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Atomic-resolution Cryo-STEM Imaging of a Structural Phase Transition in $TaTe_2$

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Layered transition metal dichalcogenides (TMD), MX_2 (M = transition metal, X = S, Se, or Te), exhibit diverse phenomena such as superconductivity, charge density waves, non-saturating magnetoresistance, and valley physics [1, 2, 3, 4]. These exotic properties typically occur below room temperature and can be dramatically altered or enhanced by adjusting sample thickness, temperature, or doping level. For instance, 1T'-$TaTe_2$ has a monoclinic (space group C2/m) structure at room temperature in which the Ta atoms form trimers and the Te atoms undergo a puckered modulation (Fig. 1a,b). Lowering the temperature reveals a semimetal-semimetal phase transition which manifests as an anomaly in the resistivity curve near 170 K (Fig. 1c) [5]. Aberration-corrected scanning transmission electron microscopy (STEM) is a powerful technique to map the atomic lattice with high precision; however, most experiments are performed at room temperature. To understand emergent, low temperature phases in TMD's, visualizing the atomic-scale structure under cryogenic conditions is necessary.

Recently, we demonstrated cryogenic STEM (cryo-STEM) with sub-Angstrom resolution and picometer precision, and visualized charge ordering in a manganite at 93 K [6]. Here, we apply cryo-STEM to unravel the low temperature phase of $TaTe_2$. From a bulk crystal, we exfoliate thin specimens and transfer them onto holey SiO_2 grids, which allows imaging in plan-view (Fig. 2a). For cross-sectional imaging, we exfoliate flakes on a Si/SiO_2 wafer and cap them with a metal ($Ni_{81}Fe_{19}$). We then prepare thin cross-sectional lamellas using the conventional focused ion beam lift-out technique. High-angle annular dark-field (HAADF) STEM is performed in an aberration-corrected FEI Titan Themis operating at 300 kV. The convergence semi-angle is 21.4 mrad, and the probe current is ~50 pA. Despite the high voltage, we do not observe knock-on damage. Low temperature imaging is performed using a double-tilt liquid-nitrogen side-entry holder. Due to reduced stability at cryogenic temperatures, we acquire images in rapid succession (0.5-1 μs/pixel) and carefully align them using rigid registration [7].

Figure 2b shows a room temperature HAADF image in the planar direction. The Ta columns appear elongated due to the stacking of Ta offsets. The Fourier transform (FT) amplitude displays Bragg peaks associated with the C2/m phase (Fig. 2b, *inset*). Upon cooling to 93 K, additional contrast modulations are apparent in the image (Fig. 2c), likely due to correlated displacements in the lattice in the low temperature phase. The formation of a superstructure is also reflected in the FT which exhibits distinct superlattice peaks. Due to the highly anisotropic structure of the compound, we also image the out-of-plane structure along the b-axis (Fig. 1b) and perpendicular to the b-axis. By quantifying subtle intensity variations and atomic offsets upon cooling, we elucidate the nature of the low temperature phase of $TaTe_2$ [8].

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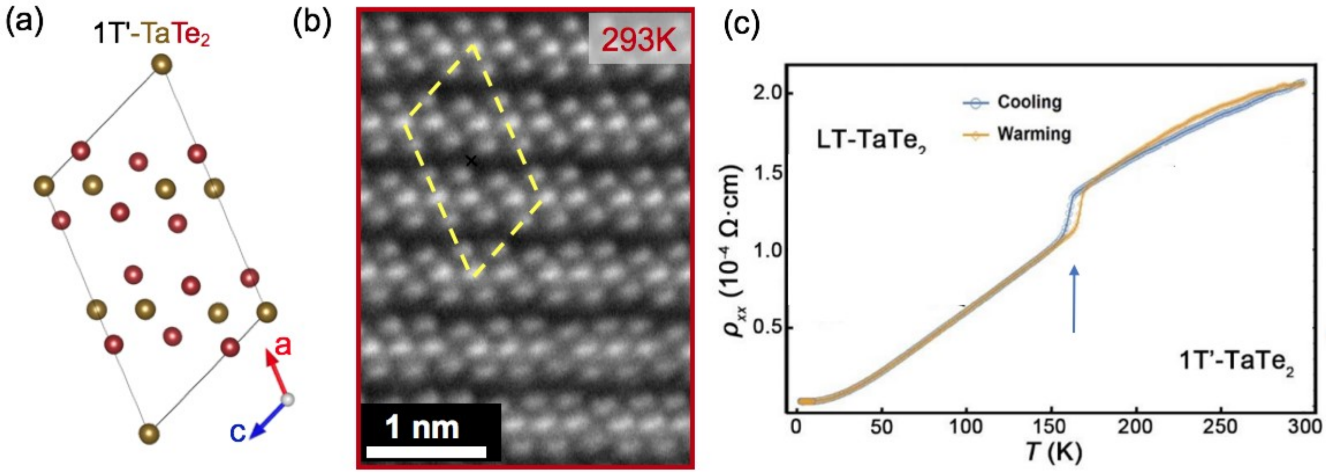

Figure 1. (a) Crystal structure of 1T'-$TaTe_2$ along the b-direction at room temperature (293 K). (b) HAADF-STEM image showing the layered structure, Ta trimers, and Te modulations. (c) Temperature-dependent resistivity shows a transition near 170 K (blue arrow). Adapted with permission from [5].

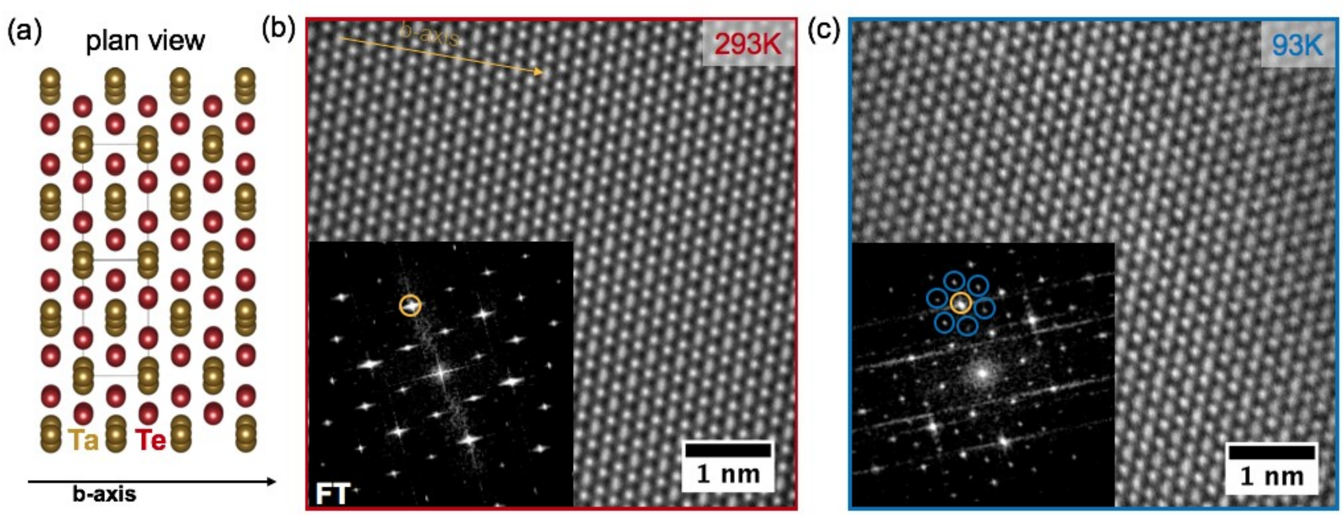

Figure 2. (a) Crystal structure of 1T'-$TaTe_2$ in plan view at room temperature. (b), (c) HAADF-STEM image at 293 K and 93 K, respectively. A periodic modulation of the contrast appears at 93 K, which reflects changes in the structure. The Fourier transform (inset) exhibits superlattice peaks at 93 K (blue circles), indicating the formation of a superstructure.

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