Two-dimensional (2D) topological insulators (TIs) are promising platforms for low-dissipation spintronic devices based on the quantum-spin-Hall (QSH) effect, but experimental realization of such systems with a large band gap suitable for room-temperature applications has proven difficult. Here, we report the successful growth on bilayer graphene of a quasi-freestanding WSe$_2$ single layer with the 1$T'$ structure that does not exist in the bulk form of WSe$_2$. Using angle-resolved photoemission spectroscopy (ARPES) and scanning tunneling microscopy/spectroscopy (STM/STS), we observe a gap of 129 meV in the 1$T'$ layer and an in-gap edge state located near the layer boundary. The system’s 2D TI characters are confirmed by first-principles calculations. The observed gap diminishes with doping by Rb adsorption, ultimately leading to an insulator-semimetal transition. The discovery of this large-gap 2D TI with a tunable band gap opens up opportunities for developing advanced nanoscale systems and quantum devices.
The quantum-spin-Hall (QSH) insulator films are characterized by a two-dimensional (2D) band gap within the film and one-dimensional (1D) metallic edge states that bridge the band gap\(^1\)–\(^3\). The edge states are spin polarized by spin–orbit coupling in a chiral configuration relative to the momentum and the edge normal; they are protected by time-reversal symmetry and thus robust against weak disorder. These edge conducting channels are ideally suited for low-dissipation transport of spin information relevant to spintronic applications\(^4\)–\(^11\). The first experimental demonstration of the QSH effect was made in HgTe/(Hg, Cd)Te quantum wells\(^6\)–\(^7\), but the system configuration was complex; furthermore, its gap was very small, and the edge spin conduction effect was observed only at very low temperatures. Nevertheless, the proof of principle has spurred a great deal of community interest in finding simple robust 2D TI systems with a large band gap. Extensive theoretical explorations have been made in various systems ranging from 2D elemental buckled lattices to transition-metal pentatellurides and to oxide heterostructures\(^8\)–\(^15\), but experimental realization of systems with properties readily amenable to applications has proven to be elusive.

Transition-metal dichalcogenides are promising for developing 2D topological insulators (TIs). These layered materials can be easily fabricated in single-layer forms. Some of them containing heavy elements with a strong spin–orbit coupling are predicted to be 2D TIs; notable examples include single-layer 1\(\text{T}\) MX\(_2\) (M = Mo or W; X = S, Se, or Te)\(^16\). Experimental work to date has mostly focused on single-layer WTe\(_2\) because the 1\(\text{T}\) phase is the stable form in the bulk\(^17\)–\(^19\), but not necessarily so for the other cases, and W has a very strong atomic spin–orbit coupling. While some of the other MX\(_2\) materials can be converted to the 1\(\text{T}\) phase by intercalation or strain, the added complexity makes it difficult to prove the QSH state\(^20\). WSe\(_2\), the material chosen for the present study, exhibits a hexagonal 2\(H\) structure in the bulk, which is of great interest for its large indirect band gap and strong spin-valley coupling\(^21\)–\(^22\), but the single layer with the 1\(H\) structure is not topological.

In this work, we show that single layers of WSe\(_2\) can be prepared instead in the 1\(\text{T}\) phase, which is topological with a gap of 129 meV based on angle-resolved photoemission spectroscopy (ARPES) experiments and 116 meV based on G\(_0\)W\(_0\) calculations. This gap is more than twice as large as that reported for 1\(\text{T}\) WTe\(_2\)\(^7\) and, furthermore, it can be tuned with surface doping to undergo an insulator–semimetal transition. Single-layer 1\(\text{T}\) WSe\(_2\) is thus an excellent candidate for developing spintronics based on the QSH effect.

**Results**

**Film structure and electron diffraction patterns.** Figure 1a shows top and side views of the structure of 1\(\text{T}\) and 1\(H\) single-layer WSe\(_2\). The observed bulk crystal structure is the 2\(H\) phase consisting of a van der Waals stack of 1\(H\) layers in an ABA sequence. The bulk 1\(\text{T}\) structure, if existent (as in WTe\(_2\)), would involve an ABC stacking of the 1\(\text{T}\) layers together with a lattice distortion along the \(x\) direction. The surface unit cells and some special points are indicated in Fig. 1b. In our experiment, films of WSe\(_2\) were grown in situ on a bilayer graphene-terminated 6\(H\)-SiC(0001)\(^23\) via van der Waals epitaxy\(^24\)–\(^25\). Reflection high energy electron diffraction (RHEED) shows that a single-layer WSe\(_2\) grown at a substrate temperature of 280 °C (Fig. 1c) exhibits a mixture of 1\(H\) and 1\(\text{T}\) phases, both with sharp diffraction patterns. At higher substrate growth temperatures, the 1\(H\) phase becomes more prevalent, and it is the only phase observed at growth temperatures above 400 °C (Fig. 1c). The 1\(\text{T}\) phase is favored.

**Fig. 1** Film structure and electronic band structure of single-layer WSe\(_2\). a Top and side views of the atomic structure of single-layer 1\(H\) and 1\(\text{T}\) WSe\(_2\). b Corresponding 2D Brillouin zones with high symmetry points labeled. c RHEED patterns taken from a 1\(H+1\text{T}\) sample and a pure 1\(H\) sample. d Core level scans taken with 100 eV photons. The 1\(H+1\text{T}\) sample shows mixed core level signals. e ARPES maps along \(\Gamma\) and \(\Gamma\) taken from the two samples at 10 K.

**TABLE**

| Energy (eV) | Intensity |
|-------------|-----------|
| -60         | -40       |
| -30         | -10       |

This table shows the energy levels and corresponding intensities for the ARPES maps. The energy levels are presented in eV, and the intensity is shown in a range from -60 to -30 eV.
at lower growth temperatures, and it becomes the only phase observed at a growth temperature of 130 °C; however, the film quality is poor as evidenced by a fuzzy RHEED pattern (not shown here). Scans of the core levels (Fig. 1d) show splittings of the Se 3d and W 4f states in the mixed phase due to the inequivalent structures; an analysis of the W core level line shape indicates that the 1H and 1T phases have a coverage ratio of 1.8 on the surface.

**Band gap determined from ARPES and calculation.** ARPES maps taken from the single-layer samples at 10 K along the ΓK direction are shown in Fig. 1e. The pure 1H phase shows a sizable gap below the Fermi level; it is therefore a semiconductor similar to the bulk case.21 The top valence band at Γ splits into two branches toward K because of the strong spin–orbit coupling of W. The valence band maximum is at K, consistent with prior studies of this phase24,26. For the mixed sample, the 1T phase gives rise to additional valence bands of very different dispersion relations, and the topmost valence band reaches near the Fermi level (Fig. 3a). Interconnection between the 1T and 1H phases have a coverage ratio of 1.8 on the surface.

**Fig. 2** Band structure and band gap of 1T′ WSe2. a Brillouin zones of 1T′ WSe2 with three domains separated by 120°. b Calculated band structure of 1T′ WSe2. c Detailed band structure along ΓY with the indirect gap E_g labeled. The Se 4p and W 5d weights for the two topmost bands near the zone center are indicated by the red and blue dot sizes, respectively. d Two ARPES maps taken along ΓY and ΓX with the sample at 10 K. The overlaid red and cyan curves are computed bands for the mixed-domain configurations. The experimental E_g is indicated.
other (Fig. 3b) at a doping level of $N_C = 6 \times 10^{13}/\text{cm}^2$, beyond which the system becomes a semimetal with a negative gap (Fig. 3c). Moreover, the band shapes become noticeably different. An implication is that Rb deposition leads to, in addition to surface electron doping, structural modiﬁcations through incorporation or intercalation of Rb in the lattice\textsuperscript{22,27}. The Rb 3$d$ core level line shapes (Fig. 3d) reveal multiple components indicating different Rb sites that vary in population with increasing Rb coverages. The tunability of the QSH gap can be a useful feature relevant to applications. The insulator-semimetal transition at $N_C$ offers a mechanism to switch off the QSH channels.

**Band gap and edge conductance measured by STM/STS.** The scanning tunneling microscopy/spectroscopy (STM/STS) measurements in a different chamber performed on a sample with a coverage of 1/2 layer made in the ARPES system transferred under a capping layer through air (see Methods section) reveal single-layer islands of $1H$ and $1T'$ structures and some two-layer islands (Fig. 4a). STS scans reveal a large band gap for the interior of $1H$ islands (Fig. 4d) and a much smaller gap for the interior of $1T'$ islands in agreement with the ARPES data, although the STS data are expected to be thermally broadened at the measurement temperature of 77 K relative to the ARPES data taken at 10 K. Figure 4b, c shows atom-resolved images of the $1T'$ and $1H$ phases, respectively. The orientation of the triangular $1H$ phase is the same as the underlying bilayer graphene, and the image exhibits a Moiré pattern\textsuperscript{24}. The $1T'$ phase has a rectangular lattice instead (Fig. 1a); all three domain orientations separated by $120^\circ$ are observed.

Figure 4e is an STM image with a $1T'$ island covering the right half only. Figure 4f shows STS curves taken at a point (A) in the island very close to the island edge (green curve A) and another point (B) in the island still near but farther away from the edge (blue curve B); the two points A and B are indicted by the correspondingly color-coded dots in Fig. 4e. With the Fermi level aligned relative to the included ARPES map\textsuperscript{28}, the gap is indicated by the two vertical red dashed lines. Also shown is a reference red curve C taken from a point deep inside the island. It shows a clear gap; the small residual tunneling density of states (DOS) within the gap can be attributed to tunneling/coupling to the underlying bilayer graphene. Similar nonzero DOS in the gap is evident in single-layer WTe\textsubscript{2}\textsuperscript{17}. Curve A, relative to curve C, shows much higher DOS within the gap, suggesting contributions from edge states\textsuperscript{13,14,17}; this extra DOS is much reduced for curve B. Details regarding the edge-state contributions through the island boundary are shown in Fig. 4g, where the differential conductance is plotted as a function of $x$ (defined in Fig. 4e) and energy, with the gap indicated by two horizontal dashed lines. A red dashed rectangle in Fig. 4g highlights the enhanced edge conductance within the gap near the island edge. Note that STS can be affected by quasiparticle interference (QPI) effects, as indicated in Fig. 4g; similar effects have been seen in WTe\textsubscript{2}\textsuperscript{29}. For curve A in Fig. 4f, the strong peak at about $-170$ meV, which might seem strange, is caused...
The QSH gap in WSe2 originates from band inversion of the W 5d and Se 4p states near the zone center and anti-crossing of the bands caused by spin–orbit coupling (Fig. 2c). A similar theoretical analysis for WTe2 shows that the relevant reverse-ordered states near the zone center are both dominated by the W 5d states. The Te states play a relatively minor role. The self-hybridization of the W 5d states is actually weaker, leading to a smaller QSH gap. Most single-layer MX2 materials, including WSe2, are stable only in the 1H phase, which gives rise to ordinary semiconductors. Our demonstration of the successful creation of single-layer 1T' WSe2 with a sizable QSH gap offers an important example of materials engineering. Its QSH gap of 129 meV is more than five times the thermal energy $k_B T$ at room temperature, suggesting that it is suitable for QSH electronics at ambient temperature. While the system is only metastable, its demonstrated stability up to ~280 °C is more than sufficient. For comparison, 1T' WTe2, which has garnered much attention, actually has a smaller QSH gap not conducive to ambient-temperature spintronics. Unlike exfoliated materials, the 1T' WSe2 films grown by molecular beam epitaxy, as demonstrated herein,
should be readily adaptable to large-scale fabrication of devices such as topological field effect transistors. Other materials such as superconductors can be added by molecular beam epitaxy, thus offering opportunities to realize additional functionality and novel properties including Majorana fermions. Our work expands the family of large-gap QSH materials and inspires further experimental exploration of novel QSH systems.

Methods

Experimental details. Thin films of WSe₂ were grown in situ in the integrated molecular beam epitaxy (MBE)/ARPES systems at beamlines 12.0.1 and 10.0.1 (Advanced Light Sources, Lawrence Berkeley National Laboratory). Substrates of 6H-SiC(0001) were flash-annealed for multiple cycles to form a well-ordered bilayer graphene on the surface. Films of WSe₂ were grown on top of the substrate at a rate of 30 min per layer by co-evaporating W and Se from an electron-beam evaporator and a Knudsen effusion cell, respectively. The growth of different phases of WSe₂ is controlled by the substrate temperature. The 1T′ phase starts to form at 130 °C but the film quality is poor at low growth temperatures. The best 1T′ phase is obtained at near 280 °C, but the 1H phase also forms and completely dominates at temperatures near 400 °C. ARPES measurements were performed with an energy resolution of <20 meV and an angular resolution of 0.2°. Each sample’s crystallographic orientation was precisely determined from the symmetry of constant-energy-contour ARPES maps. The surface electron density with Rb doping is determined from the symmetry of constant-energy-contour ARPES maps. The data that support the findings of this study are available within the article or from the corresponding author upon request.

Received: 7 February 2018 Accepted: 24 April 2018
Published online: 21 May 2018

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Acknowledgements

This work is supported by the US Department of Energy, Office of Science, Office of Basic Energy Sciences, Division of Materials Science and Engineering, under Grant No. DE-FG02-07ER46383 (to T.-C.C.), the National Science Foundation under Grant No. EFMA-1542747 (to M.Y.C.), and the Ministry of Science and Technology of Taiwan under Grant No. 104-2112-M-002-013-MY3 and the Center of Atomic Initiative for New Materials, National Taiwan University (to W.-W.P.). The Advanced Light Source is supported by the Director, Office of Science, Office of Basic Energy Sciences, of the US Department of Energy under Contract No. DE-AC02-05CH1231. The work at Academia Sinica is supported by a Thematic Project.

Author contributions

P.C. and T.-C.C. designed the project. P.C. with the aid of C.-Z.X., A.-V.F., and T.-C.C. performed MBE growth, ARPES measurements, and data analysis. Y.-H.C. and M.Y.C. performed first-principles calculations. N.-W.P., W.-L.S., and D.-S.L. conducted STM/STS experiments. T.-C.C., P.C., W.-W.P., and M.Y.C. interpreted the data. T.-C.C. and A.-V.F. jointly led the ARPES project. P.C. and T.-C.C. wrote the paper with input from other co-authors.
