Phase Contrast Imaging of Weakly-Scattering Samples with Matched Illumination and Detector Interferometry – Scanning Transmission Electron Microscopy (MIDI-STEM)

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Aberration-corrected scanning transmission electron microscopy (STEM) has made an enormous impact on materials science, with recent examples including observations of nanometer-scale polar vortices in oxide superlattices [1], atomic-scale chemical imaging [2], atomic-resolution 3D tomography [3], and many others. However, the majority of these studies image chemical species with intermediate or high atomic numbers. Samples composed primarily of low atomic number species are more difficult to study in STEM due to their poor scattering efficiency. This is why the vast majority of biological research in electron microscopy (EM) uses phase contrast plane-wave methods, including CTF-corrected cryo-EM [4] phase-plate HRTEM [5], etc. Phase contrast methods can also be used in STEM to improve contrast in weakly-scattering samples, such as ptychography [6, 7]. These methods improve contrast, but require significantly more involved processing of the experimental data.

In this study we describe a new form of phase contrast imaging, called matched illumination and detector interferometry (MIDI)-STEM. The setup of a conventional STEM experiment is shown in Figure 1a, where a converged electron beam is moved over the sample surface in a grid pattern to form an image using an annular detector (shown in Figure 1c). The MIDI-STEM experimental modifications, shown in Figure 1b, are twofold; first we place a phase plate in the probe-forming aperture above the sample. Secondly we use a high-speed direct electron detector (the Gatan K2 IS) to record a diffraction-space image of the probe at each position on the sample surface, shown in Figure 1d. The MIDI-STEM imaging mode is very simple, and is performed independently at all prove positions; we apply a virtual detector to each diffraction space image of the probe, specifically we measure the difference in electron intensities between the sum of all even rings and the sum of all odd rings.

For a given imaging process, the contrast transfer function (CTF) describes how all spatial frequencies are transferred into the recorded image. The CTFs of a MIDI-STEM experiment depend on the geometry of the phase-plate used in the probe-forming aperture. Several examples of MIDI-STEM phase plates are shown in Figure 1e, with the corresponding CTFs plotted in Figure 1f. The ideal CTF is generated by applying a phase shift of $\pi/2$ to the unscattered center disk of the electron probe, similar to how a phase plate is used in HRTEM [5]. This configuration cannot be realized in reality, but can be closely approximated by almost any MIDI-STEM geometry, demonstrated in Figure 1f. The only requirements for MIDI-STEM are that the probe consists of two equal area regions with phase shifts of 0 and $\pi/2$, and that the geometry is known to high precision, so that a matching virtual detector can be applied.

We have performed a proof-of-principle MIDI-STEM experiment, using the probe and fitted virtual

detector shown in Figure 1g to image gold nanoparticles on an ultrathin amorphous carbon substrate [8]. In this experiment we have compared a MIDI-STEM image (Figure 1h) to a conventional STEM image (Figure 1i). Both images show good contrast for the strongly-scattering nanoparticles. The weakly-scattering substrate is essentially invisible in the conventional STEM image, but produces strong contrast in the MIDI-STEM image. Figure 1j and corresponding line traces in Figures 1k-1m directly compare the contrast in the amorphous sample regions between MIDI- and conventional STEM. [9]

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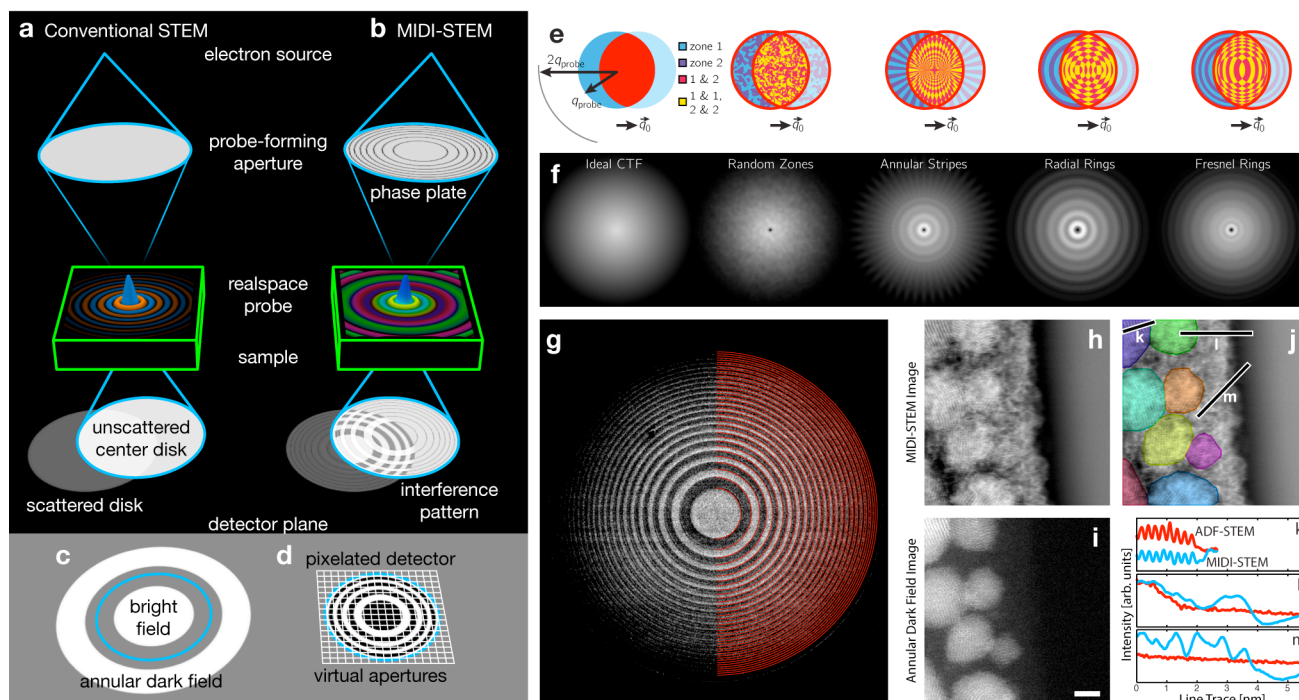[8] C Ophus, J Ciston, J Pierce et al., article in press, Nature Comm. (2016).

Figure 1. Setup of (**a**) conventional STEM experiments, and (**b**) MIDI-STEM experiments. (**c**) Typical annular detectors used for STEM experiments. (**d**) High-speed, pixelated detector capable of recording the whole diffraction pattern at each electron probe position, used for MIDI-STEM or ptychography experiments. (**e**) Some possible MIDI-STEM probe phase-plates, and (**f**) their corresponding CTFs in two dimensions. (**g**) Experimental mean diffraction image of MIDI-STEM probes, with virtual detector edges shown on right. (**h**) Simultaneously recorded MIDI-STEM image compared to (**i**) conventional annular dark field STEM image of gold nanoparticles on an amorphous substrate. (**j**) Same image as (**i**), with nanoparticles highlighted and line traces shown in (**k**), (**l**) and (**m**). (**g**)-(**m**) reproduced from [**8**].