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Defect Dynamics in 2D Transition Metal Dichalcogenide Monolayers

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Transition-metal dichalcogenide (TMDC) monolayers are promising atom-thin semiconductors for flexible nanoelectronic and optoelectronic applications [1, 2]. Intrinsic defects, such as vacancies, dislocations and boundaries, are known to have profound influence on the electronic and optical properties of these materials. For instance, chalcogen vacancies are the most common point defects in TMDC monolayers, and largely contribute to the n-type conductivity in CVD-grown MoS_2 monolayers [3]; 60˚ grain boundaries in semiconducting TMDC monolayers (Fig.1A) have been shown to be metallic and quench the local photoluminescence [4,5]. In order to fully develop the potential of TMDC monolayer, it is useful to recognize and understand the dynamical behavior of structural defects in these monolayers.

Here we show that the evolution of defects in TMDC monolayers not only can be directly monitored and explained at the atomic scale, but can also be utilized to create new nanostructures with desired properties. The experiments were performed on an aberration-corrected Nion UltraSTEM-100, operated at 100 kV. We first use a focused electron beam to generate and excite Se vacancies in the monolayer $MoSe_2$, and monitor their dynamics through sequential atomic-scale annular dark field (ADF) imaging. We find that Se vacancies are first randomly created by mild ionization damage and then preferentially agglomerate into line defects under the energy transferred from the electron beam. Successive evolution of these line defects induces nucleation of distinct triangular inversion domains within the $MoSe_2$ layer and generates conductive 60° grain boundaries within the semiconducting matrix (Fig.1B). Using density functional theory (DFT) calculations, we explain the driving force of the formation mechanism: the aggregation of chalcogen vacancies induces large lattice shrinkage, which causes the nearby metal atoms to undergo displacements and subsequently nucleates inversion domains in order to release the lattice strain. Migration of the grain boundaries can be further activated by deformation of the peripheral lattice, giving rise to the growth of the inversion domain [6].

Extensive irradiation generates both metal and chalcogen vacancies and activates their complex dynamics. Prolonged illumination of a small region of the TMDC monolayer with an intense focused electron probe can eventually lead to formation of small holes. By selectively patterning the holes, we recently reported the direct fabrication of ultrathin MX (M=Mo, W; X=S, Se) nanowires, including their ramified junctions, connecting designated points within a semiconducting TMDC monolayer (Fig. 2) [7]. These nanowires are intrinsically metallic, thus serving as promising candidates for ultrasmall interconnects within the 2D integrated circuit. Moreover, we show that these nanowires maintain extreme structural flexibility, as they remain conducting when being twisted or kinked. We will show that the chemical composition of such nanowires can be tuned by successfully fabricating intermixed $MoS_{1-x}Se_x$ and $Mo_{1-x}W_xS$ nanowires. The concentration of S and Se in the $MoS_{1-x}Se_x$ nanowire is controlled by the acceleration voltage during the electron-beam sculpting. The transport properties of these intermixed nanowires are further studied by first-principles calculations [8,9].

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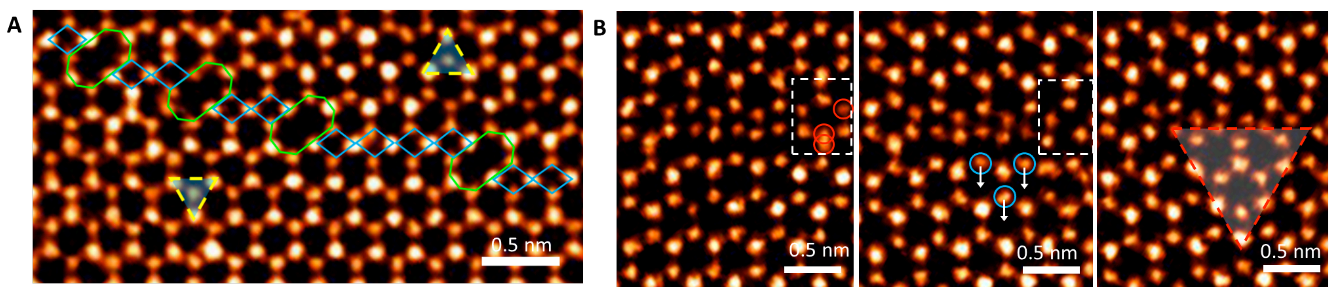

Figure 1. Nucleation process of the inversion domain in monolayer $MoSe_2$. (A) Atomic resolution ADF image of an intrinsic 60˚ grain boundary in CVD-grown monolayer $MoSe_2$. (B) Sequential ADF images showing the nucleation process of an inversion domain in monolayer $MoSe_2$ induced by Se vacancies.

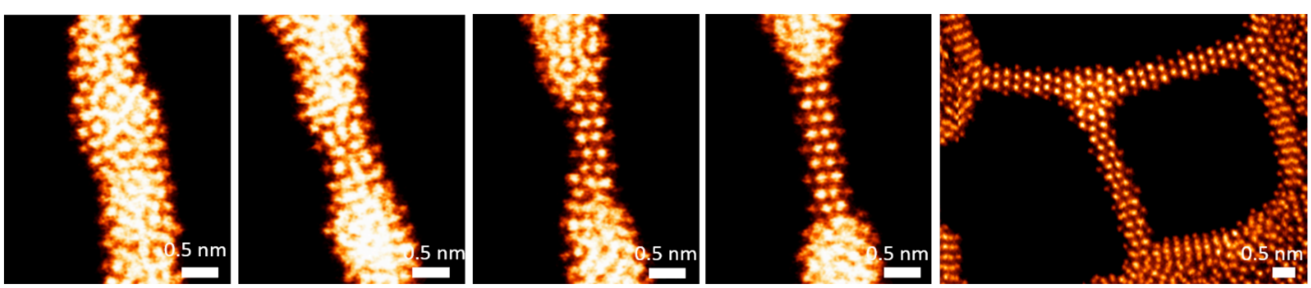

Figure 2. *In-situ* fabrication of nanowires from a TMDC monolayer. Sequential ADF images showing the fabrication process of an individual MoSe nanowire entirely within the semiconducting $MoSe_2$ monolayer. A Y-junction is shown on the right.

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