Laser-beam patterned topological insulating states on thin semiconducting MoS$_2$

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Identifying the two-dimensional (2D) topological insulating (TI) state in new materials and its control are crucial aspects towards the development of voltage-controlled spintronic devices with low power dissipation. Members of the 2D transition metal dichalcogenides (TMDCs) have been recently predicted and experimentally reported as a new class of 2D TI materials, but in most cases edge conduction seems fragile and limited to the monolayer phase fabricated on specified substrates. Here, we realize the controlled patterning of the 1T'-phase embedded into the 2H-phase of thin semiconducting molybdenum-disulfide (MoS$_2$) by laser beam irradiation. Integer fractions of the quantum of resistance, the dependence on laser-irradiation conditions, magnetic field, and temperature, as well as the bulk gap observation by scanning tunneling spectroscopy and theoretical calculations indicate the presence of the quantum spin Hall phase in our patterned 1T" phases.

Two-dimensional (2D) topological insulating (TI) states have been mainly investigated in HgTe/CdTe or InAs/GaSb quantum well systems (1-3). In the 2D TI state the quantum spin Hall (QSH) effect emerges thanks to the simultaneous presence of a bulk energy gap and gapless helical edge states protected by time-reversal symmetry, namely, opposite and counter-propagating spin states forming a Kramers doublet. Interestingly, 2D TI states were first theoretically predicted for graphene (4-6), but experimentally reported in only few related systems (7-9) such as low-coverage Bi$_2$Te$_3$ nanoparticle-decorated graphene (8). Moreover, control of the QSH phase in graphene-based systems remains a challenge.

Recently, a family of atom-thin transition metal dichalcogenides (TMDCs) materials has also been predicted to exhibit the QSHE (10-12), having its origin in the natural band inversion of the 1T' phase (one of the phases of TMDC; see Supplementary Material (SM) 1) and the spin-orbit coupling (SOC)-induced band-gap opening. Moreover, the TI state has been experimentally verified in the case of WTe$_2$ (13-15) thanks to the stability and high-quality of WTe$_2$ monolayers carefully formed on bilayer graphene/atom-thin hBN. Various signatures of the TI state have been demonstrated in this
material (13, 15), including the latest observation of a half-integer quantum value of resistance \( R_Q/2 = h/2e^2 = 12.9 \Omega \), where \( h \) is Planck’s constant and \( e \) is the charge on the electron) (14).

However, the TI phenomenon in WTe\(_2\) is rather sensitive to the substrates, synthesis process, and the chemical environment, making its controlled use in practical applications challenging. Moreover, although the (metastable) 1T’ phase can be found or induced in other TMDCs (23,25), nobody has demonstrated the existence of the QSH\(_E\) in these other TMDCs. The conditions under which helical edge states can exist at the 1T’-2H interfaces is a crucial problem which should be mastered for both TI physics and its applications. Here, we pattern a metallic 1T’-phase (SM 1) embedded into the non-topological and semiconducting 2H phase of thin MoS\(_2\) flakes, one of the TMDC family, via a facile protocol based on laser beam irradiation (23) and, via transport measurements, reveal that the embedded 1T’ phase exhibits the TI state.

In the present experiments thin MoS\(_2\) flakes are obtained by mechanical exfoliation of the bulk material and transferred onto a SiO\(_2\)/Si substrate. Layer thicknesses \(~17\) nm have been confirmed by atomic force microscopy (AFM) and optical microscopy (OM). OM and AFM images of a flake with two different patterns created by laser beam irradiation are shown in Figs. 1A and 1B (SM 2). The large rectangular pattern (with two electrode probes) is analogous to that previously used for graphene under high-magnetic fields (9) and for monolayer WTe\(_2\) (14). In contrast, the H-letter like pattern is analogous to that used in HgTe/CdTe quantum wells (I) and in our previous Bi\(_2\)Te\(_3\)-nanoparticle decorated graphene system (8). The presence of helical edge states was confirmed in all these systems. In the OM image, the color of the irradiated patterns drastically changes to semi-transparent (Fig. 1A). The cross-sectional AFM image of the irradiated part reveals a decrease in the thickness of about 10 nm (Fig. 1B). These observations are consistent with previous reports in multilayer MoTe\(_2\) (23), which demonstrated a layer thinning effect caused by the burn-out of individual monolayers due to in-plane heat accumulation from the laser beam irradiation. Heat introduced by the laser radiation causes, in turn, a 2H-1T’ transition in the upper layers (Fig. 1C) ((23); SM 3).

Typical Raman spectra are shown in Fig. 1D and we can use it to estimate the number of 1T’ layers formed. For the non-laser-irradiated region (2H phase), the Raman peaks are evident for multi-layer (>five layers) MoS\(_2\), showing the large and characteristic \(E_{2g}\) and \(A_{1g}\) peaks, while the pattern of the peaks for the laser-irradiated region (1T’+2H) has definitely changed. Both peaks are still visible, which can be attributed to the 2H layers remaining underneath the 1T’ layers (SM 3), while the other peaks can be certainly attributed to the 1T’ phase. Because the thickness of the laser-irradiated part is \(~7\) nm, as mentioned above, and the minimum thickness expected for the remaining 2H layers underneath the 1T’ ones is 5 layers \(\times\) \(~0.7\) nm = \(~3.5\) nm (the minimum thickness to get Raman signal), we estimate a maximum thickness for the 1T’ layers of \(~3.5\) nm. Since we expect the laser irradiation to slightly damage the top-most 1T’ layers, the number of pristine 1T’ layers is expected to be at most a few.

Photo luminescence (PL) signals of the laser-irradiated parts are shown in Figs. 1E and 1F. These reveal that the peak positions shifted to lower wavelengths (i.e., higher energies) and the peak intensities decreased with laser-irradiation time. This is also compatible with the above-mentioned interpretation. When the upper layers are transformed into the 1T’ phase, these layers cause no PL signals (16), whilst the remaining bottom semiconducting layers still yield a (reduced) signal (Fig. 1C). Furthermore, X-ray photoelectron spectroscopy (XPS) of the laser-irradiated part demonstrates the two types of hybridization of Mo 3d orbitals associated with the 1T’ and 2H phases (Fig. 1G).

Resistance (R) measurements as a function of back gate voltage (\(V_{bg}\)) of the samples shown in Fig. 1A are shown in Fig. 2. Au/Ti electrodes are in contact only with four corners of the rectangular pattern and each branch of the H-letter-like pattern so as to measure charge-spin transport only in the 1T’ region.
(insets of 2A and 2C). Because of the presence of a Schottky barrier at the 1T’/2H junction (> ~0.2 eV; see Fig. 3C for the semiconducting band gap in 2H region) (16,17), the 2H semiconducting layers below the 1T’ layers are expected to give a negligible contribution to these measurements, particularly at low temperatures.

For the rectangular pattern, the two-terminal resistance between electrodes 1,3 and 2,4 is measured as a function of $V_{bg}$ by flowing a constant current between electrodes 1,3 and 2,4 (see insets). The results of two samples formed after different irradiation times to each point (20 s and 30 s) are shown in Figs. 2A and 2B, respectively. Individual figures demonstrate two $R$ peaks. At high $V_{bg}$, $R$ peaks of $R_{34} \approx R_Q/2$ are confirmed in both figures (at $V_{bg} \approx +25V$ and +19V in Figs. 2A and 2B, respectively). Larger $R$ peaks are also observed at negative $V_{bg}$’s ($R_{34} \approx R_Q$ and $\approx 3R_Q/2$ at $V_{bg} \approx -5V$ and $\approx -10V$ in Figs. 2A and 2B, respectively). For the $H$-letter like pattern, when a constant current flows between electrode pair 1-2, the non-local resistance ($R_{NL}$) between electrode pair 3-4 ($R_{34}$) is measured as a function of $V_{bg}$ (see inset of Fig. 2C). Figure 2C shows the result. A $R_{NL}$ plateau of $R_{34} \approx R_Q/4$ at high $V_{bg}$ ($\approx +20-25V$) is confirmed with a $R_{NL}$ peak of $R_{34} > R_Q/2$ at low $V_{bg}$ ($\approx +5V$) also appearing. As usually occurs, large $V_{bg}$ values need to be applied to significantly tune the Fermi level via the SiO$_2$/Si substrate ((24); SM 4).

The $R_Q/2$ and $R_Q/4$ of $R$ peak values observed at high $V_{bg}$ suggests the appearance of the QSH phase without and with dephasing in metal electrodes, respectively (1,8). First, the $R$ peak values $\sim R_Q/2$ confirmed for the 1T’ rectangular patterns (Figs. 2A and 2B) are consistent with the presence of helical edge modes without dephasing. In this case, the two counter-propagating spin channels can be preserved at two different quasi-chemical potentials between the electrodes, leading to a net current flow along the edges with $R$ equal to $R_Q/2$ (as based on the Landauer-Büttiker (LB) formalism). Such a two-terminal resistance plateau $\sim R_Q/2$, reported in both rectangular mono-layer WTe$_2$ (14) and in large rectangular graphene under high magnetic fields with a similar electrode connection to the present one (insets of Fig. 2(A,B)) (9) was presented as evidence for helical edge states. Moreover, it should be noticed that the $R$ peak values $\sim R_Q/2$ observed in Figs. 2A and 2B are slightly larger than $R_Q/2$ ($\approx 0.55-0.6R_Q$). This deviation agrees well with the $R$ plateau value of 0.56$R_Q$ observed in Ref. (9), attributed to the microscopic details of the edges (e.g., local charge puddles) (3,8,18). Indeed, a somewhat irregular boundary (i.e., topological/non-topological phase junction) can be observed at some points of our 1T’-2H junction at the top-most surface layer (SM 7), which is noticeably different from clean interfaces of other parts, and may lead to local charge puddles (18). Ref. (14) also reported deviations from $\sim R_Q/2$ to larger values in long-channel samples ($>>$ a few 100nm). Remarkably, in our case, $R$ does not deviate significantly from $R_Q/2$ even for ~1 µm channel lengths, possibly because the high uniformity of the 1T’ phase formed by our highly uniform laser beam (SM 5).

Second, the $R_{NL}$ plateau value $\sim R_Q/4$ (Fig. 2C) corresponds to the case of helical edge modes with dephasing in the metal electrodes in the $H$-letter like pattern with four metal electrodes (inset) and nicely agrees with the result in Refs. (1) and (8). Once the helical edge electrons enter the voltage electrodes, they interact with a reservoir containing an infinite number of low-energy degrees of freedom so that time-reversal symmetry is effectively broken by the macroscopic irreversibility. In particular, a resistance plateau can be confirmed in Fig. 2 (C). The plateau shape, as opposed of a peak, is attributed to the channel width being narrower (~1 µm) than that in the rectangular pattern (~2 µm) and the usage of highly uniform laser beam as mentioned above (SM 6). This is also consistent with the results in (1). Consequently, the two counter propagating channels equilibrate at the same chemical potential, determined by the voltage of the metallic electrodes, leading to dissipation and, thus, emergence of integer fractions of $R_Q$. Therefore, these $R$ peaks (i.e., $\sim R_Q/2$ in the rectangular pattern and $\sim R_Q/4$ in the $H$-letter like pattern) evidently suggest the presence of the helical edge states in the
patterned 1T′ layers. These $R$ peaks without influence of dephasing will originate from either monolayer or a few layers existing below the top-surface layer, while it is speculated that the other $R$ peaks ($\sim R_0$ and $\sim 3R_0/2$ in Figs. 2A and 2B, respectively), which are typically larger than those observed at high $V_{bg}$, are derived from the top-surface 1T′ layer with edge defects (see SM 7,8) (20).

Perpendicular magnetic-field ($B_{\perp}$) dependence measurements for the samples shown in Figs. 2B and 2C are demonstrated in Fig. 3A. As $B_{\perp}$ increases, the conductance $G$, corresponding to the inverse of three $R$ peak values ($R_0/2$ red symbol) and $3R_0/2$ blue observed at high and low $V_{bg}$, respectively, in Fig. 2B and $R_0/4$ at high $V_{bg}$ in Fig. 2C (pink), exponentially decrease. $G$ values corresponding to the inverse of two other $R$ peaks (Fig. 2A and Fig. 2D) remain almost unchanged. These results are in good agreement with those in the QSHE observed in WTe$_2$ (14) and our Bi$_2$Te$_3$ decorated graphene (8), and evidently support that the first three $R$ peaks can be attributed to helical edge states. Only when the Fermi level is set to the Kramers degeneracy point, $R$ values can well reflect the band gap opening due to Zeeman effect caused by applied $B_{\perp}$, resulting in the observed exponential decrease in $G$. The pink symbol observed in the $H$-letter like pattern show a saturation at lower $B_{\perp}$ (~3 T), because the channel width (~1 µm) is smaller than the rectangular pattern and other edge effects may disturb the exponential $G$ decrease.

As far as zero-$B_{\perp}$ temperature dependence is concerned, $G$ corresponding to inverse of the three $R$ peak values ($R_0/2$, $3R_0/2$, and $R_0/4$) remain constant up to the transition temperatures ($T_{c1}$) of $\sim 40$ K (red symbol), $\sim 25$ K (blue), and $\sim 30$ K (pink), respectively (Fig. 3B). At temperatures higher than the individual $T_{c1}$, $G$ increases as temperature increases, following the thermal activation formula (i.e., a linear dependence in the Arrenius plot of Fig. 3B) with activation energies of $\sim 15$ meV (red symbol), $\sim 10$ meV (blue), and $\sim 7$ meV (pink), respectively. Moreover, $G$ increases agains above $T_{c2}$ $\sim 60$ K (red), $\sim 50$ K (blue), and $\sim 60$ K (pink), respectively, with larger slopes. The $T_{c1}$, which are lower than 100 K as reported in WTe$_2$ (14), are attributed to bulk gaps being smaller than that of WTe$_2$ (19). In contrast, $G$ increases at temperatures $> T_{c2}$ have not been observed previously (8, 14). This can be attributed to the thermally activated carriers flowing into the 1T′ QSH phase region from the 2H semiconducting region over the Schottky barrier (or band discontinuity) at the 1T′/2H layer interface, because the barrier height is much larger than the bulk gaps (17). The largest activation energy (i.e., barrier height or band discontinuity) for the pink symbol is consistent with the smallest gap value of $\sim 7$ meV as mentioned above.

Scanning tunneling spectroscopy (STS) spectra of the non-laser-irradiated 2H region and the irradiated rectangular 1T′-pattern are shown in Fig. 3C-E (SM 9). For (C), it demonstrates evident gaps $\sim 0.6$ eV, which are in good agreement with n-type semiconducting gap of thin MoS$_2$ with number of the layers larger than five. This gap is large enough to embed the present topological gaps with $\sim 10$ meV order. For (D), in the two bulk points, STS gaps of $\sim 25$ - 35 meV are confirmed, while the gap disappears at an edge. The bulk gap values almost agree with the values estimated from the temperature dependence of resistance peaks as mentioned above. Although they are smaller than the 45 meV gap reported in 1T′-WTe$_2$ (15), they are appropriate for 1T′-MoS$_2$. For (E), as $V_{bg}$ runs away from Kramers point (i.e., $V_{bg}$ $\sim -10$V), the dI/dV increases and the dI/dV dip disappears, resulting in just a metallic behavior of 1T′ phase. This supports that disappearance of the bulk gap in (D) is attributed to edge current due to Kramers point. Consequently, all results suggest that the laser-created 1T′ phases can be in QSH phases with helical edge modes.

The existence of helical edge states at the 1T′/2H interface is further supported by theoretical calculations. We first examine the electronic states of a heterostructure formed by two interfaces dividing a MoS$_2$ monolayer into three regions, 2H/1T′/2H (Fig. 4A). The three regions are infinite along the interfaces direction, but the system is finite along the perpendicular one. The relaxed atomic
positions of the unit cells near the interfaces (detail shown in Fig. 4B) are obtained from density functional theory (DFT) structural relaxation, in which we have kept a flat structure away from the interfaces as to simulate the underlying MoS$_2$ layers. We have corroborated our modeling by investigating other atomistic models of the 2H/1T' interface, starting from different initial configurations, also considering an asymmetric periodic model, and relaxing them using different constraints (see SM11,12). The DFT results in all these investigated cases converged to a qualitatively similar picture in terms of structural stability and band structure, which supports the robustness of the model here proposed.

Fig. 4C shows the overall band structure for the system shown in Fig. 4A. Our choice of a centrosymmetric system with two interfaces makes all the bands doubly-degenerate. One can clearly identify the bulk band inversion associated with the 1T' phase near $\Gamma$ and the gap opened by the SOC (and also by the quantum confinement due to the finite width of the 1T' region). Importantly, we learn from these calculations that the 1T' phase gap is in the middle of the much larger gap of the 2H phase (indicated by a yellow shade), which should enable the manifestation of the protected helical states along the interfaces. The high complexity exhibited by the bands of the 2H/1T'/2H heterostructure, even within the gap of the 2H phase, stems, in part, from the native edge states of the zigzag edges of this phase (22). The inset in Fig. 4C shows a zoom of the band structure in the relevant low-energy sector in half of the Brillouin zone. The size of the blue dots represents the relative weight of the states near the interface. The bands without dots or with very small ones are either 1T' bulk bands or trivial 2H edge states on the passivated outer edges of the heterostructure. The number of crossings (denoted by black dots at three chosen energy values) within the gap in half of the Brillouin zone is odd, although the gap opening due to the quantum confinement and hybridization of the interface and outer edge bands makes this count difficult. To make this point clearer we show in Fig. 4D the density of states projected on the 1T' side of a single interface, as obtained from the recursive Green's function method. An odd number of bands (now spin-split due to the lack of inversion symmetry) is now clearly visible crossing the gap.

The controlled patterning of the 1T'-phase on the 2H-phase thin semiconducting MoS$_2$ by laser beam irradiation was demonstrated. Integer fractions of the $R_0$ on $R$ peaks, the dependence on laser-irradiation conditions, $B$, and $T$, as well as the STS observation and theoretical calculations indicated the presence of the QSH phase in the patterned 1T' phases. It is evident that using the multi-layers bring many advantages to the present laser-beam irradiated experiments (e.g., protection from laser damage and oxidation) (SM10). Further optimization of the conditions for laser irradiation allows the on-demand patterning of 2D (or 1D) topological phases onto desired positions of non-topological phases of TMDCs and, thus, their various application to topological quantum computation (22).

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19. Three possible origins are considered for the deviation from $(R_Q/2)^1$, $(R_Q/4)^1$, and $(3R_Q/2)^1$ at $T > T_{c1}$: (1) thermally activated spins over the bulk gap, (2) spin fluctuation in helical edge spins, and (3) induced dephasing in metal electrodes. The approximate agreement with the STS gaps suggests that (1) is the dominant, consistent with the fact that the activation energy observed at $B = 0$ can correspond to the bulk gap.
20. The top-surface layer has multiple defects introduced by the high-power (i.e., ~17 mW) laser irradiation and presumably presents a different stoichiometry (a Kramers point different from other layers) and conductivity properties, including more defects along the edges of the 1T′/2H interface (SM 7), which can play as dephasing centers. This layer may be weakly coupled to the pristine 1T′ layer(s) existing underneath, due to excess in-plane heat accumulation. Indeed, these peaks disappear when low-power laser beam is applied (Fig. 2D) (see SM 7,8 in details).
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**Fig. 1** (A) Optical microscopy image of the 1T'-phase rectangular (right) and H-letter-like (left) patterns formed onto a thin 2H-MoS₂ flake by laser beam irradiation (see methods). **Inset:** Schematic cross section of a crystal structure of 1T'-MoS₂ mono-layer with distortion. (B) AFM image of a cross section of the laser irradiated part. (C) Schematic cross section of 1T' phase part created by laser-beam irradiation onto few-layer MoS₂, corresponding to (B). (D) Example of Raman spectra for non-laser irradiated region (2H phase; blue curve) and irradiated region (1T’ phase on 2H phase; red curve). Individual peaks correspond to $E_{2g} \approx 382$ nm$^{-1}$ and $A_{1g} \approx 408$ nm$^{-1}$ for 2H phase, $J_1 \approx 155$ nm$^{-1}$, $J_2 \approx 225$ nm$^{-1}$, and $J_3 \approx 330$ nm$^{-1}$ for 1T’ phase. (E,F) PL spectra of the laser-beam irradiated points plotted for wavelengths (E) 530-710 nm and (F) 640–700 nm. The numbers on the graph are the irradiation time for each plotted line and are common to both (E) and (F). (G) X-ray photoelectron spectroscopy (XPS) of the sample after laser irradiation. Red and blue lines are data fits for spectra of the 1T’ and 2H phases, respectively.
**Fig. 2 (A,B)** For the 1T'-rectangular patterns formed by two different laser irradiation times on each points ((A) 20 s and (B) 30 s; 17-mW power); two-terminal resistance measured between electrodes 1,3 and 2,4 as a function of $V_{bg}$ by flowing a constant current between electrodes 1,3 and 2,4 (insets). Contact resistances with 1T’ metallic-layer resistances are subtracted. (C) For the 1T’ H-letter like pattern; Non-local resistance ($R_{NL}$) observed for electrode pairs 3-4 as a function of $V_{bg}$, when a constant current flow between electrode pairs 1-2 (inset). (D) For the 1T’-rectangular pattern formed by reduced laser power (10mW). Equivalent circuits are shown in insets of (A,B). **Inset of (B)**; Channel length dependence of $R$ peak values in high $V_{bg}$ regions. Error bars are for the results of each three samples.
Fig. 3 (A) Out-of-plane magnetic-field ($B_\perp$) dependence of conductance corresponding to inverse of the three $R$ peaks (blue and red symbols for Fig. 2B (rectangular pattern) and pink for high $V_{bg}$ of Fig. 2C (H-letter like pattern)) and two off-$R$ peak values (green and orange symbols for Fig. 2B). (B) Temperature dependence of conductance corresponding to the three $R$ peaks in (A) in Arrhenius plot format. Dashes lines are guide to eyes. (C-E) STS spectra for non-laser-irradiated 2H region (C) and irradiated 1T' region (D,E) (SM 9). (D) The two bulk signals (blue and green lines) were measured near the center of the 1T'-rectangular pattern, and the edge signal (red line) was measured near the boundary of the 1T'/2H phases. (E) The edge signals for different three $V_{bg}$ values in sample of (D). The 1T' region was formed by irradiation with the condition for Fig. 2B.
Fig. 4 (A) Top and side view of the passivated 2H/1T'/2H heterostructure. (B) Top and side view of the atomic detail of one interface. (C) Overall band structure of the heterostructure. The yellow-shaded energy window represents the energy range within the 2H phase gap. Inside this range and near the center of the Brillouin zone, the bulk band inversion of the 1T' phase and the gap opened by the SOC can be seen along with edge and interface bands. The inset shows a zoom in the relevant energy window. The size of the blue dots represents the relative weight near the interface. The number of crossings of the relevant bands at various energies (solid lines) is odd, as expected from the presence of protected interface states. (D) Density of states projected on the 1T' side of a single interface obtained from the recursive Green's function method showing an odd number of bands crossing the gap.