Strain-Induced Metastable Topological Networks in Laser-Fabricated TaS$_2$ Polytype Heterostructures for Nanoscale Devices

Jan Ravnik,† Igor Vaskivskyi,§ Yaroslav Gerasimenko,‡ Michele Diego,† Jaka Vodeb,† Viktor Kabanov,† and Dragan D. Mihailovic†,‡,§

†Complex Matter Department, Jozef Stefan Institute, Jamova 39, SI-1000 Ljubljana, Slovenia
‡Center of Excellence on Nanoscience and Nanotechnology – Nanocenter (CENN Nanocenter), Jamova 39, SI-1000 Ljubljana, Slovenia

ABSTRACT: The stacking of layered materials into heterostructures offers diverse possibilities for generating deformed moiré states arising from their mutual interaction. Here we report self-assembled two-dimensional nanoscale strain networks formed within a single prismatic (H) polytype monolayer of TaS$_2$ created in situ on the surface of an orthorhombic 1T-TaS$_2$ single crystal by a low-temperature laser-induced polytype transformation. The networks revealed by scanning tunneling microscopy (STM) take on diverse configurations at different temperatures, including extensive double stripes and a twisted 3-gonal mesh of connected 6-pronged vertices. The resulting phase diagram can be understood to be a consequence of thermally driven minimization of discommensurations between the H and 1T layers. Nontrivial dislocation defects of embedded 2- and 4-gonal structures are shown to be associated with local inhomogeneous strains. The creation of metastable heterostructures by laser quench at cryogenic temperatures in combination with STM manipulation of local strain demonstrates nanoscale control of topological defects in transition metal dichalcogenide heterostructures may be utilized in the fabrication of nanoscale electronic devices and neural networks.

KEYWORDS: 2D materials, TaS$_2$, polytype transformation, self-assembled networks, topology

INTRODUCTION

Nonepitaxial layer stacking of two-dimensional systems performed by exfoliation and manual restacking has recently been shown to lead to new emergent properties such as gate-tunable enhanced-temperature superconductivity in twisted graphene bilayers, for example. An alternative approach to heterostructure construction in layered transition metal dichalcogenides (TMDs) that is perhaps suitable for some applications is to perform in situ polytype transformations with the prospect of achieving clean polytype interfaces. While the strain angle may be better controlled by manual restacking, controlled laser-induced strained structures involve spontaneous self-assembly over large areas which may be useful for investigation of extensive moiré states. Such problems with mismatched periodic potentials appear in numerous physical systems, such as atoms on surfaces, colloidal monolayers on periodic substrates, more recently in a Bose–Einstein condensate subject to an optical lattice, and graphene on boron nitride, leading to novel electronic and optical properties.

Among metallic TMDs, tantalum disulfide (TaS$_2$) is a particularly versatile candidate for investigating discommensuration phenomena and is often considered a prototype layered dichalcogenide. It is found in the form of many different polytypes that show a variety of charge density wave (CDW) states, superconductivity, and an unusual quantum spin liquid state, among others. The 1T-TaS$_2$ polytype has attracted additional attention recently due to the discovery of a photoinduced metallic hidden state, with a temperature tunable lifetime. This has led to a demonstration of a number of diverse practical devices, including ultrafast memory, an oscillator, and gated memristor devices driven by charge injection through electrical contacts. Voltage pulses from an STM tip have revealed domain formations that follow certain topological rules, suggesting that further new electronically ordered states may be found by polytype heterostructure fabrication.
polypeptide is a metastable structure that forms upon quenching from ~900 °C in the synthesis process. It forms a $\sqrt{13} \times \sqrt{13}$ commensurate (C) superlattice structure when cooling below $T_{C-C} = 180$ K but has an intrinsic discommensurate or “nearly commensurate” (NC) order above this temperature which persists up to $T_{NC-C} = 350$ K, whereupon it forms an incommensurate (IC) phase. Upon heating from a low-temperature C phase, an intermediate triclinic (Tr) phase is observed above $T_{C-Tr} = 220$ K, before the system undergoes a phase transition to NC order at $T_{Tr-NC} = 283$ K. A $T \rightarrow H$ polytype transformation at $T = 523$ K of a single surface layer was shown to be possible by thermal annealing in a vacuum, leading to an enhanced superconducting temperature ($T_c = 2.1$ K) with respect to the bulk. The authors attributed the increase in temperature to charge-transfer doping from the 1T substrate. However, an even higher critical temperature can be reached in the monolayers on Si/SiO$_2$ substrate ($T_c = 2.5$ K) or hexagonal boron nitride ($T_c = 3.4$ K). Such a transformation was previously observed locally after the application of a voltage pulse from an STM tip. Similar local transformations with the STM tip were observed also in other TMDs, such as TaSe$_2$ and NbSe$_2$, but in the opposite direction from $2H \rightarrow 1T$. The Ta coordination is prismatic in the 2H and orthorhombic in the 1T polytype with the unit cells being trigonal and hexagonal, respectively (Figure 1a). In this paper we report on an STM study of strain-induced mesoscopic modulations observed by STM in a laser-fabricated H-TaS$_2$/1T-TaS$_2$ heterostructure which displays self-organized spatial topographic modulations in the form of double stripes and twisted 3-gonal networks.

## RESULTS AND DISCUSSION

A single H layer was created on the freshly cleaved surface of 1T-TaS$_2$ single crystals. The sample was first slowly cooled (<1 K/min) to 77 K inside an ultrahigh-vacuum chamber of the STM (Figure 1b). It was then exposed to a single ~500 ms train of 50 fs laser pulses with a repetition rate of 100 kHz and a peak fluence of ~10 mJ/cm$^2$. The sample temperature was kept constant thereafter or heated for subsequent high-temperature measurements. We have estimated the upper bound for the lattice temperature after a burst of pulses using the two-temperature model of surface temperature that has been previously calibrated for this compound. Taking into account the accumulation of heat after exposure to a 500 ms burst of 50 fs pulses separated by 10 µs, the model gives $T = 1000$ K for a peak fluence of ~10 mJ/cm$^2$ (Figure 1c). This significantly exceeds $T_{Tr-H}$. We must note that the model does not take into account the coupling to the heat bath and thus overestimates the temperature particularly for bursts of longer duration.

The H polytype can easily be distinguished from the 1T phase due to the absence of the $\sqrt{13} \times \sqrt{13}$ CCDW that forms below $T_{C-C}$ on cooling. The exposure may result in a full or partial transformation, depending on fluence. In partially transformed samples we observe either triangles of the H phase embedded within the 1T phase (Figure 1d), or vice versa (Figure 1e). The ratio of the transformed area depends on the total energy injected into the given area and changes with the scanning position with respect to the peak of the beam and with the exposure time. This observation suggests a nucleation–growth mechanism, where the proportion of surface coverage is dependent on the local laser fluence.

Sometimes during the process cracks appear right after laser exposure and are accompanied by deposits of material which are aligned with the direction of the crack. When the transformation is complete, the presence of the H phase was confirmed by the 3 × 3 modulation of the CCDW at 4 K (Figure 1f). In agreement with previous work, we believe this is a single layer effect due to the observed weak $\sqrt{13} \times \sqrt{13}$ modulation from the layer underneath. However, further investigation by Raman spectroscopy or nano-ARPES is necessary to understand the interaction of the top H layer.

![Figure 1](image_url)
with the underlying bulk crystal. In this work we focus on the topological properties of fully transformed H monolayers measured by STM.

The most striking feature observed in large area STM scans (1 × 1 μm²) on fully transformed H polytype layers are one-directional double stripes with a period of \( \sim 18 \pm 0.2 \) nm shown in Figure 2a. The double-stripe modulations are found to be uniform over \( \gg 1 \) μm². In higher resolution scans (Figure 2b,c) we also see the characteristic \( \sqrt{3} \times \sqrt{3} \) CCDW modulation corresponding to the 1T layer underneath. The stripes around the hole get deformed and curved (pink box) or end abruptly (green box). The 6-fold vertices formed by intersection of three double stripes are observed. By performing Fourier transforms on the stripe and the CCDW modulations (Figure 2d,e), we obtain an angle \( \phi = 13.9^\circ \) of the CCDW with respect to the atomic lattice of the H layer. This implies that the 1T and H atomic lattices are aligned, since at 77 K the CCDW is at the same angle (13.9°) with respect to the lattice also in the 1T phase. The stripes are not aligned with either the lattice or CCDW direction but are at an angle of 2.3° with respect to the CDW (16.2° with respect to the atomic lattices). The stripes remain stable and unchanged if no external stimuli are present (verified up to 1 week at 77 K).

In an attempt to manipulate the stripes and elucidate their origin, we applied various voltage pulses from an STM tip. While lower voltage pulses do not affect the pattern at all, a 10 V, 0.1 ms pulse is sufficient to make a hole and locally transform the H layer back to 1T polytype and introduce strains along its rim (Figure 2f). In the back-transformed region we also see the appearance of domain walls, such as are observed in the photoinduced and STM-induced mosaics in 1T-TaS₂ previously reported. This reverse transformed layer is not very uniform, and both chiralities of the CCDW at \( \pm 13.9^\circ \) with respect to the 1T lattice are present. A cross-
No step-edge in height is observed at the interface between the mosaic and the stripe phase, implying that the layer is continuous and confirming that a ring around the hole was transformed back to the 1T structure. The height profile shows the expected ∼100 pm height modulation due to the CDW, while the stripes show an ∼40 pm height modulation. The appearance of domain walls in the mosaic state (indicated by the green triangle) is consistent with previous observations.\textsuperscript{19,20,22}

The first hint of the origin of the stripes is revealed by examining the striped area surrounding the hole. We can see that as a result of the application of the STM pulse the stripes are no longer straight but become curved, presumably compensating for the strain around the 1T-polytype ring around the hole (Figure 2f, purple box). At the edge of the ring we observe the formation of six-pronged vertices (Figure 2f, red box) or stripes that end abruptly by merging together (Figure 2f, green box). These observations suggest that the stripe structure can be manipulated with an external electrical pulse from an STM tip and suggest that they have nontrivial topological structure.

Figure 4. (a, b) A moiré pattern of two lattices with a lattice mismatch in both directions and in a single direction, showing a honeycomb and a stripe pattern, respectively. For clarity, an exaggerated mismatch of 8% is shown. (1) and (2) in (b) represent the two different vertical stackings of atoms, explained in (c). (c) Top view of stacking with an exaggerated mismatch along one direction. The blue and gray lattices represent the top (H) and bottom (1T) atomic lattices, respectively. We can see that the atoms in the top layer can line up with the atoms in the bottom layer in two different ways, marked with (1) and (2). (d) Schematic side view of the H/1T heterostructure. The mismatch between the atoms in the top two layers can be described by a mismatch vector field $D$. (e) Enlarged view of the atomic mismatch with mismatch vector $D$. 

sectional profile scan across the different regions is shown in Figure 2g (following the dotted indicated line in Figure 2f).

Upon heating above 77 K, the uniform double-stripe state starts to form complex domain structures (Figure 3). At ∼120 K, we start to observe the spontaneous appearance of six-pronged vertices strung up in a line, which separate neighboring double-stripe textures (Figure 3a,b). The vertices first start to appear on seemingly random positions, usually next to defects. This transformation does not appear to arise due to the uniform motion of the vertices or homogeneous contraction of the stripes, but rather by the spontaneous generation of new vertices, as shown in Figure 3c. With increasing temperature, the stripes become shorter and the vertices start appearing closer to each other. On approaching 220 K they form an irregular polygonal lattice of twisted 3-gons that cover large portions of the surface (Figure 3d). At slow heating above 220 K, the state becomes unstable. This coincides with the CDW transition temperature $T_{C-1T} = 220$ K from the C to the triclinic phase of 1T-TaS$_2$ on heating. At this temperature, cracks start to appear and matter starts to accumulate in lumps on the surface (Figure 3e), but no reverse transformations to 1T polytype are observed while increasing the temperature up to 300 K. Because the surface is highly uneven due to a larger number of cracks and material deposits, the STM measurements are increasingly difficult at higher temperatures. The heating was done with a platinum resistor, where the heating rate never exceeded 1 K/min.

An important role for the stability of the present system might be expected to be played by the CDW ordering within the H and 1T-TaS$_2$ layers. Because the laser transformation is performed at 77 K, which is above the 2H-TaS$_2$ CDW transition temperature ($T_{2H} = 70$ K), only the 1T-polytype CCDW is present, and there is no interaction between the 2H
and 1T CCDWs. However, a discontinuous jump of the in-plane lattice constant of $\delta a/a \approx 0.06\%$ at $T_{c-T} = 220$ K on heating associated with the transition from the C to the triclinic (T) phase of the 1T layer apparently triggers the observed breakup of the 3-gonal H layer network above 220 K.

Thermal expansion may also explain why the strain networks have not been reported in H/1T-TaS$_2$ heterostructures fabricated by high-temperature thermal annealing where the polytype transformation of the top layer occurs at 352 K, and the lattice mismatch is thermally relaxed on cooling. In our laser-fabricated heterostructure the H layer is created while the lattice mismatch is thermally relaxed on cooling. In our experiment, the polytype transformation of the top layer occurs at 532 K, and domain walls start to appear. In 1D, this is often described by the Frenkel–Kontorova model in the context of a linear chain of nearest-neighbor interacting atoms on a periodic substrate with solitonic solutions in the continuous limit. The competition between the trigonal and the stripe phases is a general phenomenon in the discommensuration transition in 2D, which has been observed in various systems, such as krypton on graphite or bilayer graphene, and can be experimentally controlled by temperature or modeled by changing the interaction energy between the layers.

The complex domain structures that may appear in 2D systems have been addressed by a variety of models that describe phase transitions of adsorbate ordering on a bulk substrate, taking into account domain walls and vertices. Gránz et al. discussed the ordering of particles interacting via a long-range dipolar potential $V_0$, subject to a square lattice potential. Minimization of energy at different values of the amplitude of $V_0$ results in a phase diagram of multiple configurations of adsorbrates in the top layer. Peculiarly, under certain conditions the monolayer perfectly adapts to the lattice in one direction, while a mismatch is observed in the other direction resulting in stripe-like ordering. Further increasing $V_0$ leads to the monolayer ordering taking on the symmetry properties of the bulk.

To understand better the phase transition in the present context, which takes place between a structured 3-gonal graph of vertices and a striped phase, we can consider the minimization of a Frank–van der Merwe rigid wall model by Villain, writing a free energy of the form

$$F = 2\Delta \nu + \mu L - T \log(g(\nu, L))$$

(1)

where $L$ describes the total length of the stripes, $\nu$ is the number of vertices, $\mu$ is an effective chemical potential, and $g(\nu, L)$ is the number of possible configurations, for a given $\nu$ and $L$. The third entropy term favors vertices at higher $T$ and is introduced to make the model consistent with the observations. We assume the stripe crossing energy $\Lambda > 0$, for which the model describes the commensurate to incommensurate transition for positive $\mu$ and commensurate to distorted (stripe) phase transition for negative $\mu$, where $\mu$ is proportional to lattice mismatch $p = a_{1T}/a_{H}$. We shift $\mu$ by the equilibrium value at $p = 1$, obtaining the final $\mu = \alpha (p - 1)$, where $\alpha$ includes the entropy of stripe meandering and other possible contributions. Employing the shift, we achieve consistency of the sign of $\mu$ with the discussion by Villain, and we are solving equations for a system around equilibrium value, where the lattices are equal. Using $l_i = L/\nu$, we rewrite the free energy as

$$F = \nu \left[ 2\Delta + \mu (l_x + l_y + l_z) + T \log \left( \frac{2}{3} \left( \frac{1}{l_x} + \frac{1}{l_y} + \frac{1}{l_z} \right) \right) \right]$$

(2)

Minimizing the free energy (2) gives us the equilibrium values for $l_i$ where a symmetrical solution for $l_x = l_y = l_z$ gives a regular lattice for high $T$. For low $T$ the solution is dependent on the value of $\mu$, which we keep at a negative value, where the model describes a distorted structure for low temperatures. In this case the minimum of free energy $F = -\infty$ is predicted for $l_i = \infty$, which describes a uniform infinite one-dimensional stripe structure with no intersections. This case was theoretically deemed by Villain to be unrealistic but appears to be borne out experimentally, since the stripes are the embodiment of an infinite elongation of the polygonal graph along one direction.

A phase diagram adopted after Villain is shown in Figure 5. Unlike hexagonal ordering on a graphite substrate, for which the original theoretical prediction was made, a trigonal network...
is considered, as our materials have a triangular lattice structure. The observed transition corresponds to varying the temperature at negative $\mu$. The sides of experimentally observed 3-gons are not straight, as described in the model, but are curved. This might be explained by using a more complex fluctuating wall model, where the entropy is contained not only in the vertex positions but also in wall fluctuations. This model yields very similar results, although the problem is approached from a different perspective.

The rules for the self-assembly of strain networks in 2-dimensional crystals arise from the symmetry properties of the underlying lattice and the different possible ways that domains can be assembled which is consistent with the underlying symmetry. The topology of the observed patterns can be described in terms of a cyclic groups $Z_n$. The group depends on the number of directional variants $m$ of $D$, described by the cyclic group $Z_m$, and the number of possible antiphase domain variants at the domain walls $n$, described by the cyclic group $Z_n$ (Figure 6a), such that $Z_k = Z_m \times Z_n$.

A single layer of the H polytype belongs to the $P6m2$ ($D^{4}_{3h}$, #187) hexagonal symmorphic space group. Such a single-layer H structure can display $m = 3$ directional variants corresponding to the cyclic group $Z_m$ and $n = 2$ antiphase variants of the $Z_n$ group. This leads to 6-valent vertices $Z_6 = Z_3 \times Z_2$ vertices with $l = m \times n$. The loop that describes the edge of each domain has 3 faces. Such a 6-valent graph of 3-gons can be colored by 6, 3, 2, or 1 color(s), corresponding to different $D$. A 6-color map is shown in Figure 6, where the antiphase boundaries and directional variants are highlighted in Figure 6b by dashed lines and arrows, respectively.

The STM topography images reveal the presence of domain walls and a 3-gonal graph structure but cannot detect the magnitude or direction of $D$ within individual domains, so the “colors” in Figure 6 cannot be distinguished. The bright stripes observed by STM and twisted 3-gonal networks in Figures 2 and 3, respectively, reveal the boundaries between the phases where $D$ changes value.

We note that a topologically related system to the present H/T interface is iron-intercalated $2H$−Fe$_{1/3}$TaS$_2$, but this has three possible antiphase boundaries, corresponding to the $Z_3$ cyclic group and two directional variants ($Z_2$) associated with two types of structural chirality in the corresponding chiral $P6_{3}22$ space group, resulting in $Z_3 \times Z_2 = Z_6$ domains, in contrast to the present case. The system thus displays a graph with 6-valent vertices, but with even $N$-gons.

Nontrivial topological defects within the H layer network are observed in areas of nonuniform strain (Figure 7). In the twisted 3-gonal domain structure (Figures 7a−c), which is predominantly composed of 3-gons, the presence of a vertex dislocation defect can be identified by emphasizing the vertex centers (Figure 7c). The defect consists of a 2-gon and a 4-gon pair surrounded by 3-gons (Figure 7b). Remarkably, there is no change in valence of the vertices: both the 2-gon and the 4-
The formation of a 4-gon, as indicated in Figure 7d,e. A defect is observed as a combination of a 4-fold vertex (red circle) and a 4-gon area enclosed by four vertices (green). (f) The same defect is represented as a dislocation.

In the stripe phase (Figure 7d–f), the presence of a vertex dislocation is similarly identified but is topologically different (Figure 7f). The thermally induced transformation from stripes to a twisted 3-gonal network can now be understood as a vertex dislocation climb process, which requires the addition of a single 4-valent vertex and the formation of a 4-gon, as indicated in Figure 7d,e.

## CONCLUSIONS

We have demonstrated laser-induced single-layer $T \rightarrow H$ polytype transformations in 1T-TaS$_2$. In contrast with previously observed laser-induced transformations in this material, where only the electronic structure is modulated$^{14,23}$ here the polytype transformation of the top layer introduces new topological properties arising from interlayer strains. In the former case only the electrons are strongly excited, but the lattice is only weakly heated$^{14}$, which makes the transition primarily electronic. In the present case periodic laser excitation results in much more pronounced lattice heating which is followed by surface polytype transformation that involves the movement of atoms$^{14,23}$. Such transformations at different base temperatures and underlying substrate strains in various materials can be expected to result in different metastable phases. Because the interface is never exposed to air or a vacuum, the T/H interfaces created are chemically “clean”, which leads to the appearance of intricate topological strain patterns arising from the lattice mismatch. The strain causes a local change of the electronic band structure$^{14,23}$, making its presence observable by methods that detects the local electronic density of states, such as the STM reported here. The periodicity of the stripe structure can be understood as a moiré interference between the H and T layers, where the H layer is deformed along one direction. The topology of the complex strain patterns and the thermal transition from stripes to twisted trigonal graph can be understood to arise from the 2-dimensional accommodation of discommensuration strain and consideration of the symmetry properties of 6-fold vertices and domain walls. Although the model does not describe the curvature of the polygonal edges, it suggests a phase diagram for the different possible ordered phases. On the practical side, domain wall crossings can be created by localized electromagnetic perturbation with an STM tip, indicating a possible path to the manipulation of network topology and eventually nanoscale topological strain network engineering of electronic structure. The laser-induced polytype transformation may be useful for applications that require large area heterostructures. Such nanostructures offer the possibility of constructing electronic devices such as nanoscale strain sensors and neural networks manipulable by strain. Nontrivial defects such as dislocations in the network structure offer an opportunity to study topologically protected localized electronic states that may be used in quantum devices based on local strain.

## METHODS

### Sample Preparation

The 1T-TaS$_2$ samples were grown in our lab by P. Šutar and A. Mrzel as reported previously$^{14}$. First, TaS$_2$ powder was synthesized by using 99.9% Ta foil and 99.98% S powder with 99.98% iodine as transport agent. The ampule with powder was heated for 5 days in a multiple zone furnace at 750–850 °C. Then it was opened and resealed with C$_{60}$ powder as getter and heated in a multiple-zone furnace with a temperature gradient of 900–800 °C for 6 h and then with a reversed gradient of 750–850 °C for 8 days. Finally, the ampule was quenched into cold water. This way numerous bulk samples of sizes up to a few mm$^3$ are produced$^{14}$. We chose a crystal with the size of $2 \times 2 \times 0.05$ mm$^3$ and mounted it on a Mo holder by silver epoxy. The sample was cleaved at UHV (10$^{-10}$ mbar) conditions, transferred into the nitrogen cooled STM stage, and slowly cooled to 77 K (or 4 K). Measured samples were confirmed to have a uniform CCDW at the surface prior to laser exposure.

### Optical Switching

We used a Ti:sapphire 50 fs pulsed laser system at a wavelength of 800 nm with a 100 kHz repetition rate and a maximum CW power of 240 mW on the sample. The beam spot measured about $100 \times 200$ μm$^2$ and varied from experiment to experiment by <5%. The beam spot was elongated as a consequence of hitting the sample at an angle of 60° with respect to normal. We used a train of pulses with peak fluences up to 10 mJ/cm$^2$ to induce the surface transformation to the H polytype. Because of different

---

**Figure 7.** Topologically nontrivial defects in 3-gonal networks of vertices at 220 K (a–c) and in a network of stripes at 180 K (d–f). Both scale bars are 50 nm. (a) The defect in a network of vertices is shown as a set of two areas enclosed by two (yellow) and four (green) vertices, instead of usual three. (b, c) The same defect is represented as a dislocation. (d, e) A defect is observed as a combination of a 4-fold vertex (red circle) and a 4-gon area enclosed by four vertices (green). (f) The same defect is represented as a dislocation.
thicknesses of the sample, the spatial variation of thermal contact between the sample and holder, and the uncertainty of the beam position, the desired transformation cannot always be achieved by using the same laser parameters. Thus, we needed to vary the exposure time from 500 ms up to 10 s. Before the switching was done, we checked with STM that the sample was in $\sqrt{3} \times \sqrt{3}$ commensurate CDW state (1T polytype).

**STM Measurements.** The measurements were done using an Omicron four-probe system. Mechanically cut platinum wire tips were used in all measurements. The measurements were done at bias voltage of $-0.8$ V in constant current mode. The current was set to different values ranging from 50 pA to 1 nA for different images to achieve the highest tip stability. The scanner drift was corrected by the FFT peaks of CDW modulation of 1T-TaS$_2$ by using scanning probe imaging processor (SPIP) software, where it was applicable.

---

**AUTHOR INFORMATION**

**Corresponding Author**

*E-mail dragan.mihailovic@ijs.si.*

**ORCID**

Jan Ravnik: 0000-0002-4886-1251

Yaroslav Gerasimenko: 0000-0002-5337-284X

**Author Contributions**

J.R., L.V., and D.M. conceived the project. J.R., L.V., Y.G., and M.D. prepared and performed the STM measurements. J.R., J.V., V.K., Y.G., and D.M. performed theoretical analysis. J.R. and D.M. wrote the manuscript. All authors contributed to the manuscript.

**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

The work was supported by ERC ADG Trajectory (GA320602) and the Slovenian Research Agency (P10040 and young researcher grants P087589 and P08333). The work was supported by ERC ADG Trajectory (GA320602) and the Slovenian Research Agency (project P10040 and young researcher grants P17589 and P08333). We thank Petra Suštar and Aleš Mrzel for sample growth.

**ABBREVIATIONS**

STM, scanning tunneling microscope; TMD, transition metal dichalcogenide; CDW, charge density wave; C, commensurate; CCDW, commensurate charge density wave; NC, nearly commensurate; IC, incommensurate; Tr, trilinac; vdW, van der Waals.

**REFERENCES**

(1) Cao, Y.; Fatemi, V.; Fang, S.; Watanabe, K.; Taniguchi, T.; Kaxiras, A.; Jarillo-Herrero, P. Unconventional Superconductivity in Magic-Angle Graphene Superlattices. Nature 2018, 556, 43.

(2) Wang, Z.; Sun, Y.-Y.; Abdelwahab, I.; Cao, L.; Yu, W.; Ju, H.; Zhu, J.; Fu, W.; Chu, L.; Xu, H.; et al. Surface-Limited Superconducting Phase Transition on 1T-TaS$_2$. ACS Nano 2018, 12, 12619–12628.

(3) Kim, J.-J.; Park, C.; Yamaguchi, W.; Shino, O.; Kitazawa, K.; Hasegawa, T. Observation of a Phase Transition from the T Phase to the H Phase Induced by a STM Tip in 1T-TaS$_2$. Phys. Rev. B: Condens. Matter Mater. Phys. 1997, 56, R15573.

(4) Villain, J. Commensurate-Incommensurate Transition of Krypton Monolayers on Graphite: A Low Temperature Theory. Surf. Sci. 1980, 97, 219–242.

(5) Mangold, K.; Leiderer, P.; Bechinger, C. Phase Transitions of Colloidal Monolayers in Periodic Pinning Arrays. Phys. Rev. Lett. 2003, 90, 158302.

(6) Sun, Q.; Hu, J.; Wen, L.; Liu, W.-M.; Juzeliūnas, G.; Ji, A.-C. Ground States of a Bose–Einstein Condensate in a One-Dimensional Laser-Assisted Optical Lattice. Sci. Rep. 2016, 6, 37679.

(7) Hayashi, K.; Kawamura, A. Formation of TaS$_2$ Polytypes and Hydrogen Impurity. Mater. Res. Bull. 1986, 21, 1405–1410.

(8) Thomson, R.; Burk, B.; Zettl, A.; Clarke, J. Scanning Tunneling Microscopy of the Charge-Density-Wave Structure in 1T-TaS$_2$. Phys. Rev. B: Condens. Matter Mater. Phys. 1994, 49, 16899.

(9) Wilson, J.; Di Salvo, F.; Mahajan, S. Charge-Density Waves in Metallic, Layered, Transition-Metal Dichalcogenides. Phys. Rev. Lett. 1974, 32, 882.

(10) Nagata, S.; Aochi, T.; Abe, T.; Ebisu, S.; Hагino, T.; Seki, Y.; Tsutsumi, K. Superconductivity in the Layered Compound 2H-TaS$_2$. J. Phys. Chem. Solids 1992, 53, 1259–1263.

(11) Sipos, B.; Kusmartseva, A. F.; Akrap, A.; Berger, H.; Forró, L.; Tutiš, E. From Mott State to Superconductivity in 1T-TaS$_2$. Nat. Mater. 2008, 7, 960.

(12) Klajniček, M.; Zorko, A.; Mravlje, J.; Jagličič, Z.; Biswas, P. K.; Prelovšek, P.; Mihailovic, D.; Arčon, D.; et al. A High-Temperature Quantum Spin Liquid with Poloron Spins. Nat. Phys. 2017, 13, 1130.

(13) Law, K. T.; Lee, P. A. 1T-TaS$_2$ as a Quantum Spin Liquid. Proc. Natl. Acad. Sci. U. S. A. 2017, 114, 6996.

(14) Stojchevska, L.; Vaskivskyi, I.; Mertelj, T.; Kusar, P.; Svetin, D.; Brazovskii, S.; Mihailovic, D. Ultrafast Switching to a Stable Hidden Quantum State in an Electronic Crystal. Science 2014, 344, 177–180.

(15) Ravnik, J.; Vaskivskyi, I.; Mertelj, T.; Mihailovic, D. Real-Time Observation of the Coherent Transition to a Metastable Emergent State in 1T TaS$_2$. Phys. Rev. B: Condens. Matter Mater. Phys. 2018, 97, No. 075304.

(16) Vaskivskyi, I.; Mihailovic, I.; Brazovskii, S.; Gospodaric, J.; Mertelj, T.; Svetin, D.; Sutar, P.; Mihailovic, D. Fast Electronic Resistance Switching Involving Hidden Charge Density Wave States. Nat. Commun. 2016, 7, 11442.

(17) Liu, G.; Debnath, B.; Pope, T. R.; Salguero, T. T.; Lake, R. K.; Balandin, A. A. Charge-Density-Wave Oscillator Based on an Integrated Tallantum Disulphide-Boron Nitride-Graphene Device Operating at Room Temperature. Nat. Nanotech. 2016, 11, 845.

(18) Yoshida, M.; Suzuki, R.; Zhang, Y.; Nakano, M.; Iwasa, Y. Membrane-Type Switching in Two-Dimensional 1T-TaS$_2$ Crystals. Science advances 2015, 1, No. e1500606.

(19) Ma, L.; Ye, C.; Yu, Y.; Lu, X. F.; Niu, X.; Kim, S.; Feng, D.; Tómanek, D.; Son, Y.-W.; Chen, X. H.; Zhang, Y. A Metallic Mosaic Phase and the Origin of Mott-Insulating State in 1T-TaS$_2$. Nat. Commun. 2016, 7, 10956.

(20) Cho, D.; Cheon, S.; Kim, K.-S.; Lee, S.-H.; Cho, Y.-H.; Cheong, S.-W.; Yeom, H. W. Nanoscale Manipulation of the Mott Insulating State Coupled to Charge Order in 1T-TaS$_2$. Nat. Commun. 2016, 7, 10453.

(21) Huang, F.-T.; Cheong, S.-W. Aperiodic Topological Order in the Domain Configurations of Functional Materials. Nat. Rev. Mater. 2017, 2, 17004.

(22) Gerasimenko, Y. A.; Vaskivskyi, I.; Mihailovic, D. Dual Vortex Charge Order in a Metastable State Created by an Ultrafast Topological Transition in 1T-TaS$_2$. arXiv preprint arXiv:1704.08149, 2017.

(23) Gerasimenko, Y.; Vaskivskyi, I.; Ravnik, J.; Vodeb, J.; Kabanov, V. V.; Mihailovic, D. Ultrafast Jamming of Electrons into an Amorphous Entangled State. arXiv preprint arXiv:1803.00255, 2018.

(24) Li, L.; O’Farrel, E.; Loh, K.; Ega, D.; Ozyl roz, B.; Neto, A. C. Controlling Many-Body States by the Electric-Field Effect in a Two-Dimensional Material. Nature 2016, 529, 185.

(25) Yang, Y.; Fang, S.; Fatemi, V.; Ruhman, J.; Navarro-Moratalla, E.; Watanabe, K.; Taniguchi, T.; Kaxiras, E.; Jarillo-Herrero, P. Enhanced Superconductivity upon Weakening of Charge Density Wave Transport in 2 H-TaS$_2$ in the Two-Dimensional Limit. Phys. Rev. B: Condens. Matter Mater. Phys. 2018, 98, No. 035203.
26) Navarro-Moratalla, E.; Island, J. O.; Manas-Valero, S.; Pinilla-Cienfuegos, E.; Castellanos-Gomez, A.; Quereda, J.; Rubio-Bollinger, G.; Chirrolí, L.; Silva-Guillén, J. A. Agrait, N.; et al. Enhanced Superconductivity in Atomically Thin TaS₂. Nat. Commun. 2016, 7, 11043.

27) DiSalvo, F.; Waszczak, J. Paramagnetic Moments and Localization in 1 T-TaS₂. Phys. Rev. B: Condens. Matter Mater. Phys. 1980, 22, 4241.

28) Zhang, J.; Liu, J.; Huang, J. L.; Kim, P.; Lieber, C. M. Creation of Nanocrystals through a Solid-Solid Phase Transition Induced by an STM Tip. Science 1996, 274, 757–760.

29) Bischoff, F.; Auwärter, W.; Barth, J. V.; Schiffer, A.; Fuhrer, M.; Weber, B. Nanoscale Phase Engineering of Niobium Diselenide. Chem. Mater. 2017, 29, 9907–9914.

30) Scruby, C.; Williams, P.; Parry, G. The Role of Charge Density Waves in Structural Transformations of 1 TTaS₂. Philos. Mag. 1975, 31, 255–274.

31) Sezerman, O.; Simpson, A.; Jericho, M. Thermal Expansion of 1T-TaS₂ and 2H-NbSe₂. Solid State Commun. 1980, 36, 737–740.

32) Torres, E.; DiLabio, G. A Density Functional Theory Study of the Reconstruction of Gold (111) Surfaces. J. Phys. Chem. C 2014, 118, 15624–15629.

33) Hanke, F.; Björk, J. Structure and Local Reactivity of the Au (111) Surface Reconstruction. Phys. Rev. B: Condens. Matter Mater. Phys. 2013, 87, 235422.

34) Jin, C.; Regan, E. C.; Yan, A.; Utama, M. I. B.; Wang, D.; Zhao, S.; Qin, Y.; Yang, S.; Zheng, Z.; Shi, S.; et al. Observation of Moiré Excitons in WSe₂/WSe₂ Heterostructure Superlattices. Nature 2019, 567, 76.

35) Aldén, J. S.; Tsen, A. W.; Huang, P. Y.; Hovden, R.; Brown, L.; Park, J.; Muller, D. A.; McEuen, P. L. Strain Solitons and Topological Defects in Bilayer Graphene. Proc. Natl. Acad. Sci. U. S. A. 2013, 110, 11256–11260.

36) Lin, X.; Liu, D.; Tománek, D. Shear Instability in Twisted Bilayer Graphene. Phys. Rev. B: Condens. Matter Mater. Phys. 2018, 98, 195432.

37) Naik, M. H.; Jain, M. Ultrasoundand and Shear Solitons in Moiré Patterns of Twisted Bilayer Transition Metal Dichalcogenides. Phys. Rev. Lett. 2018, 121, 266401.

38) Coppey-Smith, S.; Fisher, D. S.; Halperin, B.; Lee, P.; Brinkman, W. Dislocations and the Commensurate-Incommensurate Transition in Two Dimensions. Phys. Rev. Lett. 1981, 46, 549.

39) Bak, P. Commensurate Phases, Incommensurate Phases and the Devil’s Staircase. Rep. Prog. Phys. 1982, 45, 587.

40) Granz, B.; Korshunov, S. E.; Geshkenbein, V. B.; Blatter, G. Competing Structures in Two Dimensions: Square-to-Hexagonal Transition. Phys. Rev. B: Condens. Matter Mater. Phys. 2016, 94, 054110.

41) Frank, F.; van der Merwe, J. H. One-Dimensional Dislocations. I. Static Theory. Proc. R. Soc. London. Ser. A: Math. Phys. Sci. 1949, 198, 205–216.

42) Chaikin, P. M.; Lubensky, T. C.; Witten, T. A. Principles of Condensed Matter Physics; Cambridge University Press: Cambridge, 1995; Vol. 1.

43) Ribeiro-Soares, J.; Almeida, R.; Barros, E.; Araújo, P.; Dresselhaus, M.; Cançado, L.; Jorio, A. Group Theory Analysis of Phonons in Two-Dimensional Transition Metal Dichalcogenides. Phys. Rev. B: Condens. Matter Mater. Phys. 2014, 90, 115438.

44) Gan, L.-Y.; Zhang, L.-H.; Zhang, Q.; Guo, C.-S.; Schwingenschlögl, U.; Zhao, Y. Strain Tuning of the Charge Density Wave in Monolayer and Bilayer 1 T-TaS₂. Phys. Chem. Chem. Phys. 2016, 18, 3080–3085.

45) Johari, P.; Shenoy, V. B. Tuning the Electronic Properties of Semiconducting Transition Metal Dichalcogenides by Applying Mechanical Strains. ACS Nano 2012, 6, 5449–5456.