Imaging the TaS_2 Charge Density Wave Transition in Real-Space with Electron Microscopy

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Principal Investigator(s): Jeeyoung Judy Cha
User(s): James L. Hart, Saif Siddique

Affiliation(s): Department of Materials Science and Engineering, Cornell University
Primary Source(s) of Research Funding: Gordon and Betty Moore Foundation, EPiQS Synthesis Investigator Award
Contact: jc476@cornell.edu, jlh478@cornell.edu, ms2895@cornell.edu
Website(s): cha.mse.cornell.edu
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Abstract:

We studied the charge density wave (CDW) and insulator-to-metal transition in the two-dimensional (2D) material tantalum(IV) sulfide (TaS_2). We highlight two key findings: first, basal dislocations strongly influence the transition, acting as both nucleation and pinning sites, and second, the bias-induced CDW switching is driven by Joule heating. Both findings are relevant to the design of next-generation electronics based on TaS_2.

Summary of Research:

Tantalum(IV) sulfide (TaS_2) is a prototypical 2D quantum material that hosts several CDW phases. At low temperature, the TaS_2 ground state is the commensurate (C) CDW phase, which is insulating [1]. Above 200 K, TaS_2 transitions to the nearly commensurate (NC) CDW phase, which is defined by a network of phase slips in the CDW order parameter, i.e. discommensurations [1]. The NC phase is metallic. Hence, the C to NC phase transition is accompanied by a large change in electrical resistance, and is promising for device applications. Moreover, the transition can be triggered with an applied electric field, with potential application for 2-terminal devices [2]. However, implementing nanoscale TaS_2 devices requires a detailed understanding of how the CDW phase transition occurs on nanoscale dimensions, which is currently lacking. Direct real-space imaging of the CDW transition is needed.

We developed a four-dimensional scanning transmission electron microscopy (4D-STEM) approach which allows the visualization of the CDW transition. In 4D-STEM, an electron beam is focused to a nanoscale probe, rastered across the sample surface, and a full diffraction pattern is captured at each spatial coordinate [3]. Our method provides significant advantages over prior attempts to image the CDW transition [4,5].

Figure 1 shows the resistance versus temperature for a TaS_2 device measured within the electron microscope. A clear insulator-to-metal transition is observed around ~ 200 K. For each of the datapoints in Figure 1, a full 4D-STEM dataset was collected and used to map out the CDW phase; select temperature scans are shown in Figure 2. For the C to NC CDW transition, the appropriate order parameter is the coherent domain size DNC. At 120 K in the insulating phase, the measured domain size DNC is > 100 nm, which is consistent with the C CDW phase. At 250 K, in the metallic phase, the DNC is < 10 nm, consistent with the NC CDW phase. Interestingly, at the insulator-to-metal transition (195 K), we find that the DNC map displays sharp lines and domains. Hence, the CDW transition is highly inhomogeneous.

To understand the nature of the sharp lines seen in the CDW map, we performed additional electron microscopy experiments. We find that all of the lines seen in the CDW map correspond to basal dislocations, i.e. both the burgers vector and the line vector lie in the ab-plane. A plan-view STEM image of one such dislocation is shown in Figure 3. We find that dislocations both nucleate and pin the CDW transition in TaS_2, thus, device engineering efforts should consider the role of dislocations.

Next, we investigate electric field-induced switching of TaS_2. Nearly a decade ago, several research groups showed that starting in the insulating C phase, application of a strong electric field can trigger a rapid transition to a metallic phase. The switching is fast, reversible, and energy-efficient, making this switching process appealing for 2-terminal devices [2]. However, the actual switching mechanism is still under debate.

To resolve this question, we operate a 2-terminal device within the electron microscope. In doing so, we can image the CDW during device operation, offering unprecedented insight to the switching process. Here, we

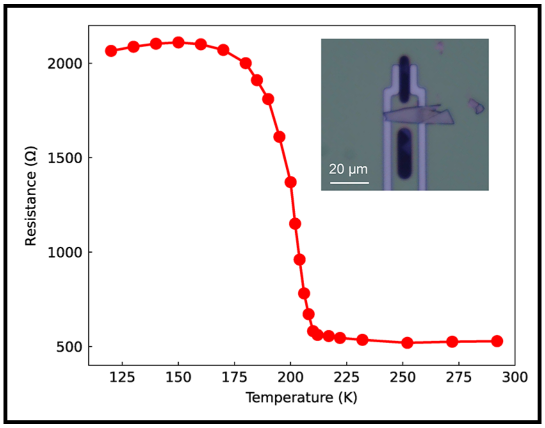

Figure 1: In situ *STEM resistance versus temperature for a TaS_2 device upon heating. Inset shows optical image of device.*

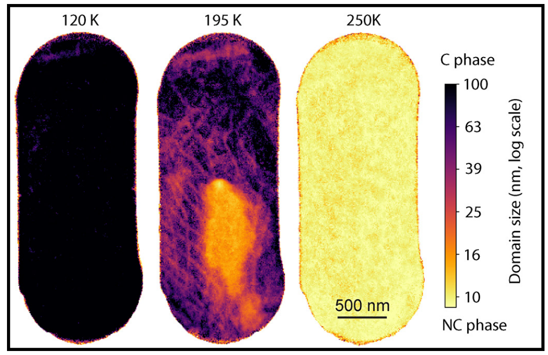

Figure 2: Select 4D-STEM maps of the CDW order parameter, captured simultaneous with the resistance data in Figure 1.

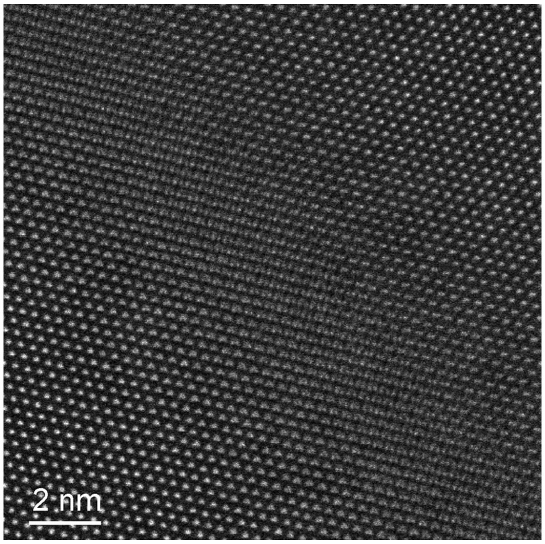

Figure 3: Atomic-resolution STEM image of a basal dislocation, imaged in the ab-plane. The dislocation line runs from the top-left to the bottom-right of the image.

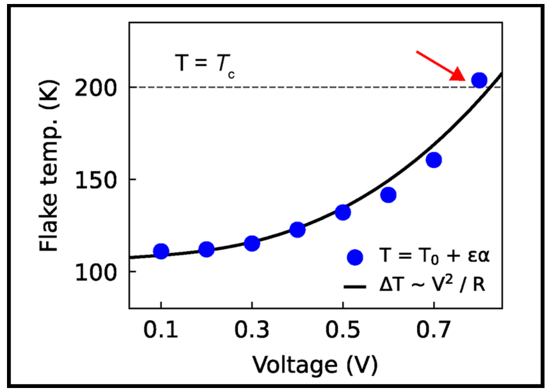

Figure 4: The measured flake temperature versus applied voltage. Resistive switching occurs when the temperature surpasses the CDW transition temperature T_c (red arrow).

highlight one key finding from these experiments, which proves that switching is driven by Joule heating. First, we apply a DC voltage to our flake, while simultaneously measuring the flake resistance and collecting diffraction data. From the diffraction data, we extract the flake strain, and using the thermal coefficient of expansion, we can then determine the flake temperature. Figure 4 shows the measured flake temperature as a function of applied voltage. For voltages from 0.1 to 0.7 V, the flake is highly resistive (> 2 kΩ). Then, at 0.8 V, the resistance drops to 400 Ω. At the resistive transition, we see that the flake temperature surpasses 200 K, which is the thermal transition point for the TaS_2 C to NC phase transition. Hence, our data shows that at the critical voltage which causes resistive switching, the Joule heating is sufficient to raise the flake temperature above the CDW transition temperature.

Conclusions:

We used in operando 4D-STEM to study the CDW behavior of the 2D quantum material TaS_2. We showed that dislocations both nucleate and pin the CDW transition, and that the bias-induced transition is driven by Joule heating. These findings are important for the optimization of next-generation TaS_2 devices.

References:

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