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Automated Electron Beam Manipulation for Controlled Materials Transformations

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Scanning transmission electron microscopy (STEM) has been a pillar for materials discovery for various fields ranging from electrochemistry to biological systems [1-3]. While the current standard for STEM operation and data collection is primarily human based, the processing time of a human operator is substantially lower than that of a computer-based process. The use of computer automation to control the electron beam and microscope parameters provides a direct pathway for experiments with higher control, lower beam damage, and decreased time per experiment.

The major benefit for automated processes in STEM is the high level of control in the electron beam location and dose. This precise control of beam position opens the avenue to probe material properties which are highly direction or shape dependent and those which are highly spatially localized. On the other hand, the control of electron dose per location allows the manipulation of matter at the atomic scale. Here we show the use of automated STEM developed in Python to control the electron beam to design material shape and change material properties with real time feedback.

To demonstrate the electron beam control available in automated experiments, holes are drilled inside rectangular nanoparticles via electron beam radiation. These nanoparticles exhibit plasmon resonances ranging from the near infrared to the ultraviolet which can be tuned during synthesis. After nanoparticle fabrication, the resonance cannot be altered; however, the use of an electron beam can modify the shape of the nanoparticle, thus changing the plasmon resonance. Figure 1 details the automated drilling process with dynamic feedback and drift correction for controlled shape modification. Once a region of interest is identified, nanoparticles of interest are selected graphically, and image segmentation identifies individual nanoparticles. The electron beam is positioned in a spiral path, effectively drilling away material for a pre-specified time. An image is collected via a high-angle angular detector (HAADF) and cross-correlated with the initial image to calculate a drift correction. This correction is critical as it keeps the electron beam location consistent. The average HAADF intensity within the drilling region is used as a feedback signal. The drilling continues if the intensity does not meet a pre-defined drop in intensity.

Figure 2 reveals the results from drilling the inside of a single nanoparticle to 0% of its initial HAADF intensity. Drilling via the electron beam is continued for 10 steps after which the intensity in the inside 30% of the nanoparticles decreases to the background value. This effectively shows the manipulation of nanoparticles with real-time feedback to automate the drilling process. Electron energy loss spectroscopy (EELS) collected at iterative steps of the drilling process allows the changes in plasmon resonance to be correlated to the drill depth.

The drilling of nanoparticles provides a framework for electron beam manipulation of other systems such as Si-doped graphene and ferroelectric $BiFeO_3$ opening avenues for tailoring structure property relationships. Furthermore, specific shapes and patterns can be created within various structures and length-scales. This work was performed at the Center for Nanophase Materials Sciences, a US Department of Energy Office of Science User Facility.

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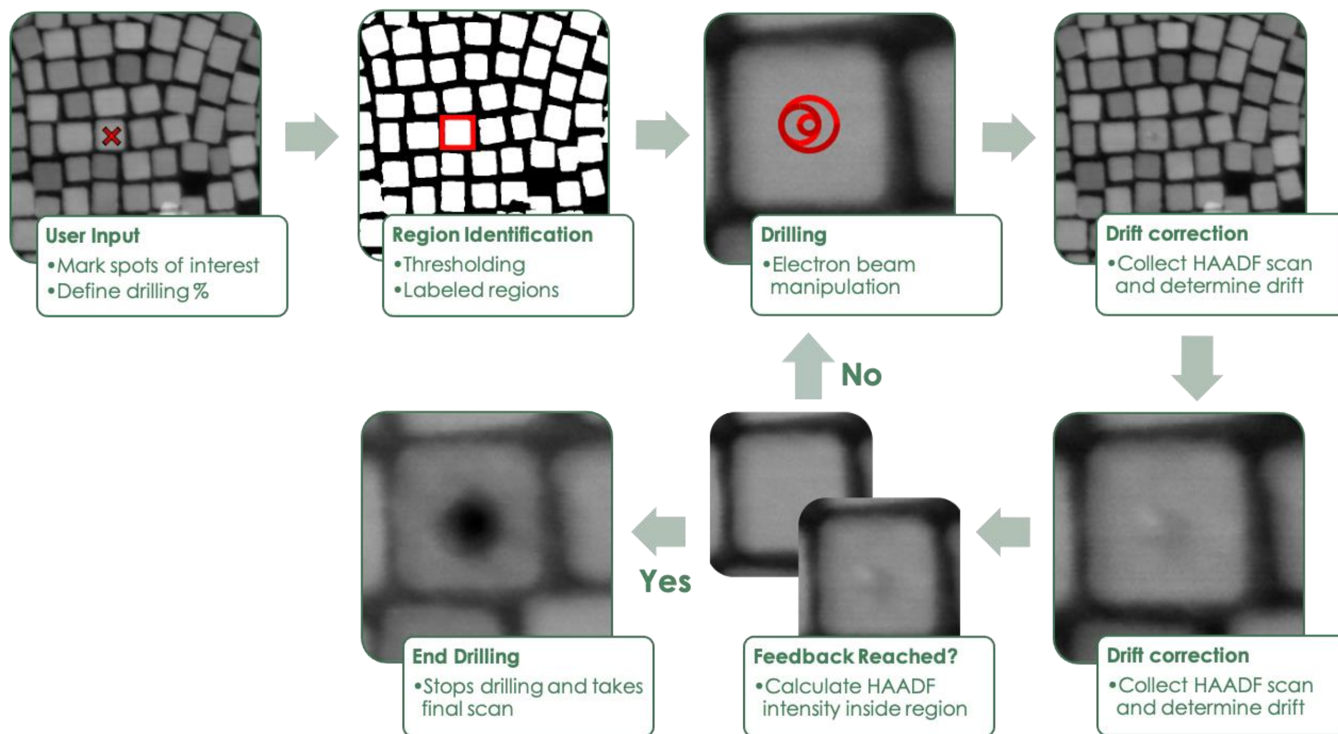

Figure 1. Workflow for automated drilling inside nanoparticles.

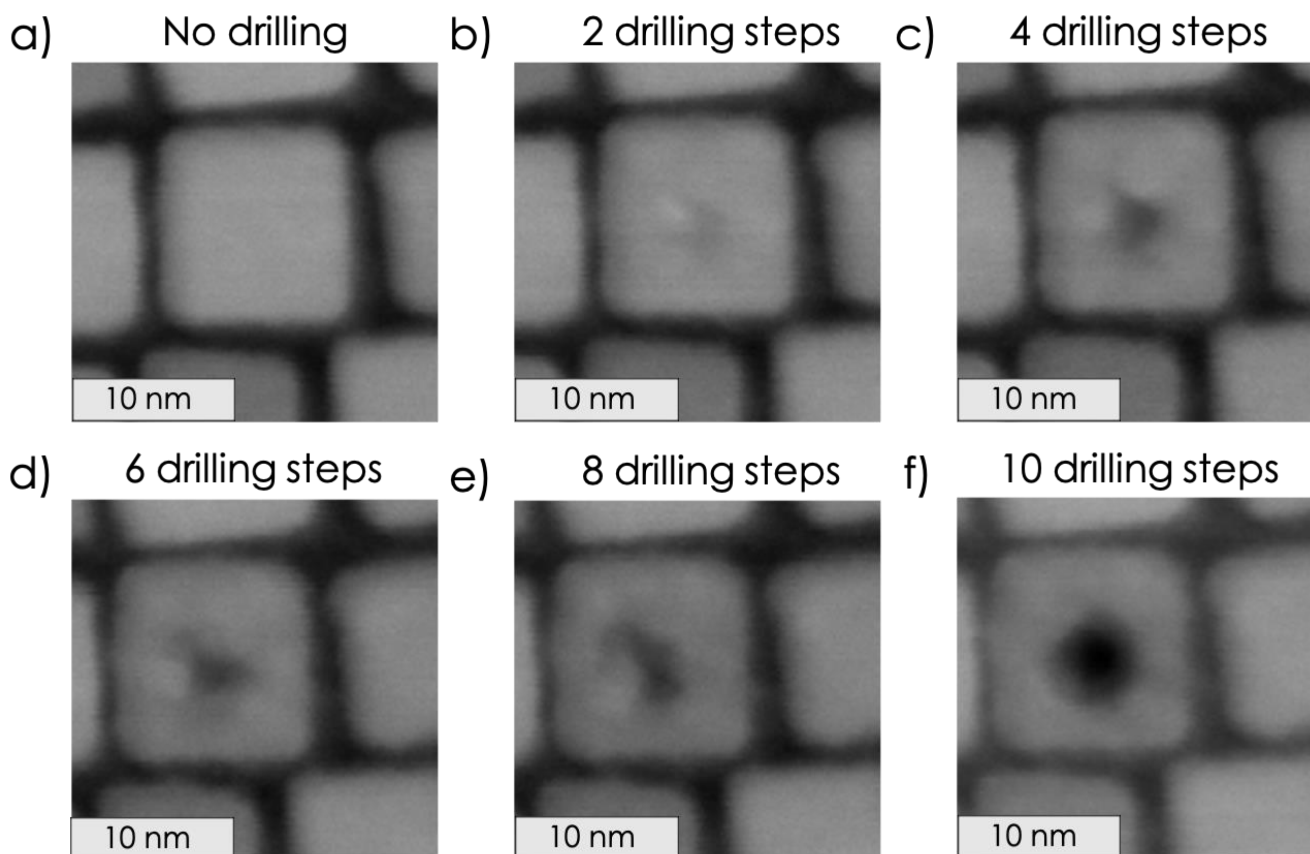

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Figure 2. HAADF images of automated drilling inside a nanoparticle (a) before drilling, after (b) 2, (c) 4, (d) 6, (e) 8, and (f) 10 drilling steps.

References

1. S. J. Pennycook, et al., in *Advances in Imaging and Electron Physics, Vol 153*, (2008), p. 327.
2. S. J. Pennycook, et al., Journal of Electron Microscopy **58** (2009), 87.
3. Y. Jiang, et al., Nature **559** (2018), p. 343.

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