Focused Ion Beam Patterning for Defect-Mediated Nucleation on 2D van der Waals Materials

V. A. Zarubin, K. Reidy, I. Charaev, J. D. Thomsen, J. P. Klein, F. M. Ross
Massachusetts Institute of Technology, Cambridge, MA 02139
verazar@mit.edu

Y. Yu
Raith America Inc., Troy, NY 12180

The ability to assemble arrays of three-dimensional (3D) metallic and semiconducting nanostructures is key to development of nanoelectronics and optoelectronics. In particular, 3D nanoislands on two-dimensional (2D) van der Waals bonded materials display intriguing functionalities that enable applications in plasmonics, single photon emission, exciton coupling, and superconducting junctions.[1] When 3D nanoislands are grown on conventional, bulk substrates, "directed self-assembly" is a useful approach where growth takes place spontaneously but is favored at particular nucleation sites. One strategy for directed self-assembly is to create topological features on the 2D material surface that act as nucleation sites for metals and semiconductors.[2] Ion beam patterning is particularly well suited to create topological features with high spatial resolution on 2D materials, owing to the intrinsic confinement of implanted defects in the vertical direction and reduced broadening in the lateral direction.[3]

In this work, we use focused ion beams to create local defects and mill nanopores in layered suspended 2D materials (graphene, MoS_2, WSe_2) (Fig 1a). Ions used in this work are all non-Ga species, and delivered as arrays of spots and lines using He^+ in the Carl Zeiss ORION helium ion microscope (HIM) and Si^{2+} and Au^{2+} in the Raith VELION focused ion beam-scanning electron microscope (FIB-SEM). Ion doses are calibrated and the resulting local defects and nanopores are characterized with high resolution scanning transmission electron microscopy (Fig. 2). We study the extent to which the defects act as nucleation sites for Au, and tune deposition conditions and surface cleanliness to facilitate self-assembly of specific nanoisland shapes (Fig. 1b). Recorded movies of *in situ* transmission electron microscopy heating reveal the effect of post-deposition annealing on island shape and size. The island geometry lends to automated image analysis to obtain statistics of the nucleation density and location. The technique can be generalized to create patterned structures of other materials, such as Si and Ge via chemical vapor deposition or metals via thermal and e-beam evaporation. This templating and nucleation control strategy opens routes towards directed self-assembly of 3D nanoislands on 2D materials for hybrid materials with optimized charge transfer.

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[1] S. Bae *et al.,* Nature Materials **18**, 550-560 (2019).
[2] Gherasimova, M., Hull, R., Reuter, M. C. & Ross, F. M., Appl. Phys. Lett. **93**, 23106 (2008).
[3] Kretschmer, S. *et al.* ACS Appl. Mater. Interfaces. **10**, 30827 (2018).

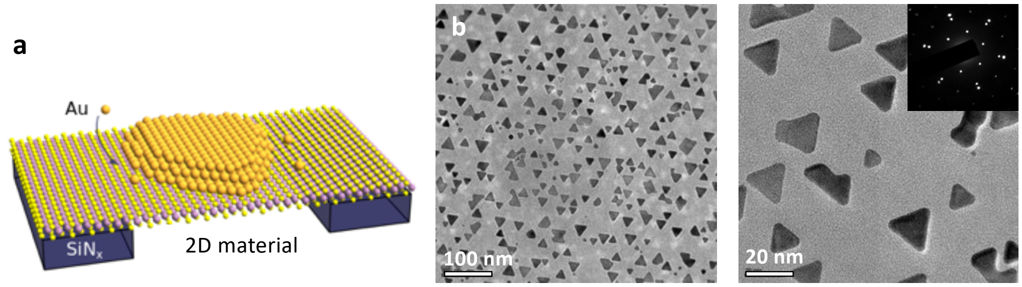

Figure 1: Formation of facetted nanoislands on suspended 2D materials **(a)** Schematic of suspended 2D material MoS_2 on TEM grid with facetted Au nanoisland deposited on the surface. **(b)** Facetted, epitaxial, Au nanoislands grown on pristine WSe_2. The Au nanoislands form equilibrium triangular shapes that are optimal for plasmonics.

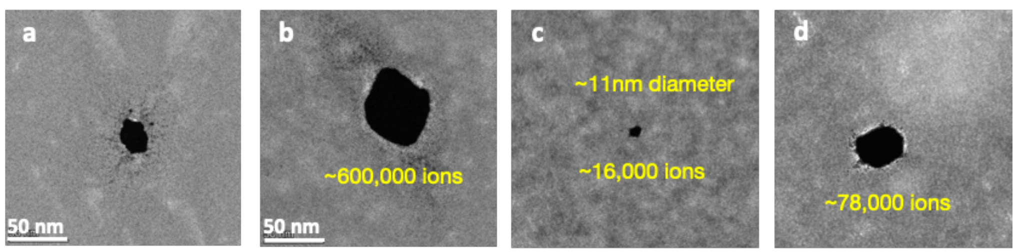

Figure 2: Dose calibration of monolayer MoS_2 (a, b) with He^+ in Zeiss ORION helium ion microscope (HIM) and (c, d) Si^{2+} in Raith VELION focused ion beam-scanning electron microscope (FIB-SEM).