Efficient Phase-contrast Imaging via Mixed-state Electron Ptychography: From Crystal Structures to Electromagnetic Fields

Zhen Chen¹, Yi Jiang², Michal Odstrcil³, Yimo Han⁴, Gregory Fuchs¹ and David Muller¹

¹Cornell University, Ithaca, New York, United States, ²Argonne National Laboratory, Lemont, Illinois, United States, ³Paul Scherrer Institut, Villigen PSI, Aargau, Switzerland, ⁴Rice University, Houston, Texas, United States

Phase-contrast imaging plays a crucial role in imaging many types of materials, especially those with weak scattering potentials or low damage-threshold doses. With the recent advances in detector technology, phase-contrast imaging techniques in scanning transmission electron microscopy (STEM) are experiencing a renaissance. One widely used method, differential phase contrast (DPC) imaging, has been applied both for the atomic resolution crystal structures and nanometer scale electromagnetic field. However, accurate phase or field quantification via DPC requires caution as the probe shape effects can be very significant, especially at atomic resolution [1-2]. Ptychography via a Wigner-distribution deconvolution (WDD) method allows for correction of small residual geometric aberrations [3] but structural information beyond the aperture-limit has yet to be realized. Iterative ptychographic algorithms such as ePIE [4] or maximum likelihood (ML) [5,6] have the flexibility to correct a wide range of experimental uncertainties. Recently, using a high-dynamic-range electron microscope pixel array detector (EMPAD) [7] and ePIE ptychography with an in-focused probe illumination, we have achieved a 0.39 Å resolution phase-contrast image at 80 keV, 2.5 times that of the aperture-limited resolution for STEM annular dark-field (ADF) images or standard WDD [8].

Broadened illumination with a defocused probe for ptychography can usually reduce the effort in data acquisition. However, conventional ptychography algorithms assuming a coherent probe do not work well for datasets with a defocused probe (Figure 1b) and even fail to produce physical structures when using a modest scan step size. After accounting for the partial coherence of the probe via a mixed-state model [9], the reconstructed phase image improves significantly (Figure 1c). In addition, the probe shape beyond an aberration-free focused probe, such as the defocused probe shown in Figure 1d, can be readily reconstructed, which opens doors to new phase imaging techniques with more advanced probe designs. We also achieve simultaneously sub-angstrom resolution, high contrast/SNR and a large field-of-view phase contrast imaging at a low dose condition [10].

As ptychography can correct the image distortions from the probe, it is even more important for imaging of magnetic structures. Because magnetic imaging usually requires a magnetic field-free condition for preserving the intrinsic magnetic structures, and thus the main imaging forming lens, the objective lens is replaced by a more distant lens, whose longer focal length results in larger aberrations and limits the spatial resolution to a few nanometers. We show a ptychographically-reconstructed phase image in Figure 2a with a phase precision better than $2\pi/3100$ rad. The high precision enabled us to directly image the 2D discontinuities of the magnetization (Figure 2b-c). Verified by simulation, we find that ptychography can give about two times better field precision than the conventional center-of-mass imaging with a low-dose illumination (Figure 2d). [11]



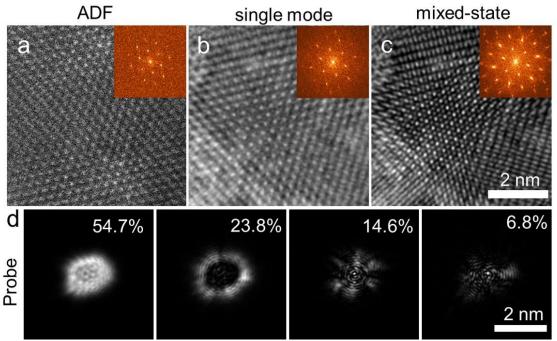


Figure 1. Figure 1. Resolution and signal-to-noise improvement via mixed-state ptychography using a MoSe₂/WS₂ bilayer sample. (a). Annular dark-field (ADF) image using a similar dose as for ptychography; (b). Phase image from single mode ptychographic reconstruction; (c). Phase image from mixed-state ptychographic reconstruction with four probe modes; (d). Reconstructed probe intensity distribution of the four orthogonal modes.

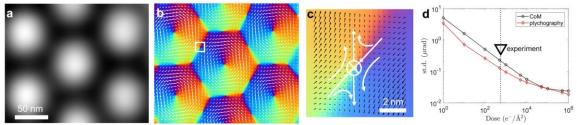


Figure 2. Figure 2. Magnetic field of a magnetic skyrmion lattice from Lorentz electron ptychography in FeGe single crystal. (a). Phase image induced by magnetic skyrmions reconstructed from ptychography; (b). Magnetic induction field calculated from the phase image in (a); (c). Enlarged image from the rectangular region marked on (b); (d). Simulated dose dependence of magnetic field precision for center-of-mass (CoM, black line) and ptychography (red line). The experimentally-measured precision from (b), shown as a triangle on (d), is 0.6 μrad.

References

- [1] K Müller et al., Nature Communications 5 (2014), p. 5653.
- [2] MC Cao, et al., Microscopy, 67 (2018), p. i150-i161.
- [3] H Yang, et al., Nature Communications, 7 (2016), p. 12532.
- [4] A Maiden, et al., Ultramicroscopy, 109 (2009) p. 1256-1262.
- [5] P Thibault, et al., New Journal of Physics, 14 (2012), p. 063004.
- [6] M Odstroil, et al., Optics Express, 26 (2018), p. 3108-3123.
- [7] MW Tate, et al, Microscopy and Microanalysis 22 (2016), p. 237.

- [8] Y Jiang, et al, Nature 559, (2018), p. 343.
- [9] P Thibault, et al., Nature, 494 (2013), p. 68-71.
- [10] Z Chen, et al., under review on Nature Communications (2020).
- [11] We thank M.-H. Chiu, Lance Li, M. J. Stolt, and S. Jin for samples. Research supported by US NSF (grants DMR-1539918, DMR-1429155, DMR-1719875) and US DARPA (TEE-D18AC00009).