Assessing Ptychographic Methods for Maximum Low Dose Performance

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For beam sensitive materials, the dose that can be applied and the dose efficiency of the imaging method often sets the achievable resolution, rather than just the capabilities of the optical system. This is crucial in biology but also for many materials science samples such as metal organic frameworks (MOFs), zeolites, and organic perovskites. While scanning transmission electron microscopy (STEM) has come to dominate high resolution materials science, for the most beam sensitive materials TEM [1] has remained predominant due to the dose efficiency of its phase contrast imaging. Recent technological advances have, however, started to challenge this status quo for low dose imaging, through the combination of fast 4D STEM and ptychography. Ptychography can exceed the dose efficiency of TEM but has been limited until very recently by the relatively slow speed of cameras, which makes 4D STEM exceedingly slow for high probe position count datasets. This bottleneck has recently been overcome using event driven camera technology [2], and conventional framing cameras are also increasingly closing the gap between 4D STEM scan speeds and high-speed conventional STEM, greatly facilitating drift-free and low dose 4D data acquisition.

However, there are many forms of ptychography and so far, sufficient broad comparison of the low dose performance of the various ptychographic methods has been lacking. Ptychographic methods can be grouped into iterative and direct algorithms. Iterative methods are most often performed with a defocused probe, as the higher signal per diffraction pattern facilitates convergence, but with a careful choice of reconstruction parameters they can also produce useful phase images in a low dose focused probe configuration. Here, we present a comparison of a broad spectrum of these algorithms, both iterative and direct, for reconstructing low dose data sets.

Figure 1 compares ptychographic reconstructions of 2.5 nm thick Methylammonium lead iodide (MAPbI3) at a dose of 50 e-/A², using a convergence angle of 13 mrad, with a focused probe. The iterative gradient descent (GD) algorithm converges, but to a relatively poor quality image showing very little atomic structure. The rPIE, ePIE [3], ER and WASP [4] iterative and direct SSB algorithms, all perform better under these conditions, revealing relatively similar atomic structures from which the iodide and the Pb sites can be located. On the heavy Pb atomic sites, contrast reversals are present. These reversals can be easily corrected using our new phase offset method [5], which can greatly improve the interpretability of ptychographic data.

Switching to a defocus of 20 nm, as shown in Figure 2, improves the GD result significantly, however it remains dominated by low frequency features of limited utility, with high resolution information still less clear than the other iterative methods. Although the SSB becomes blurred when the probe is defocused, as expected due to the optical sectioning effect, this can be corrected post collection as we show in the figure.

In addition to the choice of algorithm, the parameters used are critical to obtaining the best solutions. For iterative schemes, probe step size and defocus are linked in their importance to the amount of overlap of adjacent illuminated regions. The convergence angle determines which frequencies will be transferred most strongly and sets the upper limit on conventional image resolution. Although ptychography can provide super-resolution and exceed this limit, at very low doses there is insufficient scattering to higher angles for super-resolution to provide significant benefits; rather, at low doses, correction of residual aberrations is expected to play a greater role.

Graphic:

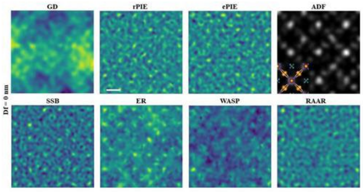

Fig. 1. GD, rPIE, ePIE, ER, WASP and SSB reconstructions at 50 e^-/A^2 dose using a 13 mrad convergence angle shown alongside the annular dark field (ADF) images using a focused probe. Scale bar is 3 Å.

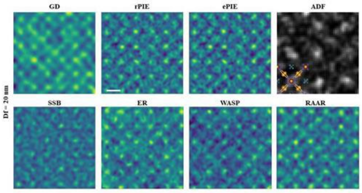

Fig. 2. GD, rPIE, ePIE, ER, WASP and SSB reconstructions at 50 e^-/A^2 dose using a 13 mrad convergence angle shown alongside the annular dark field (ADF) images using a defocused probe of 20 nm. Note that the SSB image for the defocused probe has had the defocus corrected post collection. The iterative methods produce noticeably stronger atomic contrast with the defocus. Scale bar is 3 Å.

Keywords:

Ptychography, Low-dose, 4D STEM

Reference:

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