doi:10.1017/S1431927622008911
Microsc. Microanal. 28 (Suppl 1), 2022
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# In-situ Imaging of Anisotropic Layer-by-layer Phase Transition in Few-layer $MoTe_2$

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In-situ transmission electron microscopy (TEM) has been an important technique to monitor the phase transition of materials at atomic scales, providing valuable information on how materials respond to external stimuli in real time. Two-dimensional (2D) materials offer great potential because of their layered structure, which enables direct observation of phase transition along the in-plane directions. Here, we use graphene (Gr) encapsulation to create an enclosed, in-situ reaction cell for TEM studies of few-layer $MoTe_2$ on a MEMS-based heating holder. We choose $MoTe_2$ because it exhibits multiple structural phases (2H, 1T', and $T_d$) [1] that can be easily manipulated with different stimuli [2, 3], making it a promising phase change material. Whereas typically, graphene encapsulation protects $MoTe_2$ from sublimation induced by in-situ heating or electron beam damage [4] while being highly electron transparent, allowing atomic resolution studies of $MoTe_2$ down to a single unit cell thick. In our work, we combine dark-field TEM (DFTEM) and aberration-corrected scanning transmission electron microscopy (STEM) to image the phase transitions of few-layer $MoTe_2$ from the micro- to atomic scales. We find that $T_d$-to-2H phase transitions initiate at 2H-$T_d$ interfaces and propagate primarily along the b-axis of the nearby $T_d$ grain, indicating an anisotropic, layer-by-layer phase transformation.

First, we fabricate a stack of hBN/Gr/$MoTe_2$/Gr/hBN by encapsulating a few-layer exfoliated $MoTe_2$ flake with graphene and hBN. The encapsulated $MoTe_2$ was then irradiated by a laser to locally convert part of the 2H phase into the metastable $T_d$ phase. The laser-irradiated, Gr-encapsulated $MoTe_2$ stacks are transferred onto MEMS chips for in-situ heating after removing the thick hBN layers by $XeF_2$ dry etching. We characterize the crystal structures and orientations by selected-area electron diffraction (SAED), DFTEM, and aberration-corrected STEM (Figure 1). In order to capture the dynamics of $T_d$-to-2H phase transition at both micro and atomic scales, we apply heat pulses as short as 0.5s using a MEMS-based heating holder to "freeze" the growth front of the 2H phase at different temperatures. This allows us to acquire both low-magnification DFTEM images that highlight the 2H phase region and atomic-resolution annular dark-field (ADF) STEM images at the interface after each heat pulse (Figure 2). We find that the $T_d$-to-2H phase transition can occur at temperatures as low as 275 $^{o}$C (much lower than the previously reported 1000 $^{o}$C [4]), where it initiates at 2H-$T_d$ interfaces and anisotropically propagates along the b-axis of the $T_d$ grain. We individually measure and compare the phase front of each separate layer and find that the linear growth rate of the 2H phase at 275 $^{o}$C varies from 5 to 14 nm/sec between different layers, which suggests that other factors such as strain and local defect densities may affect the energy barrier of phase transition. This technique can be applied to other 2D materials and help unveil their phase transition kinetics [5].

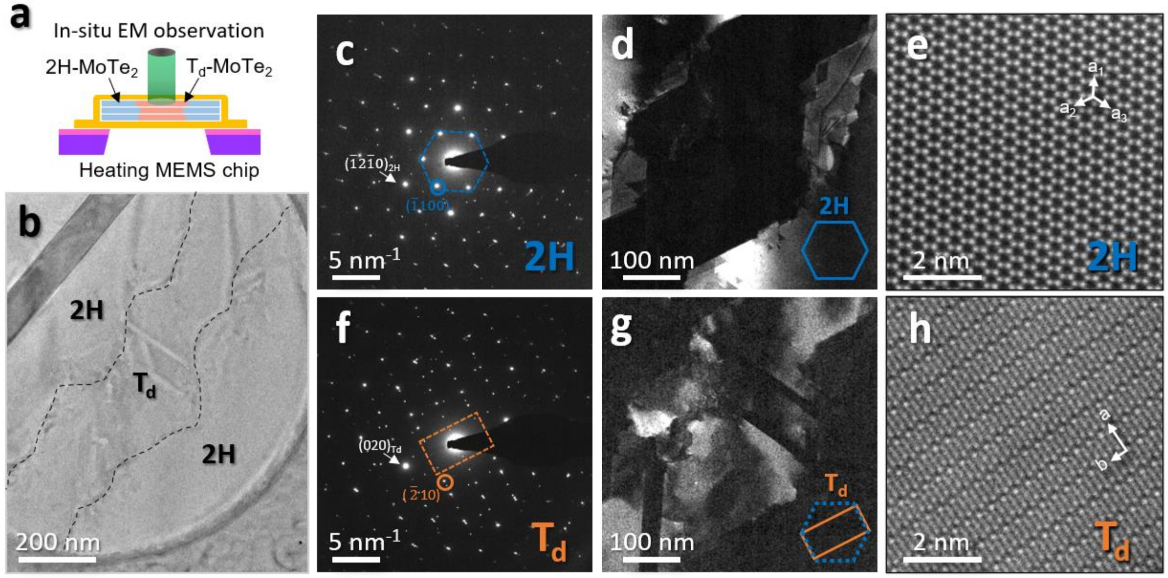


**Figure 1.** Imaging phase transformations in $MoTe_2$. (a) Schematic of the sample geometry for in-situ electron microscopy experiments. The laser-irradiated $MoTe_2$ flake is encapsulated by graphene layers and transferred onto the MEMS chip for in-situ heating. (b) Bright-field TEM image of suspended $MoTe_2$ containing both 2H and $T_d$ grains. The $T_d$ phase region was delineated by the laser trajectory and outlined by the dashed lines. (c, f) SAED pattern, (d, g) dark-field TEM, and (e, h) aberration corrected ADF-STEM images for 2H and $T_d$ phase, respectively. The DFTEM images (d, g) are formed by selecting the $(\underline{1}100)_{2H}$ and $(\underline{2}10)_{Td}$ Bragg spots in (c) and (f) with the objective aperture.

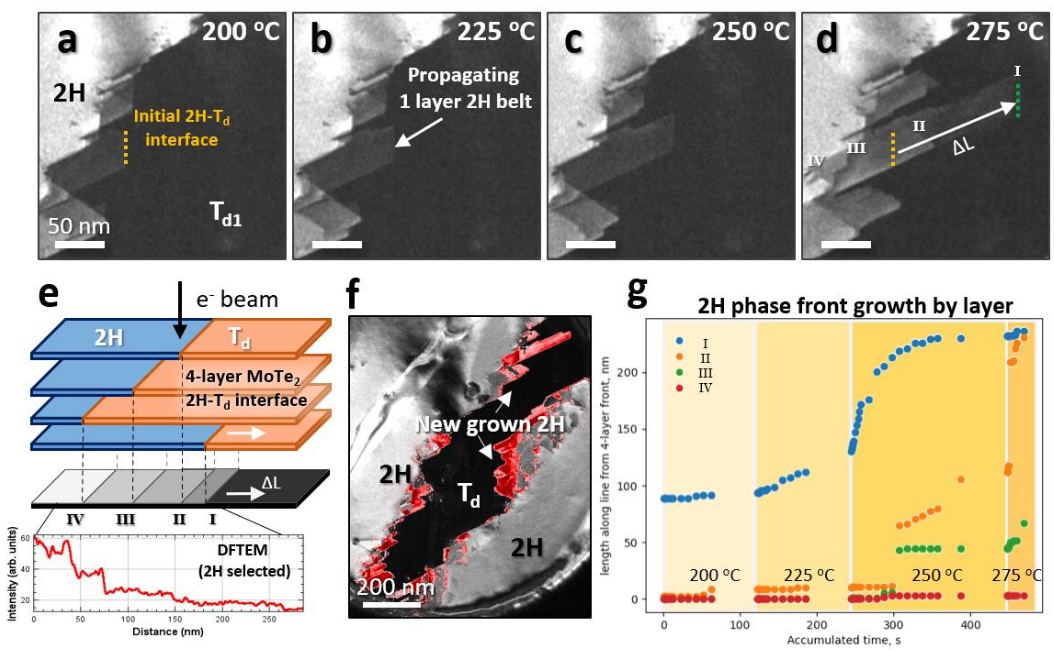


**Figure 2.** Anisotropic layer-by-layer, $T_d$-to-2H phase transition. (a-d) DFTEM images show the 2H phase front propagating after each pulse (200 to 275 $^oC$). The 2H-$T_d$ interface of different layers can be individually identified by their intensity difference. (e) Schematic of the intensity differences of 4-layer

https://doi.org/10.1017/S1431927622008911 Published online by Cambridge University Press

$MoTe_2$ 2H-$T_d$ interfaces in DFTEM. The mono-, bi-, tri-, and quad-layer 2H phase fronts are labeled as I, II, III, and IV respectively. (f) Overlay of a low magnification DFTEM image with newly formed 2H region marked in red, showing that the $T_d$-to-2H phase transition occurs only at 2H-$T_d$ interfaces. (g) Plot of 2H phase front positions of different layers as a function of accumulated heating time. The propagation rates are extracted by the slope of the curves, which show strong temperature dependence.

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[5] This work was supported by the DOE award number DE-SC0020190. This work was carried out in part in the Materials Research Laboratory at UIUC.

https://doi.org/10.1017/S1431927622008911 Published online by Cambridge University Press