Tracking the TaS$_2$ 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

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 ongoing, 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.
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References:
