doi:10.1017/S143192761801005X
Microsc. Microanal. 24 (Suppl 1), 2018

Charge-Density Wave Localization and Co-Existence in 1T-TaS$_2$

Spencer A. Reisbick[1] and David J. Flannigan[2]

[1.] Department of Chemistry, University of Minnesota, Minneapolis, MN.
[2.] Department of Chemical Engineering and Material Science, University of Minnesota, Minneapolis, MN.

Several strongly electron-correlated materials are known to display charge-density waves (CDWs) under various conditions (*e.g.*, low temperatures) [1]. In 1T-TaS$_2$, for example, three distinct CDW phases of differing levels of commensurability are known to exist, the nature of each having been shown to depend on temperature. Above 350 K, an incommensurate phase is present, which is a semi-metallic state consisting of a randomly-oriented lattice distortion [2,3]. Below 350 K, this phase is thought to transform into a nearly commensurate (NCCDW) phase, which further transforms into a commensurate (CCDW) phase below 180 K. The NCCDW and CCDW phases consist of 6 and 12 Ta atoms, respectively, displaced toward a single central atom. In reciprocal space, the NCCDW phase is identified by the presence of an array of hexagonal satellite spots midway between each Bragg spot. The CCDW phase appears as a smaller hexagon of satellite spots surrounding each Bragg spot, which represents a repeating array of "Star of David" configurations [3]. Further, the satellite spots associated with each phase occur at distinct scattering angles. Conversely, in the relatively high-temperature semi-metallic phase, only Bragg spots are observed, which arise from an undistorted hexagonal lattice. Dynamics manifesting as changes in features of the satellite spots for both the NCCDW and CCDW phases have been studied using dedicated femtosecond X-ray and electron diffraction techniques, with scattering signal accumulated from relatively large specimen regions [4,5]. However, the behavior of localized CDW phases within discrete micrometer to nanoscale regions remains poorly understood.

Here, we describe our use of *in situ* TEM to characterize local CDW phase formation and transitions in micrometer-scale regions of 1T-TaS$_2$. As shown in Figure 1, selected-area diffraction patterns (DPs) acquired from a micrometer-scale specimen region contain satellite spots arising from both the CCDW and NCCDW phases, despite the specimen temperature being below 180 K. This suggests that some other parameter, in addition to temperature, dictates the formation of the CDW phases. Further, this casts doubt on dynamics derived from monitoring intensity changes of the satellite spots (*i.e.*, melting of CDW phases), as this could instead arise from a local change in relative phase concentration rather than a global phase transition. Importantly, we have also found that, from flake to flake, the temperature at which super-lattice spots fade and vanish varies and depends on location, topology, and thickness. In all specimens, we find that the bulk CDW signal intensity follows the temperature-dependent 1T-TaS$_2$ phase diagram [6].

In addition to region-specific and flake-to-flake variations, we find that different regions within a single 1T-TaS$_2$ specimen display different CDW scattering profiles when comparing normalized super-structure intensities to the structural Bragg signature. Specifically, there is an apparent significant decrease in CDW symmetry near the edges of individual flakes, indicating a disruption of the phase formation, again due to specimen morphology rather than temperature alone. In this case, it may be that the free edge produces a structural distortion that is energetically favorable compared to formation of a CDW phase (and associated lattice distortion). Importantly, methods averaging signal from relatively large specimen areas have reported observing first-order transitions between the CDW phases [6], but we observe no such behavior for local, selected-area diffraction experiments. The higher-order nature of the transitions observed here suggests that large-scale measurements observe phase changes only once a threshold concentration is reached. Moving forward, we expect additional insight into accessible temporally-resolved states will be gained by studying localized responses to *in situ* optical excitation and conducting comparisons to thermal studies [7][8].

References:

[1] K. Rossnagel, J. Phys.: Condens. Matter **23** (2011), p. 213001.
[2] J. A. Wilson *et al*, Adv. Phys. **50** (2001), p. 1171.
[3] A. W. Tsen *et al*, Proc. Natl. Acad. Sci. U.S.A. **112** (2015), p. 15054.
[4] M. Eichberger *et al*, Nature **468** (2010), p. 799.
[5] C. Laulhé *et al*, Phys. Rev. Lett. **118** (2017), p. 247401.
[6] M.Yoshida *et al*, Sci. Rep. **4** (2014), p. 7302.
[7] I. Vaskivskyi *et al*, Sci. Adv. **1** (2015), p. e1500168.
[8] This work was supported partially by the National Science Foundation through the University of Minnesota MRSEC under Award No. DMR-1420013 and partially by the Arnold and Mabel Beckman Foundation through a 2015 Beckman Young Investigator Award.

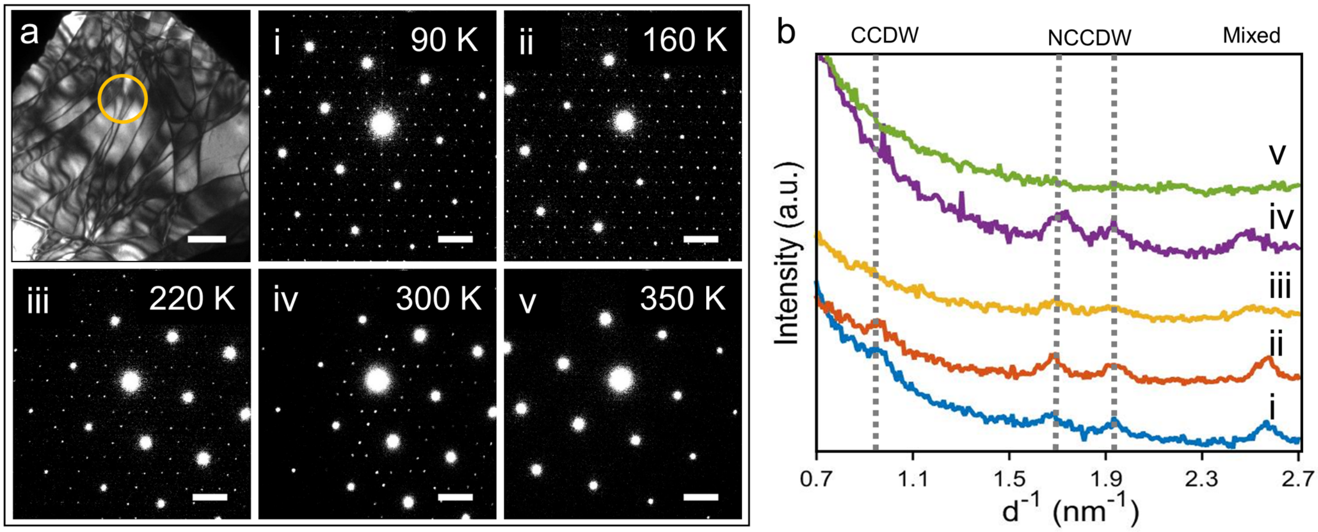

Figure 1. Localization and co-existence of CCDW and NCCDW phases below 180 K within a micrometer region of a multi-layer 1T-TaS_2 specimen. (a) Bright-field TEM image and associated selected-area diffraction patterns (DPs) of 1T-TaS_2 at temperatures ranging from 90 to 350 K. The position and size of the selected-area aperture is indicated by the yellow circle in the image (scale bar = 1 μm). The black features around the image periphery are the Cu support grid. The temperature at which the DPs were acquired are labeled in the upper-right corner of each panel (scale bars = 2 nm^{-1}). (b) Select radially-integrated scattering-angle regions from the DPs shown in (a). Each plot is labeled according to the pattern from which it was generated (*e.g.*, i = 90 K, ii = 160 K, *etc.*). Expected peak positions arising from the CCDW and NCCDW satellite peaks are labeled, as is a region labeled Mixed, which contains unresolved intensity from both phases.