Electronic Supplementary Information

Thickness-driven phase selection for epitaxial helical tellurium on a van der Waals superconductor

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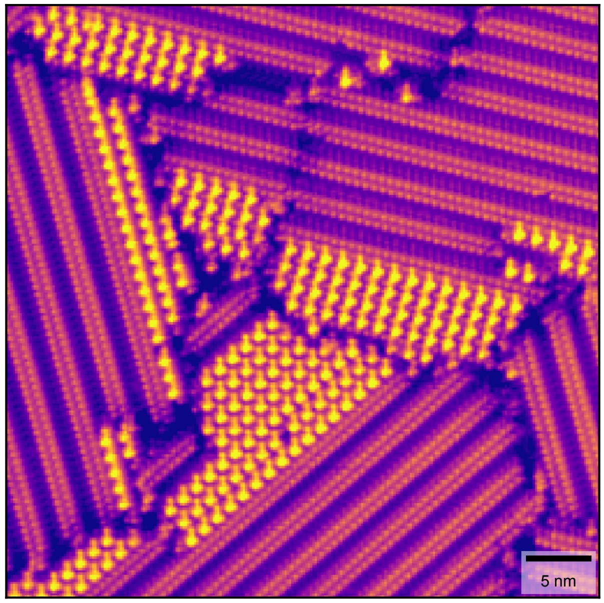

Fig. S1 Rotational domains of the 1L stripe phase. High-resolution STM topography of 1L Te on $NbSe_2$. The film exhibits extended domains in three equivalent orientations separated by 120°, aligned with the high-symmetry lattice directions of the hexagonal substrate (V_{bias} = -0.8 V, I_{set} = 100 pA).

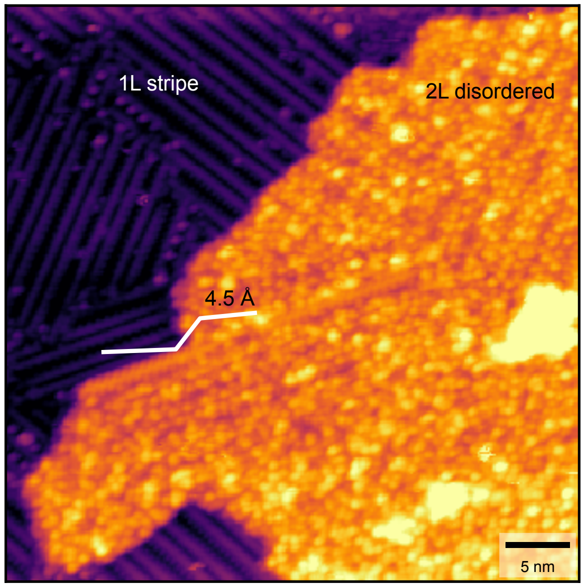

Fig. S2 Disordered 2L islands. High-resolution STM image of a representative 2L island situated atop the striped 1L film. In contrast to the underlying 1L template, the surface of the 2L island lacks long-range periodic order (V_{bias} = 1.5 V, I_{set} = 10 pA).

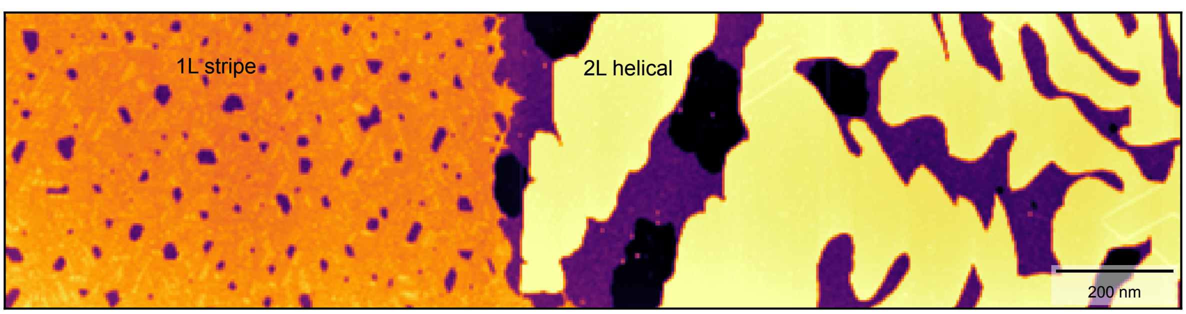

Fig. S3 Persistence of the striped 1L phase in regions free of 2L islands. Large-area STM topography from a different region on the same sample shown in Fig. 2b of the main text. The 1L striped regions that remain untransformed are absent of 2L islands, supporting the conclusion that 2L precursors are required to nucleate the phase transformation (V_{bias} = 1.5 V, I_{set} = 10 pA).

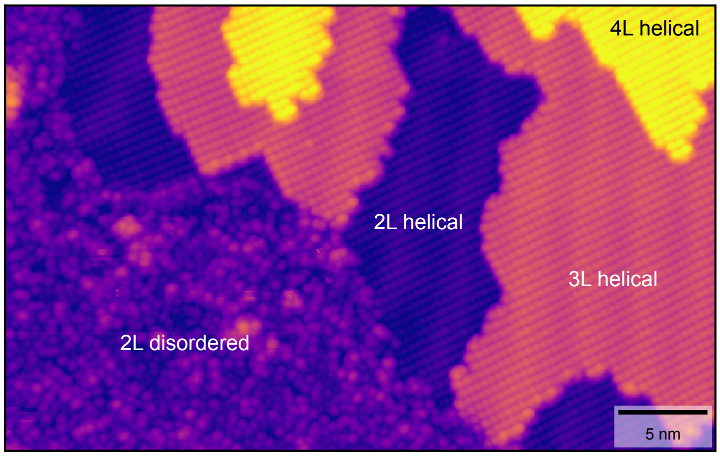

Fig. S4 "Helical-core,disordered-shell" 2L morphology during transformation. Zoomed-in STM image of a partially transformed 2L island. The crystalline helical Te core, identified by its rectangular surface lattice, is surrounded by an atomically disordered shell. This captures the propagation of the crystallization front during the phase transition (V_{bias} = -1.0 V, I_{set} = 20 pA).

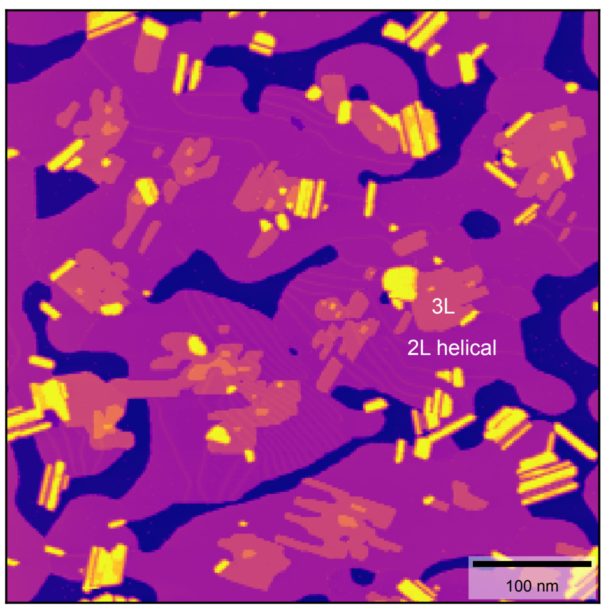

Fig. S5 Morphology evolution at high coverage. Large-area STM image of the ≈1.5 ML coverage sample (corresponding to Fig. 2d in the main text) after holding at RT for 4 hours. The film has fully transformed into helical islands, which are decorated by higher, rod-like structures (V_{bias} = -1.5 V, I_{set} = 10 pA).

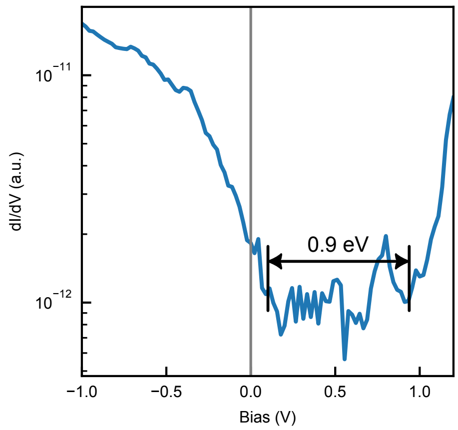

Fig. S6 Typical dI/dV spectrum acquired on a 2L Te island at 77 K. The spectrum shows a band gap of ≈ 0.9 eV, with the Fermi level residing in the valence band.

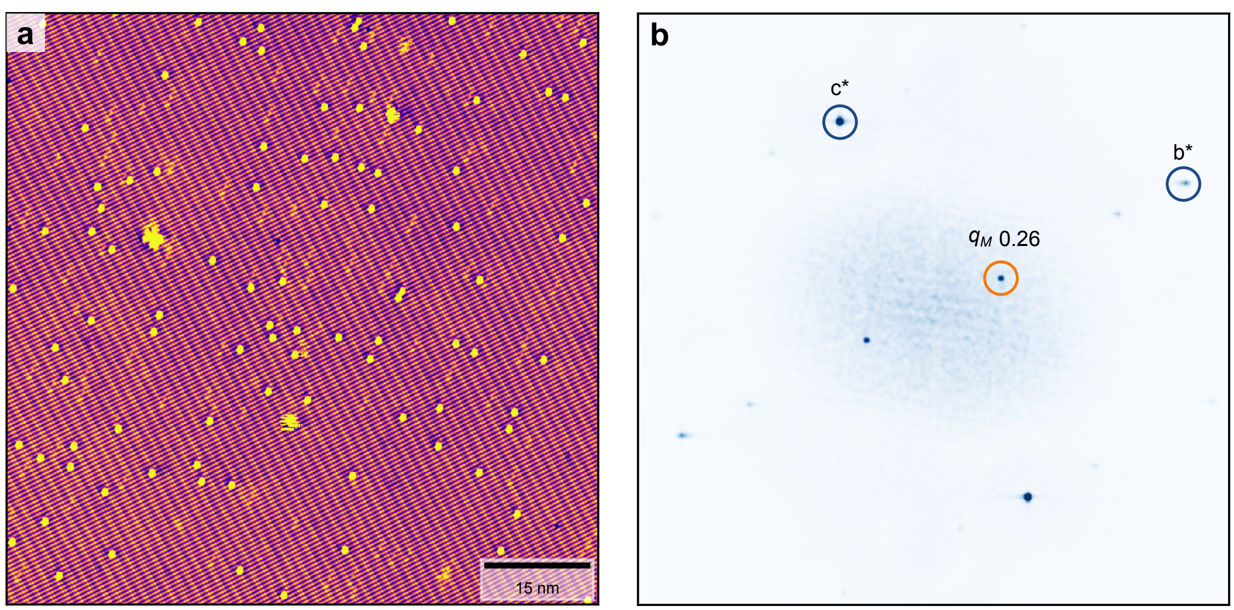

Fig. S7 Large-scale STM characterization of the moiré pattern. a, Large-area (83 nm × 83 nm) STM image showing the continuous distribution of the one-dimensional moiré modulation (V_{bias} = -1 V, I_{set} = 20 pA). b, FFT of a, showing sharp superlattice spots consistent with the main text (Fig. 4).