, \(k = 2.2\). The gauge was attached to an unstrained device of the design described above using the same epoxy as for the samples, and was then connected to a home-built balanced Wheatstone bridge circuit in a “quarter bridge” configuration. Together with a second (passive) gauge, this constituted one arm of the bridge. The second arm consisted of two 390 Ω resistors, and a variable resistor (10 Ω) was used to balance the circuit. A source voltage of \(V_s = 5 \text{ V}\) was applied. Once balanced, the gauge was...
strained using the device and the output voltage was recorded with a Keithley digital multimeter. The typical output voltage induced by strain ($V_a = 4$ mV) was well above the noise level (50 μV). The output voltage was converted to a strain value via:

$$\epsilon = \frac{4V}{kV}$$

where $\epsilon$, the total strain, is the sum of bending (tensile) strain and perpendicular (compressive) strain. In such a strain geometry, the perpendicular strain is considerably smaller than the bending strain, hence “strain” in the main text refers to the tensile bending strain. To separate further these components requires additional gauges to be placed on the underside of the device, which is impractical given the geometry and small size. From the above relation, we obtained the strain characteristics of the device. A maximum strain of up to 0.2% was initially recorded during tightening due to plastic deformation of the CuBe bridge. This relaxed to around 0.1% once all screws were tight, which is therefore the maximum strain that could be applied to the sample using this particular device and Al block combination.

**Data availability**

Data are available from the corresponding authors upon reasonable request.

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