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4D-STEM at atomic resolution: past, present and future

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The development of high-speed pixelated and segmented detectors for the scanning transmission electron microscope (STEM) has allowed for the emergence of the technique of 4D-STEM. In this technique, the STEM detector plane (which is in two-dimensional reciprocal space) is recorded at each of a set of STEM probe positions (in real space). Here, we use the term 4D-STEM to refer to the use of segmented detectors, such as a quadrant detector, and not restricted to fully pixelated cameras. Several closely related techniques have emerged that can process the 4D-STEM dataset. Here we will focus on ptychography, but making appropriate links to other approaches where appropriate.

Electron ptychography was proposed in the late 1960s by Hoppe as a method to solve the phase problem in electron scattering. Its first practical implementation led to a demonstration of resolution beyond the diffraction limit [1] in phase imaging. With more recent developments in detector technologies, dramatic improvements in spatial resolution have been demonstrated [2]. The influence of partial coherent on ptychography is very different to phase imaging in a conventional TEM [3] and we will review why this enables high-resolution, but more importantly why the deleterious effects of chromatic aberration are much less severe in ptychography enabling high-resolution at low beam energies without a monochromator [4]. This discussion will lead to an examination of sampling in the detector plane and how many detector pixels are required. We will also explore a mathematical framework to enable comparison between different imaging modes which shows that ptychography can never be as beam efficient as a perfect Zernike phase plate microscope.

Looking ahead, we will explore the difficulties associated with quantification of phase images, suggest some possible strategies for such quantification and examine some of the new opportunities that may arise. The importance of multimodal approaches will be highlighted. The presentation will be highlighted by a range of applications including recently to Li-ion battery cathodes [5] and polymer crystals [6].

References:

[1] Nellist et al., *Nature* **374** (1995) 630.
[2] Chen et al., *Science* **372** (2021) 826.
[3] Nellist et al. *Ultramicroscopy* **54** (1994) 61.
[4] Pennycook et al., *Ultramicroscopy* **196** (2019) 131.
[5] Song et al., *Joule* **6** (2021) 1049.
[6] Hao et al., *Polymer* **284** (2023) 126305.

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