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A Facile Method for Improving Quantitative 4D-STEM

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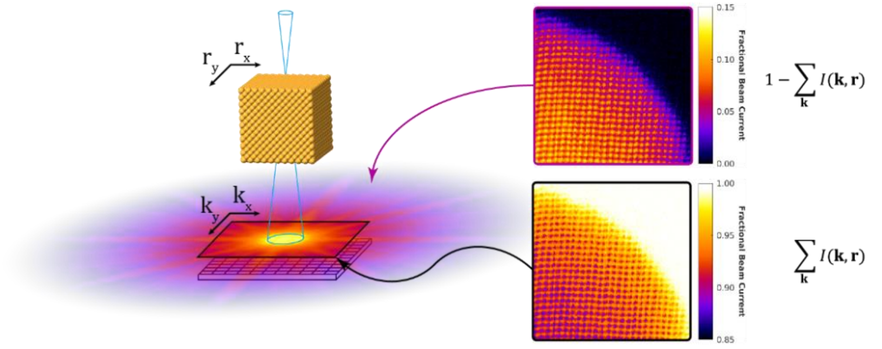
As the scanning transmission electron microscope (STEM) continues to benefit from advances in hardware, such as higher-order aberration correctors, more sensitive detectors, and more powerful computational resources, there is an ever-growing interest in extracting as much quantitative information out of each experiment as possible. The relatively recent development of high-speed direct electron detectors [1–6] has made it feasible to collect diffraction patterns as a function of probe position, generating four-dimensional (4D-STEM) datasets: two real space (r_x, r_y) and two reciprocal space dimensions (k_x, k_y), as seen in Figure 1. Along with such detectors has come the development and advancement of numerous data processing techniques such as electron ptychography, center-of-mass imaging, differential phase contrast, symmetry STEM, S-matrix reconstruction, and many more.[7] All of these processing techniques make use of the additional data collected in 4D-STEM yielding improvements in spatial resolution, measurement sensitivity, and experimental flexibility. Electron ptychography, for example, has demonstrated improved spatial resolution well beyond the limit of the numerical (probe forming) aperture, [8] while the symmetry STEM technique visualizes changes in structural symmetries at the picometer scale, which is otherwise impossible with conventional STEM detectors. [9]

One fundamental method of particular interest is quantitative STEM, whereby the scattered intensity of the electron probe is related to specific specimen properties such as structure, composition, and thickness through comparison with rigorous electron scattering simulations incorporating carefully measured experimental parameters. Such measurements enable nanometer- to pico-scale insights into structure-property relationships and are broadly applicable across the spectrum of materials applications. [10–14] Significant progress has been made in quantitative STEM imaging with conventional integrating detectors, mostly in the high-angle annular dark field (HAADF) regime. This can be attributed to the contrast mechanism in HAADF-STEM, which is strongly dependent on atomic number (often called Z-contrast). While conventional annular detectors have been used to great effect, they can suffer from several limitations, especially detector non-uniformity; whereas, 4D-STEM detectors are sensitive to individual electron strikes without suffering from background noise. Unfortunately, many modern 4D-STEM detectors have relatively few pixels (128x128 or 256x256), which forces a compromise between angular resolution and numerical aperture (maximum scattering angle collected on the detector).

In this paper, we demonstrate a simple method for the acquisition of precisely quantified ADF-STEM images simultaneously with low angle scattered electrons acquired at high angular resolution and dynamic range. This is achieved by recovering electron flux that has been scattered beyond the 4D-STEM detector, and using it to synthesize annular dark field (ADF) STEM images. The left-hand side of Figure 1 shows a schematic representation of 4D-STEM of a metallic nanoparticle, whereby experimental conditions result in a considerable fraction of the electron flux being scattered beyond the limits of the detector. The experimental data presented in the right-hand side of Figure 1 show that the beam current on the detector, $\sum_{\mathbf{k}} I(\mathbf{k},\mathbf{r})$, falls to approximately 85% of the normalized beam current when passing through the [100]-oriented Au nanoparticle specimen. Figure 2 shows the integrated beam current for 20 nm thick [100]-oriented Au specimen (simulated: 15 mrad convergence angle; 300 kV). This demonstrates that for datasets collected with detector numerical apertures less than approximately 200 mrad, there can be significant reduction in detected electron flux. By establishing a robust method for normalizing beam current for 4D-STEM data, the electron flux that falls beyond the detector, $1 - \sum_{\mathbf{k}} I(\mathbf{k}, \mathbf{r})$, can be recovered and used to synthesize ADF-STEM images. We show that such synthetic ADF-STEM images result in higher contrast images that are more reliable for quantitative STEM when compared to images formed strictly by angles subtended by the detector. Additionally, we discuss the impacts of shot noise in the limit of low beam current for dose sensitive applications. We apply this approach to the quantitative measurement of nanoparticle structures [15].



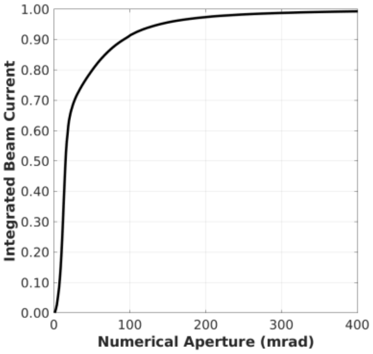


Figure 1. Schematic representation of 4D-STEM *(left)*, where diffraction patterns $\mathrm{I}(\mathbf{k_x}, \mathbf{k_y})$ are collected as a function of probe position $(\mathbf{r_x}, \mathbf{r_y})$, resulting in a 4D dataset $\mathrm{I}(\mathbf{k}, \mathbf{r})$. Experimental conditions can result in the measured normalized electron flux, $\sum_{\mathbf{k}} I(\mathbf{k}, \mathbf{r})$, dropping considerably below unity *(right)*. This flux can be recovered and used to synthesize ADF-STEM images with improved contrast for more robust quantitative STEM comparisons with simulation.

Figure 2. Integrated beam current on [100]-oriented Au as a function of numerical aperture of the detector. Simulated for an aberration-free beam with convergence angle of 15 mrad at 300 kV and a specimen thickness of 20 nm.

References:

[1] MW Tate et al., Microscopy and Microanalysis **22** (2016), p. 237.
[2] P Pelz et al., Microscopy and Microanalysis **27** (2021), p. 188.
[3] H Ryll et al., Journal of Instrumentation **11** (2016), p. P04006.
[4] I MacLaren et al., Microscopy and Microanalysis **26** (2020), p. 653.
[6] GW Paterson et al., Microscopy and Microanalysis **26** (2020), p. 944.
[7] C Ophus, Microscopy and Microanalysis **25** (2019), p. 563.
[8] Y Jiang et al., Nature **559** (2018), p. 343.
[9] M Krajnak and J Etheridge, Proceedings of the National Academy of Sciences **117** (2020), p. 27805.
[10] BD Esser et al., Physical Review Letters **117** (2016), p. 1.
[11] SD Findlay et al., Materials Science and Technology **24** (2008), p. 660.
[12] JM LeBeau et al., Journal of Physics: Conference Series **371** (2012), p. 2.
[13] JM Lebeau et al., Nano Letters **10** (2010), p. 4405.
[14] KHW van den Bos et al., Physical Review Letters **116** (2016), p. 1.

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