

Near-Ideal Direct-Electron Focused-Probe 4D-STEM Data for Open-Source Phase Reconstructions

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The availability of direct-electron cameras with high dynamic ranges and very fast detection speeds is revolutionizing the ability of scanning transmission electron microscopy (STEM) to make use of every electron for virtual imaging and advanced computational phase reconstructions. State of the art detectors can now acquire four-dimensional (4D) data at STEM pixel dwell times of over 100,000 diffraction patterns per second while counting each electron. At the same time, the proliferation of open software packages to make use of this data has made such analyses widely accessible, and due to a convergence to the Python programming language, easy to compare in terms of computational efficiency and reconstruction quality. The first commercially available Dectris ARINA detector [1] has been installed in the Nion UltraSTEM100 instrument in Vienna, where an ultra-stable sample stage and flexible electron optics are ideally suited to 4D-STEM. For our initial comparisons, we use an atomically focused probe (34 mrad convergence semi-angle) and choose a camera length optimized for maximum signal in the bright-field and the first-order Bragg disks. In this contribution, we present some of the first data acquired on this new detector, namely convergent-beam electron diffraction maps of pristine monolayer graphene, which is a near-ideal dose-robust uniform atomic phase object. The ability to reliably count electrons at such speeds (the detective quantum efficiency is 0.85 at 60 keV [1]) also enables the variation in beam current to be easily measured and, if desired, corrected for, which we find has an appreciable impact on the bright-field signal and reconstructions that make use of it (most notably parallax imaging [2]). A pixel exposure time of 100 μ s provided a high signal for phase reconstructions without needing to resort to multi-frame averaging. The ARINA is able to bin the native 192 \times 192 detector array in hardware for faster acquisition, and we find that further software binning up to four times does not harm reconstructions, whereas a dense real-space sampling below 0.08 \AA per pixel (512 \times 512 px scan over the 2 \times 2 nm² field of view) was noticeably helpful. The graphic shows the concurrently acquired high-angle annular dark-field (HAADF, 80–300 mrad) and virtual ADF images (\sim 40–80 mrad), as well as a range of open-source phase reconstructions from the binned 4D dataset:

single-sideband (SSB) and Wigner distribution deconvolution (WDD) [3], as well as iterative differential phase contrast (DPC), parallax-corrected bright-field imaging, and batched iterative gradient descent single-slice ptychography [2]. Apart from modest scan distortions, visual inspection of the phase images reveals deviations from the expected uniform atom contrast, and notable differences in phase magnitudes. Computational times also vary greatly depending on the algorithm and the binning. The quality of the phase images is assessed by evaluating the variation of atomic phase shifts using a robust parameter-based quantification method [4] and compared to data simulated with the *abTEM* code [5] and reconstructed with the same algorithms. These quantitative comparisons will be presented at the meeting, where the data and code will also be provided. Further results on defocused-probe datasets and the prospects for live reconstructions will be discussed.

- [1] P. Zambon et al., *Frontiers in Physics* **11**, 1308321 (2023).
- [2] G. Varnavides et al., *arXiv* 2309.05250 (2023).
- [3] T.J. Pennycook et al., *Ultramicroscopy* **151**, 160-167 (2015).
- [4] C. Hofer and T.J. Pennycook, *Ultramicroscopy* **254**, 113829 (2023).
- [5] J. Madsen and T. Susi, *Open Research Europe* **1:24**, 13015 (2021).