Tracking the TaS2 Charge Density Wave Transition with Electron Microscopy and Electric Biasing

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Primary CNF Tools Used: Nabity Lithography System, Zeiss Supra SEM, CHA Thermal Evaporator

MATERIALS

Abstract:

Our project's aim is to better understand the charge density wave (CDW) and associated metal to insulator transition (MIT) in 2-dimensional crystals of tantalum disulfide (TaS_2). We use *in situ* cryogenic scanning transmission electron microscopy (STEM) with *in situ* electric biasing to correlate changes in the CDW properties and sample resistance as a function of temperature. While this project is still on going, we have reached several milestones, including detecting the CDW and MIT with electrical resistance measurements, and developing a novel STEM strategy to track the CDW transition in real space. This project involves electron beam lithography and thin film deposition performed at the CNF.

Summary of Research:

Tantalum disulfide (TaS_2) crystals with the 1T crystal structure undergo a CDW transition at ~ 150 K from the so-called nearly commensurate (NC) to the fully commensurate (C) phases, and there is an associated MIT with a ~ 10-fold increase in resistance. Importantly, the transition can be driven via an electric pulse, which allows for the operation of 2-terminal devices for neuromorphic computing [1]. However, our understanding of how the CDW transition occurs in real space is very limited, both for transitions driven by thermal cycling and with electric pulses. To address this question, we use a novel variable temperature STEM holder, which allows for continuously variable temperature from ~ 100-1000 K, and also permits measurement of sample resistance *in situ* [2]. In principle, this approach will allow us to visualize changes in the CDW structure as a function of temperature, which we can then correlate with changes in the sample resistance. Moreover, we will be able to visualize the CDW during electric pulsing.

The first step towards this goal is to fabricate TaS_2 devices and measure the CDW / MIT *ex situ* (meaning outside of the STEM). To do so, we first exfoliated TaS_2 flakes onto a SiO_2 / Si substrate. We then used the Nabity electron lithography package on the Supra scanning electron microscope (SEM) to lithographically pattern electrodes onto the flake. Lastly, we used the CHA thermal evaporator to deposit Cr / Au electrodes. An example device is shown in Figure 1.

Electrical resistance versus temperature data is shown in Figure 2 for another device. The measurement was performed using a physical property measurement system. The thermally induced CDW transition is clearly observed.

Having successfully measured the CDW transition in flakes *ex situ*, our next goal was to observe the transition with *in situ* STEM. For these experiments, we used specialty SiNx substrates with pre-patterned electrodes and through-holes for STEM observation. Figure 3 shows a TaS_2 flake which we transferred onto the pre-patterned electrodes. Note the hole underneath the flake, which is used for STEM imaging. We then studied this device within the STEM. Figure 4 shows a STEM image of the same flake, and the inset shows an electron diffraction pattern, which encodes information related to the CDW structure. By analyzing the diffraction data, we are able to determine the nature of the CDW phase.

Conclusions and Future Steps:

In this project we have fabricated TaS_2 electronic devices, and observed the CDW transition using electrical measurements. We have also developed a STEM method to observe the CDW transition in real space with nanoscale spatial resolution. Next steps for this project will include imaging the CDW phase with STEM, both as a function of temperature and applied electric field.

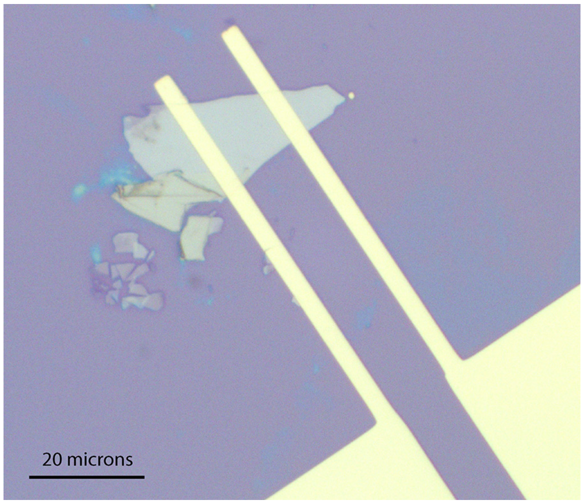

Figure 1: Optical image of a TaS_2 flake exfoliated onto SiO_2/ Si, with Cr / Au electrodes patterned on top of the flake.

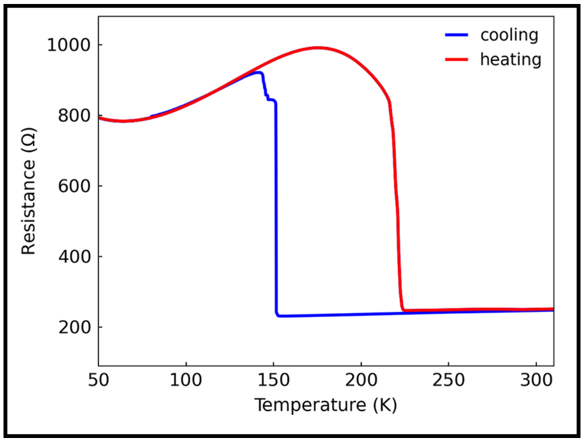

Figure 2: Resistance versus temperature data for a 2-terminal TaS_2 device.

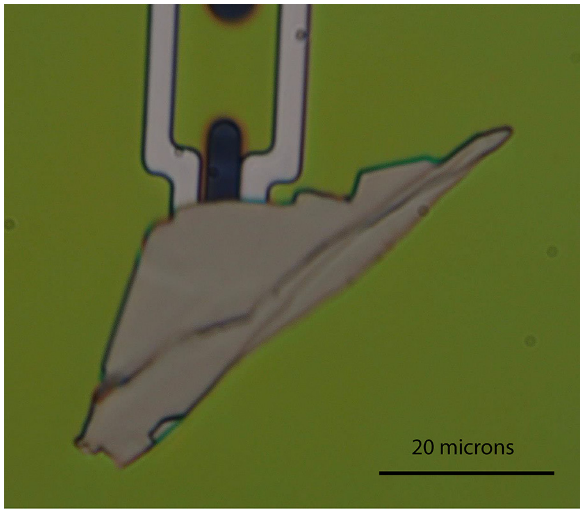

Figure 3: Exfoliated TaS_2 transferred to a chip that allows for in situ *STEM measurements. The dark hole beneath the flake allows for STEM observation, and the electrodes allow for* in situ *biasing.*

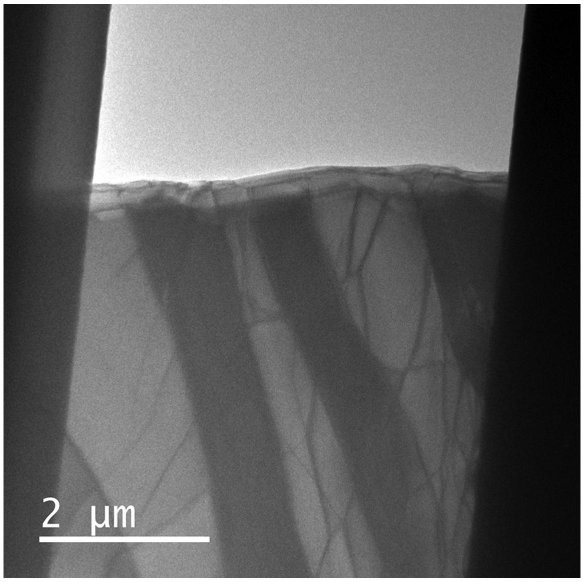

Figure 4: Same TaS_2 flake shown in Figure 3, but imaged within the STEM. The dark bands on the left and right hand sides of the image are the electrodes, and the empty region at the top of the image is the through-hole. The inset shows an electron diffraction pattern, and the weaker spots correspond to the CDW.

References:

[1] Hollander, M. J.; Liu, Y.; Lu, W.-J.; Li, L.-J.; Sun, Y.-P.; Robinson, J. A.; Datta, S. Electrically Driven Reversible Insulator-Metal Phase Transition in 1T-TaS_2. Nano Lett. 2015, 15 (3), 1861-1866.

[2] Goodge, B. H.; Bianco, E.; Schnitzer, N.; Zandbergen, H. W.; Kourkoutis, L. F. Atomic-Resolution Cryo-STEM Across Continuously Variable Temperatures. Microsc. Microanal. 2020, 26 (3), 439-446.