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Exploring the Validity Limits of Direct Ptychographic Methods to Analyse 4D Scanning Transmission Electron Microscopy Datasets

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In a focused-probe scanning transmission electron microscope (STEM), using a pixelated detector allows us to collect an information-rich 4D dataset: recording a 2D scattering pattern for each probe position in a 2D raster scan. Compared to a conventional bright-field or dark-field STEM scan, this 4D-dataset is densely sampled and thus interrogation of this data can reveal more information about the specimen through which the scattered electrons have been transmitted.

Among the information to which this data enables access, is the complex specimen transmission function. This is significant as important specimen features such as electric and magnetic fields manifest in the phase of the specimen transmission function and are challenging to detect otherwise [1]. The complex transmission function can also reveal details such as light (weakly-scattering) atoms, which can be difficult to visualise by other means, particularly if they are near to a heavy column [2].

Direct (i.e. non-iterative methods) to retrieve the complex specimen transmission function include integrated centre-of-mass (iCoM), single sideband (SSB) ptychography and Wigner distribution deconvolution ptychography (WDD). Each of these methods makes a phase object approximation, which is to neglect dynamically scattered electrons. This may be an accurate model for some very thin samples, but is increasingly inaccurate for thicker and denser materials. Realistic specimens studied in the STEM can easily fall into this inaccurate regime and so it is critical that the behaviour of these imaging methods is studied in cases where the phase object approximation is an imperfect model. In this work, we study each of these three analysis methods through a simulated thickness series of 4D-STEM datasets. We find that the ptychographic methods are more robust than other imaging tools, likely due to the more efficient use of the information available within the data.

One way the breakdown of the phase object approximation can be measured is in the phase structure of the overlap region within the Fourier-transformed data. Illustrated in Fig 1 are example overlap regions. If the weak phase object approximation holds completely true, the phase in this region is flat. The left-most subfigures show overlap regions in data from a very thin sample – the phase varies only very slightly here (note the narrow range of the colour bar limits). In thicker samples (the central and right-most figures), the phase varies much more rapidly within the overlap region. We further demonstrate that by optimization of focal condition, the range of applicability of these methods can be broadened (cf. upper and lower row of Fig 1).

We further demonstrate that these methods are also applicable in a four-dimensional TEM dataset, through application of the principle of reciprocity, as indicated in Fig 2, and discuss the range of cases in which this may be a preferred method for acquiring multidimensional information about a specimen.

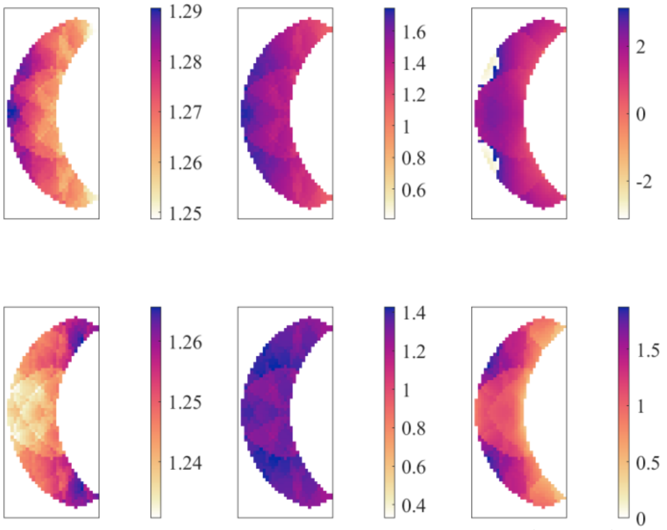

Figure 1. Illustration of phase interference in overlap regions for 3 Å, 31 Å and 175 Å thick samples (left to right) in the top surface focus case (upper panel) and the mid-plane focus condition (lower panel). Colour bars in radians, perceptually uniform colour map following Ref [3].

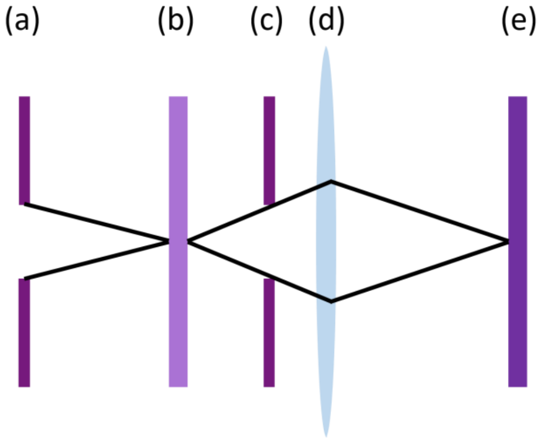

Figure 2. Schematic to illustrate the principle of reciprocity. One can read the diagram left-right as a TEM: (a) condenser aperture, (b) specimen, (c) objective aperture, (d) objective lens, (e) detector; or right-left as a STEM: (e) source, (d) objective lens, (c) aperture, (b) specimen, (a) detector.

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