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Atomic-scale Fabrication of 1D-2D Nano Hetero-structures within 2D Materials through Automated Tracking and Electron Beam Control

Matthew G. Boebinger[1], Ayana Ghosh[1], Sudhajit Misra[1], Kevin Roccapriore[1], Kai Xiao[1], Andrew R. Lupini[1], Panchapakesan Ganesh[1], Maxim Ziatdinov[1], Sergei V. Kalinin[2], Stephen Jesse[1] and Raymond R. Unocic[1]

[1.] Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, TN, USA.
[2.] Department of Materials Science and Engineering, University of Tennessee, Knoxville TN, USA.

Direct atomic control over the fabrication of nanoscale architectures remains the central goal of nanotechnology. With the advent of aberration-corrected scanning transmission electron microscopy (STEM), the sub-Å sized electron (e)-beam has now become available to use as a tool for precision atomic fabrication of materials [1-3]. Prior work using the e-beam as a fabrication tool has shown the ability to position dopant atoms in 2D and 3D materials [1, 3, 4] and form specific edge structures or nanopores through controlled e-beam irradiation [5, 6]. Different nanostructures, such as nanopores and atomic edge configurations, within 2D materials, have been shown to locally alter the functional properties to enhance electronic, magnetic, optical, and catalytic properties within individual monolayers [5-6]. Control over the types of edge can allow for the local tailoring of properties for specific purposes. However, the mechanisms determining the formation of these larger structures are not fully understood.

In this work, we extend beyond dopant manipulation to *atomic transformations* in 2D materials to investigate how the symmetry of the probe trajectory affects the material's transformation. We explore if it is possible to deterministically control the atomic edge structure bounding a single nanopore. This goal is accomplished through the use of a feedback-controlled e-beam system that was specifically developed to enable more precise control over the atomic fabrication process with simultaneous *in situ* visualization and feedback [7]. This system also allows for control over the position, size, dose and shape of the scanning e-beam pathway profile. Several e-beam scanning pathway profiles (circular, square, triangular) were generated then used to irradiate 2D MoS_2 along specific crystallographic orientations as shown in Figure 1. This allows us to determine what influence e-beam induced transformation have on the formation of unique atomic edge structure configurations (e.g. Mo_6S_6 nanowire (MoS-NW)) as a function of scan direction and crystal orientation. In Figure 1(a), a circular beam path results in the MoS-NW forming along the edges that terminates normal to the zigzag sulfur direction (ZZS), however the circular pathway forces the disordering of the bottom edge of the frame, rounding out the nanopore. For the square beam pathway shown in Figure 1(b), there was a similar tendency to form MoS-NW edges along the ZZ directions but the edge of the nanopores were irregularly shaped. When the triangular beam path was used and aligned such that all edges terminated in the ZZS direction in Figure 1(c), the MoS-NWs were shown to form reliably and controllably. These results demonstrate that there is a preferential formation of the MoS-NW structure to form on the ZZS terminating edge. We infer that precise alignment of the e-beam triangular path on this direction could be used to reliably fabricate these structures. These experimental results were combined with theoretical simulation efforts to determine the formation energy of MoS-NW edge structures on the different terminating directions within MoS_2 monolayers.

To gain further insight into the resultant atomic transformation mechanisms during the initial stages, a deep convolutional neural network (DCNN) was constructed to assist in atomic identification, tracking

and reveal trends in the atomic re-ordering process [8-9]. In Figure 2, several frames were extracted prior to the nanopore formation to reveal the initial stages of atomic re-ordering in efforts to provide insights into the telltale signatures of nanopore formation and then MoS-NW formation. Initial work has been conducted that allows for the decoding and tracking of the different atomic species throughout the milling process as seen in Figure 2 (a-c). Individual atoms are then tracked throughout the milling process (Figure 2d) and statistical analysis was used to identify trends in the atomic re-ordering in different milling processes. Through the application of this DCCN model combined with the automated scanning e-beam control system, in-depth studies of the e-beam matter reactions in 2D materials can be conducted that can lead to a greater degree of control to fabricate nanoscale devices using automated STEM techniques [10].

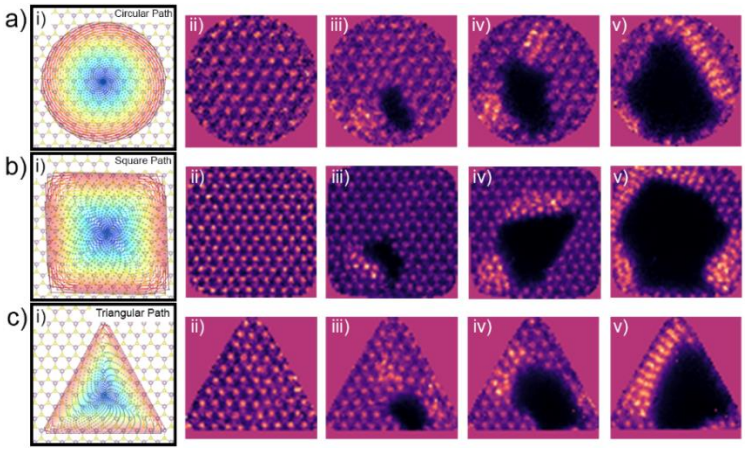

Figure 1: Controlled fabrication of nanopores with MoS-NW edge reconstructions in MoS_2 as a function of scanning e-beam pathway profile: (a-c) The circular, square and triangular beam path used for milling, respectively: (i) schematic of scanning beam pathway, (ii-v) frames from the milling process showing the nucleation of the nanopore to the formation and growth of the MoS-NW edge structures.

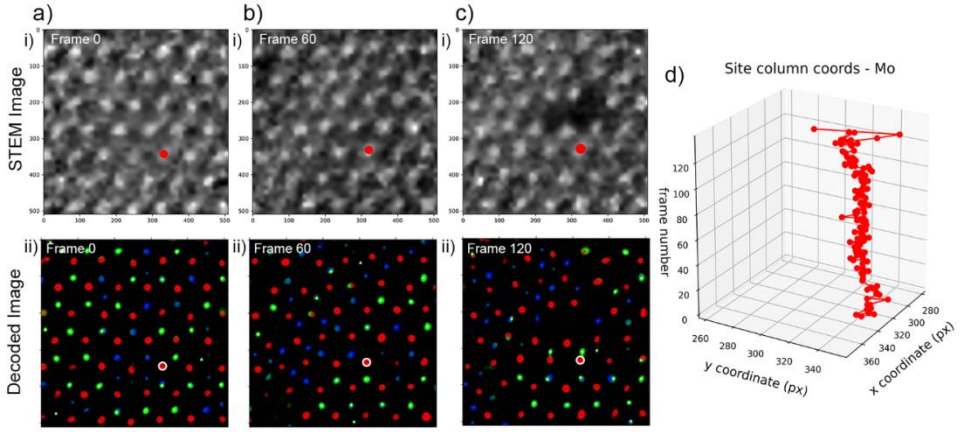

Figure 2: DCNN assisted atomic identification and tracking: (a-c) Frames 0, 60 and 120 extracted from a milling experiment. (i) The experimental data frame taken during the milling process with a single Mo atom highlighted. (ii) The decoded frame from above with all atoms in frame classified as Mo (red), 2S sites (green), 1S (blue), with the same Mo atom highlighted. (d) Tracking plot of the Mo atom position that can be acquired from the milling dataset.

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