

Supporting Information

Complete Strain Mapping of Nanosheets of Tantalum Disulfide

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Bulk sample synthesis

High-quality single crystals of 1T-TaS₂ were grown by the chemical vapor transport (CVT) method with iodine as a transport agent. The high-purity Ta (3.5 N) and S (3.5 N) were mixed in chemical stoichiometry and heated at 850°C for 4 days in an evacuated quartz tube. The harvested TaS₂ powders and iodine (density: 5 mg/cm³) were then sealed in another quartz tube and heated for 2 weeks in a two-zone furnace, in which the source zone and growth zone were fixed at 900°C and 800°C, respectively. The tubes were rapidly quenched in cold water to ensure retaining of the 1T phase.

Nano flake preparation

Thin 1T-TaS₂ flakes were micromechanically exfoliated onto the scotch tape. The 10 μm-thick silicon substrates were carefully inverted and placed atop large areas of the crystal. Subsequently, the substrate was slowly peeled off from its longer edge and examined under an optical microscope to identify suitable patches of crystal that had adhered to the substrate, taking into account the sample size, shape, and uniformity. We measured the thickness of one 1T-TaS₂ flake with the Park NX20-SPM Atomic Force Microscope using the non-contact mode, and calibrated it against the 80 nm-high Pt fiducial mark on the same silicon wafer. The thickness of all other flakes was determined using the Ta L-edge fluorescence intensity collected from the Vortex detector during the synchrotron experiment.

X-ray nanodiffraction (nanoXRD)

Nanodiffraction measurements were performed using nano-Mii (Nanoscale Multimodal Imaging Instrument) at the Hard X-ray Nanoprobe Beamline of the NSLS-II.^{1, 2} The 12 keV coherent incident X-rays are focused using a Fresnel zone plate with an outer ring width of 40 nm and outer diameter of 240 μm. This gives an incident X-ray convergence of 2.58 mrad, or 0.15°. The X-ray beam focused by the Fresnel zone plate (FZP) was prepared using a central beam stop and an order sorting aperture (OSA). Under the condition of this experiment, the focused beam size was about 80 nm. The photon flux is estimated around 2×10⁹ photons/μm²/sec at 12 keV. A pixel-array detector (Merlin, 55 μm/pixel, 512×512 pixels), positioned 0.5 m downstream of the sample, was used to collect the diffracted x-rays. A schematic drawing of the experimental setup as well as the diffraction geometry (including the θ , δ and γ angles) is displayed in Fig. 1(a) of the main text.

While the exfoliated 1T-TaS₂ often has edge facets indicating high-symmetry crystalline orientations, we need to self-consistently determine the crystal orientation during the nanoXRD experiments. This is achieved using a wide-angle Dexela CMOS X-ray detector (Dexela 1512NDT), mounted behind the Be exit-beam window of nano-Mii. The detector was pre-calibrated with Si wafer so that we could convert each pixel into the δ and γ angles. By rotating (rocking) sample θ we captured both the crystal Bragg peaks and charge ordered superlattice peaks on the detector with the corresponding θ , δ and γ angles. We first filter the nanoXRD peaks by their two-theta values (i.e. d-spacings) and diffraction intensities, and then identify the crystal orientation by indexing the measured reflections using SPEC.³ Nanodiffraction measurements with higher angular resolution and better detection sensitivity were carried out after removing the Dexela detector, allowing the diffracted x-rays to be transmitted to the Merlin detector. The nanoXRD data are collected using the fly-scan method as described in ref. 4. The data collection

time per voxel is set to 0.1 seconds to prevent radiation damage. At least two consecutive fly-scans were taken at the same sample area to ensure there was no observable damage from the X-ray. Over the course of the 0.1 seconds, for a 300 nm step size, it takes 7.5 μ s for the piezopositioners to move ~300 nm. Ta L-edge fluorescence was collected using a Vortex detector positioned at 90° relative to the incident X-rays.

We collected simultaneously both the nanoXRD patterns (as those plotted on the area detector shown in Fig. 1 (a)) and the fluorescence X-rays from the sample while performing 2D imaging scans. X-ray fluorescence data were analyzed using PyXRF.⁵

Estimating Young's modulus

Following the definition in Fig. 3(j) inset of the main text, the surface stress f per unit length at the circumference of the nanobubble of radii r and height h follows

$$f \sin \theta \cdot 2\pi r = \Delta p \cdot \pi r^2$$

where R is the surface curvature, with $\sin \theta = r/R$. The strain is determined in Fig. 3 and is dominated by the off-diagonal term $\epsilon \sim 10^{-3}$. For a nano bubble with a thickness of d , Young's modulus is therefore

$$G = \frac{f}{\epsilon d} = \frac{\Delta p R}{2\epsilon d}$$

Here $\Delta p \sim 0.7$ bar, $R \sim 0.3$ mm, $d \sim 100$ nm. Thus $G \sim 100$ GPa.

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