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# Applications of Phase-Contrast STEM as Dose Efficient Method for High-resolution Imaging of Soft Materials

Roberto dos Reis[1], Anahita Pakzad[2], Paul Smeets[1] and Vinayak Dravid[1]

[1]Northwestern University, Evanston, Illinois, United States, [2]Gatan Inc, Pleasanton, California, United States

Radiation damage is a constant concern in structure analysis at high resolution using energetic beams [1]. In addition, for soft materials, high-energy electrons are predominantly only scattered once in the crystal structure (conditions described as a weak-phase object) and, consequently, only a minuscule phase shift is introduced to the electron wave that is further collected [2]. Thus, making the most of the acquired signal while preserving sample integrity is crucial to obtain structural information from soft materials.

Conventional transmission electron microscopy (CTEM) - a plane wave illumination - has been vastly employed to observe contrast from such low scattering samples. CTEM is typically performed with a combination of defocus and spherical aberration that optimizes contrast and it is still the preferred technique in fields such as molecular biology. For example, the popularization of cryo-EM, highlighted by the 2017 Nobel Prize in Chemistry, has stimulated the pursuit for enhancements in phase-contrast imaging; e.g. by exploring different phase plates configurations [3]. On the other hand, the analytical capabilities of STEM mode, such as the annular dark field (ADF) and spectroscopic signals, have made STEM an indispensable characterization tool in physics and materials science. Therefore, the ability to perform phase imaging in STEM can take advantage of its multimodal characteristics.

A major advance in data acquisition for phase imaging in STEM has come from the development of direct-detection cameras that offer fast acquisition with electron counting, consequently improving the signal to noise SNR. Such characteristics enable the recording of weaker signals at higher scattering angles in each Ronchigram within a ptychographic dataset. Ptychographic phase-contrast imaging in STEM, for example, has been applied to obtain simultaneous images from soft-hard interfaces [4-7]. In addition, it has been shown that for weakly scattering specimens, ptychography can provide superior dose efficiency compared to CTEM for high-resolution imaging [8,9].

In this work, we leverage the potential of ptychography as a dose efficient method for direct imaging of soft materials, in particular, MOFs, COFs, and biomolecules. To this end, we explore different acquisition geometries in ptychography using focused (Fig. 1a) and defocused (Fig. 1b) probes. Acquisition of datasets with focused probes has the advantage of keeping the conditions required for incoherent imaging, allowing for simultaneous acquisition of HAADF images and chemical information. However, it requires a larger number of probe positions consequently increasing the exposure time and electron dose-rate applied. The highly convergent beam used in STEM broadens into discs in the far-field (Ronchigram). The phase information is found where these discs overlap (Fig. 1a)

On the other hand, defocused probe ptychography has the advantage of largely reduce the electron dose rate [9,10] achieved by the use of larger probes, in detriment of the multimodality aspect of STEM. The probe is defocused (equivalently to move the specimen up or down in relation to the beam focus). Such configuration results in a combination of near-field ptychography and the focused probe setup. The central disc (ronchigram) is essentially a Gabor hologram. Since defocused ptychography covers a large field of view, errors in the probe positions that denigrate the reconstruction of the phase are easily detected within the ronchigram.

Figure 2 shows examples of experimental datasets using both configuration geometries using recently released K3 IS in counting mode. The acquisition of 4D-STEM datasets in counting mode opens the opportunity to further reduced the electron dose rate necessary to obtain meaningful information from a ptychographic dataset in both configurations. Fig. 2 (a) shows a single CBED acquired from a MOF structure using a focused probe (df = 0nm) with a convergence angle of 27.1 mrad. Fig. 2 (b) shows single defocused CBED (df = 200nm) acquired from a mosaic COF film using a similar probe size (27.1 mrad).

We will present variations in the experimental setup (e.g. total dose, dose-rate, number of overlapping probes) to evaluate the optimal condition to acquire ptychographic datasets for imaging of soft materials.[11]

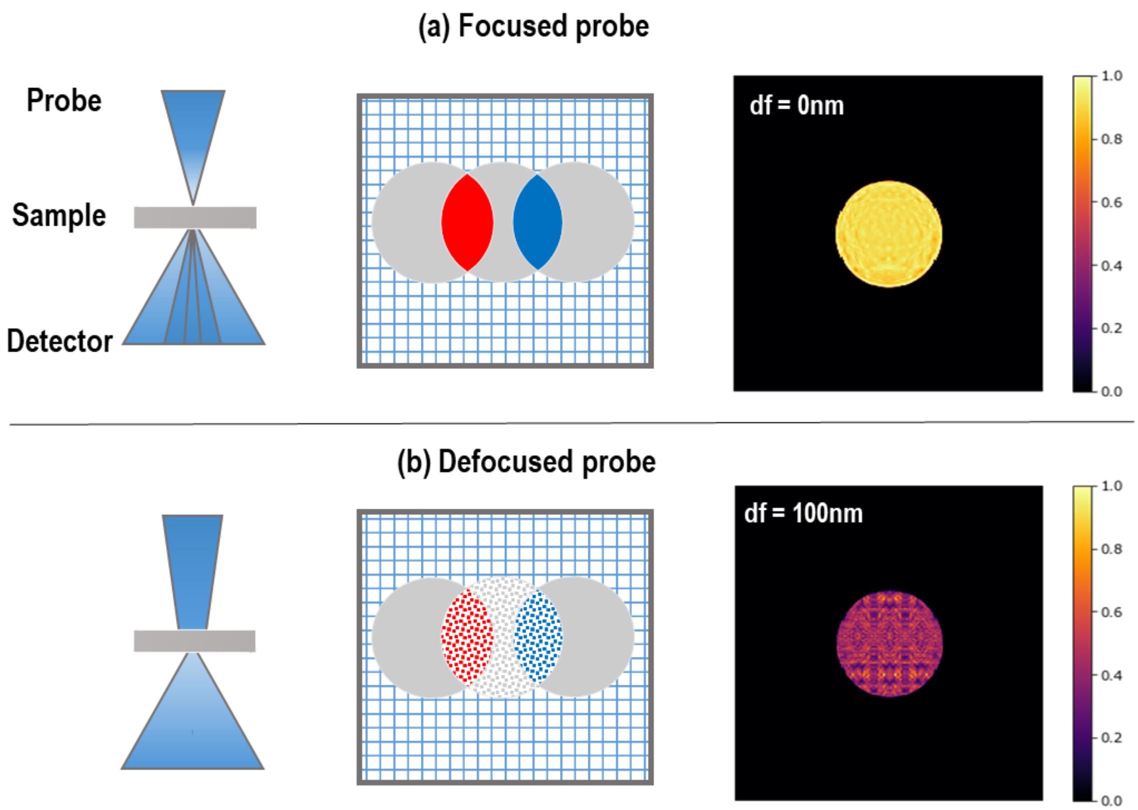


**Figure 1.** Geometries for acquisition of ptychographic datasets. In a focused probe geometry (a), the beam-crossover is formed in the sample plane, producing a ronchigram in the far field. The phase information is found where these discs overlap (red/blue areas). In a defocused probe geometry (b), an undistorted near-field image of the sample is observed within the central disc. Far right column shows simulated ronchigrams to represent both configurations.

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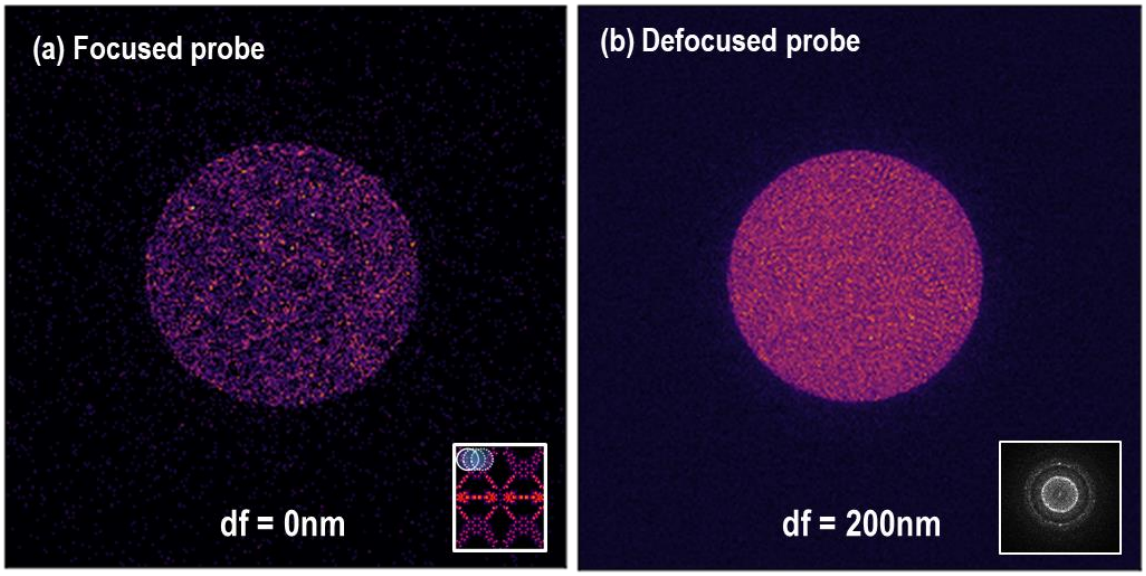


**Figure 2.** Experimental single ronchigram obtained using two different configurations, focused (b) and defocused (b) probe. Inset in (a) represents a typical density of overlapped discs needed to obtain high-resolution phase image. Inset in (b) shows Fast-Fourier-transform of the structure within the ronchigram

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