Lithography for *in situ* Cryogenic Scanning Transmission Electron Microscopy

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Primary CNF Tool Used: Nabity System for Supra SEM, PDMS Casting Station - Rm 224

Abstract:

Charge density waves (CDWs) of two-dimensional (2D) layered materials such as tantalum disulfide (TaS_2) and rare-earth tri-tellurides (RTe_3; R = rare-earth) can be used to make energy-efficient memory devices. To realize potential device applications, the CDW phase transitions of these materials should be studied at the nanoscale in the length scales of devices, and the changing transport properties must be correlated with the CDW phase transitions. In this project, we fabricate nanodevices of TaS_2 and RTe_3 on transmission electron microscopy (TEM) *in situ* chips such that we can operate the device inside a TEM, directly visualize the CDW phase transitions, and measure the transport properties during the CDW phase transition as a function of temperature between 100 K and 300 K.

Summary of Research:

We have successfully induced the CDW phase transitions of TaS_2 nanodevices during *in situ* cryo STEM experiments by both cooling down to 100 K (Figure 1) and by electric field. We have directly visualized the nucleation and growth of the nearly commensurate (NC) CDW out of the commensurate (C) CDW in TaS_2 nanodevices (Figure 2) and established that the nucleation of the NC-CDW state is defect-mediated (Figure 3). We have also proved that the electric-field induced phase transition from the C-CDW phase to the NC-CDW phase in TaS_2 is via Joule-heating, rather than strictly field-induced. We have also observed surprising CDW behaviors in exfoliated RTe_3 flakes due to stacking disorders present in these nanoscale samples.

Conclusions and Future Steps:

Our experiments constitute the first demonstrations of real-space direct imaging of electronic phase transitions in layered materials and the important role layer stacking disorders play on these electronic phase transitions. Three manuscripts are in preparation for submission this summer, which summarize our findings: (1) Defect-mediated CDW phase transition in TaS_2, (2) Field-induced CDW phase transition in TaS_2 is via Joule heating, and (3) Suppression of CDW phases in exfoliated RTe_3 flakes.

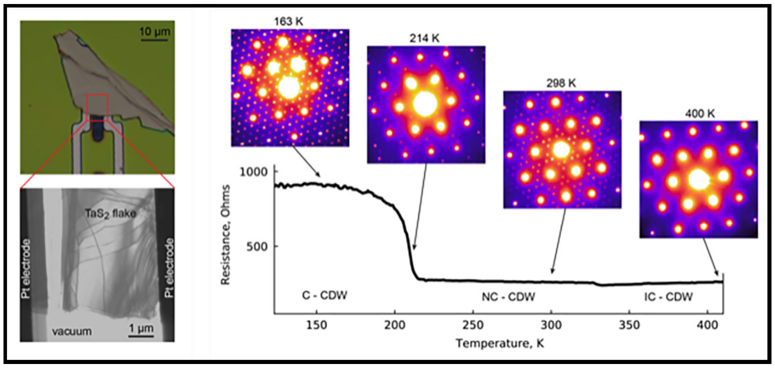

Figure 1: (Top, left) Optical micrograph of TaS_2 flake nanodevice with two electrodes. (Bottom, left) TEM image of the same TaS_2 flake, with stacking disorders shown as dark lines. (right) in situ *resistance measurement as well as* in situ *4D STEM acquisition of the TaS_2 nanodevice as a function of temperature down to 110 K. The C-CDW and NC-CDW phases can be distinguished by the superlattice patterns in the electron diffraction.*

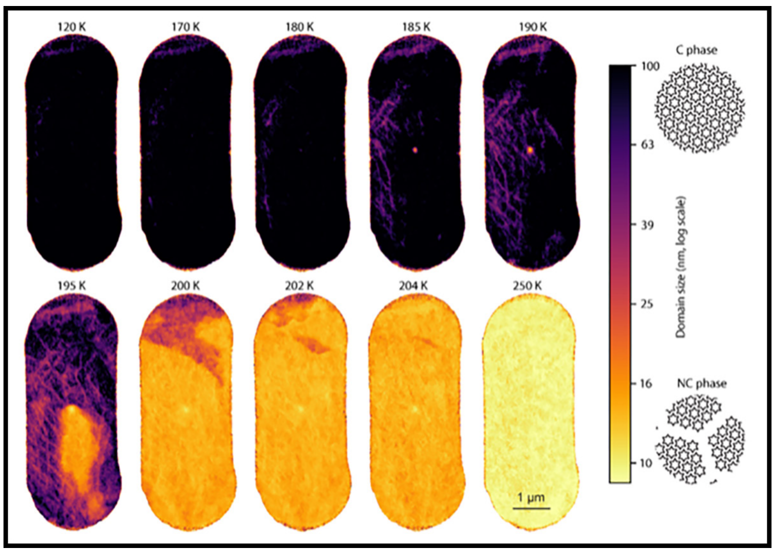

Figure 2: Nucleation and growth of the NC-CDW phase out of the C-CDW phase in the TaS_2 device as the temperature increases from 120 K to 250 K.

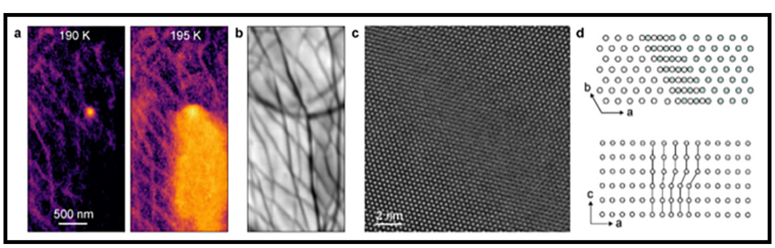

Figure 3: The nucleation of the NC-CDW phase (a) coincides with the location where the layer disorder defects (dark lines) merge (b). These dark lines were imaged in atomic resolution (c) to reveal the layer stacking disorder (d).