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In Situ Laser Synthesis of 2D WSe_2 Within TEM

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Non-equilibrium synthesis and processing enables a wide variety of materials systems with tuned metastable phases and properties. Here we report how a prototype setup allows laser illumination to be coupled into transmission electron microscopes (TEM) for real-time observations of two-dimensional (2D) materials synthesis and processing. The configuration and ability of *in situ* photon delivery system was reported previously[1-2]. Amorphous atoms and molecules were deposited at room temperature on TEM grids by pulsed laser deposition. They were subsequently irradiated by a laser built within a high-resolution TEM (HRTEM) and then crystallized through various pathways observed by *in situ* TEM imaging. In addition, post-growth high-angle annular dark field (HAADF) Z-contrast scanning TEM (STEM) measurements are conducted for characterization of crystal phases, defects and grain boundaries at the atomic scale.

Two-dimensional WSe_2 was synthesized from amorphous precursors within the TEM with laser irradiation. The final morphology and the growth pathways were found to be dependent on the substrate properties. Figure 1a shows the post-growth atomic resolution HAADF image of 2D WSe_2 grown on graphene by laser irradiation (wavelength = 785 nm, pulse width = 10 msec). The final 2D WSe_2 crystals are polycrystalline, evident by the selected area electron diffraction (SAED) pattern and also the fast Fourier transformation (FFT) of an overview STEM image. Conversely, WSe_2 crystals grown on chemical vapor deposition (CVD) $MoSe_2$ monolayer substrate by laser present a perfect alignment with the $MoSe_2$, confirmed by the SAED pattern and FFT image (Figure 1b). We attribute this improvement in van der Waals epitaxy to the reduced lattice mismatch from WSe_2/graphene (25%) to WSe_2/$MoSe_2$ (0.2%). STEM images also show the twisted angles between WSe_2 and $MoSe_2$ heterobilayer are 0^o (3R), 60^o (2H), or $\leq 5^o$.

The crystallization pathway of 2D WSe_2 by laser irradiation within the TEM is shown in Figure 2. On graphene (Figure 2a), prior to become the 2D planar structure, 3D metastable WSe_2 was achieved in a Se-rich environment. Similar 3D metastable phase has been found during the formation of 2D MoS_2 flakes from the thermolysis of ammonium thiomolybdates on Si_3N_4 membrane using in situ heating TEM[3]. The structural evolution was found to largely depend on Se to W ratio during laser irradiation. On $MoSe_2$ (Figure 2b), prior to form epitaxial WSe_2, crystallites nucleate with different orientations first and then register into the lattice of $MoSe_2$. The temperatures of the substrate corresponding to different laser energies were measured by the shifts of their exciton peaks using electron energy loss spectroscopy (EELS) at cryogenic temperatures in a mono-chromated TEM. The minimum crystallization temperature for WSe_2 was measured to be 300 °C.

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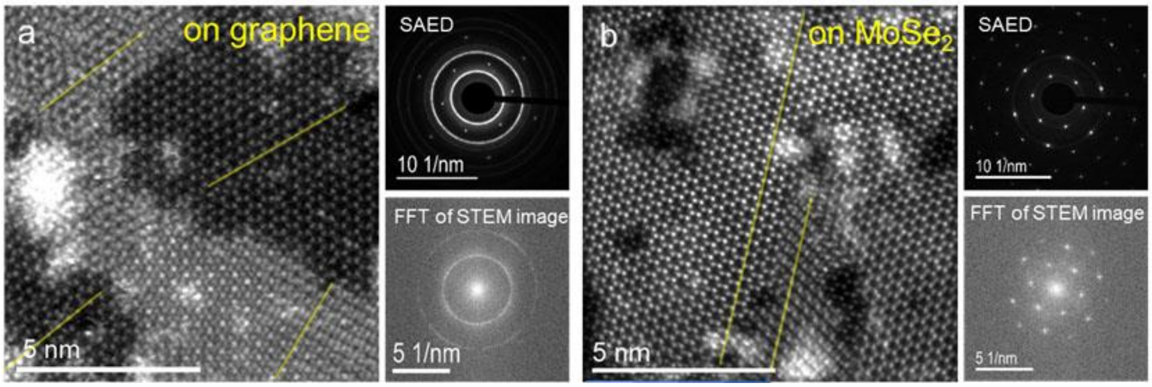

Figure 1. Post-growth morphology of 2D WSe2 grown by laser irradiation within the TEM on (a) graphene and (b) CVD grown monolayer MoSe2

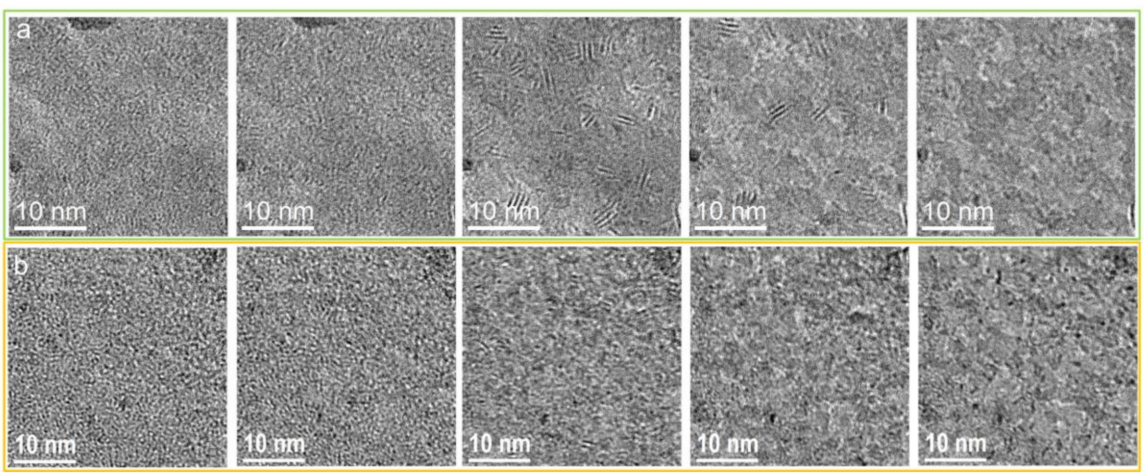

Figure 2. Growth of 2D WSe2 by laser irradiation within the TEM evolved with increasing laser energy on (a) graphene and (b) CVD grown monolayer MoSe2

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