Dual vortex charge order in a metastable state created by an ultrafast topological transition in 1T-TaS$_2$

Yaroslav A. Gerasimenko,* Igor Vaskivskyi, and Dragan Mihailovic†

CENN Nanocenter, Jamova 39, SI-1000, Ljubljana, Slovenia and
Department of Complex Matter, Jozef Stefan Institute,
Jamova 39, SI-1000, Ljubljana, Slovenia

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Abstract

Many body systems in complex materials undergoing non-equilibrium phase transitions may self-organise into ordered metastable emergent states with new and unexpected functionalities$^{1–7}$. Here, using large-area scanning tunneling microscopy we reveal an intricate chiral vortex structure and complex tiling of charged electron domains in the metastable metallic state in 1T-TaS$_2$ created by a non-equilibrium topological transition initiated by a single femtosecond optical pulse. A Moiré analysis shows that the interference of non-equilibrium nested Fermi surface (FS) electrons leads to the creation of charge vortices $\vec{D}$ on a length scale of $\sim$70 nm. On a much smaller scale of $\sim$5 nm, domain configurational patterns appear, which show bound vortex-antivortex pairs, discommensurations, domain wall (DW) crossings and DW kinks, consistent with a rapidly quenched Berezinskii-Kosterlitz-Thouless (BKT) -transition. Revealing the detailed mechanism for the transition leads the way to design of long-range charge-ordered metastable states with intricate emergent properties under controlled non-equilibrium conditions.

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Photoexcited metastable states in crystals have lifetimes which usually range from picoseconds to microseconds\cite{1-5}, which makes it hard to investigate their structure in sufficient detail to reveal transient mesoscopic or microscopic order\cite{8}. Consequently, the detailed mechanisms for photoinduced metastability have not been very well understood till now. Even though it is of fundamental importance to prove the principle of the existence of LRO created of a transient emergent state, no direct experimental evidence has so far been presented in any system exhibiting a photoinduced phase transition. Uniquely, as a model system, 1T-TaS$_2$ (TDS) has a metastable state with a temperature-tunable lifetime, which is usefully long at low temperatures\cite{6,9} for detailed experimental investigations. This opens the possibility of studying not only the fine structure, but also the origin and mechanism of the metastability with high resolution scanning tunnelling microscopy (STM). In this layered di-chalcogenide system (Fig. 1a) the competition of lattice strain, a FS instability and Coulomb interactions lead to a number of different phases and a multidimensional multi-parameter phase diagram, where strain, chemical pressure, doping and laser photoexcitation fluence play different roles\cite{10-12}. In equilibrium, the high-temperature metallic state of TDS is unstable towards a three-pronged quasi-two dimensional FS nesting instability\cite{13} causing a transition at $T_{IC} = 540$ K from a single-band metal to a uniform three-directional incommensurate (IC) charge density wave (CDW) state. On cooling this transforms into a 'nearly-commensurate' (NC) state at $T_{NC} = 350$ K as a result of incommensuration strain, giving almost commensurate patches separated by smooth discommensurations (DCs)\cite{14}. Eventually the state becomes fully commensurate (C) and insulating below $190 \sim 220$ K, as shown schematically\cite{14,15} in Fig. 1b. The localized electrons in the C state have a remarkable star-of-David (SD) 'polaron lattice' structure, (Fig. 1c) where exactly one electron is localised on the central Ta of the SD and the 12 surrounding Ta atoms are symmetrically displaced towards it. Photoexcitation of the material by a single ultrafast optical pulse was shown to cause an insulator-to-metal transition (IMT) to a hidden metastable (H) state within a narrow range of photoexcitation fluences and pulse lengths. An indication of possible long range order (LRO) came from the narrowness of the collective amplitude mode of the CDW whose frequency shifts by 3\% at the transition\cite{6}. IMT switching caused by direct charge injection through electrodes was recently demonstrated in TDS\cite{16-22} while switching by STM tip revealed that the uniform CCDW can be locally broken up into non-periodic domains. No LRO was observed however, in either case\cite{18,19}. A hint of new states was found
in ultrafast electron diffraction (UED) experiments at significantly larger photoexcitation densities, but the $k$-space resolution was insufficient to resolve the ordering vector or the domain structure, so the fundamental question whether metastable LRO can form under non-equilibrium conditions remains unanswered. Here, for the first time we reveal LRO in an transient emergent state in a non-thermal transition using in-situ ultrafast optical switching in combination with a low-temperature high resolution STM. The studies reveal a remarkable dual vortex structure of polaron ordering in the emergent state, quite unlike any of the other states in this system, or in any other known material.

Results

We photoexcite freshly in-situ cleaved single crystals of TDS with focused 50 fs single pulses at 400 nm within a UHV STM chamber. A scanning electron microscope (SEM) is used to select a homogeneous region of the crystal without strain and for precise positioning the tip and the beam on the sample (Fig. 1e). The pulse fluence was adjusted to just above switching threshold at 0.9 mJ/cm$^2$, taking care to avoid heating the lattice to the NC state (see supplementary Fig. 5). To determine both LRO and local structure of charge modulation, STM images of C, NC and H states were measured on different length scales (Fig. 2). The large area scan shows a regular polaron lattice in the C ground state at 4.2 K (Fig. 2a). In the NC state this breaks up into a modulated structure of domains and "soft" DWs (Fig. 2c). The corresponding atomic-scale images are shown in Fig. 2b and d respectively. In the photoexcited H state at 4.2 K (Fig. 2e,f) the uniform C lattice is transformed into domains of diverse sizes and shapes, separated by sharp DWs. The actual pattern is different every time the experiment is performed, indicating that the domain structure is determined by fluctuations rather than predetermined by sample defects and imperfections.

Fourier transforms (FTs) of the large-area scans demonstrate a set of peaks shown in Fig. 3 with high reciprocal space resolution, allowing us to discern fine details of the long range charge modulations described by the vectors $Q_{NC}^{(i)}$, $Q_{C}^{(i)}$ and $Q_{H}^{(i)}$, for the NC, C and H states respectively. Here $i = 1, 2, 3$ refer to the three CDW directions at 120° to each other. FTs of atomic resolution scans (Figs. 3a,d) allow us to calibrate the charge modulation vectors with respect to the underlying atomic lattice vectors $a^*$ and $b^*$. 
In the C state, CDW wavevectors $\pm Q_C^{(i)}$ appear as the six brightest peaks in the FT in Fig. 3a. Less intense atomic peaks coincide exactly with the linear combination of the CDW vectors, $a^* = 3Q_C^{(1)} - Q_C^{(2)}$, defining the commensurability of the C CDW with the underlying lattice. The NC state has additional domain structure, giving rise to peak splitting. Fig. 3b shows one bright CDW superlattice peak surrounded by several less intense satellite peaks. The bright peak corresponds to fundamental CDW wavevectors $Q_{NC}^{(i)}$, while the satellites characterise the domain periodicity, $k_{domain}^{(i)}$ shown in Fig. 3c. The appearance of the satellites in the NC state is well understood in the plane wave picture of the CDW, where anharmonic interactions produce secondary distortions - described in terms of harmonics of the fundamental wave $Q_{NC}^{(i)}$. Their wave vectors are related to the latter as $Q_{sat}^{(1)} = n(a^* + Q_{NC}^{(2)}) - (3n - 1)Q_{NC}^{(1)}$, and in the NC state the order is limited to $n = 1^{14}$. The interference of fundamental and the first harmonic wave results in “beatings” - large-scale periodic modulations of the distortion amplitude. The interference pattern has hexagonal domains with inverse period $k_{domain}^{(i)} = Q_{NC}^{(i)} - Q_{sat}^{(i)}$ separated by smooth domain walls. The CDW is locally commensurate with the lattice inside the domain, and its only distinction from the C-state is a decreased CDW amplitude near the domain walls (see SI).

In the photoexcited state, we first ascertain the relation between the atomic lattice peaks and the CDW lattice peaks from a FT of atomic resolution STM image of several domains (Fig. 3d). From the position of the second order CDW peaks (for higher accuracy) we obtain $\phi = 13.45^\circ \pm 0.35^\circ$, which differs by $0.45^\circ$ to the C state, and $0.35^\circ$ from the nearest reported NC state (see Supplementary Information for error analysis). This is the first indication that the H state has different LRO than the C or NC states.

The periodicity of the CDW and the domains in the H state can be analyzed similarly to NC state from FTs of large area scans over $200 \times 200$ nm$^2$. We see six high intensity peaks (Fig. 3e). They result from the largest amplitude modulation in the topographic image, which comes from individual polarons, and are associated with the fundamental CDW vectors, $Q_{H}^{(i)}$. Their FWHM angular width is $0.31^\circ$ (cf. Fig. 3g) which shows that the H state $Q_{H}^{(i)}$s are homogeneously rotated w.r.t. the C state over a macroscopically large area ($\sim 200 \times 200$ nm), which is a clear indication that the H state has distinct LRO.

In the H state the single satellite characteristic of the NC state is converted into a diffuse streak spanning a range of angles $1.7^\circ < \Theta < 9.5^\circ$ (Fig. 3e,f) which reflects the distribution of the domain sizes and domain shapes. The cross-section $AA'$ along the streak (Fig. 3h)
reveals that FT intensity is not random in $k$-space, but rather there are approximately equally separated peaks with different intensities, showing that some domain sizes are more favourable than others. If we zoom into aggregates of the small or large domains, individual peaks can be discerned (see SI for more detailed analysis). The fundamental vector $Q_H^{(i)}$ stays the same, independent of the domain size, as a consequence of the LRO.

The existence of different domain sizes can be qualitatively described by extending the harmonic picture. The first peak position in the streak is at $\Theta \approx 1.7^\circ$ and fits rather well the expected position of $Q_{sat}$ with $n = 1$, while peaks at larger $\Theta$ appear as higher harmonics. Their positions can be fit by varying angle $\phi$ and length $Q_H/a^*$, which gives $\phi = 13.51^\circ$ and $Q_H/a^* = 0.278$ for $n = 1 \ldots 5$ (see SI for fitting and real-space reconstruction). We note that these values are close to the first metastable state observed under optical excitation in time-resolved UED data\textsuperscript{22}. There it was tentatively associated with the triclinic CDW appearing in the equilibrium phase diagram, but our higher resolution allows us to associate it with the emergent photoinduced state (see Supplementary Fig. 12 for comparison).

We continue by examining the charge displacements within domains and the associated DWs. To characterize the topological structure, we define a misfit displacement vector $\vec{D}$ as the charge displacement in the H state with respect to the C lattice (see Fig. 4a for definition). Locally, the structure is composed of irregular domains with different $\vec{D}$ (Fig. 4b-d). There are 12 possible discrete displacement vectors and $\vec{D} = 0$ (Fig. 4a), which can be separated by 6 different types of DWs. Commonly, three DWs meet at a $Z_3$ vertex resulting in pairs of vortices and antivortices (labeled $Y$ and $\bar{Y}$ ) such as shown in Fig. 4b\textsuperscript{23}. For each individual $Y$ defect the misfit displacement vector sum is nonzero, e.g. $D_0 + D_2 + D_6 \neq 0$, but for each pair it must be zero: $D_0 + D_2 + D_6 + D_5 = 0$. The latter condition sets the local topological rules for the misfit displacement vectors. In addition, a significant number of $X$ wall crossings (Fig. 4c) and kinks ($K$s) (Fig. 4d) are observed, higher energy non-trivial defects created by photoexcitation which are not expected in equilibrium hexagonal tiling (Fig. 4b). A contour encircling an $X$ defect results in a non-zero sum of the misfit vectors, $D_{10} + D_2 + D_5 \neq 0$, which makes this a non-trivial topologically protected defect. The $K$ defect corresponds to a change of domain wall type without a change of the misfit field. It contains one incomplete polaron hexagram, a nontrivial defect with zero Burgers vector (see Suppl. Fig. 18).

In addition to the local vortex structure, there exists a long wavelength structure which
is even more intriguing (Fig. 5). Plotting the magnitude $|\vec{D}|$ and angle $\alpha$ of the vector field $\vec{D}$ in Fig. 5b and c respectively as a Moiré interference pattern between the C lattice and the H lattice (see Fig. 5a), we observe a clear hexagonal lattice of vortices with a period $L_H = 2\pi/(|Q_H - Q_C|) \sim 70$ nm (Fig. 5b-d). Repeated experiments show that the winding number $n_w$ of the large-scale vortices is either $n_w = -1$ or $n_w = +1$, but never both simultaneously, implying that there is a macroscopically spontaneously broken mirror symmetry in the formation of the LRO $\vec{D}$ vector field. We also observe an accompanying weak, but unambiguous modulation of the domain size with a similar wavelength shown in Fig. 5e. The domain size distribution explains the streaks in the Fourier transform shown in Fig. 3e,f.

While the long-wavelength order in the H state apparently requires tessellation of different domain sizes, the NC state (above $\sim 200$ K) displays a regular domain structure with a period $L_{NC} = 2\pi/(|Q_{NC} - Q_C|) \sim 15$ nm and uniform domain sizes (Fig. 2c), separated by “smooth” domain walls (or discommensurations). Comparing with other reported states, samples only a few layers thick deposited on substrates which are inherently strained show either no CDW at low temperature, or the presence of a “supercooled” metallic state with randomly distributed domains as reported by Ma et al. On the other hand, thick samples such as the one used here, or unstrained free-standing monolayer samples do not readily show a supercooled phase, so we cannot make any empirical connection between any supercooled state and the H state. On the other hand, the STM tip-induced state at low temperatures displays a local domain structure with four types of $Z_3$ vortices and domains of different size separated by sharp DWs, but there is no evidence of the intricate long range order observed in the H state (see SI for analysis).

Discussion

The mechanism for the transition is quite unlike previously studied non-equilibrium second-order symmetry-breaking FS nesting transitions such as the one in TbTe$_3$, and the trajectory of the C→H transition cannot be discussed in terms of conventional non-equilibrium symmetry-breaking with a Landau order parameter that vanishes at some critical time $t \to t_c$. It requires consideration of the competing effects of FS hot-electron interference, Coulomb interactions and particularly the incommensurability strain which
force the ordering of the system on different timescales. The state is created under very specific conditions of laser fluence $0.8 < \mathcal{F} < 2.5 \text{ mJ/cm}^2$, within a time and temperature window defined by the pulse length range $30 \text{ fs} < \tau_p < 5 \text{ ps}$\(^6\). During this time, the lattice remains below the transition temperature to the NC state, while the peak electronic temperature is estimated $\sim 1700 \text{ K}$\(^6\), thermalizing with the lattice on a timescale of $\sim 7 \text{ picoseconds}$ to $150 \sim 185 \text{ K}$ (see Fig. 5 in SI for lattice temperature estimates). Transient photodoping is an essential component: photoemission experiments show that short pulse photoexcitation causes melting and a rapid $< 50 \text{ fs}$ transient shift of the chemical potential above the switching threshold $\mu_c^{33,34}$, which implies that FS nesting is modified while the system is out of equilibrium. The hot nested electrons interfere to condense into a transient incommensurate charge modulated structure with a wavevector $Q_{IC}^*$, reflecting the non-equilibrium FS. The accompanying change in low-energy electronic structure occurs on a timescale $< 400 \text{ fs}$, as recently shown by coherent phonon spectroscopy\(^35\). Thereafter, as the incommensurate electronic CDW tries to adjust to the nearest commensurate lattice $Q_C$, individual domains spontaneously form (Fig. 4e). However, local topological defects such as $K$s, and $Y$ vortices without associated nearby anti-defects\(^29,36–38\) also appear, presumably as a consequence of the rapid quench through the transition. On a longer timescales\(^40,41\), a continuous tiling emerges with a LRO $Q_H$, and the $K$ and $Y$ defects become bound to form $X$ defects and $Y – \bar{Y}$ vortex-anti-vortex pairs (see Fig. 4f) following local topological rules\(^19,23\). The process is remarkably similar to superconductors where the binding of unbound vortices leads to the formation of a vortex lattice in a non-equilibrium Berezinsky-Kosterlitz-Thouless transition\(^39\). The long-wavelength chiral vortex lattice of the misfit displacement vector field $\vec{D}$ and particularly the periodic modulation of domain size with a wavelength of $\sim 70 \text{ nm}$ may be considered as an emergent phenomenon associated with the quench and interference of nonequilibrium nested electrons.

Finally, we remark on the implications of the fact that strictly speaking, LRO cannot exist in two dimensions\(^39\). Out-of-plane stacking of domains and orbital ordering along may critically contribute to the stability of the H state\(^42\). STM experiments show that domain walls on subsequent layers avoid each other (see Supplementary Fig. 13), indicating that inter-layer Coulomb interactions may lead to interlayer ordering of the $Z_3$ vortices. At the same time the very large out-of-plane versus in-plane resistivity persisting into the H state\(^43\), implies a non-trivial role for out-of-plane interactions in establishing the observed long range
vortex order.

The validation of the concept of formation of emergent LRO through many-body interactions under non-equilibrium conditions, and the underlying topological transformation mechanism revealed by these experiments represents a large step towards understanding, and whence designing new emergent metastable states in complex materials. The present system, with its associated M-I transition may lead to advances in novel non-volatile all-electronic memory devices without the involvement of ions or magnetism, through controllable switching between electronic topologically ordered states.

Methods

The results presented here were measured in the custom-built Omicron Nanoprobe 4-probe UHV LT STM with base temperature of 4.2 K and optical access for laser photoexcitation (Fig. 1e). Crystals of 1T-TaS$_2$ were synthesised by the iodine vapor transport method. Samples were cleaved in situ at UHV conditions and slowly cooled down to 4.2 K.

Photoexcitation of 1T-TaS$_2$ single crystals was performed at 4.2 K (C-state) with a single 50 fs optical pulse at $\sim 400$ nm (second-harmonic generation from 800 nm Ti-Sapphire laser) focused onto a 100 $\mu$m diameter spot within STM UHV chamber. This ensures highly spatially homogeneous excitation on the scale of the STM scans ($\sim 200$ nm). It is important that photoexcitation energy density is carefully adjusted to be slightly above the threshold value ($\sim 0.9$ mJ/cm$^2$) for switching to the hidden state. Significantly higher excitation energies result in heating of the lattice above the phase transition temperature $T_{NC-C}$, which we want to avoid.

The surface was characterised each time before photoexcitation to confirm a defect-free initial C state. The beam was aligned at low power to hit the apex of the STM tip. Then the tip was retracted and a high-power single pulse was applied (Fig. 1e). After approaching the tip, an area of the order of $500 \times 500$ nm$^2$ was checked for homogeneity.

Contrast adjustment in Fourier transforms is routinely used for analysis of STM data in 1T-TaS$_2$ and is described thoroughly in literature. Here all FT images has max/min ratio of $\approx 2.5$, approximately an order of magnitude of absolute FT amplitude. Only parts of the images used for FT are shown in Fig. 2. Full-scale versions of those for the hidden state are shown in Supplementary Fig. 19. Raw images were corrected to match triangular lattice
using standard procedure (see Supplementary Information).

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Author contributions

Y.G. and I.V. performed the measurements, Y.G. and D.M. carried out the analysis and wrote the paper. All authors discussed the results and contributed to the manuscript preparation.

* Electronic address: yaroslav.gerasimenko@ijs.si
† Electronic address: dragan.mihailovic@ijs.si

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**Figures**
FIG. 1: The structure and charge ordering of equilibrium and non-equilibrium states of 1T-TaS$_2$ and a schematic diagram of the experiment. a The crystal structure of 1T-TaS$_2$. b The regions of stability for charge-density wave states under equilibrium conditions on cooling and heating, and charge density simulations in the incommensurate (IC), nearly commensurate (NC), triclinic (T) or commensurate (C) states respectively (see Supplementary Information). c The polaron structure in the C state, where twelve atoms are displaced towards the central Ta with the one extra electron, as indicated by the arrows. d The lifetime of the H state created by an ultrafast optical pulse in the C state is indicated by the colour.$^{16}$ e Schematic view of the combined STM and SEM system with optical access for ultrafast laser pulse excitation. f The SEM image shows a cleaved a-b surface of TDS single crystal in contact with the sharp tungsten STM tip. After beam alignment the tip is retracted and the area underneath is illuminated with a focused single ultrafast laser pulse.
FIG. 2: Raw STM images of the hidden and equilibrium CDW states in 1T-TaS$_2$:  

a) Large-area STM image ($V_t = -800$ mV, $I = 1.5$ nA) of the C state CDW showing a uniform periodic structure (polaron voids appear as defects). Bright dots correspond to polaron positions. The height is indicated by the colour bar in each case. 

b) Atomic resolution STM image of the C state, revealing the hexagram structure shown in Fig. 1 b and c. (a) Bias and current were adjusted to highlight the top sulfur layer without any admixture of the tantalum orbitals ($V_t = 100$ mV, $I = 200$ pA). The CDW modulation is larger than the atomic size and has a peak in the group of three S atoms, positioned above the central Ta atom of the SD hexagram (red) (the corresponding model is shown in the inset). Each star has the same atomic configuration, illustrating that CDW order is commensurate with the atomic lattice. 

c) Large-area STM image of the nearly commensurate CDW state at 300 K ($V_t = -250$ mV, $I = 150$ pA), showing a periodic array of brighter areas – domains – separated by darker, soft domain walls. 

d) Atomic resolution STM image of the NC state ($V_t = -200$ mV, $I = 1.5$ nA), revealing the sulfur triangles with a slight admixture of tantalum orbitals. The dotted red line shows a single atom shift of the CDW modulation upon crossing a domain wall. 

e) An extensive-area STM image ($V_t = -200$ mV, $I = 100$ pA) of the H state after switching with a single ultrafast optical pulse showing a complex sharp network of domain walls. 

f) Atomic resolution STM image of several domains in the H state ($V_t = -800$ mV, $I = 1.5$ nA). The CDW is commensurate with the atomic lattice within the domains. Dashed red lines highlight the phase shifts of polaron positions on crossing a domain wall.
FIG. 3: Reciprocal space analysis of the metastable H state in relation to equilibrium states: a A Fourier transform (FT) of the atomically resolved STM image of C state (Fig. 2b) shows commensurability in reciprocal space: peaks of atomic (black) and CDW (red) lattices coincide at certain positions. The two lattices are rotated by $\phi = 13.9^\circ$. $a^*$ and $b^*$ are unit vectors of the atomic reciprocal lattice and $(Q_C^{(i)}, i = 1...3)$ – of the CDW reciprocal lattice. b FT of the STM image of NC state (Fig. 2c) shows the CDW reciprocal unit cell. c Expanded image of panel (b) shows the highest-intensity $Q_{NC}^{(2)}$ peak (red), surrounded by 3 neighbours (blue). The domain modulation vector is given by $\vec{k}_\text{domain}^{(2)} = \vec{Q}_{NC}^{(2)} - \vec{Q}_{sat}^{(2)}$. (see Supplementary Fig. 3 for more details). Note that the FT amplitude is zero between the satellite peaks. d FT of the atomic resolution STM image in the H state (Fig. 2f). The black (red) hexagon shows the reciprocal unit cell of the atomic (CDW) lattice. The CDW rotation angle is $\phi = 13.45 \pm 0.35^\circ$. e FT of the large-scale STM image of the hidden state (Fig. 2e) reveals a CDW unit cell with six bright peaks – fundamental vectors (red) – and three-fold streaks protracting over $\Theta \approx 9.5^\circ$. f Detailed structure of the H state streaks within the black square of panel (e). g, Azimuthal cross-section data of the $Q_{H}^{(1)}$ fundamental CDW peak in the FT of the hidden state (e), compared with the $Q_C (T = 4.2K)$, and the NC state, $Q_{NC} (T = 205K$, third-order peak, right axis). The lines show gaussian fits. FWHM = 0.31° for the H state. h A cross-section of the H state FT in the panel (f) along the AA’ line with a 5-pixel lateral averaging. The arrow shows the $n = 1$ CDW harmonic (see text).
FIG. 4: Local topological structure in the H state, and a schematic sequence of events in its formation.  

a, The twelve possible ⃗D vectors connecting 0 and 1 − 12 atomic sites of a David star. The thirteenth is the identity operation ⃗D = 0. 

b, Z₃ vortex-antivortex pair separate 4 domains. The paths defining the winding number are indicated by arrows. Colour shading shows the misfit vector in the adjacent domains according to the scheme in the panel (a). 

b, X crossing of two domain walls separates four domains. A contour encircling it results in a non-zero sum of the misfit vectors, D₁₀ + D₂ + D₅ ≠ 0. 

d, A kink K is a defect site where a change of the domain wall type occurs without a change of ⃗D. 

e-f A proposed sequence of events in the formation of the H state. Initially, disconnected domains with 6 kinks (Ks) are formed within the intermediate IC* CDW state. Continuous tessellation emerges when Ks bind into X crossings and Y − ¯Y pairs. During this process the transient IC* state is converted into the H state, following both local rules and long-range vortex ordering shown in Fig. 5.
FIG. 5: Long range topological order in the H and NC states. a The definition of $|\vec{D}|$ and its direction $\alpha$ w.r.t. the C lattice. b,c, Large scale real space images (from Fig. 2e) of the H state showing $|\vec{D}|$ and $\alpha$ for each polaron with respect to its position in the initial C lattice. The image exhibits long range hexagonal vortex lattice on a scale of $\sim 70$ nm. The vortices (winding number -1) are indicated by white arrows. d, The Moiré pattern between $Q_H$ and $Q_C$ lattices calculated on the same grid. The vortices of the misfit vector field are shown by white arrows. A cartoon in the bottom right corner schematically shows the actual observed hexagonal tessellation of domains: instead of continuous change, $\vec{D}$ (shown by the black arrows) is fixed inside a domain and changes on crossing a domain wall. e, The modulation of the domain size in the H state on a scale of $\sim 70$ nm, emphasized by shading.
Supplementary Information for “Dual vortex charge order in a
metastable state created by an ultrafast topological transition in
1T-TaS₂”

Yaroslav A. Gerasimenko,* Igor Vaskivskyi, and Dragan Mihailovic†
CENN Nanocenter, Jamova 39, SI-1000, Ljubljana, Slovenia and
Department of Complex Matter, Jozef Stefan Institute,
Jamova 39, SI-1000, Ljubljana, Slovenia
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Supplementary Note 1: simulation of CDW states using Nakanishi-Shiba model

Below we summarize how various CDW states in 1T-TaS$_2$ are built in real and reciprocal space. The fundamental CDW vectors ($Q^{(1)}, Q^{(2)}, Q^{(3)}$) form the triangular basis, which creates star of David distortion of the Ta atomic lattice. They appear in the IC state aligned with atomic lattice vectors, whereas in NC and C states they are shrunk and rotated. One can thus describe them as the following product:

$$
\left( Q^{(1)} \quad Q^{(2)} \quad Q^{(3)} \right) = \gamma \cdot \begin{pmatrix}
1 & 0 \\
-1 & 1 \\
0 & -1
\end{pmatrix} \cdot \begin{pmatrix}
a^* \\
b^*
\end{pmatrix} \cdot \begin{pmatrix}
\cos(\phi) & -\sin(\phi) \\
\sin(\phi) & \cos(\phi)
\end{pmatrix},
$$

where $a^*$, $b^*$ are reciprocal vectors of Ta triangular atomic lattice, $\gamma = Q^{(i)}/a^*$ is star of David size and $\phi$ is the rotation angle between CDW and atomic lattices.

In CCDW state the fundamental $Q^{(i)}_C$ is determined by the commensurability condition:

$$Q^{(1)}_C = \frac{1}{13}(3a^* + b^*)$$

or, equivalently,

$$3Q^{(1)}_C - Q^{(2)}_C = a^*. \tag{Eq. 1}$$

The above commensurability condition allows to calculate the ideal length of CDW vector, $Q^{(i)}_C/a^* \approx 0.277$, and its rotation w.r.t. atomic lattice, $\phi \approx 13.9^\circ$.

Real space images of IC and C state can be readily built as the following sum:

$$Z = \Re \left[ \sum_i \exp(iQ^{(i)}R) + \exp(ia^*R) + \exp(ib^*R) \right],$$

where $R$ is in-plane real-space vector. The sum term gives CDW modulation, the last two – atomic modulation. Suppl. Fig. 1a-d shows the real and reciprocal space models of IC and C states. In the commensurate case each star of David is centered at a certain Ta atom position, resulting in the same atomic structure of all CDW units across the sample (Suppl. Fig. 1c). This feature can be checked experimentally in atomic resolution STM images and is indeed observed in C, NC and H states (main text, Fig. 2b,d,g). In contrast, violation of commensurability will result in irregular atomic configuration for each star of David, as illustrated in Suppl. Fig. 1a.
**Supplementary Figure 1:** Model of IC and C states: a, c, Real space models. Star of David distortion is shown by red polygon. In the commensurate state DS centers coincide with certain Ta atoms and, in contrast, have random atomic surrounding in the incommensurate state. b, d, Reciprocal space (FT) models, illustrating the relation between atomic (black) and CDW (red) lattices.

In NCCDW state the periodic domain structure appears, a model example of which is shown in Suppl. Fig. 2a. Domains appear as brighter areas, which are separated by darker domain walls. CDW inside the domains is commensurate, but has different phase in neighboring domains. The phase change occurs inside the domain walls. To describe such state it is necessary to add the vectors describing a domain network to $Q_{NC}$ basis of CDW.

It can be modelled with simple approach used to interpret the early X-ray results, which considers domain structure as a result of the interference of two triple CDWs. One of them is given by the three fundamental NCCDW vectors, whereas the other has smaller amplitude and is given by three additional vectors chosen in such way, that their linear combination with fundamental ones will be equal to some vector of reciprocal atomic lattice. This gives the new commensurability condition:

$$Q_{sat}^{(1)} = -2Q_{NC}^{(1)} + Q_{NC}^{(2)} + a^*.$$  \[Eq. 2\]
The corresponding vectors are shown in Figure 2b,c. This equation gives only one possible
commensurability condition for reciprocal lattice vector $G = a^*$, but it can be extended to
the case of so-called higher harmonics $G = na^*$, $n = 1, 2, \ldots$, as shown in the main text.

It is worth noting, that the domain wall is always two stars of David thick, independent
of $Q_{NC}^{(i)}$, as long as (Eq. 2) is fulfilled. This model also gives slowly varying modulation of
height, peaking at the domain centre (cf. Suppl. Fig. 2a). Real space image is then given
by:

$$Z = \Re \left[ \sum_i \left( \exp(iQ_{NC}^{(i)}R) + \exp(iQ_{sat}^{(i)}R) \right) + \exp(ia^*R) + \exp(ib^*R) \right].$$

Full details of the nearly commensurate state are captured with Nakanishi-Shiba theory\(^1\),
which considers NC state as a product of CCDW and periodic domain structure overlayed
on top of it:

$$Z = \sum_i \exp(Q_C^{(i)}R)\Psi^{(i)}(R),$$

where $Q_C^{(i)}$ are fundamental CCDW wavevectors and

$$\Psi^{(i)}(R) = \sum_{l,m,n \geq 0 \ l \cdot m \cdot n = 0} \Delta_{lmn} \exp(iq_{lmn}^{(i)}R)$$

is the modulation responsible for the domain structure.

Modulation periods are given by:

$$q_{lmn}^{(i)} = lk_{domain}^{(1)} + mk_{domain}^{(2)} + nk_{domain}^{(3)} + q^{(i)}.$$ \hspace{1cm} \text{Eq. 3}

Here the last term is the discommensuration vector:

$$q^{(i)} = Q_C^{(i)} - Q_{NC}^{(i)}.$$ \hspace{1cm} \text{Eq. 4}

with $Q_{NC}^{(i)}$ being the fundamental NCCDW wave vector (see Suppl. Fig. 2d). $q^{(i)}$ therefore
represent the deviation from commensurability. The correct domain periodicity should
result in the commensurate CDW inside the domains and thus can be calculated using
commensurability condition similar to that for fundamental vectors (Eq. 1):

$$k_{domain}^{(1)} = 3q^{(1)} - q^{(2)}.$$ \hspace{1cm} \text{Eq. 5}

The above equations are illustrated in Figure 2d.

These two approaches are equivalent and connected by the relation (Suppl. Fig. 2c):

$$k_{domain}^{(1)} = Q_{NC}^{(1)} - Q_{sat}^{(1)}.$$ \hspace{1cm} \text{Eq. 6}
Finally, additional satellites can appear from linear combinations of $Q_{NC}^{(i)}$ and $Q_{sat}^{(j)}$ where $i \neq j$. This is equivalent to several of $(l, m, n)$ coefficients being non-zero in NS model (Eq. 3). For example, the satellite $-k_{domain}^{(3)}$ will be seen near $Q_{NC}^{(2)}$ as a result of the following vector equation (see Suppl. Fig. 2e):

$$-k_{domain}^{(3)} = -Q_{NC}^{(1)} - Q_{sat}^{(3)}.$$  

Eq. 7

Below we will refer to these satellites as $-k_{domain}^{(i \pm 1)}$, where $i$ determines the fundamental peak $Q_{NC}^{(i)}$, near which the satellite $(i \pm 1)$ is observed. They are often seen in FT of STM images obtained in NC state, though their intensity is much smaller compared to that of $k_{domain}^{(i)}$ (cf. Fig. 3c in the main text). The relation between these satellites and domain packing is illustrated in Suppl. Fig. 3.

One can readily see that David stars have different spacing inside the domain walls, depending on the combination of satellites used in simulation. The interference pattern produced by multiple satellites slightly deviates from the pure commensurability, so it is hard to determine which atomic sites David stars occupy inside the domain walls. To get a qualitative understanding, we have calculated pair distribution functions (PDF) for David star centers (cf. Suppl. Fig. 4b) obtained in simulations and compared them to the one for Ta atoms (cf. Suppl. Fig. 4b). The results are shown in Suppl. Fig. 4a, where the first three panels correspond to David stars PDF and the last one is atomic. In the latter, peaks are marked by the respective atomic configurations, i.e. $2 + 1$ is the sum $2\vec{a} + \vec{b}$. In the case where just one satellite, $k_{domain}^{(i)}$, the largest DS peak coincides with the $3 + 1$ configuration. Adding more satellites, $-k_{domain}^{(i \pm 1)}$, shifts it slightly, as one can expect from the change of the vector sum length. More importantly, each configuration produces additional peaks located close to atomic other than the $3 + 1$, going through the possible $2, 2 + 1$ and $3$ shifts, in nice correspondence to the theoretical analysis by Ma et al.4.


Supplementary Figure 2: Real and reciprocal (FT) space models of NCCDW: a, Real space model of NCCDW state with overlaid tantalum lattice. Angle between CDW and atomic lattices used in the simulation is $\phi = 11.7$ and periods ratio is $Q_{NC}^{(i)}/a^* = 0.283$. Star of David distortion is shown with red polygons and is commensurate with Ta lattice inside domains. b, Reciprocal space model showing relation between atomic and CDW lattices. Atomic vectors are shown in black, $Q_{NC}^{(i)}$ – in red, $Q_{sat}^{(i)}$ – in blue. This model uses commensurability condition (Eq. 2) in the NC state to calculate satellites, $Q_{sat}^{(i)}$, position. c, Zoom-in of the reciprocal space model, showing in details the CDW unit cell. This panel demonstrates the relation between satellites and domain structure vectors, $k_{domain}^{(i)}$ (green). d, Zoom-in of the $Q_{NC}^{(2)}$ area of the reciprocal space model, demonstrating the relation between Nakanishi-Shiba model and satellite structure. Discommensuration vectors of Nakanishi-Shiba model, $q^{(i)} = Q_{C}^{(i)} - Q_{NC}^{(i)}$; (Eq. 4) are shown in black. Their vector sum determines the domain period, $k_{domain}^{(i)}$ according to the equation (Eq. 5). e, Possible satellite positions near the fundamental peak for the case of $Q_{NC}^{(2)}$. One of them corresponds to $k_{domain}^{(2)}$, other two are described by $-k_{domain}^{(j)}$, where $j = 1, 3$ in this case and result from the interference of $Q_{NC}^{(i)}$ and $Q_{P}^{(j)}$ (see (Eq. 7)).
Supplementary Figure 3: Relation between satellites and packing of hexagonal domains in NCCDW: a, Simulation with $Q_{NC}^{(i)}$ and $k_{domain}^{(i)}$ satellites showing the hexagonal domains with clearly visible domain walls in between. This image is characteristic to NCCDW state. Top inset shows the FT. White square emphasizes the domain wall. b, c, Simulation with $Q_{NC}^{(i)}$ and $-k_{domain}^{(i+1)}$ (in (b)) or $-k_{domain}^{(i-1)}$ (in (c)) satellites. Whereas domains stay almost in place, the domain walls structure is different. None of (b) or (c) alone are realized experimentally in NCCDW state. d, e, Simulation with $Q_{NC}^{(i)}$, $k_{domain}^{(i)}$ and $k_{domain}^{(i-1)}$ (in (d)) and $k^{(i)}$ and $-k_{i}^{(i+1)}$ (in (e)). These images are also characteristic to NCCDW state, though compared to (a) they have different phase shift inside the domain wall, resulting in different star of David arrangements (see areas inside white squares). f, Simulation with $Q_{NC}^{(i)}$, $k^{(i)}$ and both $-k_{domain}^{(i+1)}$ and $-k_{domain}^{(i-1)}$ is equivalent to the (a) case.
Supplementary Figure 4: Pair distribution functions for interference patterns produced by multiple satellites: a, Pair distribution functions for simulations with $Q_{NC}^{(i)}$ and $k_{domain}^{(i)}$ (panel 1), plus $k^{(i-1)}$ (panel 2) or $-k^{(i+1)}$ (panel 3). Panel 4 contains atomic pair distribution function. b, c, Point patterns with numerically determined centers of David stars (b) and Ta atoms (c) used for PDF calculations.
Supplementary note 2: fluence dependence of amplitude mode temperature

The peak lattice temperature is determined from the frequency of the amplitude mode (AM) (Suppl. Fig. 5a), whose temperature dependence is determined from independent low-fluence measurements (Suppl. Fig. 5b). A plot of $T_{AM}$ vs. fluence (Suppl. Fig. 5c) shows that at threshold, the temperature of the AM reaches $150 \pm 10$ K. The AM is the mode which is most strongly coupled to the electrons, so its temperature is inevitably by far the highest of all the lattice modes.

Supplementary Figure 5: The determination of the lattice temperature after photoexcitation:  

a. The time-dependence of the AM frequency measured after photoexcitation by time-resolved coherent phonon reflectivity response at different excitation fluences and b, its temperature dependence with low fluence.  

At threshold fluence for switching (0.85 mJ/cm$^2$) $T_{AM}$ is $150 \pm 10$ K (arrow).  

d. Electron and lattice temperatures calculated within the two-temperature model for 1T-TaS$_2^{7,8}$ for the optical pulse fluence of 1 mJ/cm$^2$ applied at $T = 4.2$ K.
Supplementary note 3: error estimation in HCDW $\phi$ angle measurement

To determine the $Q_H$ vector length and angle from FT of the atomic-resolution image (main text, Fig. 3d) we used peak positions for atomic and second order CDW reflexes. The latter have higher pixel resolution, compared to the first order ones. The values obtained this way were averaged among the three independent directions, $Q^{(i)}_H$, $i = 1, 2, 3$. With the FT resolution available in atomically-resolved scans the error is limited by the pixel size. The error bars are thus set as an error introduced by the single pixel shift of the reflex position on the average value of either angle or length.

Supplementary Figure 6: Parametric diagram of CDW states: the behavior of the fundamental CDW vector with temperature (equilibrium: NC and C) or after optical excitation (H) is shown in terms of the length ($Q^{(i)}/a^*$) and the angle between atomic and CDW lattices ($\phi$). Hidden states parameters we measured are shown by two red points: one from atomic resolution (larger error), another – from satellite positions and real space analysis (error is equal to experimental width of the fundamental peak). Blue dot marks the $Q_{NC}$ position in NC state measured with STM on the same sample at $T = 205$ K. Literature data for the lowest-temperature values of $Q_{NC}$ measured with STM is shown by black ($T = 215$ K)\textsuperscript{5} and gray ($T = 240$ K)\textsuperscript{6} points. For the latter, the length is deduced from the approximate period of 1.2 nm\textsuperscript{6}. For reference, yellow line and diamonds show NC states observed with X-ray diffraction\textsuperscript{3} down to $T = 195$ K. At lower temperatures, CDW vector changes discontinuously to C state (blue diamond). Yellow dotted line shows the continuation of the NC trend. Red dashed line connects NC and C state. No stable states were observed in this region of parameters before. Large discrepancy of STM and X-ray data likely originates from the distortion of NC structure\textsuperscript{6} as the transition to C-state is approached.
Supplementary Figure 7: Single $Q_H$ in the optically switched hidden state: a Part of STM image (main text, Fig. 2e) of the optically switched hidden state. The areas with large and small domains are marked with squares I and II. b, c, FT of the areas I and II: the fundamental vector (red) $Q_H^{(1)}$ remains the same, while the satellite positions (blue) changes in accord with the real-space domain sizes. Therefore, in the FT of the full image multiple satellites correspond to the same fundamental vector.

Supplementary Method 1: Fitting FT peaks position in hidden state with Nakanishi-Shiba model

In order to get high value of satellite spread angle $\Theta$ and small deviation of the CDW rotation angle $\phi$ from its C-state value within the NS model, we use $k_{\text{domain}}^{(i)}$ harmonics (see also Suppl. Fig. 16d). The fitting procedure of the real data with NS model is given below.

In the first step we estimate the required number of harmonics. To this end, we find the smallest angular distance between the fundamental, $Q_H^{(1)}$, peak and the satellite peaks around, which in this case is $\Theta_1 \approx 1.7^\circ$. This value appears the same, independent of the fundamental peak (i.e. $i = 2, 3$) It gives a rough estimate of $k_{\text{domain}}^{(1)}$ length and the number of groups as $\Theta_{\text{max}}/\Theta_1 = 5$. Indeed, one can see from the cross-section that the peaks along $k_{\text{domain}}^{(1)}$ direction can be separated into five groups (Suppl. Fig. 8).

In the second step, we determine the averaged peak positions for each group. Peaks along the $k_{\text{domain}}^{(i)}$ direction in the experimental data (Suppl. Fig. 9a,b) are additionally split in the radial direction by 2-3 pixels. This gives the characteristic scale of the additional modulation equal to 1/3 of the scan size or $\sim 70$ nm. The only periodicity that can be
Supplementary Figure 8: 7-line average FT cross-section through satellites near $Q_H^{(1)}$: the highest peak corresponds to the fundamental vector $Q_H^{(1)}$, whereas smaller peaks show that satellites are grouped near five positions.

found on such scale in real space image corresponds to the distance between groups of small domains, separated by groups of larger domains and vice versa. This kind of modulation cannot be described by NS model. We collapse them into 'average' single peak using the simple centroid procedure described below.

To this end we estimate seed positions, that should be located at multiples of $k_{domain}^{(1)}$ and hence lie on a straight line. Then we take rectangular area centered at seed centers and $1.7^\circ$ wide along $k_{domain}^{(1)}$ and 10 pixels in the radial direction and calculate the weighted centroid for each of these areas. All the real experimental FT points included in the averaging, not just detected peaks. The resulting centroid FT coordinates are used as average peak positions to be fitted with NS model.

Fit is done using the least squares method. The quantity minimized is the deviation of all five NS harmonics of $k_{domain}^{(1)}$ from the average experimental points. In the NS model we vary the $Q_H^{(1)}$ parameters $\phi$ and $Q_H^{(1)}/a^*$ and thus change direction and length of $k_{domain}^{(1)}$. The result of the fit is shown in Suppl. Fig. 9b. The comparison of the STM data with the real-space simulation of the individual harmonics is presented in the Suppl. Fig. 9c-f and shows nice correspondence of the domain sizes.

The emergence of higher harmonics underlines the differences between hidden and equilibrium states. The distinction that takes place in the reciprocal space can be illustrated on the three-dimensional phase diagram, which shows simultaneously the fundamental CDW vector position, $(\phi, Q^{(i)}/a^*)$, and corresponding harmonics position, $\Theta$ (Suppl. Fig. 10). It appears, that with temperature and photodoping 1T-TaS$_2$ explores different axes of this
Supplementary Figure 9: Fitting FT peaks position in the hidden state with NS model: a, Part of the Fourier transform (Fig. 3e) of the large-scale STM image of hidden state showing $Q_H^{(1)}$ peak (red), its satellites (blue) and their position with respect to the origin. b, Zoom-in of the $Q_H^{(1)}$ peak area in (a) revealing the details of the Nakanishi-Shiba model: fundamental peak is shown in red, satellite peaks – with blue circles, fitted $k_{\text{domain}}^{(1)}$ harmonics – in green. c, d, Close-up of the two regions in Fig. 2e (main text), demonstrating the domains most similar to the real-space model of the third, e, and of the fifth, f, harmonics of $k_{\text{domain}}^{(i)}$. Contrast was adjusted in the model images to emphasize the domains.

Phase diagram. Indeed, within the phenomenological model presented before$^{7,12}$, the full domain wall length is linked to the difference between the photodoping concentration of electrons and holes, $n_d = n_e - n_h$, and their chemical potentials being equal at the same time, $\mu_d = \mu_e = \mu_h$. Given the domain walls are arranged into the regular hexagonal structure, one could extract the CDW vector $Q_H \sim Q_C - \pi n_d$. For high values of $n_d$, which is the case in the experimentally observed H state, deviation of $Q_H$ from $Q_C$ should be large, in contrast to the measured values. The system thus chooses to rotate $Q_H$ slightly, but excite multiple harmonics instead. We expect such changes to emerge from different nature of the transition, as discussed in the main text.
Supplementary Figure 10: Parametric \((\phi, Q^{(i)}/a^*, \Theta)\) diagram of CDW states illustrating the difference between the equilibrium and hidden states in 1T-TaS\(_2\): Blue surface shows the first harmonic, \(Q_{sat}\), satellite positions \(\Theta\) within the Nakanishi-Shiba model (respective angles and vectors are shown on the sketch). Solid lines show experimentally observed equilibrium trajectory of NCCDW (blue) state\(^3\) upon cooling, which then changes discontinuously to CCDW (green) state as temperature is lowered. Hidden state (fitted \(\phi\) and its value from atomic resolution scan are close) is located in between C and NC states, and does not lie on the continuation of NC trajectory (dotted line). Most importantly, hidden state satellites (domain structure) follow different axis: instead of rotating CDW vector, higher harmonics are excited.
Supplementary Discussion 1: breakdown of Nakanishi-Shiba model at low temperatures

Nakanishi-Shiba model used to describe the NC state in 1T-TaS$_2$ is based on the modulation of CDW amplitude by the interference of two waves. The first one is fundamental with the period $Q_{NC}$ and the second is $n = 1$ harmonic with the period $Q_{sat}$ (see (Eq. 2)). Modulation of the amplitude can be clearly seen at high temperatures, where David star distortion is the largest in the center of a domain and decreases towards its edge (main text, Fig. 2c).

The model predicts that higher harmonics $n > 1$ should emerge as the temperature is decreased. Their phases are adjusted in such a way that the amplitude modulation is flattened, and the overall domain pattern remains periodic. Such flattening is indeed observed at low temperatures, but the domain periodicity is broken and no higher harmonics are observed. Such behavior can be explained as the breakdown of the amplitude modulation in favor of phase modulation – emergence of solitonic domain walls.

In order to check, whether the higher harmonics observed in the H state are associated with the domain flatness, we apply a frequency-selective inverse Fourier transform. An example of two flat domains separated by the sharp domain wall is shown in Suppl. Fig. 11a. The height profile (Suppl. Fig. 11c) confirms that David star distortion is the same independent of the distance to the domain wall ($x = 7$ nm). Next, we apply band pass filter to the Fourier transform of the image: the band is a ring including only the first CDW Brillouin zone and all the Nakanishi-Shiba harmonics in it. Suppl. Fig. 11b shows the same two domains as before but in the bandpassed inverse Fourier transform. The respective height profile (Suppl. Fig. 11c) clearly shows, that David star distortion becomes smaller towards the domain wall. Thus, the harmonics observed in the H state are not related to the domain flatness. This result also shows that the latter comes from the increased intensity in the higher Brillouin zones of CDW.
Supplementary Figure 11: Absence of relation between domain flatness and harmonics in the H state: a, Zoom of a domain boundary in the STM image of the optically switched state. b, Zoom of the same area as in (a) but in the inverse of the bandpassed FT of the STM image. The band includes the first CDW Brillouin zone only. c, Height profiles along the cross-section shown by the dashed red line in (a) and (b). Black and red lines are guides for the eye.
Supplementary Figure 12: Triclinic CDW state: a, STM image of a triclinic CDW state measured on heating at 240 K ($V_t = -800$ mV, $I_t = 100$ pA) demonstrating domains (stripes) elongated in one direction (orange arrow) and b, its Fourier transform. CDW unit cell is shown in red. Only 4 of 6 CDW peaks are clearly split, whereas two other ($\pm Q_T^{(1)}$) has no satellites. This type of loss of hexagonal symmetry of the domain structure (not CDW itself) is the signature of the triclinic state\textsuperscript{5}. Amplitude modulation peaks are marked green. c, zoom-in of the central part of FT (b) showing that hexagonal symmetry of amplitude modulation is also lost in the triclinic state. (b)-(c) allows to tell unambiguously triclinic state from hidden. d, the same zoom-in for FT of NCCDW state at 300 K (from Fig. 3c in main text), showing the hexagonal symmetry of the amplitude modulation.
Supplementary Figure 13: Domain wall behavior in adjacent layers: STM image of the hidden state measured with large setpoint current \((V_t = -800 \text{ mV}, I_t = 1.5 \text{ nA})\), which allows to get information about the TaS\(_2\) layer beneath. Three domain walls in the top layer (dark grooves approx. 2 DS wide) are marked with dashed yellow lines. Domain walls beneath can be identified by the darker areas within the domains and are marked with dashed red lines. Crossing of domain walls in the adjacent layers appears as a dip (see arrow). It is clearly seen, that the domain walls in the adjacent layers tend to avoid each other. This observation suggests that the role of interlayer interactions cannot be ruled out.
Supplementary Discussion 2: analysis of hidden state obtained by electrical switching with STM tip

Switching from CCDW to metastable metallic hidden state can be done also with electrical pulse applied between two contacts on top of 1T-TaS$_2$ crystal. Alternatively, the pulse can be applied between the crystal and STM tip, and the resulting state can be imaged in situ. Below we perform the same analysis of electrically switched state as the one presented in the main text for the optically switched state.

Here the best switching results were obtained for small tip-sample separation with a low pulse voltage ($V_p \sim 3.5V$). Suppl. Fig. 14a shows that the homogeneous CCDW state breaks down into an irregular array of domains separated by sharply defined walls (they appear bright in the scan due to different sign of tip bias, compared to the figures in the main text), apparently similar to the optically switched hidden state (o-HCDW). Fourier transform (Suppl. Fig. 14b) reveals the CDW unit cell with the six peaks and their satellites. The spread of the latter, $\Theta_{max} \approx 5.5^\circ$, is almost twice smaller compared to that of o-HCDW, which reflects smaller domain walls density in case of electrical switching. The zoom-in of the area around one of the peaks shows that satellite structure is smeared and intensity along $-k_{domain}^{(i\pm1)}$ directions is vanishingly small, in contrast to more well defined structure in the o-HCDW state. Even though FT resolution is somewhat worse due to smaller size of the scan (80 nm vs 200 nm for o-HCDW), the fundamental peak is strongly smeared, FWHM = 0.754°, as can be seen from angular cross-section in Suppl. Fig. 14c. The above results present strong evidence towards less homogeneous transition to hidden state and absence of the true LRO in case of electrical switching.

Smaller $\Theta_{max}$ in case of electrical switching suggests that optical pulse excites the electronic system more strongly. The different kind of domain walls (absence of $-k_{domain}^{(i\pm1)}$ satellites) implies that the nature of excitation also plays role in determining the resulting domain structure. The above arguments allow us to discuss optical switching separately from the electrical one.
Supplementary Figure 14: Electric switching to hidden state with STM tip: a, Large-scale STM image of the domain state obtained after in situ switching with STM tip at 4.2 K ($V_t = 800$ mV, $I_t = 500$ pA); b, Fourier transform of the STM image in (a), showing the CDW unit cell (red). The length of satellite tail is $\Theta_{max} \approx 5.5^\circ$, which is smaller compared to 9.5° observed in the optically switched hidden state. Inset shows the zoom-in of the $Q_{electric}^{(2)}$ peak area, which appears quite smeared with structure barely seen. The satellites corresponding to $-k^{(\pm 1)}_{domain}$ are absent. c, Angular cross-section of the $Q_{electric}^{(2)}$ peak along $q_x$ (red) and $q_y$ (blue) FT axes. Solid lines are Gaussian fits.

To illustrate the real space details of the domain structure, we have performed the mapping of David stars misfit vectors. The results are shown in Suppl. Fig. 15. Misfit vector field shows no distinct structure. Its amplitude changes randomly in space (Suppl. Fig. 15b), and its direction jumps between very different, sometimes opposite, values (Suppl. Fig. 15b). Such a behavior is qualitatively different from the Moiré-like vortex structure observed for the long-range ordered H or NC states (see main text): gradual rotation of the direction (dark-blue to dark-red) and gradual increase of the amplitude from 0 (green) to 1 (cyan) and then 1 + 1 (blue). This allows us to conclude that the H state obtained by electrical switching does not have long range order.

It should be noted, that domain walls in this state also show $X$ and $K$ crossings in addition to the standard $Y - \bar{Y}$ ones characteristic of the pure hexagonal structure. This resemblance could suggest similar mechanism of transition.
Supplementary Figure 15: Misfit vector map in the hidden state obtained by electrical switching with STM tip: a, large-scale STM image of the H state obtained after in situ switching with STM tip at 4.2 K ($V_t = 800$ mV, $I_t = 500$ pA); b, the map of the misfit amplitude on the $(0, 1, 1 + 1)$ scale in atomic units for the image in panel (a); c, the map of the misfit direction for the image in panel (a); d, the legend to the panels (b) and (c), showing the definition of the misfit vector $\vec{D}$, its amplitude and direction.
Supplementary discussion 3: Nakanishi-Shiba model of supercooled NCCDW

Since NS theory describes well the domain structure of NCCDW state, we can use it to model various outcomes that could be found for supercooled NCCDW state. Here we consider two relevant scenarios: (i) “frozen” domains with single $Q_{NC}$ and (ii) sum of “frozen” domains corresponding to different $Q_{NC}$.

The first scenario is equivalent to the trajectory, where the system is heated to some temperature above C–NC transition and then is quenched to low temperatures. The model FT corresponding to this scenario is shown in Suppl. Fig. 16a, where the parameters are chosen to obtain large $\Theta$ angle (length of the streak) similar to that in experiment. This corresponds to the temperatures as high as $T = 300 \text{ K}$, since at lower temperatures $\Theta$ is known to be smaller. In FT picture we should observe single fundamental $Q_{NC}^{(i)}$ set of peaks and with one $k_{\text{domain}}^{(i)}$ satellite, determined by the commensurability condition (Eq. 2). Two domains are unlikely to merge into larger one, given $Q_{NC}$ is fixed. Indeed, the mismatch between commensurate and real NCCDW lattices increases with size, resulting in energy loss, not gain. Hence, only $k_{\text{domain}}^{(i)}$ corresponding to $l + m + n = 1$ in (Eq. 3) are allowed. The only path of merging two domains into larger one can occur with simultaneous rotation of $Q_{NC}$ inside the domain, i.e. another $Q_{NC1}$ will appear in FT (see Suppl. Fig. 16b). This situation is discussed below.

The second scenario corresponds to the trajectory, where high-temperature state is quenched at intermediate rates and different $Q_{NC}$ are present in real-space picture. Another option is that single-$Q$ rapidly quenched state has relaxed at low temperatures, as described above. In both trajectories the FT of such image will result in a number of $Q_{NC}^{(i)}$ and $k_{\text{domain}}^{(i)}$ peaks (see Suppl. Fig. 16c). This scenario has two distinct features in the FT picture. First, fundamental peak will be smeared and satellites will no longer be equidistant (due to random choice of local fundamental vectors). Second, satellites will not lie on the same line with fundamental peak. Both these features are in contrast to experimentally observed FT picture.

For comparison the model CDW state with 5 harmonics used to fit experimental data for the hidden state is shown in Suppl. Fig. 16d. The features of model FT of supercooled NC state are absent here and the pictures are qualitatively differen. Thus hidden state cannot be considered as a supercooled NCCDW state.
Supplementary Figure 16: Comparison of FT pictures for five-harmonic CDW (hidden) and for various scenarios of supercooled NCCDW:  

a, First scenario: quenched high-temperature NCCDW state with $\phi = 11.7$ and $Q_{NC}^{(i)}/a^* = 0.286$. No shorter $k_{domain}$ are allowed. 
b, HT NC state relaxes partially to a configuration closer to CCDW. Additional $Q_{NC1}^{(i)}$ (red dot) appears with e.g. $\phi = 13.5$ and $Q_{NC1}^{(i)}/a^* = 0.279$ closer to that of CCDW ($13.9^\circ$, 0.277). The corresponding satellites are shown with blue dots. Green dotted lines show the direction of all the $k_{domain}$ for the original $Q_{NC}^{(i)}$ vector. 
c, Second scenario: domains with different $Q_{NC}$ (red dots) are frozen during quench ($\phi = (11.7 \times 13.5)$, $Q_{NC}^{(i)}/a^* = (0.286 \times 0.279)$). Each of $Q_{NC}$ has its own set of satellites with different $k_{domain}$, shown with blue dots. The satellites no longer lie on the green dotted line, which connect the highest-temperature $Q_{NC}$ and its satellites. 
d, Five-harmonic CDW used to fit experimental data. Red vector shows the experimentally determined $Q_{H}^{(i)}$ with $\phi = 13.5$ and $Q_{H}^{(i)}/a^* = 0.279$. Blue dots illustrate the five harmonics of $k_{domain}^{(i)}$. All of them correspond to either of three lines, which originate from fundamental peak (compare to panel (c)).
Supplementary Method 2: mapping misfit vectors in the H state

David stars positions in the H state can be determined with high accuracy using image processing methods. Each David star appears as a well-defined peak in STM images. The accuracy of finding the peak could be compromised mostly by the noise. The noise spatial scale can be divided into two categories: (i) slowly varying background on the scale of several DS and (ii) spikes smaller than DS. Type-1 noise makes detection of DS inside domain walls hard. This problem is solved by using gradient-based algorithms, e.g. Laplacian of Gaussian. Type-2 noise can be overcome using the known separation of neighboring DS. Still algorithm performance can be improved by preprocessing the image, based on the known global and local structure.

Both noise sources can be most substantially reduced in the bandpassed inverse Fourier transform of an experimental STM image. To this end, the band is selected to include only the fundamental and harmonic peaks. It is important that such procedure preserves the whole domain wall structure (see Suppl. Fig. 17a,b for comparison). Another approach is to build cross-correlated image using the simulated David star shape and template matching algorithm. We have checked that different preprocessing methods result only in quantitative differences of several pixels in peak position, whereas qualitative behavior is the same. Below we use the maximum filter based on the dilation algorithm\textsuperscript{13} applied to bandpassed inverse FT image (Suppl. Fig. 17c).

To map the misfit vectors of David stars in the H state with respect to their original positions in the C state, we have to know the latter. Here we note, that C order is locally present in the single domain. Therefore, we create the simulated C lattice which exactly coincides with that in the largest domain we can find in the experimental image (Suppl. Fig. 17d). In this way two arrays containing H and C state centers are obtained. Misfit vectors $\vec{D}$ are then calculated by finding the nearest C neighbor for each H state David star and measuring length and angle of the shift between them (Suppl. Fig. 17d,e). Angle is then corrected by the difference between atomic axis and experimental $x$ axis. Each H state David star is then given two discrete indices: one shows the shift on $(0, 1, 1+1)$ atomic scale and another — the angle on the 12-category scale (see Suppl. Fig. 17f,g). To assign the shift index we assume that detected position can have the error up to ±2 pixels. In the next step angle index is assigned based on angle and the known shift index. Here we assume that
angle can have error up to ±30 degrees.

Supplementary Figure 17: Mapping misfit vectors $\vec{D}$ in the H state: a, zoom of the original image of the H state; b, zoom of the bandpassed inverse Fourier transform of the original image (a) with: c, David stars marked with the red dots; d, overlayed David star centers of the simulated C lattice (black dots); e, overlayed displacement vector field $\vec{D}$; f, overlayed discrete shift index – the length of $\vec{D}$ on (0,1,1 + 1) atomic scale; g, overlayed interpolated angle index – the direction of $\vec{D}$;
Supplementary Figure 18: Inner structure of the $X$ and $K$ defects: a, b, Schematic drawing of David star positions on a Ta lattice in (a) the $X$ crossing and (b) the $K$ kink (see main text). Black arrows indicate the misfit vectors. In the case of the $X$ crossing their sum along the closed contour is nonzero. In the case of the $K$ kink there is no change in the misfit vector, rather the domain wall changes type (number of atoms shared by David stars touching each other). However, the edge David star (yellow background) is sharing atoms with three neighbors, rather than two, as every other ones. c, Twelve possible misfit vectors $\mathcal{D}$ connecting $0$ and $1-12$ atomic sites of a David star. The thirteenth is the identity operation $\vec{D} = 0$.

Supplementary note 4: Correction of STM images.

STM fine X-Y piezoscanners were calibrated at low temperatures within ten percents before measurements. Later each picture was corrected with FFT peaks to match the triangular lattice of stars of David, i.e. using 2 fundamental CDW vectors. Correction for scanner calibration, drift and X-Y crosstalk was done either by calculating the affine transform between the observed and ideal lattice. In all the cases $1.174\text{nm}$ was taken for CDW period, but this choice affects neither ratio between CDW and atomic periods nor the angles. The ratios were checked for consistency with positions of atomic peaks in FT whenever possible. The ratios were checked for all – H, C, NC – states.
Supplementary Figure 19: Full images of the optically switched hidden state used for analysis: a – large-scale and b – atomic resolution. For clarity, only parts of these images are show in the main text in Fig. 2e,f. Fourier transform is done on the full images.

* Electronic address: yaroslav.gerasimenko@ijs.si
† Electronic address: dragan.mihailovic@ijs.si

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