Qualitative Phase Contrast Imaging using Interferometric 4DSTEM

Andrew Ducharme^{1,2*}, Philipp Pelz³, Levi Brown¹, Peter Ercius², Ben McMorran^{1,2}

- ^{1.} Department of Physics, University of Oregon, Eugene, OR, United States.
- ² National Center for Electron Microscopy, Lawrence Berkeley National Laboratory, Berkeley, CA, United States.
- ^{3.} Material Science and Engineering Department, University of California, Berkeley, CA United States.
- * Corresponding author: aducharm@uoregon.edu

Phase contrast imaging has been implemented inside the TEM column in the last decade at atomic resolution through differential phase contrast (DPC) and ptychography [1-2]. This imaging mode has higher contrast than more direct TEM imaging techniques, unlocking measurements of low Z and 2D materials and nanoscale electric fields [3-4]. Another route to phase contrast imaging is through structuring the electron beam prior to sample interaction, such as in MIDI-STEM [5], or our technique, interferometric 4DSTEM (ISTEM).

Ptychography and DPC are the current workhorses in this area, but ISTEM similarly works at atomic resolution without the post-imaging computational and theoretical complexity of the former, while producing an absolute phase measurement, unlike the later [6]. Additionally, the ISTEM contrast transfer function is unitary for low spatial frequencies, unlike single-sideband ptychography and MIDI-STEM, which lose all contrast in the same regime [7]. ISTEM uses a diffraction grating to split the electron beam, creating a N-beam Mach-Zehnder interferometer measuring the interference of diffraction orders passing through vacuum with those passing through the specimen. Fig. 1(a) depicts this experimental geometry. Fourier analysis of this N-beam interference pattern, seen in Fig 1(b), produces a qualitative phase at each pixel in the sample plane. Two and three beam interference, one of many terms comprising the overall interference, contains quantitative phase information. Isolating these particular patterns is an active area of research.

Here, we demonstrate the technique by imaging phosphorus nanoribbons inside carbon nanotubes, previously studied with conventional electron imaging [8]. Figs. 2(a) and 2(b) demonstrate the standard HAADF and bright field image acquired simultaneously with the ISTEM image in Fig. 2(c). The phase contrast clearly elucidates the nanotubes against the vacuum far more strongly than typical approaches with less dose. We extract this qualitative view by Fourier transforming the measured interference pattern at each location in a 4DSTEM scan. We have recently implemented a rapid Fourier analysis algorithm, outlined in [9], to accurately compute the phase of the Fourier transformed interference pattern with spatial sampling possible for direct electron detectors and without interpolation [10].

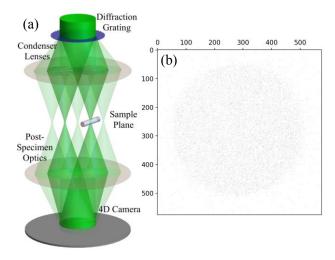


Figure 1. (a) Experimental geometry of interferometric 4DSTEM measurement. (b) Individual frame of a measured ISTEM interference pattern corresponding to a single pixel in final phase contrast image. Axes units are detector pixels.

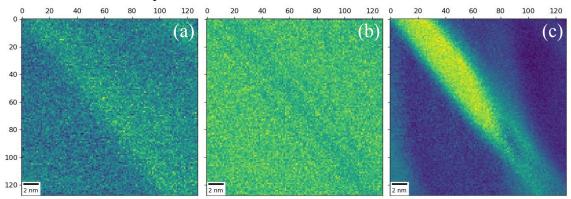


Figure 2. Simultaneously acquired (a) HAADF, (b) bright field, and (c) ISTEM phase contrast images of a carbon nanotube. Axes units are pixels.

References:

- [1] N Shibata et al., Nat. Phys. **8** (2012), p. 611.
- [2] H Yang et al., Nature Commun. 7 (2016), p. 12532.
- [3] A Akshay et al., Nano Lett. **21** (2021), p. 7131.
- [4] JG Lozano et al., Nano Lett. 18 (2018), p. 6850.
- [5] C Ophus et al., Nat. Commun. 7 (2016), p. 10719.
- [6] FS Yasin et al., Nano Lett. **18** (2018), p. 7118.
- [7] TR Harvey et al., Phys. Rev. Appl. **10** (2018), p. 061001.
- [8] J Zhang et al., Nano Lett. **20** (2020), p. 1280.
- [9] C Offelli and D Petri, IEEE TIM **39** (1990), p. 363.
- [10] The experiments were performed at the Molecular Foundry, Lawrence Berkeley National Laboratory, which is supported by the U.S. Department of Energy under contract no. DE-AC02-05CH11231. This material is based upon work supported by the National Science Foundation under Grant No. 2012191.