

Kinetic pathways to hidden quantum states via laser-engineered topological defects

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Kinetic pathways to hidden quantum states via laser-engineered topological defects

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The deterministic control of quantum phases via ultrafast light-matter interaction lies at the frontier of modern physics [1, 2]. While light can trigger hidden phases inaccessible through equilibrium pathways, these photo-induced metastable states are typically transient or highly volatile, limiting their practical utility [2–5]. Here, we demonstrate a hybrid optical-thermal strategy that utilizes photo-induced topological defects as kinetic gateways to unlock a robust, hidden charge density wave (CDW) phase in the van der Waals antiferroelectric (AFE) In_2Se_3 . Using complementary ultrafast electron diffraction and microscopy, we show that an initial optical quench induces a metastable state characterized by long-lived topological defects via the Kibble-Zurek mechanism [6, 7]; upon thermal cooling, these defects act as heterogeneous nucleation seeds, lowering the kinetic barrier for the system to evolve thermodynamically into a hidden CDW state. Remarkably, this phase transition is fully reversible: a single laser pulse resets the system to a defect-engineered intermediate phase, preserving the topological landscape for subsequent switching. Our results establish a paradigm for using photo-induced topological defects to imprint kinetic pathways, providing a blueprint for the discovery of hidden quantum landscapes and the development of high-speed, non-volatile memory technologies.

Thermodynamic ground states, which possess the low-

est free energy, represent the most stable configuration of nature under equilibrium conditions. Navigating the complex free-energy landscapes of quantum materials to achieve desired quantum phases, through external stimuli such as chemical doping, pressure, or strain, poses a central challenge in modern condensed matter physics. Beyond these conventional tuning knobs, ultrafast optical excitation has opened a powerful alternative route, revealing a vast hidden topography of metastable phases that are entirely inaccessible via conventional thermal pathways [8–12]. These light-induced states, ranging from transient superconductivity and magnetism to Floquet-engineered topological phases, hold immense scientific and technological promise [13–17]. However, such exotic metastable phases are typically unstable, relaxing back to equilibrium states within picoseconds or nanoseconds [10–23]. Harnessing these metastable states as functional intermediates to achieve a long-lived, robust hidden phase remains an elusive goal, representing a key bottleneck in quantum phase engineering [8, 9, 24–26].

Topological defects-localized imperfections that arise from spontaneous symmetry breaking during phase transitions-offer a promising avenue to address this challenge [6, 7, 27]. These structural motifs are ubiquitous in nature, appearing as vortices in superfluids and superconductors, dislocations and disclinations in liquid crystals, skyrmions in magnetic materials and even as cosmic strings hypothesized in the early universe [6, 7, 28–31]. Unlike ordinary excitations, their stability is protected by topology, rendering them robust against continuous deformations and capable of persisting for extended durations. A fundamental question therefore

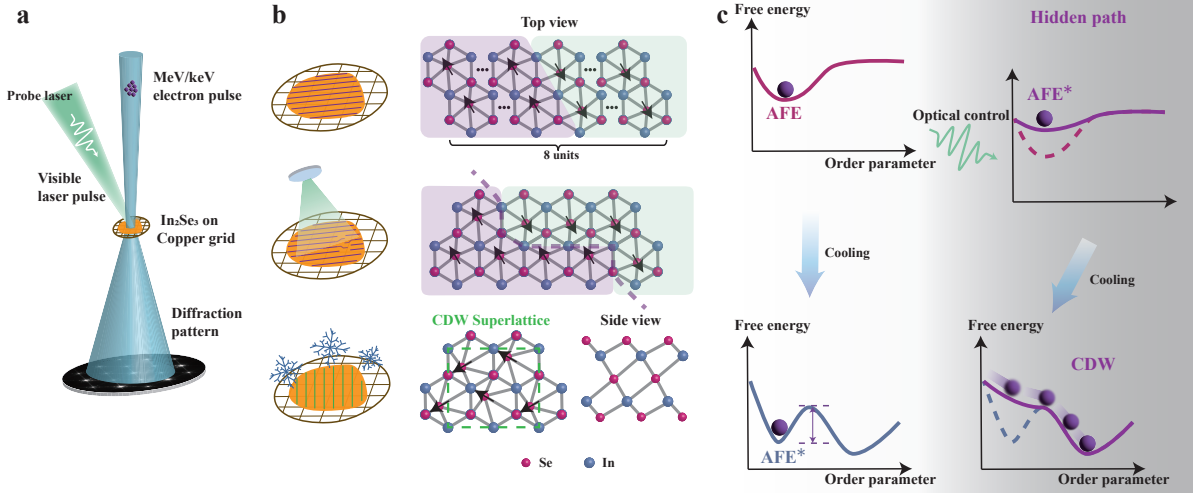


FIG. 1: (a) Schematic of the UEM and UED. Our single crystal samples were all exfoliated down to thin film with 50 nm thickness and mounted on copper grids. UEM and UED operate in transmission geometry, allowing us to map (HK0) plane of the reciprocal lattice with (HKL) denoting the Miller indices. The energy of the pump laser is above band gap (~ 2 eV). (b) Sketch of the hybrid optical-thermal strategy and corresponding structures. Top: AFE state with an 8-unit periodicity. Middle: laser-engineered intermediate AFE* state characterized by topological defects (e.g., domain walls or phase slips, where stripes are misaligned). Bottom: hidden CDW state obtained by thermodynamically cooling the AFE* state. The green box shows the local unit of the $2 \times \sqrt{3}$ CDW. (c) Free energy landscape of the material for direct cooling (left) and light-induced hidden path (right). Left panel shows the AFE potential well becomes sharper and a CDW potential well develops as the temperature decreases, but the material is trapped in the deep AFE potential well. Right panel shows that the laser lowers the energy barrier through generating topological defects, enabling the CDW formation upon further cooling.

arises: can we engineer these long-lived topological defects to sculpt the energy landscape, and thereby deterministically guide the system towards an otherwise inaccessible hidden quantum state [32, 33]?

Inspired by the Kibble-Zurek mechanism, which originally described the formation of cosmic strings during the rapid cooling of the early universe, we propose and demonstrate a hybrid optical-thermal protocol (optical quench - thermal cooling) for robust quantum phase control. Using complementary ultrafast electron microscopy (UEM) and ultrafast electron diffraction (UED) (Fig. 1a), we directly visualize this protocol in the van der Waals material $2\text{H-In}_2\text{Se}_3$, revealing a robust hidden phase. An ultrafast optical quench disrupts the initial antiferroelectric (AFE) phase and generates a high density of topological defects via the Kibble-Zurek mechanism. These defects act as persistent nucleation sites within a light-induced metastable AFE* state, effectively lowering the kinetic barrier as verified by density functional theory (DFT) calculations. Upon controlled thermodynamic cooling, the system evolves through these topological gateways, allowing the $2 \times \sqrt{3}$ charge density wave (CDW) order to crystallize (Fig. 1b). Remarkably, this hidden state is not only stable over a broad temperature range but also fully reversible, with a single laser pulse resetting the system to AFE* phase that preserves the topological landscape for subsequent switching. We also

find that the hybrid protocol is strictly path-dependent, a hallmark of the added non-commutative control dimension in our approach.

The van der Waals chalcogenide $2\text{H-In}_2\text{Se}_3$ serves as a prototypical platform for exploring phase competition and realizing phase switching due to its rich polymorphism ($\alpha, \beta, \beta', \gamma$) [34–42]. Among these, the β' phase is of particular interest for quantum phase engineering, as it hosts a variety of ferroic order, including antiferroelectricity and ferroelasticity [43–45]. At high temperature (> 525 K), the material exists in the paraelectric β phase (space group $P6_3mc$) with no distortions. Upon cooling below 525 K, it undergoes a symmetry breaking transition into the β' phase, characterized by a nanostrapped superstructure. This superstructure arises from antiparallel displacements of Se atoms between neighboring stripes, establishing an AFE order [43–45]. The nanostrapped can align along three equivalent crystallographic directions, forming three AFE domains. Although theories suggest a CDW phase with lower energy, the system becomes trapped in the AFE phase upon further cooling because of the deep potential well, unless external perturbations are applied [39–41, 46]. In the presence of an external stimulus, it is possible to alter the free energy and realize a new state of matter as shown in Fig. 1c.

We first established the equilibrium behavior of the system to define the conventional phase landscape. At

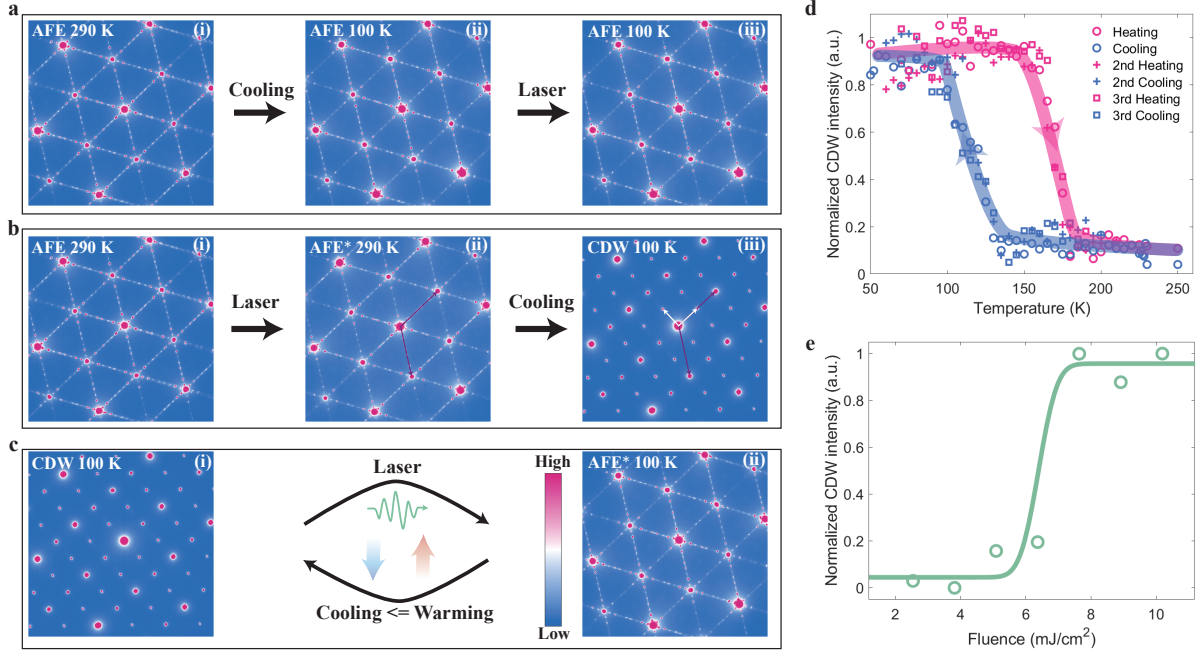


FIG. 2: (a) Path a (reversed operation sequence): (i) AFE state at 290 K, (ii) cooling down to 100 K, (iii) pumping at 100 K with AFE order remaining (it could be AFE* with higher fluence). (b) Path b (hybrid optical-thermal protocol): (i) AFE state at 290 K, (ii) laser excitation at room temperature, (iii) cooling to 100 K with the emergence of CDW. (c) Reversibility: (i) a single laser pulse converts CDW at 100 K to AFE* state, while (ii) thermodynamic evolution (warming and cooling) converts the AFE* back to the CDW state. (d) Temperature dependence of CDW intensity. The CDW is robust and persists for multiple heating-cooling cycles. Red and blue curves represent heating and cooling paths, respectively. (e) Fluence dependence showing the threshold for CDW generation.

room temperature, the diffraction patterns show intense Bragg peaks respecting the three-fold symmetry of the crystal lattice. Between these, we observed weak satellite peaks, which are the signature of the AFE nanostriped superstructure, reflecting the associated periodicity of the Se antiparallel displacement (Fig. 2a(i)). While the samples contain a mosaic of all three domain orientations, the high spatial resolution of UEM allows us to isolate and characterize single-domain regions (Supplementary Note 1). Upon cooling the pristine AFE phase from room temperature to 100 K, we observe a gradual increase in the intensity of both Bragg and superlattice peaks, consistent with the thermal suppression of Debye-Waller factors and enhanced structural correlation. Crucially, the symmetry and positions of these diffraction features remain invariant throughout the cooling process (Fig. 2a(ii)). This confirms that, under equilibrium conditions, the material remains trapped in the AFE phase, with no further symmetry breaking or transition to a CDW state. This observation establishes the AFE phase as a deep, kinetically stable potential well that shields the system from reaching the hidden ground state via purely thermal pathways (see left panel of Fig. 1c).

Remarkably, the hybrid optical-thermal strategy unlocks an exotic phase. First, we pump the material

at room temperature with a single ultrafast laser pulse (520 nm, 300 fs). This pulse drives the system into a metastable intermediate AFE* state (Fig. 2b(ii)). While the diffraction pattern of AFE* qualitatively resembles the pristine phase, it is characterized by a significant broadening of superlattice peak linewidths, indicative of reduced correlation lengths and increased defect density (more details in SI and will be illustrated further in Fig. 3b). Next, we cool this AFE* state to base temperature at a controlled rate (~ 1 K/min; varying the cooling rate slightly does not affect the results). When the temperature drops below 150 K, a striking transformation occurs: the AFE superlattice peaks diminish and an orthogonal superstructure emerges. By 100 K, the AFE order is completely replaced by this new superstructure as shown in Fig. 2b(iii). Crucially, reversing the operation sequence, i.e. cooling the pristine AFE phase first followed by laser excitations, fails to induce the hidden state as illustrated in Fig. 2a.

The light-induced hidden phase corresponds to a $2 \times \sqrt{3}$ CDW. This induced CDW phase exhibits remarkable stability: it persists through multiple temperature cycles between 100 K and 200 K, with clear thermal hysteresis between warming and cooling paths (Fig. 2d). This broad hysteresis loop suggests that the transition from

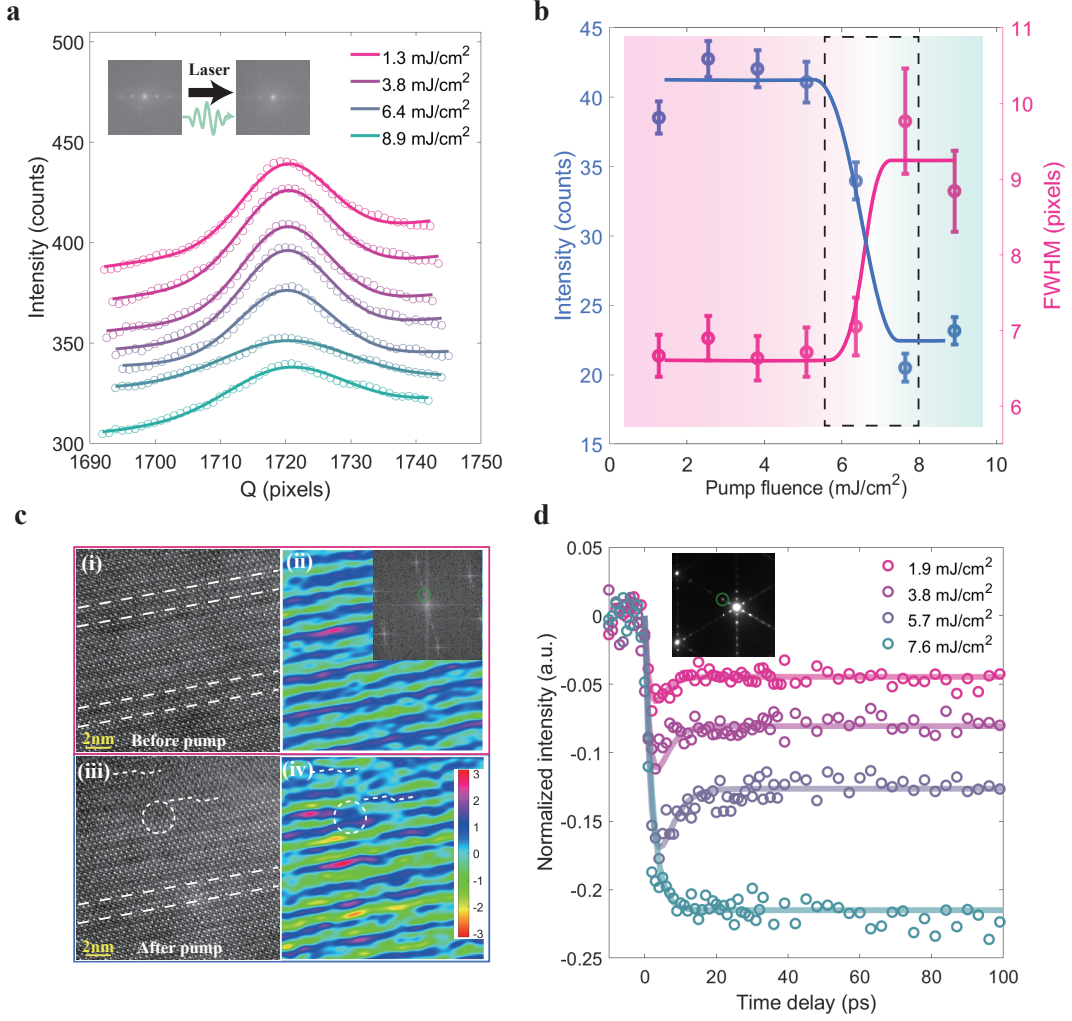


FIG. 3: (a) Intensity of AFE superlattice peaks as function of momentum along (0 -1 0) at various pump fluences. Each trace is vertically offset for clarity. The data is fitted with Gaussian functions to extract the intensity and linewidth. (b) Intensity and full width at half maximum (FWHM) as a function of pump fluence. The error bars represent standard deviation. Linewidths exhibit notable broadening above ~ 7 mJ/cm². (c) Spherical aberration-corrected electron microscopy images. Before pumping (i), the AFE pattern is straight, while after pumping (iii) some AFE stripes become misaligned. Corresponding phase distributions of the stripes are shown in (ii) and (iv), respectively. White thick dashed lines are guides to the eye, indicating the well-ordered AFE stripes before pumping. Topological defects such as phase slips and domain walls are labeled by a circle and thin dashed lines, respectively. (d) Relaxation dynamics of the AFE superlattice peak measured at room temperature for various pump fluences.

the AFE* state to the CDW is first-order and likely involves a nucleation process. This explains why reversing the operation sequence does not yield CDW: the system lacks the thermal energy required for nucleation and growth of the CDW order. Interestingly, the system exhibits deterministic reversibility (Fig. 2c). The CDW state can be erased by a single pulse laser, resetting the system to the AFE* metastable state. Warming the sample to 200 K and re-cooling it to 100 K restores the CDW phase. The repeatability of this protocol has been confirmed across multiple samples. Together, these results underscore the complementary roles of laser excitation as

a kinetic trigger to the AFE* state and thermal history as a thermodynamic path to the CDW phase, enabling precise control over quantum phases.

The laser fluence study indicates that AFE* formation requires the laser pulse to exceed a critical threshold of approximately 6.5 mJ/cm² (Fig. 2e). This threshold suggests that the optical pulse needs to overcome an energy barrier to drive the system into this metastable state AFE*. This energy matches that required to reach the 550 K paraelectric phase, suggesting that the AFE* formation necessitates a transient restoration of the high symmetry parent state (Supplementary Note 2). In-

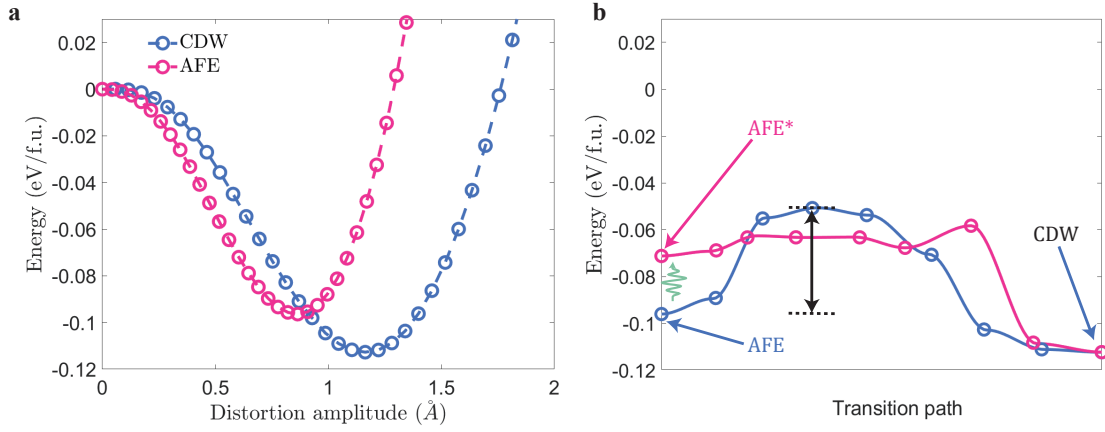


FIG. 4: (a) Total energy as a function of distortion amplitude for the AFE and CDW phases. The high temperature paraelectric phase with no distortion is taken as the zero-energy reference. The depths of the AFE and CDW energy wells are 96 and 113 meV per formula unit (f.u.), respectively, indicating that the CDW phase is energetically more favorable than the AFE phase. However, the AFE energy decreases more rapidly with increasing distortion, suggesting that the AFE phase may form more readily than the CDW phase. (b) Energy profile along the transition pathway connecting the 1×8 AFE phase, light-induced AFE* phase (AFE with misaligned stripe domains), and the $2 \times \sqrt{3}$ CDW phase. The energy barrier between the AFE and CDW phases is approximately 45 meV/f.u. The introduction of topological defects (such as phase slips or domain walls) by laser pulses raises the energy of the system and consequently reduces the transition barrier to 13 meV/f.u..

triguingly, while the room-temperature diffraction patterns of the pristine AFE and photo-induced AFE* states are nearly indistinguishable, their low temperature evolutions are fundamentally different. In the absence of optical excitation, the system remains trapped in the AFE phase, whereas AFE* metastable state becomes a deterministic gateway to the CDW ground state upon cooling. This bifurcation in the phase trajectory confirms that the laser does not merely produce transient heating, but instead creates a distinct metastable precursor. In the following, we identify that this AFE* state is a defect-engineered template that reshapes the material's energy landscape.

To elucidate the microscopic nature of the AFE* state, we employed a multimodal approach combining UEM, UED, spherical aberration-corrected electron microscopy, and Raman spectroscopy. A key signature of the AFE* precursor is the significant broadening of the AFE superlattice peaks in momentum space once the laser fluence exceeds the critical threshold (Fig. 3a and Fig. 3b). Such broadening indicates a drastic reduction in the structural correlation length ξ after optical excitation, a feature typically associated with topological defect generation [21, 47, 48]. While point defects of crystal structures can cause modest broadening of superlattice peaks, this possibility is ruled out because the Bragg peaks remain resolution-limited after pumping (Supplementary Note 3). Moreover, the pristine state can only be restored by annealing above 550 K to the paraelectric phase, consistent with topologically protected defects that require a full reset of the order parameter (Supplementary Note 4).

Direct evidence for these light-induced topological defects is provided by spherical aberration-corrected electron microscopy. After pumping, the neat AFE pattern fragments into a dense mosaic of nanodomains. We observe the emergence of disordered regions at the junctions of these nanodomains, where the AFE order parameter is locally suppressed (Supplementary Note 5). Crucially, some regions exhibit misaligned striped AFE patterns as shown in Fig. 3c. The spatial phase distribution reveals pronounced phase offsets of the stripes after photoexcitation compared with the uniform phase map of the pristine sample (Supplementary Note 6). These regions represent phase slips or domain walls, which are typical topological defects that provide heterogeneous nucleation sites necessary for the subsequent CDW transition. Raman scattering further supports this defect-only scenario: nearly identical phonon modes between the pristine and AFE* states confirm that the lattice symmetry is unchanged, instead only the existing displacement pattern is reconfigured (Supplementary Note 7).

Consistent with this picture, time-resolved UEM measurements of the metastable state show the relaxation dynamics of the superlattice that corresponds to the formation and evolution of topological defects (Fig. 3d). At low pump fluence, light only slightly perturbs the AFE order, leading to lattice displacements that relax back to equilibrium positions rapidly (a few picoseconds). As the pump fluence increases, the relaxation time of the superlattice peaks becomes progressively longer, and eventually diverges at a critical fluence of 7.6 mJ/cm^2 (Supplementary Note 8). This fluence is in accordance with the threshold for linewidth broadening, indicating that

a light-induced metastable state characterized by topological defects forms above this threshold [21, 47]. In addition, high-precision UED experiments, which show a much slower relaxation time for the superlattice compared to Bragg peaks, further reinforce this scenario (Supplementary Note 9).

The physical basis for the creation of these defects lies in the Kibble-Zurek mechanism. The ultrafast laser pulse acts as a non-adiabatic quench (optical quench), effectively melting the AFE order parameter and transiently returning the system to the high-symmetry paraelectric phase (~ 100 fs). The subsequent structural recovery (~ 1 ps) occurs much faster than the intrinsic timescale for establishing long-range order. According to Kibble-Zurek mechanism, as the system is quenched through the phase transition, independent regions of the material choose the phase of their order parameter locally. Because the speed of information propagation is finite, these regions cannot coordinate, leading to the spontaneous formation of topological defects (e.g. domain walls and phase slips) at mismatched boundaries. In our hybrid optical-thermal strategy, these photo-induced defects are not merely sources of disorder; they serve as kinetic gateways that locally lower the activation barrier for the CDW phase. This allows the system to bypass the kinetic trap of the AFE phase during cooling, providing a deterministic pathway to a region of the energy landscape that is otherwise inaccessible.

The above picture is supported by our DFT calculations. As shown in Fig. 4a, the total energy as a function of the order parameter (distortion amplitude) indicates that the AFE and CDW phases are lower in energy than the paraelectric phase by approximately 96 and 113 meV/f.u. (formula unit), respectively. Although the CDW phase possesses lower energy, the AFE phase emerges first upon cooling from the paraelectric phase during sample synthesis. This can be explained by a faster dynamical tendency for AFE formation, as evidenced by the steeper energy decrease with increasing order parameter. The transition energy barrier from AFE to CDW is of 45 meV/f.u., which traps the system in the AFE state upon further cooling, in agreement with our experiments. However, the generation of topological defects by laser excitation increases the energy of the system and efficiently reduces the transition barrier. For example, the phase slips formed by the misalignment of AFE stripes raises the energy of the system by 25 meV/f.u and lowers the barrier to only 13 meV/f.u. as shown in Fig. 4b (Supplementary Note 10). Such a reduced barrier can be overcome easily by thermal fluctuations, enabling the CDW formation upon subsequent cooling. Given that domain walls generally carry a positive formation energy, we expect our mechanism to apply broadly to other types of topological defects as well, albeit with quantitative variations.

Our findings culminate in a comprehensive model of

the $2\text{H-In}_2\text{Se}_3$ energy landscape (Fig. 1c). At equilibrium, the AFE phase is a deep and kinetically stable potential well. Although our DFT calculations confirm that the $2 \times \sqrt{3}$ CDW phase is the global energy minimum (the true ground state), it is shielded by a high activation barrier that prevents its formation during conventional cooling. The ultrafast laser pulse serves as a non-adiabatic optical quench that bypasses this barrier. By generating a high density of topological defects via the Kibble-Zurek mechanism, the laser sculpts the energy landscape, creating a metastable AFE* state. These defects act as heterogeneous nucleation seeds that locally lower the activation energy, providing a kinetic stepping stone toward the hidden ground state. During the subsequent cooling process, the system is guided through these topological gateways, allowing the CDW order to crystallize. This hybrid optical-thermal strategy reveals a fundamental hierarchy in phase control: the ultrafast laser pulse provides kinetic selection by populating the landscape with topological defects, while the controlled cooling provides thermodynamic selection by stabilizing the global minimum.

The concept of using topologically protected defects to sculpt the free-energy landscape introduces a new paradigm for quantum phase control. Rather than relying on sustained external stimuli such as pressure and strain, deterministic engineering via laser-generated topological defects provides a non-volatile route to access and stabilize hidden quantum states. We anticipate that this approach can be directly applied to many other ferroic or strongly correlated materials, including defect-engineered superconductivity. This shifts the focus of ultrafast materials science from the transient manipulation of existing phases to the deliberate engineering of kinetic pathways. As such, it offers a blueprint for discovering and stabilizing exotic quantum states across a wide range of correlated materials, opening a new frontier in the rational design of functional quantum matter.

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AUTHOR CONTRIBUTIONS

X.Z. and D.X. conceived and supervised the project. Z.Z. collected the UED data with assistance from Q.X., J.X., Y.L., L.C., Y.L. on the MeV-UED instrument which was constructed and maintained by T.J., P.Z. and X.Z. Z.Z. collected the UEM data with assistance from J.L. and S.S.. Z.Z. analyzed the data with important insights from C.X., D.Z., Y.L., A.K. and A.Z.. C.X. and Y.Z. prepared the sample with suitable phase. H.T. performed the DFT calculations. S.W. and S.J. performed the Raman scattering measurements. Z.Z., H.T., D.X. and X.Z. wrote the paper with critical input from A.K. and A.Z. and all other authors.

COMPETING INTERESTS

The authors declare no competing interests.

METHODS

Crystal growth and sample preparation

The 2H β' -In₂Se₃ flakes were obtained by quenching 2H α -In₂Se₃ (purchased from HQ Graphene) from 300°C to room temperature. Ultrathin 2H-In₂Se₃ flakes were mechanically exfoliated from bulk crystals using adhesive tape and transferred onto melted Crystalbond adhesive. After solidification, the adhesive was dissolved in water to obtain free-standing thin films (~ 50 nm), which were subsequently transferred onto standard 2000-mesh copper TEM grids for ultrafast electron diffraction and microscopy measurements.

Ultrafast electron microscope

Ultrafast electron microscopy (UEM) experiments were performed at the Synergetic Extreme Condition User Facility using a modified thermionic-emission transmission electron microscope (JEM-2100Plus, JEOL) equipped with multiple imaging modes. A 520 nm pump pulse was focused onto the sample with a full width at half-maximum (FWHM) spot diameter of approximately 40 μ m to photoexcite the specimen. The probe electron pulses were generated by illuminating a LaB₆ photocathode with 260 nm laser pulses with spot size ~ 50 μ m. Both the pump and probe pulses were derived from a 1040-nm femtosecond laser system (Spirit1040-4, Spectra-Physics) with a pulse duration of 300 fs (FWHM). Frequency conversion was achieved using BBO crystals. The temporal delay between the pump and probe pulses was controlled by a motorized optical delay stage (DDS600, Thorlabs). Ultrafast measurements of relaxation dynamics were performed at a repetition rate of 50 kHz. A single shot mode was chosen for generation of topological defects and switching of CDW phase. Temperature-dependent measurements were carried out using a double-tilt liquid-nitrogen cooling holder (Model 636, Gatan). Most diffraction data were acquired using UEM, benefiting from its high spatial resolution to target single domains; this includes the data presented in Fig. 2a-b-c and Fig. 3a-b-d.

Ultrafast electron diffraction

Ultrafast electron diffraction (UED) experiments were performed at Shanghai Jiao Tong University using a mega-electron-volt UED (MeV-UED) instrument. A Ti:sapphire regenerative amplifier system (Vitara and Legend Elite Duo HE, Coherent Inc.) delivered 30 fs laser pulses centered at 800 nm (1.55 eV) with a repetition rate of 400 Hz to generate both the UV pump laser pulse and the electron probe pulse. For electron probe generation, the laser pulses were frequency-tripled using nonlinear optical crystals and directed onto a photocathode to generate electron beams with a charge of approximately 10 fC per pulse. The electron beams were generated in a 2.33-cell photocathode RF gun (2856 MHz), where the intense RF field rapidly accelerates the electron beam to relativistic velocity ($v \approx 0.99 c$) with a kinetic energy of about 3 MeV. A double-bend achromat lens, consisting of two dipole magnets and three quadrupole magnets, is further used to compress the electron pulse width to approximately 30 fs (FWHM) [49]. The temporal delay between the optical pump and electron probe pulses was controlled using a motorized optical delay stage. The 400 nm pump pulse was obtained using a BBO crystal. Diffraction patterns were obtained by imaging a P43 phosphor screen onto an electron-multiplying charge-coupled device camera (EMCCD). Sample temperature was controlled using a closed-cycle helium cryostat. The high temporal resolution of UED enabled accurate dynamic studies. Furthermore, the lab-built setup allowed comprehensive fluence and temperature dependent measurements (Fig. 2d-e).

Spherical aberration-corrected electron microscopy

Spherical aberration-corrected electron microscopy experiments were performed at Shanghai Jiao Tong University using a double aberration-corrected Spectra 300 transmission electron microscope manufactured by Thermo Fisher Scientific. Atomic-resolution high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) imaging was carried out under aberration-corrected conditions to resolve the atomic structure of the samples with sub-angstrom spatial resolution. The microscope was operated at an acceleration voltage of 300 kV. The convergence semi-angle and HAADF detector collection angles were optimized to enhance atomic-number-sensitive (Z-contrast) imaging. Fast Fourier transform (FFT) and inverse FFT analyses of the atomic-resolution images were performed using standard Digital Micrograph routines and custom image-processing scripts.

Raman scattering

Non-polarized Raman spectra were acquired in reflection geometry using a home-built confocal optical microscope integrated with a closed-cycle helium cryostat (attoDRY 2100). A 532 nm solid-state laser served as the excitation source. The laser beam was focused along the c-axis onto the In₂Se₃ samples through a low-temperature microscope objective (Attocube, 0.8 numerical aperture). The reflected radiation was collected with the same objective and directed into a spectrometer equipped with a 1200 grooves/mm diffraction grating and a Si CCD (Princeton Instruments PIXIS: 100B eXcelon). A combination of one reflective Bragg grating and three Bragg notch filters effectively removed most of the laser sidebands, enabling measurements of Raman shifts down to 20 cm^{-1} . The laser power was kept below 500 μ W to avoid significant laser heating of the samples.

Density function theory

All calculations for In_2Se_3 were performed using the Vienna *ab-initio* Simulation Package. The electron-electron exchange-correlation interaction was treated within the generalized gradient approximation. The plane-wave basis set was expanded with an energy cutoff of 300 eV. Structural relaxations were carried out using the conjugate gradient algorithm until the residual forces on each atom were smaller than 1 meV/Å, and the total energy was converged to within 10^{-5} eV. The Brillouin zone was sampled using a Γ -centered k-point mesh with an in-plane reciprocal-space resolution of 0.15 Å⁻¹ or finer. Phonon spectra were calculated using the finite-displacement method as implemented in Phonopy. Perturbed structures were constructed in a 5×5 supercell with a displacement amplitude of 0.01 Å. For the self-consistent calculations of these supercells, the total energy convergence criterion was set to 10^{-5} eV, and a Γ -centered 2×2 k-point mesh was employed.

The transition pathway between the 1×8 AFE and 2×√3 CDW phases was investigated using the variable-cell nudged elastic (NEB) band method. To construct a physically meaningful initial path, the 1×2 AFE and FE phases were chosen as intermediate reference configurations, as their distortion patterns share key structural features with both the 1×8 AFE and CDW phases. Starting from the end-point structures (1×8 AFE and CDW), three intermediate images were generated by linear interpolation between the 1×8 AFE phase and the 1×2 AFE (or FE) phase, and another three images between the 1×2 AFE (or FE) phase and the CDW phase. This procedure yields a total of seven intermediate images (including the 1×2 AFE or FE configuration) along the reaction pathway. The NEB calculations were then performed based on this initial path to obtain the minimum-energy transition pathway and the associated energy barriers.

DATA AVAILABILITY

The data that supports the findings of this study are present in the paper and/or in the supplementary information, and are deposited in the Zenodo repository. Additional data related to the paper is available from the corresponding authors upon reasonable request.

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