Automated high-resolution phase-contrast scanning transmission electron microscopy

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Scanning transmission electron microscopy is a key tool for characterizing material structure and composition at atomic scales [1]. More recently, the development of high-speed pixelated electron detectors has enabled the collection of full 2D convergent beam electron diffraction patterns for every scan position in a normal 2D raster scan, producing 4D datasets. As well as being used to generate standard STEM images with variably defined collection angles, new 4D-STEM techniques such as center-of-mass [2], ptychography [3], nanoscale strain mapping [4] and STEM holography [5] are now also available.

Despite its widespread usage, however, taking STEM data remains a mostly manual process, limiting the efficiency of microscope sessions and introducing subjective bias into the selection of regions of interest (ROI). With the processing power of modern desktop computers and the availability of open-source computer vision and image analysis software, computers are now capable of deciding which ROIs are worth interrogating during an experiment and where to move a microscope stage next to take more useful data. A major challenge is developing workflows that are sufficiently customizable for the range of samples and experiments conducted in EM investigations.

In this presentation, we demonstrate the operation and applications of a custom-built automation program to acquire high-resolution data on an aberration-corrected STEM [6]. This program is integrated with a high-speed direct electron detector capable of acquiring large 4D-STEM scans with 87,000 frames per second [7]. The program's pipeline architecture allows rapid development of custom workflows based on basic methods, making it highly customizable for different samples and experiments. Figure 1 shows HAADF-STEM images of CdSe/CdS core/shell NPs acquired using this system. ~11,000 NPs were imaged in one 8.5-hour session and ~18,000 NPs were imaged with consistent settings over multiple sessions. Figure 2 shows a 3x3 grid of 4D-STEM datasets optimized for phase contrast taken of dose-sensitive core/shell SrYbF5/CaF¬2 NPs using this system, generating 1.2 TB of data in 15 minutes. This data was suitable for generating atomic-resolution phase contrast STEM images of the sample based on

the center of mass of the probe. This shows the potential to study large amounts of materials at high resolution without human intervention leading to statistically meaningful results. [6]

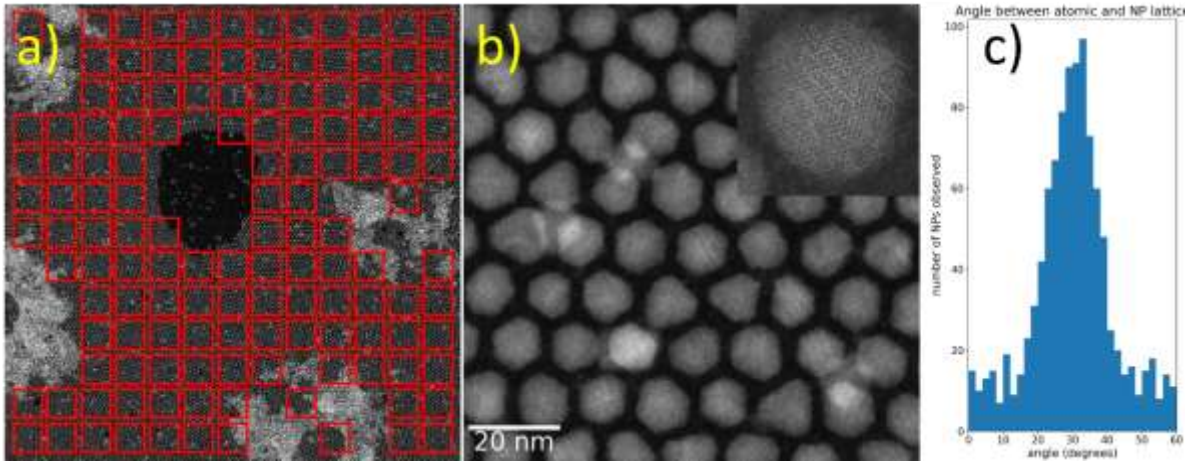

Figure 1: Images from automated HAADF-STEM imaging of CdSe/CdS core/shell NPs. a) Tiling pattern for acquiring images from single-layer regions of the superlattice. b) Atomic-resolution image of superlattice of nanoparticles (inset shows single NP) c) Histogram of the relative in-plane angle for 1081 NPs with the closest [0001] zone-axis alignment within full set of imaged NPs.

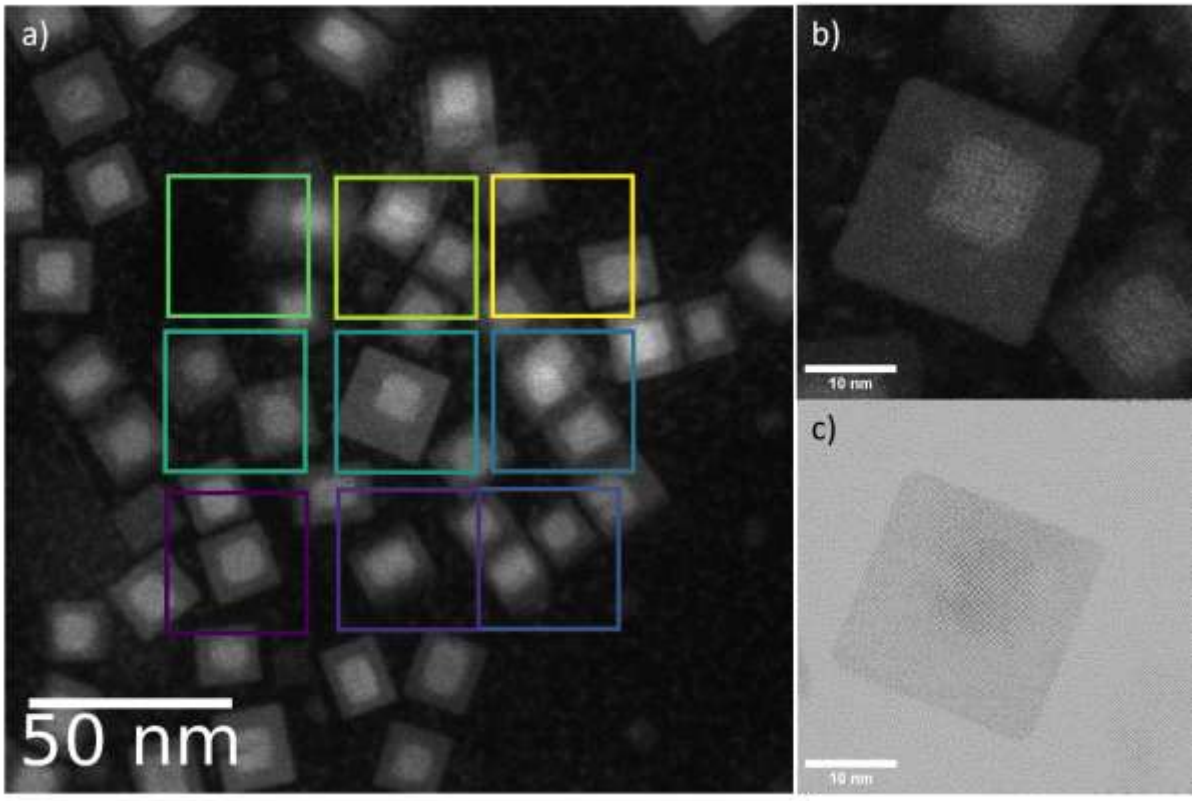

Figure 2: Images from automated 4D-STEM imaging of core/shell $SrYbF_5/CaF_2$ NPs. a) 3x3 grid of acquired 4D datasets overlaid on a low-resolution survey image. b) Simultaneously acquired HAADF-

STEM image from central square in a). c) Phase-contrast STEM image from central square in a) generated from 4D-STEM data.

[1] Tsuda, K, et al., Appl. Phys. Lett. 103, 082908 (2013)
[2] Tate, MW, et al., Microscopy and Microanalysis, 22 (2016)
[3] Nellist, PD, McCallum, BC and Rodenburg, JM, Nature 374 (1995)
[4] Ozdol, VB et al., Applied Physics Letters 106 (2015)
[5] Harvey, T, et al., Physical Review Applied, 10 (2018)
[6] Pattison, A.J. et al., Nanotechnology, 35 015710 (2024)
[7] Ercius, P. et al., arXiv:2305.11961 (2023)
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