

Ultra Low Dose Phase Contrast and Retrieval Imaging in Scanning Transmission Electron Microscopy via Electron Counting

by

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2025

Declarations

I declare that this thesis has not been submitted as an exercise for a degree at this or any other university and it is entirely my own work.

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A handwritten signature in black ink, reading "Julie M Bek". The signature is fluid and cursive, with the first name "Julie" and the last name "Bek" being more prominent than the middle initial "M".

Julie Marie Bekkevold

Abstract

Scanning transmission electron microscopy (STEM) is an incredibly powerful tool for characterising materials to inform product development and solve mysteries in materials and biological sciences alike. In STEM, a focused electron beam is scanned across a region of interest on a sample and signals from the electron interactions with the sample material are collected to characterise it [1]. In recent decades, STEM has been used to probe both structural and functional properties of materials enabling important innovations in technologies for a more sustainable future. Traditionally, these insights have been gained by collecting the electrons that have been scattered to higher angles by the sample material, typically termed high-angle annular dark field (HAADF) STEM. However, the scattered intensity is proportional to $Z^{1.7}$, where Z is the atomic number, hampering the visibility of lighter elements. Additionally, the collection efficiency is poor making HAADF unsuitable for imaging beam sensitive materials where the electron dose impacting the sample must be reduced.

A candidate for high collection efficiency detection is four-dimensional scanning transmission electron microscopy (4D-STEM), where an image of the convergent beam electron diffraction (CBED) pattern is captured at every scan position [2]. This yields a four-dimensional dataset with a 2D image for every position in a 2D image, which contains an immense amount of information about the sample. However, most of the information about weakly scattering materials, like battery materials, metal organic frameworks (MOFs), and biological specimens, is contained in the phase modulation imposed on the electron beam. Since physical detectors are unable to detect complex information, all phase information in the electron wavefunction is lost upon detection. There are several techniques for retrieving this phase information from the detected signal, the simplest of which utilises the fact that the center of mass (COM) of the CBED pattern is proportional to the gradient of the relative phase shift applied by the sample. Thus, the integrated center of mass (iCOM) retrieves the phase information and, due to the intensity being linear in Z and the high collection efficiency, enables characterisation of materials at significantly lower doses than traditional HAADF [3].

For ultra-low-dose imaging it is useful to scan the same area on the sample many times using a very fast scan speed so that the beam exposure is spaced out in shorter bursts to delay sample damage [4]. While 4D-STEM is very powerful, it significantly

bottlenecks the practicable scan speed due to the high read-out overhead associated with reading out a full image at each scan position. To address this, this thesis focuses on the use of segmented detectors for estimation of the COM signal while scanning with extremely low dwell times below 500 ns. At these speeds, the finite detector response function to each electron detection event may cause artefacts in the recorded images. Therefore, a signal digitisation hardware is used to digitise the detector signal in real-time, such that the temporal position of each electron detection event – and thus the integrity of the recorded image – is maintained.

Finally, this thesis investigates the usefulness of segmented detectors and simple detector geometries for phase retrieval using iterative ptychography. The investigations presented paves the way for robust characterisation, with ultra-high resolution and dose efficiency, for beam sensitive materials.

Preface

This thesis marks the conclusion of three years of a postgraduate research journey at Trinity College Dublin. The experimental work for this thesis was carried out in Trinity College Dublin, and at the University of Tokyo and Stanford University.

This work would not have been possible without the support of the staff at the Advanced Microscopy Laboratory. In particular, I wish to thank Clive Downing for his patient guidance at the Titan — whether I had accidentally crashed the vacuum, or simply needed someone in the room for moral support. Clive also fabricated the DPCable STO sample that became the foundation of all my experimental work, giving me valuable experience with a real sample to test parameters on. I am truly thankful for your generosity and expertise, Clive.

Most of the experimental data presented in this thesis were collected at the Electron Microscopy & Materials Science Laboratory, University of Tokyo. I am deeply grateful to Professors Yuichi Ikuhara, Naoya Shibata, and Ryo Ishikawa for hosting me and for helping me do the digitisation experiments with their detectors.

I am equally grateful to Dr. Stephanie Ribet and Dr. Georgios Varnavides at Lawrence Berkeley National Laboratory, and Andrew Barnum and Professor Colin Ophus at Stanford University for doing the experiments for segmented detector ptychography with me. Furthermore, I wish to thank Dr. Danielle Douglas-Henry for preparing the PtSe₂ samples for me, and for all her great advice.

This project was funded by Research Ireland under Grant No. 19/FFP/6813. Additional travel support was provided by the Royal Microscopical Society, the Microscopy Society of Ireland, the European Microscopy Society, and the Institute of Physics. Without these travel grants, I would not have been able to attend and present at so many conferences — experiences that shaped both the scientific content and the scope of this thesis.

With all my thanks and appreciation,



Julie Marie Bekkevold, 18th July 2025

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The Norwegian singer Ingrid Bjørnov once dedicated a full hour on the radio to beginnings, and I can relate. I love beginnings — new ideas, new lab books, new samples, new data, new code, new notebooks. The possibilities are endless with a brand new notebook and a fresh start. Finishing, however, has always been harder. But even the most chaotic code can run just fine, and I’ve come to learn that progress very rarely looks polished: more often than not it’s a long train of unfinished thoughts, and the meaning of it all becomes clear later.

A PhD is a long time to spend with your own unfinished thoughts. So first and foremost I wish to extend a huge thank you to those who gently reminded me to finish what I started, and who saw value even in the not-quite-polished pieces.

I’m especially grateful to my supervisor, Professor Lewys Jones, who always knew how to pull a narrative thread through my chaos. Thank you for being my guide, champion, and voice of clarity. From our very first conversation — when you said, “We need your expertise” — to nominating me as an invited speaker at M&M 2023 and sending me off to the University of Tokyo to collaborate independently. You always showed immense confidence in me, which also helped me to have confidence in myself. Your strategic instinct for wrapping ideas into coherent stories helped me see when it was time to let go, write up, and move on. Thank you for your guidance, and thank you for giving me the space to find my own research interests.

To my parents, thank you for being my foundation and for always having my back. *I know you were on my side, even when I was wrong.* And I can never thank you enough for that! You’ve always believed in me, whether I was calling triumphantly from the other side of the planet or crying on the couch at the cabin. I am immensely grateful that I can always hop on a plane back home to you — “det er aldri lenger enn bak en sky heim”. Your quiet strength and encouragement has carried me through more than you know.

I would not have been where I am today if it had not been for the love and support that I got from family, friends, and the postgraduate support system in Trinity College Dublin. Amongst many friends who have been important to me along the way, I would like to extend a particular and huge thanks to Marthe — my conference buddy, karaoke partner, and voice note confidante — I’m so glad I

got to share the chaotic and wonderful experience of my first international conference with you. The memories from Minneapolis 2023 will live on forever! To Tigran, thank you for being a grounding presence in the early days, and for reassuring me — a hundred times over — that I was doing just fine. To Patrick, thank you for being my rubber duck. And to Steph, thank you for all the rubber ducks — and for helping with the final experiments.

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Last, but certainly not least: To my dearest George — thank you for coming into my life while I was still bright-eyed and bushy-tailed. But thank you even more for staying when I slowly fell apart. Thank you for believing in me when I couldn't see the point, for gently encouraging me to keep going when I wanted to give up, and for making space for both my joy and my overwhelm. I'm endlessly grateful for the calm you brought into my life, and for the home you and Kate gave me across the ocean. A home filled with care, laughter, venting sessions, and excellent dinners. A home that gave me room to finish this final chapter while slowly healing. This thesis would have been finished eventually, but not like this. Not with this much peace. Not with this much dignity. Thank you both, from the bottom of my heart, for everything.

There is a Norwegian saying: “Veien blir til mens du går” — The road is made by walking it – and I walked it, often without knowing where it would lead. There were days when I wanted to rewrite the whole map, and days when I forgot why I set out in the first place — but in time, the pieces came together.

This thesis is far from perfect — but it is mine. And I finished it.

(...Ready for it?)

Communications

Publications

2025

Julie Marie Bekkevold*, Matthew Mosse*, Matthew Geever, Jonathan J. P. Peters, and Lewys Jones

About Time: Moving Past Pure Poisson Noise by Simulating Low-Dose STEM Images with Temporal Detector Response and Time-Dependent Noise

(In Preparation)

Julie Marie Bekkevold*, Georgios Varnavides*, Jonathan J. P. Peters, and Lewys Jones

Aberration robust ptychography using segmented and few-element array detectors

(In Preparation)

Georgios Varnavides, **Julie Marie Bekkevold**, Stephanie M. Ribet, Mary C. Scott, Lewys Jones, and Colin Ophus

Beyond Contrast Transfer: Spectral SNR as a Dose-Aware Metric for STEM Phase Retrieval

(Submitted to Microscopy and Microanalysis, pre-print available at doi.org/10.48550/arXiv.2507.19476)

Georgios Varnavides*, **Julie Marie Bekkevold***, Stephanie M. Ribet, Mary C. Scott, Lewys Jones, and Colin Ophus

Relaxing Direct Ptychography Sampling Requirements via Parallax Imaging Insights

(Submitted to Microscopy and Microanalysis, pre-print available at doi.org/10.48550/arXiv.2507.18610)

Julie Marie Bekkevold, Stephanie M. Ribet, Mary C. Scott, Lewys Jones, Colin Ophus, and Georgios Varnavides

Evaluating the Transfer of Information in Phase Retrieval STEM Techniques

Elemental Microscopy (2025), pre-print available at doi.org/10.69761/ehch7395

Julie Marie Bekkevold, Jonathan J. P. Peters, Ryo Ishikawa, Naoya Shibata, and Lewys Jones

Auto-thresholding for Unbiased Electron Counting

Microscopy (2025) dfaf025

*Denotes equal contribution.

2024

Julie Marie Bekkevold, Jonathan J. P. Peters, Ryo Ishikawa, Naoya Shibata, and Lewys Jones

Ultra-fast Digital DPC Yielding High Spatio-Temporal Resolution for Low-Dose Phase Characterisation

Microscopy and Microanalysis **30** 5 (2024) 878-888

Jonathan J. P. Peters, Tiarnan Mullarkey, **Julie Marie Bekkevold**, Matthew Geever, Ryo Ishikawa, Naoya Shibata, and Lewys Jones

On the temporal transfer function in STEM imaging from finite detector response time

Ultramicroscopy **267** (2024) 114056

Taichi Kusumi, Shun Katakami, Ryo Ishikawa, Kazuaki Kawahara, Tiarnan Mullarkey, **Julie Marie Bekkevold**, Jonathan J.P. Peters, Lewys Jones, Naoya Shibata, and Masato Okada

New Poisson Denoising Method for Pulse-count STEM Imaging

Ultramicroscopy **264** (2024) 113996

Presentations

2025

Microscopy and Microanalysis Meeting 2025, Salt Lake City, UT, USA

Oral Presentation

Segmented Detector Digitization and the Role of Denoising for Increasing Achievable Temporal Resolution in Phase Characterization

Microscopy Society of Ireland Symposium 2025, Dublin, Ireland

Oral Presentation

Iterative Ptychographic and iCOM Phase Reconstruction with Segmented and Fewer Element Array Detectors

2024

Materials Research Society Fall Meeting 2024, Boston, MA, USA

Oral Presentation

High Spatio-Temporal Resolution Differential Phase Contrast Imaging via Detector Signal Digitisation

European Microscopy Congress 2024, Copenhagen, Denmark

Oral Presentation

Accessible STEM-DPC Imaging Using ADF Detector Allowing in-situ Magnetic Reversal Studies

European Microscopy Congress 2024, Copenhagen, Denmark

Poster Presentation

High Spatio-Temporal Resolution Differential Phase Contrast Imaging via Detector Signal Digitisation

4th Annual Women in Microscopy Conference, NUANCE Center (Online)

Oral Presentation

Accessible Magnetic Reversal Studies Using ADF Detector For STEM-DPC

Microscopy Society of Ireland Meeting 2024, Limerick, Ireland

Thesis-in-three Presentation

Retaining Precision in Differential Phase Contrast Imaging at High Frame Rate, Low Dose Conditions by Electron Event Counting

2023

The 20th International Microscopy Congress 2023, Busan, Korea

Oral Presentation

Optimising the camera length for DPC imaging using segmented detectors

Microscopy and Microanalysis Meeting 2023, Minneapolis, MN, USA

Invited Talk

Retrofitting and Reconfiguring Existing Microscopes for Digital DPC: an Accessible Approach to Low-Dose Phase Mapping

IOP Spring Meeting 2023, Armagh, Northern Ireland

Poster and Rosse Medal Finalist Presentation

Camera Length Optimisation for DPC Imaging Using Segmented Detectors

Microscopy Society of Ireland Meeting 2023, Cork, Ireland

Poster Presentation

Camera Length Optimisation for DPC Imaging Using Segmented Detectors

List of Acronyms

4D-STEM	four-dimensional scanning transmission electron microscopy
ABF	annular bright field
ADF	annular dark field
API	application programming interface
BF	bright-field
CBED	convergent beam electron diffraction
CCD	charge-coupled device
CDF	cumulative distribution function
CMOS	complementary metal-oxide-semiconductor
CNR	contrast-to-noise ratio
COM	center of mass
CTF	contrast transfer function
DDC	direct digital controller
DPC	differential phase contrast
FFT	fast Fourier transform
FOV	field of view
HAADF	high-angle annular dark field
HRTEM	high-resolution transmission electron microscopy
iCOM	integrated center of mass
MAPS	monolithic active pixel sensor
MOF	metal organic framework
PAD	(hybrid) pixel array detector
PDF	probability distribution function
PMT	photo-multiplier tube
PSF	point spread function
PSNR	peak signal-to-noise ratio

ROI	region of interest
SAAF	segmented annular all-field
SSIM	structural similarity index
STEM	scanning transmission electron microscopy
SSB	single side-band
SNR	signal-to-noise ratio
STO	strontium titanate
TEM	transmission electron microscope
TTF	temporal transfer function
UV	ultraviolet
WPOA	weak phase object approximation

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Chapter 1

Introduction and Motivation

Microscopy is the technical field of viewing and imaging objects that are too small to see with the unaided eye. Historically, the ability to image and observe the function of microscopic systems has been crucial for technological advances and discoveries. Where visible light microscopy reaches its limits, other types of microscopy and spectroscopic techniques have emerged to give further insight and understanding into microscopic physics, chemistry and biology. Electron microscopy is one of these techniques which allow us to study and modify materials at the nanoscale — atom by atom.

This chapter begins by relating traditional visible light microscopy to electron microscopy, and then the underlying theory governing electron microscopy will be presented. Of specific importance for this thesis is the phase problem that arises because real detectors are unable to detect complex-valued waves. It is worth noting that this is a general problem for scientists in many fields, not just microscopy. The theory behind two different phase retrieval methods in scanning transmission electron microscopy (STEM), which both have strengths and weaknesses as solutions to the phase problem, will be presented. Additionally, the sample damage mechanisms that motivate low-dose technique development will be introduced. Finally, the reasons why phase retrieval techniques are dose efficient will be touched upon briefly, and will be discussed further in subsequent chapters.

1.1 Electron Microscopy

Visible light microscopes use photons to image samples. The resolution of such a microscope is given by

$$R = \frac{1.22\lambda}{2NA} \quad (1.1)$$

where λ is the wavelength of the light and $NA = n \sin \theta$ is the numerical aperture [5, 6]. Here, θ is the opening angle of the limiting aperture in the imaging system, limited by the geometry of the microscope, and n the refractive index of the imaging medium. This expression arises from the interaction of the light with

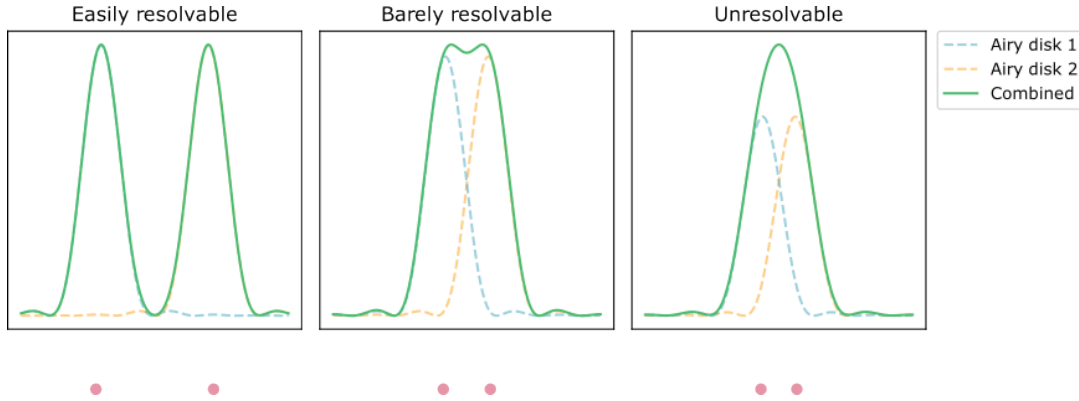


Figure 1.1: Illustration of the Rayleigh criterion for resolution of two points through an imaging system.

the optical system, commonly described through a point spread function (PSF) explaining how every point in the object plane is smeared to a disk in the image plane. This expression arises from the interaction of the light with the optical system, where every point in the object becomes an Airy disk in the image, see Figure 1.1. The Rayleigh criterion states that two points are resolvable if their separation is larger than where the peak of their Airy disks overlap with the first minimum of the other. An illustration of this may be seen in Figure 1.1.

For visible light microscopes, the numerical aperture is typically around 1 at best, meaning it has little ability to resolve any features smaller than roughly 0.5λ [6]. As a result, the highest possible resolution for visible light microscopes — using blue light of wavelength ~ 400 nm — is around 200 nm, and if we want to study things that are smaller we have to think differently [1]. The natural solution to enhance the resolution is to lower the wavelength of the radiation used for the imaging, which is done with ultraviolet (UV), extreme-UV and X-ray imaging. Another solution is to use electrons instead of photons.

Electrons, like all other particles, possess both wave and particle properties, and the de Broglie wavelength is given by

$$\lambda = \frac{h}{p}, \quad (1.2)$$

where h is Planck's constant and p is the momentum [7]. The electrons in a transmission electron microscope (TEM) are accelerated by an acceleration voltage, V_{acc} , such that their kinetic energy becomes $K = eV_{acc}$. Already at acceleration voltages on the order of 10 kV will the electrons travel at roughly $0.2c$, which is high enough to warrant a relativistic treatment, see diversion of the classical velocity from the relativistic velocity in Figure 1.2(a). This illustrates why relativistic corrections to electron velocity, and thus also wavelength, are necessary for an extended range of beam energies around the typical range used in STEM. From the relativistic total

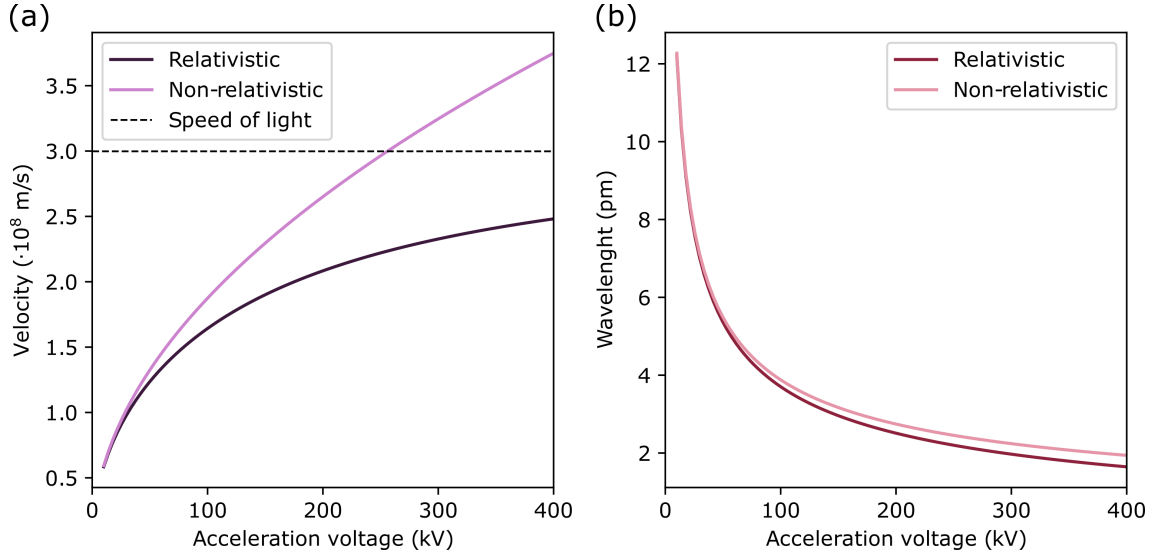


Figure 1.2: (a) The relativistic and non-relativistic velocity of electrons vs acceleration voltage applied, and (b) their corresponding relativistic and non-relativistic wavelength.

energy

$$E^2 = p^2 c^2 + m^2 c^4 \quad (1.3)$$

and the knowledge that the kinetic energy is the total energy minus the rest energy;

$$K = E - mc^2, \quad (1.4)$$

the relativistic momentum becomes [8]

$$p = \sqrt{2mK \left(1 + \frac{K}{2mc^2} \right)}. \quad (1.5)$$

From combining this with Equation 1.2, the relativistic wavelength of electrons accelerated by V_{acc} becomes

$$\lambda = \frac{h}{\sqrt{2mV_{acc}e \left(1 + \frac{V_{acc}e}{2mc^2} \right)}}. \quad (1.6)$$

The relativistic and non-relativistic wavelength for electrons accelerated by V_{acc} in the normal TEM operating range 10 – 400 kV is shown in Figure 1.2(b). From this, it is clear that the relativistic correction to the electron wavelength is fairly minor at the relevant velocities, and if we assume the resolution follows Equation 1.1 sub-pm features should be easily resolvable. Note that this is not to say that relativistic correct is unnecessary, in fact, it must be used when doing TEM and STEM simulations. However, in reality the very best resolution achievable falls short of this because of imperfections in the electromagnetic lenses in the TEM.

Similarly to how glass lenses are used to focus the photon beam in visible light microscopes, since the change in refractive index changes the path of the light, electron microscopes use electromagnetic lenses to focus the electron beam. This is because an electron moving through an electric or magnetic field changes its path due to the Lorentz force,

$$\vec{F}_L = -e(\vec{E} + \vec{v} \times \vec{B}). \quad (1.7)$$

The force due to an electric field is used to accelerate the electrons in the electron gun. Typically, the source is made of W or LaB₆ because their low work function allow them to readily emit electrons when heating or a strong electric field is applied [1, 9, 10]. Thermionic emission, through heating of the source, produces an electron beam with a higher energy spread than cold field emission from a very fine tip source, and the consequences of this will be discussed later. Cold field emission is practicable due to the low work function of the source material and the enhanced electric field strength at an atomically sharp source tip [11–13]. The applied magnetic field from electromagnetic lenses will apply a force on the fast-travelling electrons in the beam which sends them in a spiralling motion slowly moving inwards toward the optic axis. In this way, the lenses focus the electron beam to a fine spot. The magnetic fields are generated using coils of low-resistance wires where the magnetic field is controlled by adjusting the current through the wires, however, because of the spherical symmetry required these lenses can only be made to converge the beam — never diverge it. This becomes problematic because lens aberrations that would be corrected in a visible light microscope by a diverging corrector lens is not applicable, and the aberrations severely limit the resolution achievable with the electron microscope. As a result, the theoretically possible resolution is not reached experimentally because the electromagnetic lenses that are used to focus the electron beam are far from perfect. For the remainder of this thesis we will regard how the lens aberrations affect the electron beam in STEM mode only.

1.2 Scanning Transmission Electron Microscopy

In STEM the electron beam is focused to a small probe, which is then scanned across the region of interest on the sample and some form of the electrons' interactions with the sample is recorded for each scan position. The electron beam is produced with an electron gun, in which electrons are extracted from a source and accelerated using strong electric fields. To produce the sub-Ångström sharp probe to scan across the sample, the beam is demagnified in the condenser system. The condenser aperture is used to control the semi-convergence angle, α , of the beam, that is, the maximum angle of deflection from the optic axis the beam is allowed to have. In its simplest

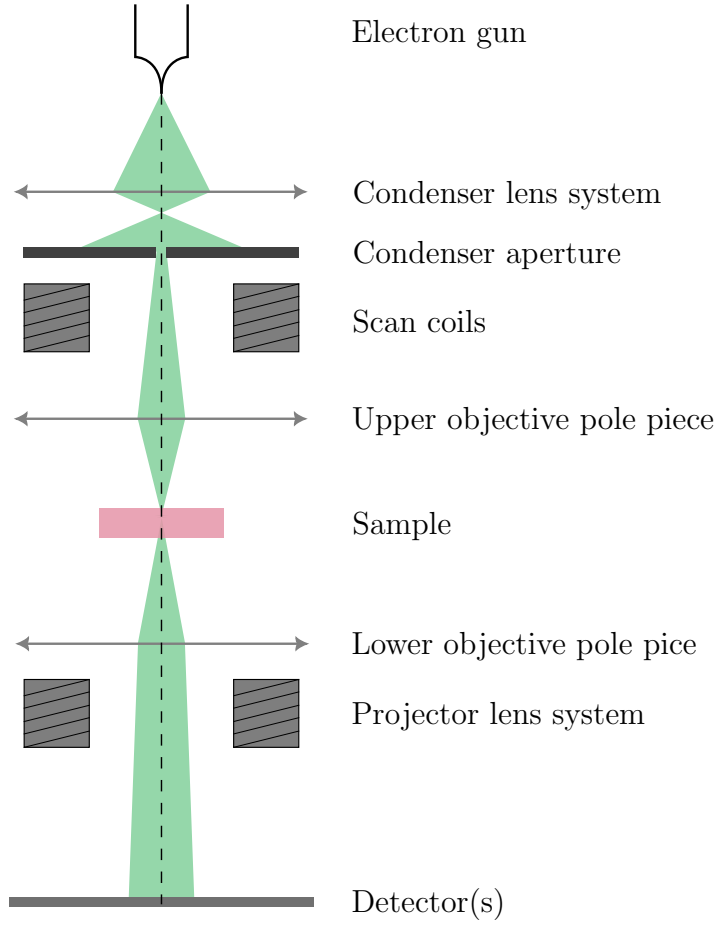


Figure 1.3: Simplified illustration of the lens system for STEM.

form, the probe in STEM is the Fourier transform of the aperture function, $A(\vec{k})$,

$$P(\vec{r}) = \int A(\vec{k}) e^{2\pi i \vec{k} \cdot \vec{r}} d\vec{k}, \quad (1.8)$$

before taking lens aberrations into account. The aperture function, $A(\vec{k})$, is mathematically expressed as a top-hat function equal to unity inside the aperture opening and zero elsewhere. Since the probe size is determined by the Fourier transform of the aperture, it follows that a larger aperture opening gives a smaller probe, and conversely a smaller aperture opening yields a larger probe size. Generally, the performance of an electromagnetic lens worsens the further from the optic axis the electrons pass through it, and therefore a higher α yields a higher proportion of aberrated electrons in the beam [14]. As a result, a smaller condenser aperture must be used when no aberration correction is applied to avoid using the heavily aberrated parts of the beam. This helps explain why aberration corrected microscopes can achieve probe sizes of 0.5 Å, whereas uncorrected microscopes only reach probe sizes of 2 Å at the very best [15].

Aberrations impose a phase factor on the probe which is incorporated by multiplication with $e^{-i\chi(\vec{k})}$ inside the integral in Equation 1.8 such that it becomes

$$P(\vec{r}) = \int A(\vec{k}) e^{2\pi i \vec{k} \cdot \vec{r}} e^{-i\chi(\vec{k})} d\vec{k}. \quad (1.9)$$

The aberration function, $\chi(\vec{k})$, may be given as

$$\chi(k, \phi) = \frac{2\pi}{\lambda} \sum_{n,m} \frac{1}{n+1} C_{n,m} (k\lambda)^{n+1} \cos[m(\phi - \phi_{n,m})], \quad (1.10)$$

by separating $\vec{k} = (k_x, k_y) = (k \cos(\phi), k \sin(\phi))$ into its radial and azimuthal components such that $k = \sqrt{k_x^2 + k_y^2}$ and $\phi = \arctan\left(\frac{k_y}{k_x}\right)$ [1, 14, 16]. The summation over the aberrations is over n, m which are the radial and azimuthal order respectively. Here, Krivanek notation is used for the aberration coefficients $C_{n,m}$, for which the names are summarised in Appendix A [14].

The first and most commonly considered aberration is defocus, given by the defocus coefficient $C_{1,0}$, which expresses how strongly the objective lens is focusing the beam and frequently denoted by Δf . Confusingly, the definition $\Delta f = -C_{1,0}$ is not uncommon, and the term “defocus” is used interchangeably¹ for both Δf and $C_{1,0}$. Throughout this thesis the term “defocus” should be taken to mean Δf to avoid such confusion here. Additionally, Figure 1.4 illustrates the practical meaning of positive and negative defocus values to further avoid confusion. By convention, a positive $C_{1,0}$ value means that the objective lens is over-focused, giving a beam cross-over before the sample. In other words, a negative defocus value, Δf , denotes over-focus. Conversely, positive defocus means the objective lens is under-focused and denotes a situation where the beam cross-over is after the sample.

As mentioned previously, electromagnetic lenses perform worse the further from the optic axis electrons pass through them. One of the effects of this is that the lenses have a significantly higher focusing power for electrons entering the lens far away from the optic axis. This is the origin of spherical aberration, denoted $C_S = C_{3,0}$, which for the spherically symmetric electromagnetic lenses in a TEM is positive [17]. Because of the spherical aberration, the electrons in the beam will be focused to a range of different focus points along the optic axis, see the ray diagram in Figure 1.5(a). The point at which the width of the electron beam is at its lowest is called the disk of least confusion, and its width is given by [1, 18]

$$d_{\text{sph}} = 0.5 C_S \alpha^3. \quad (1.11)$$

¹For example, simulation softwares like abTEM [16] typically allow giving defocus with a named parameter `defocus` which is defined as $\text{defocus} = \Delta f = -C_{1,0}$, but also as $C_{1,0}$ directly.

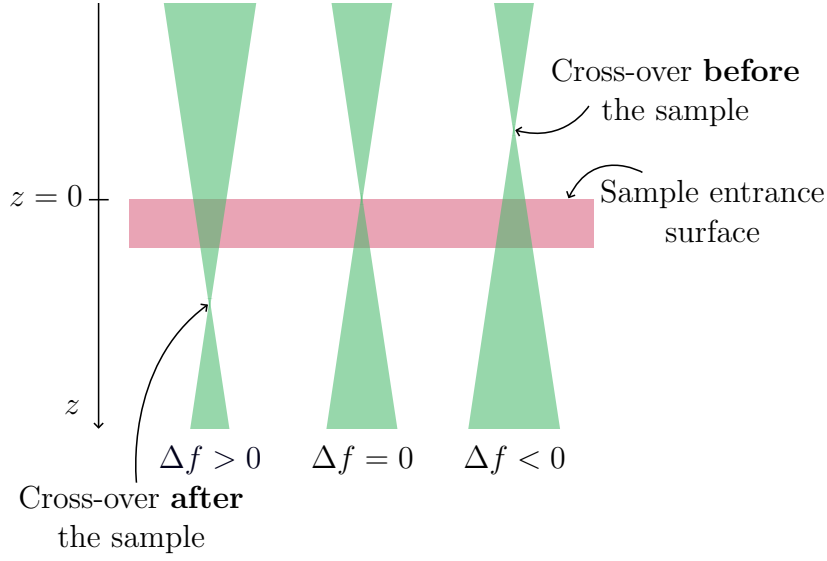


Figure 1.4: Illustration of the practical meaning of defocus. Note that the beam moves in positive z -direction, which is here defined to be downwards to align with most commonly used microscopes.

In non-aberration corrected microscopes, $C_S \approx 1.2$ mm is typical, which gives a disk of least confusion width of $6 \text{ \AA}$ for an $\alpha = 10$ mrad beam. With such high spherical aberration, the disk of least confusion is also significantly removed from where $\Delta f = 0$. The optimal defocus value to counteract the spherical aberration is known as Scherzer defocus [1, 17], and is given by

$$\Delta f_{Sch} = \left(\frac{3}{2} C_S \lambda \right)^{\frac{1}{2}}. \quad (1.12)$$

Some STEMs are equipped with spherical aberration correctors which use hexapole, octapole, or dodecapole electromagnetic lenses to produce regions of negative spherical aberration to correct the C_S in the beam [19, 20]. A set of two such multi-pole lenses significantly mitigates the effects of spherical aberration. Further, more recent efforts to mitigate the effects of spherical aberration involves the use of thin films and phase plates to apply negative spherical aberration or otherwise alter the phase of the electron beam [21]. This works by reversing spherical aberration in a similar manner as one would with a concave lens in visible light microscopy. Together, defocus, $C_{1,0}$, and spherical aberration, $C_{3,0}$ are the two most considered aberrations since they have the most profound effect on the resulting image [22]. When considering only these two, the aberration function in Equation 1.10 reduces to

$$\chi(\vec{k}) = \frac{2\pi}{\lambda} \left(\frac{\lambda^2 k^2}{2} C_{1,0} + \frac{\lambda^4 k^4}{4} C_{3,0} \right) [1, 23]. \quad (1.13)$$

In addition to the aberrations that arise due to the imperfections in the lenses, there also exists chromatic aberration due to imperfections in the electron gun, since any heating of the filament will result in an energy spread in the emitted electron beam. It follows from Equation 1.7 that electrons with different energies, and there-

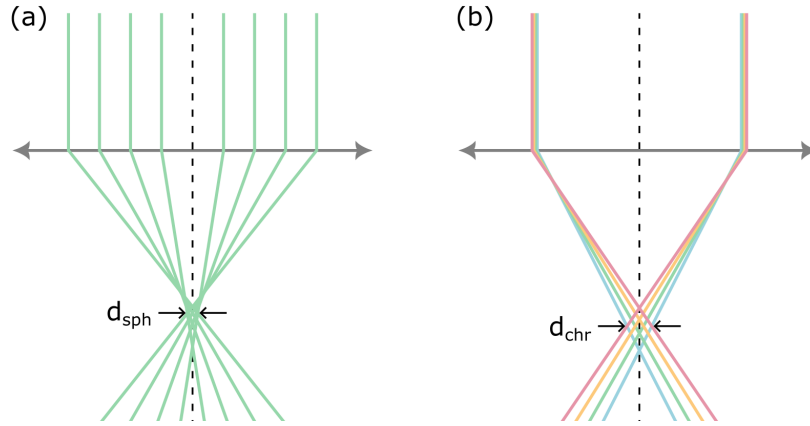


Figure 1.5: Ray diagrams showing how (a) spherical aberration and (b) chromatic aberration leads to a spread of the focal point of the electromagnetic lens. Here, the colours represent different energy electrons — with blue indicating higher energy and red lower energy electrons.

fore different velocities, will be focused differently by the lenses. Specifically, higher energy electrons are focused less strongly, such that the effective focal point is further away from the lens. Conversely, lower energy electrons are focused more strongly, resulting in a focal point closer to the lens for these electrons. This is illustrated with a ray diagram in Figure 1.5, with colours representing different energy electrons. As a result, the beam will again have a disk of least confusion, the width of which is given by

$$d_{\text{chr}} = C_c \frac{\Delta E}{E_0} \alpha, \quad (1.14)$$

where ΔE is the beam energy spread and E_0 the primary energy [24]. Additionally, there are plenty of higher order aberrations, but the ones mentioned here are the ones that mainly govern the STEM probe size. A list of all the aberrations up to the 8th order can be found in Appendix A. Finally, it follows from Equation 1.8 that any aberrations that only affect the angles beyond the semi-convergence angle do not deteriorate the probe because they are excluded by the aperture. As a result, smaller semi-convergence angles are typically used for highly aberrated probes.

The most common way of recording a STEM image is using an annular dark field (ADF) detector that collects electrons that have been scattered by the sample to higher angles than the semi-convergence angle. Using such a monolithic detector give a single intensity value per scan position which is roughly proportional to $Z^{1.7}$, where Z is the atomic number. This yields easily interpretable images where atoms appear bright on a dark background. In this thesis, the ADF image is frequently used to investigate experimental parameters such as detector offset and gain values in subsection 2.2.3 and threshold optimisation for digitisation in subsection 4.2.2, as well as for navigation on the sample during experiments.

This section has introduced the main theoretical background behind the formation of the STEM probe and the effect that the aberrations have on the achievable size of this probe. In the next section, we will introduce the sample and how it alters the electron probe to eventually form a convergent beam electron diffraction (CBED) pattern that may be detected on a real detector, in the light of the phase problem which then arises.

1.3 The Phase Problem

Wave-like signals, like the electron wave functions that are carrying the information detected in STEM, are typically complex. The phase problem arises when these signals are measured using real-world detectors, which are only able to detect intensity variations, not complex values. Instead, these detectors merely detect the intensity of the signal, meaning that all the information contained in the imaginary part of the signals is lost. Notably, this is a problem in all areas of science where wave-like signals are detected including, but not limited to, x-ray crystallography [25–27], visible light microscopy [28, 29], and astronomy [30, 31], as well as electron microscopy [32].

The general expression for the probe in STEM can be seen in Equation 1.8, and when it interacts with the specimen a phase change is imposed on the electron beam. Additionally, the specimen may scatter electrons out to higher angles, altering the resulting amplitude of the detected signal. Typically, the phase changes are much more subtle and contain a lot more information about the sample when compared with the pure amplitude signal. The electrons that are scattered to higher angles than α is the signal that is typically detected with an ADF detector. This signal produces a single intensity value for each scan position proportional to the number of electrons detected, which is roughly proportional to $Z^{1.7}$ where Z is the atomic number.

In general, a complex object may be expressed mathematically as

$$O(\vec{r}) = a(\vec{r})e^{i\varphi(\vec{r})} \quad (1.15)$$

where $a(\vec{r})$ is the amplitude and $\varphi(\vec{r})$ is the phase [1]. Both magnetic and electric fields contribute to the phase modulation of the electron wave through the sample, and so the phase of Equation 1.15 may be given as

$$\begin{aligned} \varphi(\vec{r}) &= \sigma V_{proj}(\vec{r}) && \text{(in absence of magnetic field)} \\ \varphi(\vec{r}) &= \sigma V_{proj}(\vec{r}) - 2\pi \frac{e}{h} \Phi(\vec{r}) && \text{(in general)} \end{aligned}$$

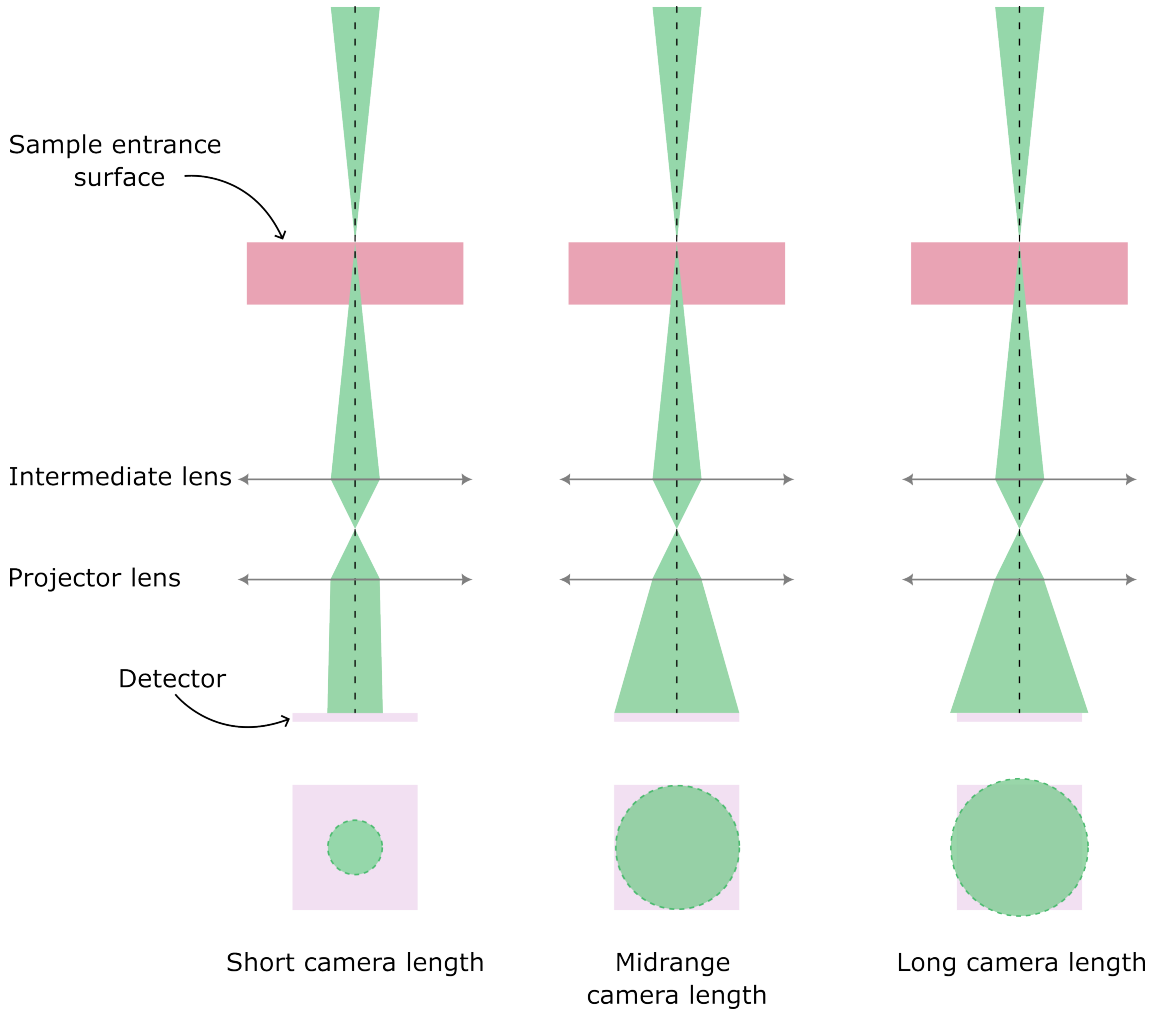


Figure 1.6: Illustration of how changing the strength of the projector lens changes the camera length — the effective distance between the sample and the detector.

where $V_{proj}(\vec{r})$ is the projected electrostatic potential and $\Phi(\vec{r})$ is the magnetic flux enclosed by two trajectories running through the sample and vacuum, and $\sigma = 2\pi e/\lambda E$ is a proportionality constant [1, 33, 34]. Notably, the magnetic contribution to the phase change is at least an order of magnitude smaller than the electric contribution, and as such, even in the presence of magnetic fields the phase change due to electric fields is much easier to detect or reconstruct [35].

Typically, it is assumed that the sample only slightly alters the phase of the incoming electrons, and does not notably alter its amplitude. This is the weak phase object approximation (WPOA), which in the absence of any magnetic fields reduces the object expression from Equation 1.15 to

$$O(\vec{r}) = e^{i\sigma V_{proj}(\vec{r})} \quad (1.16)$$

where the projected electrostatic potential $V_{proj}(\vec{r})$ typically contains contributions from the crystal potential from the atomic nuclei in the sample and electric fields

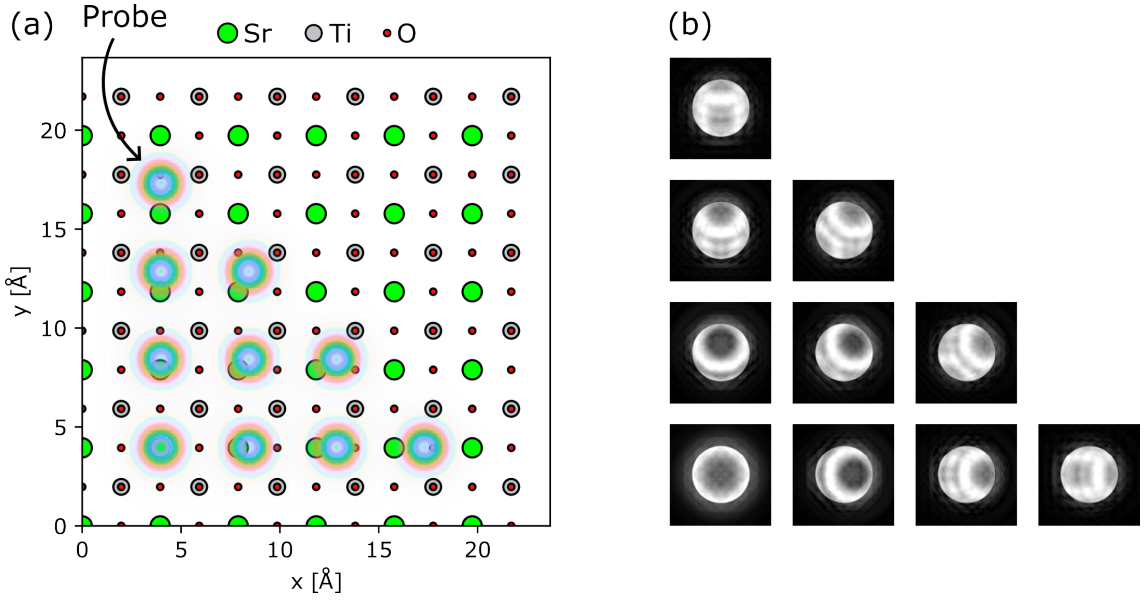


Figure 1.7: (a) Illustration of the probe, $P(\vec{r})$, on a few selected position on an strontium titanate (STO) sample, $O(\vec{r})$, and (b) the corresponding diffraction patterns, $I(\vec{k})$.

due to, for example, polarisation or pn-junctions [1, 36]. Then, the exit wave, which is the wavefunction of the electron beam after passing through the sample, is the probe multiplied with the object for each scan position $\vec{R}$:

$$\psi_{\text{exit}}(\vec{r}, \vec{R}) = P(\vec{r} - \vec{R}) \cdot O(\vec{r}). \quad (1.17)$$

Here, the scanning nature of the imaging is incorporated as a relative lateral distance, $\vec{R}$, between $O(\vec{r})$ and $P(\vec{r} - \vec{R})$ where the probe is the one moving² [37]. This exit wave contains all the imaging information about the sample — that is, both the amplitude and phase modulation imposed on the electrons by the sample [32, 36]. Interestingly, if the entire electron wave function, including both the amplitude and phase, could be recorded, it would be possible to correct aberrations in post-processing of the data [32].

Finally, after passing through the sample the beam is projected onto a detector. The space available for the expansion of the beam post-specimen can be altered by adjusting the camera length of the microscope. This adjusts the projector lenses so that the effective distance between the sample and the detector is changed, while the physical distance between them stays the same as may be seen in Figure 1.6. Effectively, increasing the camera length increases the size of the bright field disk, or CBED pattern, at the detector. For STEM measurements the Fraunhofer diffraction condition, $\frac{w^2}{L\lambda} \ll 1$ where w is the distance between scatterers, L is the camera length, and λ the electron wavelength, is typically satisfied. This means that the detector is in the far field such that the detected wave is the Fourier transform of the

²Although the object may equally well be moved relative to the probe.

exit wave. As a result, $\psi_{\text{det}}(\vec{k}) = \mathcal{F}\{\psi_{\text{exit}}(\vec{r})\}$ is the imaging wave at the detector plane. Then the diffraction pattern detected by the detector is

$$I(\vec{k}) = |\psi_{\text{det}}(\vec{k})|^2 \quad (1.18)$$

$$= \psi_{\text{det}}(\vec{k})\psi_{\text{det}}^*(\vec{k}) \quad (1.19)$$

and all phase information is lost. In the following section various methods for retrieving this phase information will be discussed.

1.4 Phase Retrieval Techniques

In the previous section the mathematical formulation of the sample object, and its effect on the STEM probe as it is scanned across the sample, was introduced. From Equation 1.18 it became clear how the phase problem arises when the CBED pattern is detected by a real detector. In this section, a few different techniques for phase contrast and phase retrieval in STEM will be introduced — with a main focus on the center of mass (COM) estimation and ptychography, which are most relevant for this work.

There exists several techniques for phase contrast and retrieval that attempt, in different ways, to recover the lost phase information. In principle, the missing phase information can be detected by interfering the electron beam with a reference beam with a known phase, typically the vacuum beam [38]. Since electrons exhibit wave-like properties they interfere with other electrons, which in turn transfers phase information to amplitude information that we can detect. This technique is known as off-axis electron holography, and can be employed in both TEM and STEM mode [39]. It requires a biprism in the column to split the electron beam into two beams, and thus requires a specialised microscope.

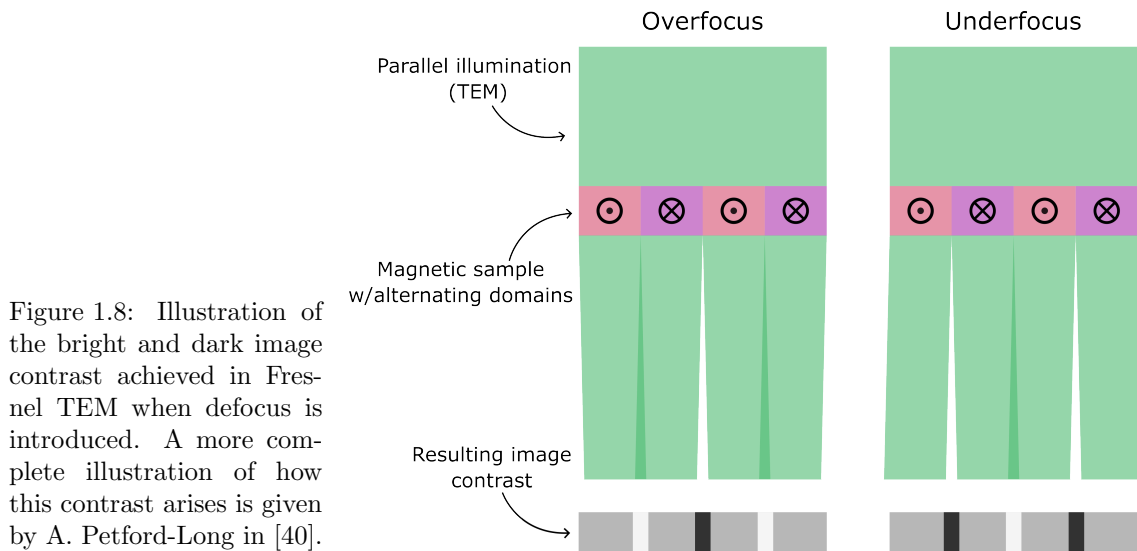


Figure 1.8: Illustration of the bright and dark image contrast achieved in Fresnel TEM when defocus is introduced. A more complete illustration of how this contrast arises is given by A. Petford-Long in [40].

A different phase contrast technique, which utilises the self-interference in the electron beam, is Fresnel TEM [33, 40, 41]. Unlike off-axis electron holography, this technique does not require a vacuum reference beam but instead uses the interference of the parallel illumination electron beam with itself at interfaces between domains with magnetic or electric fields with different orientations. These interfaces are typically referred to as domain walls, and the self-interference of the electron beam at these interfaces helps to transfer the phase information to amplitude. In order for the contrast to become visible, Fresnel TEM requires a defocused objective lens, and yields dark or bright contrast at domain walls depending on the field direction in the domains and the defocus direction [40]. With overfocus, domain walls where the beam is deflected towards the domain wall will give bright contrast, whereas for underfocus this will give dark contrast, see illustration in Figure 1.8. The opposite is true for domain walls where the beam is deflected away from the domain wall. Most commonly the Fresnel TEM technique is used to image long range magnetic or electric fields, as the resolution achievable with a defocused TEM technique is limited.

1.4.1 Differential Phase Contrast

The goal of all phase imaging techniques is to retrieve the phase change obtained by the beam electrons from passing through the sample [42]. The two contributions to this phase change originate from the electric and magnetic fields in the sample [1, 43], and may be detected in STEM as a shift in the center of mass (COM) of the CBED pattern [34, 42, 44–50]. Most prominently, electrons in the beam are deflected by positive atomic nuclei in the sample and thus the intensity distribution of the direct beam is altered, as illustrated in Figure 1.9. Magnetic fields present in the sample will also produce a shift of the electron beam, but the net force experienced by the electrons will be perpendicular to both the electrons' velocity and the magnetic field direction following Equation 1.7 [33]. An illustration of the deflection by magnetic fields in and out of the plane may also be seen in Figure 1.9.

By using position sensitive detectors, like a pixellated detector, it is possible to detect a displacement of the COM of the electron beam [42, 51, 52]. This displacement is, to a good approximation, proportional to the gradient of the phase change picked up by the electron beam [53]. It also follows from Equations 1.15 and 1.16 that the COM displacement is proportional to the gradient of the projected electrostatic potential of the sample, in the absence of magnetic fields. Note that the phase change due to magnetic fields has been included so far for completeness, however magnetic materials are not studied or discussed any further in this thesis. The relation between the electrostatic potential, $V(\vec{r})$, the electric field, $\vec{E}(\vec{r})$, and

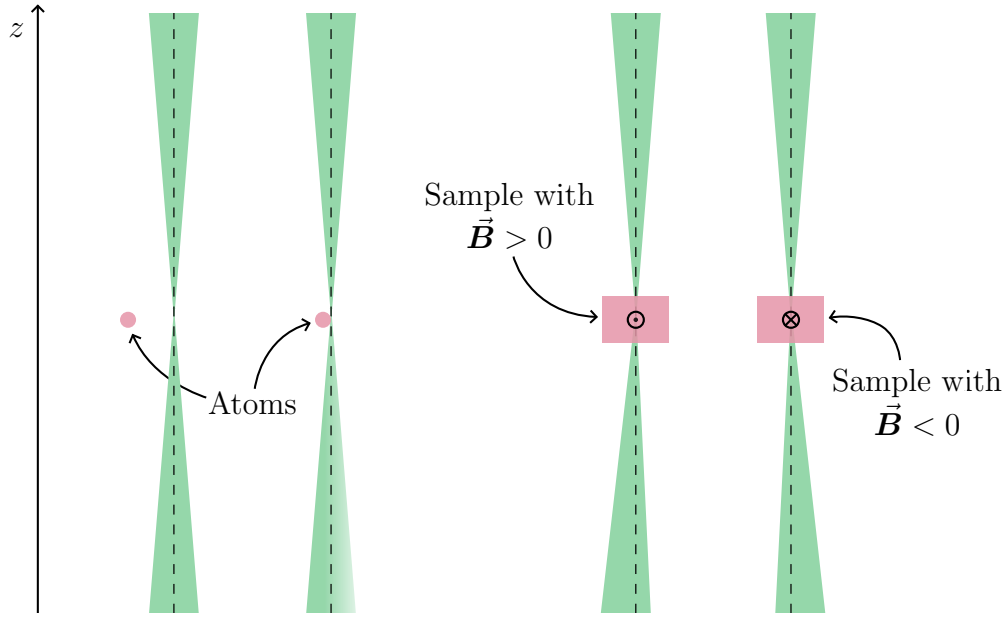


Figure 1.9: Illustration of how the electric field surrounding an atom redistributes the electrons within the beam causing a shift in its center of mass, which may be detected by a pixellated or segmented detector. For a sample with a magnetic field, the electron beam is shifted perpendicular to the magnetic field direction.

the charge density distribution, $\rho(\vec{r})$ is

$$\nabla^2 V(\vec{r}) = -\vec{\nabla} \cdot \vec{E}(\vec{r}) = \frac{1}{\epsilon_0} \rho(\vec{r}) \quad (1.20)$$

from the Poisson equation and Gauss' law [3]. Thus, the phase shift is proportional to the electric field of the sample [34, 49, 50, 54].

The displacement of the COM of the CBED pattern is necessarily a vector signal, where each image pixel will have an x- and y-displacement components typically calculated on the grid of the pixellated detector. Generally, for any detector geometry, the COM shift in x-direction as a function of image position $\vec{r}$ may be found as the sum of the intensity detected in each segment or pixel $I(\vec{r})$ multiplied by the center of mass of that segment or pixel ($\vec{k}_x$):

$$\text{COM}_x(\vec{r}) = \sum_i I_i(\vec{r}) \cdot \vec{k}_{xi}. \quad (1.21)$$

The equivalent expression for the y-direction follows trivially, and the vector COM signal is $\overrightarrow{\text{COM}} = (\text{COM}_x, \text{COM}_y)$. Importantly, any mismatch between the rotation of the scan and the rotation of the detector will show up as $\text{curl } \overrightarrow{\text{COM}} \neq 0$. The curl of a static electric field should always be zero, which means that if we assume the electric field in our sample is unchanging the relative rotation between the scan and the detector may be found by minimizing the curl of the $\overrightarrow{\text{COM}}$ signal.

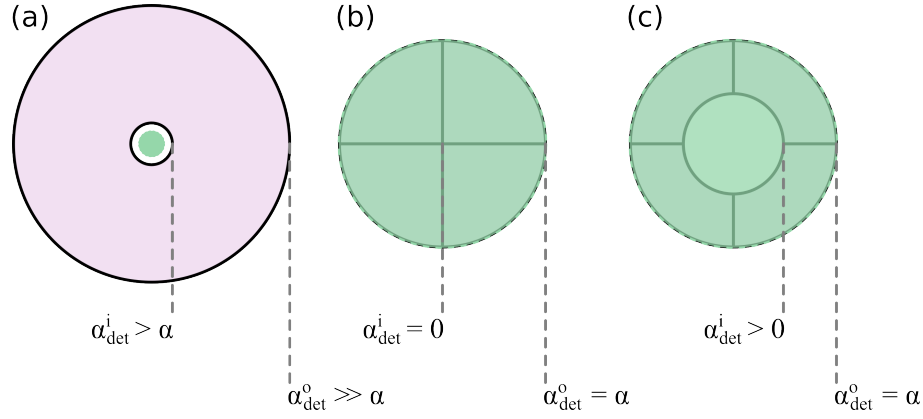


Figure 1.10: Explanation of typical detector collection angles for (a) ADF imaging, and COM estimation with segmented detector in the (b) bright-field (BF) region and (c) in the annular bright field (ABF) region.

It has been demonstrated extensively that four detector segments is sufficient to estimate the phase change obtained by the beam at atomic resolution for thin samples [3, 49, 53–58]. For a segmented detector with four equal segments only, the calculation from Equation 1.21 and the corresponding calculation of COM_y , simplifies to subtracting the intensity detected in opposing detector segments³. Thus,

$$\text{COM}_x = S_1 - S_3 \quad (1.22)$$

$$\text{COM}_y = S_2 - S_4 \quad (1.23)$$

where S_i denote the detector segment intensity values. In the next chapter various detector geometries and their benefits and shortcomings will be introduced. Then, the reasoning for why one would want to estimate the COM using a detector with only four segments will be clarified.

If the COM signal is proportional to the projected electric field, then the integrated COM signal, often termed integrated center of mass (iCOM), must be proportional to the projected electric potential of the sample following Equation 1.20. With the detected COM signal being $\overrightarrow{\text{COM}}(\vec{r})$ the integration can be performed in Fourier space,

$$\mathcal{F}\{\text{iCOM}(\vec{r})\}(\vec{k}) = \frac{\mathcal{F}\{\overrightarrow{\text{COM}}(\vec{r})\}(\vec{k}) \cdot \vec{k}}{2\pi i k^2} \quad (1.24)$$

where $\vec{k}$ is position vector in reciprocal space [3, 53]. Then, using the inverse Fourier transform we can obtain the iCOM signal, which is proportional to the projected potential of the sample. Because this signal is proportional to Z , in contrast to $Z^{1.7}$ like ADF, both light and heavy atoms may be imaged simultaneously.

³This is sometimes termed differential phase contrast (DPC) for historic reasons. Nevertheless, the calculation is simply an estimation of the COM.

Finally, the COM is typically calculated from signal within the bright field disk. That is, the inner, α_{det}^i , and outer, α_{det}^o , collection angles of the detector are in the range 0 mrad to the semi-convergence angle, α mrad, see illustration in Figure 1.10. Since the intensity detected by an annular detector falls exponentially with increasing inner collection angle beyond α mrad, placing the detector in the BF — or ABF region — is far more dose efficient than ADF. This is because far fewer electrons are scattered to the dark field than remain in the bright field [59, 60]. Importantly, the placement of the detectors in the TEM column contribute to the large difference in collection angles between ADF and DPC detectors in the illustration in Figure 1.10. Additionally, the camera length may be adjusted to reach desired collection angles, which will be revisited in section 3.3.

1.4.2 Ptychography

Another technique for recovering the missing phase information is to use the self-interference of electrons in the unscattered beam with scattered beams and retrieve the phase computationally from overlapping regions in the diffraction plane. The umbrella-term for these types of techniques is “ptychography”, which stems from the Greek words $\pi\tau\upsilon\chi\eta$ meaning “to fold” and $\gamma\rho\acute{\alpha}\varphi\epsilon\iota\nu$ meaning “to write” which picturesquely describes how the information we seek is folded into the diffraction pattern. Ptychography is a powerful technique that has been utilised in visible light, UV, extreme-UV and x-ray imaging for decades, before it emerged relatively recently in the electron microscopy field [37]. It was suggested by Hoppe [61] over 50 years ago, but it took decades before detectors became good enough for the method to become experimentally feasible.

By assuming that the illuminating probe is constant and the object is static during a scan, and that the probe is scanned across the sample with a known distance between each scan position, the phase of the object can be retrieved computationally [62]. This is possible because the overlapping regions of diffracted beams with the un-diffracted electron beam contains redundant information from which the phase and amplitude of the sample may be reconstructed by deconvolving the object and probe from Equation 1.17 when they are detected as diffraction patterns given by

$$I_j(\vec{k}) = |\mathcal{F}\{O(\vec{r}) \cdot P(\vec{r} - \vec{R}_j)\}|^2 \quad (1.25)$$

for the j th scan position in real space, $\vec{R}_j$ [63]. Several methods of ptychography exist, both direct approaches like single side-band (SSB), Wigner distribution deconvolution (WDD), and optimum bright field (OBF) as well as iterative approaches. Many open source algorithms for doing ptychographic phase reconstruction are available [64–66]. Solving for the exit wave-function when the intensities are measured is

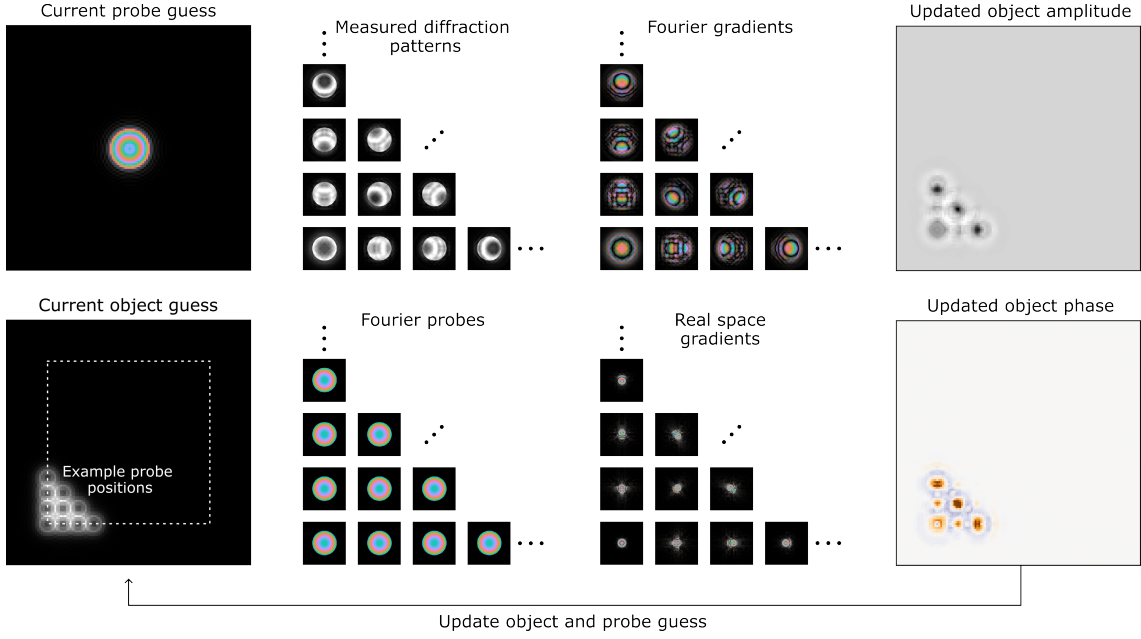


Figure 1.11: Sketch of the iterative ptychography process through the first iteration where an initial guess for the probe and an empty object is given. The gradient step is calculated using the measured diffraction patterns and the Fourier probe, and used to update the guess for the probe and the object. Then, the updated probe and object are used as the guesses for the next iteration.

a high-dimensional, inverse problem, where the goal is to obtain the two functions for the probe $P(\vec{r} - \vec{R}_j)$ and object $O(\vec{r})$ such that

$$O(\vec{r}) \cdot P(\vec{r} - \vec{R}_j) = \psi_{\text{exit},j}(\vec{r}) \quad \forall \quad j \quad (1.26)$$

where j again denotes the scan position [63]. In modern iterative ptychographic engines this is done by starting with an empty guess for the object and an initial probe. The initial guess for the probe should as best as possible reflect the experimental conditions the data was acquired with, meaning that ideally the defocus and other aberrations should be known — or somehow estimated by other means. First the probe is forward propagated and compared with the measured intensity before being inverse Fourier transformed to update the guesses for the object and probe for the next scan position, $O_{j+1}(\vec{r})$ and $P_{j+1}(\vec{r})$ [63, 67]. This procedure of transforming the current object and probe to compare with the experimental data, and then updating the object and probe, is repeated over and over again for a given number of iterations. In the end, the final object reconstruction should in theory be equal to the object that was imaged. Importantly, there are physical constraints that may be applied to help the algorithm converge to a solution faster. For example, the electrons only ever pick up a positive phase change upon traversing the sample, which means that we may demand positivity.

In contrast to DPC and Fresnel TEM, which must be used in-focus and out-of-focus respectively, ptychography can be used both in-focus and out-of-focus. The benefit of out-of-focus ptychography is that the defocused probe covers a larger area on the sample giving less beam damage to the sample by spreading out the dose over a larger area at a time. A large defocus also gives larger overlap regions in real space. This has two main benefits: firstly it allows for larger real-space scan steps to be taken, which illuminates the entire region of interest using fewer exposures and thus less dose. Secondly, the increased overlap areas increase the redundancy in the datasets which helps the ptychographic reconstruction algorithm converge faster. In later chapters the sampling requirements in both real and reciprocal space for ptychography will be investigated, and the implications for low-dose characterisation and temporal resolution discussed.

1.5 Sample Damage Mechanisms

Electrons travelling at over half the speed of light are very energetic, and as a result may cause a lot of damage when they hit the sample. Two main damage mechanisms are typically considered in STEM, namely knock-on damage and radiolysis [68–70]. The former is the result of an elastic collision between a beam electron and an atom in the sample [71, 72]. Knock-on damage is typically most prevalent in crystalline materials and is a consequence of high energy electrons knocking an atom out of its position in the crystal leaving a vacancy [72]. The “knocked out” atom either becomes an interstitial atom, that is, an atom that is placed outside of the lattice positions in the crystal lattice, migrates to another vacant position in the lattice, or is ejected from the sample. Radiolysis, on the other hand, is the result of an inelastic collision between a beam electron and a sample atom in which a core electron of an atom is knocked out. This is typically more damaging to non-conductive materials like biological specimens and samples suspended in liquid-phase because of the lack of available electrons to fill the resulting vacancy.

Collisions are considered elastic if no energy, or only a negligible amount, is lost in the collision. This typically means that all, or almost all, of the energy lost by the incoming beam electron was transferred to an atom in the sample which is then knocked out of its place as a result. The energised atom migrates through the sample until it has lost all of its kinetic energy to collisions with other atoms and found a local energy minimum to rest in, or is freed from the sample entirely. An illustration of the formation of an interstitial atom may be seen in Figure 1.12(a). Formation of a significant amount of interstitial atoms reduces the crystallinity of the sample and ultimately causes amorphisation. The energy that is required to displace an atom from its position in the sample is referred to as the threshold

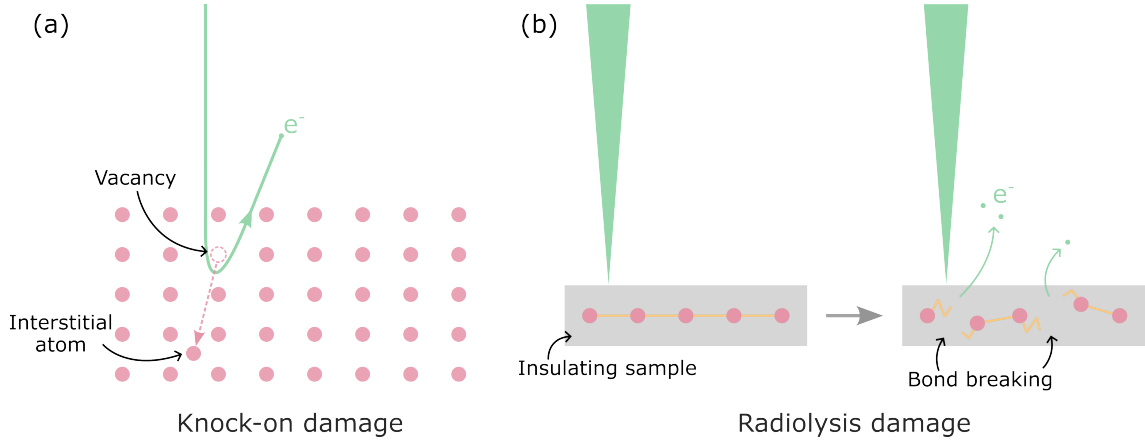


Figure 1.12: Illustration of the two main sample damage mechanisms in STEM: (a) knock-on damage and (b) radiolysis.

displacement energy, E_d , and is a material specific property that is mainly dependent on the atomic number of the atom and the strength of the bonds to neighbouring atoms [70, 73, 74]. Moreover, the maximum energy that may be transferred from a beam electron to an atom is also dependent on the mass of the nucleus of the atom, in addition to the energy of the beam electron. As a result, the occurrence of knock-on damage is not only highly material dependent, but also beam energy dependent.

For knock-on damage to occur, the beam electron has to be scattered through a sufficiently high angle, which is often high enough for the electron to be back-scattered [74–76]. The probability of this happening is the combined probability of elastic interaction between a beam electron and the sample, and the probability of an elastic collision scattering the electron through an angle higher than the minimum angle that will result in knock-on damage. Mathematically, the former may be given as

$$P(\text{elastic scattering}) = 1 - e^{-\frac{t}{\lambda}} \quad (1.27)$$

where t is the sample thickness and λ the total elastic mean free path [74], which is clearly sample-dependent. The probability that an electron is scattered through an angle $\theta \geq \theta_{min}$ is

$$P(\theta \geq \theta_{min}) = \frac{1}{1 + \left(\frac{\theta_{min}}{\theta_0}\right)} \quad (1.28)$$

in which θ_0 is the relativistic screening parameter and depends on both atomic number and electron beam energy [74]. The elastic mean free path increases with increasing beam energy, meaning that for a fixed sample thickness the probability of an elastic collision occurring decreases exponentially with increasing beam energy. For most low Z-number elements this will mean that the probability of knock-on damage reaches a peak at low to medium beam energies, and is lowered at high beam energies > 200 keV, as has been shown in numerous studies [69–71, 74].

Still, for high Z-number elements, the threshold displacement energy is so high that the beam energy needed to cause knock-on damage exceeds the typical 300 keV, especially for materials with strong bonding nature.

Inelastic electron-electron interactions occur when a beam electron transfers some of its energy to a core electron in the sample. This may cause the core electron to be emitted as a secondary electron either forwards through the sample or backwards up the TEM column. Typically, this will be followed by the relaxation of an outer shell electron via the emission of a photon. Both the emitted photons and the energy loss of the electron are used in spectroscopic characterisation in STEM [77], although these are not used, and therefore not discussed, in this thesis. For conductive samples the abundance of available free electrons helps to fill any electron vacancies in atoms in the sample, which reduces the severity of the damage caused by secondary electron emission. Still, the inelastic electron-electron interactions frequently cause damage via defect formation and induction of stress and strain in the crystal structure of conductive samples [68, 70]. On the other hand, for non-conductive samples radiolysis can cause a net positive charge locally in the sample via the emission of secondary electrons, and can also change the bonding of the sample species and thus alter and damage the sample [68, 77, 78]. When an electron vacancy created by a beam electron is not filled, due to the lack of available free electrons to fill the void, the atom becomes ionized and both strong covalent bonds and weak van der Waals bonds in the sample may be broken as a consequence. When chemical bonds are broken, the molecule changes its shape and position to adjust, which for example causes loss of crystallinity in quasi-crystalline organic samples [69].

In addition to the two damage mechanisms introduced above, the irradiation of a sample with a highly energetic electron beam also inevitably cause thermal heating, which may be mitigated by cooling the sample. For this reason, biological specimens are typically imaged at cryogenic temperatures in electron microscopy. Regardless of the damage mechanism, the fewer electrons that hit the sample, the less damage occurs. Consequently, the shorter the duration of irradiation, the less damage is induced. While there are other parameters of the microscope that may be fine tuned to avoid causing too much beam damage, the most important control is that of the total electron dose. This may be reduced by controlling electron fluence and limiting the beam exposure time. At the end of the next chapter the dose equation is presented, as well as various experimental parameters controllable to reduce the electron dose delivered to the sample.

Chapter 2

Techniques and State-of-the-Art

No work in modern physics, and especially not in microscopy, is done without relying on a significant amount of groundwork done by others. From Ernst Ruska and Max Knoll building the first TEM back in 1931 [79], to Tiarnan Mullarkey and Jonathan Peters developing and building the Pulse signal digitiser [80] in very recent years, there are many contributions by fantastic scientists that have all created the space in which it was possible for me to do the work presented in this thesis.

In this chapter, recent advances in technologies and techniques, which have been the foundation upon which this work is built, will be introduced and explained. Firstly, the concept of STEM simulation is introduced, and some important practicalities are explained. Secondly, the technologies of detectors are introduced; explaining both the working principle of scintillator-based and four-dimensional scanning transmission electron microscopy (4D-STEM) detectors, as well as the signal digitisation hardware used for this work. This chapter also mentions some practical aspects that are important to keep in mind for experiments. Finally, the practicalities of achieving ultra-low doses in STEM and basic COM/iCOM data analysis with segmented detectors is introduced.

2.1 STEM Image Simulation

Assuming that there exists an analytic expression for the sample, $O(\vec{r})$, the STEM image that can be acquired with a probe, $P(\vec{r})$, given by Equation 1.8, may be simulated. Since the interaction between the electrons and the entire sample can be difficult to simulate in one go, it is common to do a multi-slice simulation where

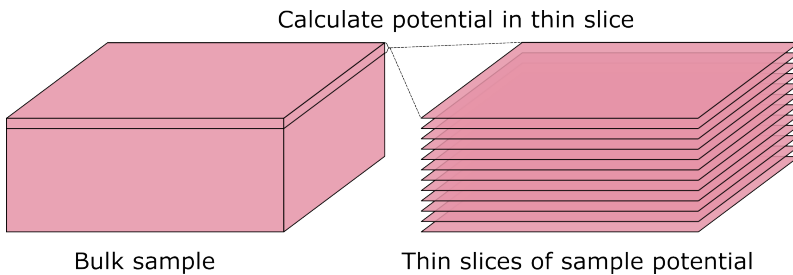


Figure 2.1: Illustration of how the bulk sample is broken up into thin slices such that the projected potential in each slice can be calculated for use in multi-slice TEM image simulation.

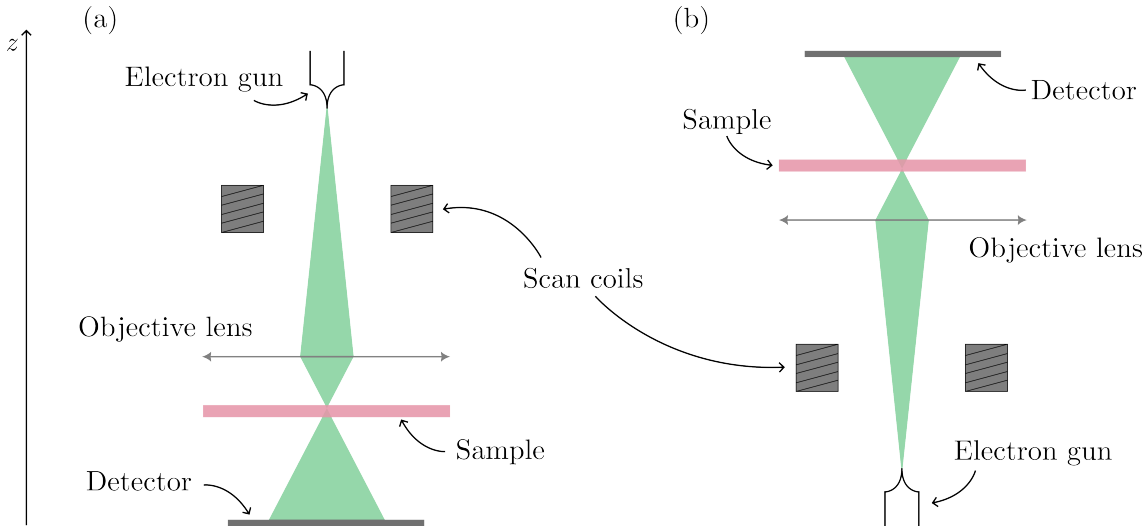


Figure 2.2: Simplified illustration of the setup of (a) Thermo Fisher Scientific and JEOL TEMs and the Prismatic image simulation software, and (b) the Nion STEM setup also used for abTEM simulations. Figure adapted from [87].

the sample potential is sliced up into many smaller projected potentials. Then, the exit wave from one slice is propagated and used as the input wave for the next slice [81, 82]. An illustration of how a bulk sample is broken up into thin slices is included in Figure 2.1. This type of multi-slice simulation is available in several TEM and STEM image simulation softwares [23, 83]. Any parameter change that changes either the probe or the object, for example changing defocus or otherwise altering the aberration surface, requires a new simulation.

At any non-zero temperature the atoms in the sample vibrate, and in crystalline samples these lattice vibrations are termed phonons. The average displacement of atoms in a crystalline sample varies periodically with a period which is much larger than the time each electron from the electron beam spends inside the crystal. Therefore, it is assumed that each electron only “sees” a frozen snapshot of slightly displaced atoms [1]. Averaging over many configurations will give a statistical representation of lattice vibration effects¹. This “frozen phonon” approximation is used in simulation to simulate the effect of phonons in crystalline samples and has been shown to yield results very similar to simulations of the full effect of lattice vibrations, at a fraction of the computational power [84–86].

Simulation softwares simulate the STEM signals using an infinite dose of electrons by default, giving simulated images that are probability images. This means that the intensity at each pixel in the simulated image is the probability of an electron being detected by the applied detector in that pixel. Both simulation softwares Prismatic [82, 83] and abTEM [16] allow for applying a high-angle annular dark field (HAADF) detector with a specified angular range, in addition to the possibil-

¹Note that this requires multiple simulations for each “frozen phonon” configuration.

ity of returning the CBED pattern at each scan position. In addition to this, abTEM allows applying segmented detectors directly to the simulation. Regardless of what virtual detector is applied, electrons may be scattered outside the detector range, which will lower the total detected intensity. Finally, a major difference between the two simulation softwares is that Prismatic defines a positive z -direction — and the direction of travel for the electron beam — down through the sample, akin to how one would normally draw the schematic for a STEM from Thermo Fisher or JEOL. Conversely, abTEM defines positive z -direction, and electron beam travel direction, to be up through the sample equivalent to the way Nion STEMs operate, see illustrations in Figure 2.2.

When a dataset has the image of the full CBED pattern for every scan position, it is typically called a 4D dataset because each pixel in the two-dimensional real-space image contains a two-dimensional image of the reciprocal space diffraction pattern. Importantly, the real-space extent of a simulated sample constrains the sampling in reciprocal space, with a larger real-space sample allowing for smaller pixel-sizes in the reciprocal space diffraction pattern. Pixellated detectors capable of acquiring 4D-STEM datasets experimentally are available, and in increasing use around the world despite being expensive to acquire and time-consuming to use [52, 56, 88], see Figure 2.4. A 4D-STEM simulation only excludes electrons that are scattered beyond the maximum collection angle specified for the 4D output, so most electrons may be collected, giving a very high collection efficiency.

In the following, the role of the detector in STEM imaging will be discussed further.

2.1.1 Detector Geometries

Both in experiment and in simulation, it is necessary to have some form of detection mechanism in order to visually interpret the data. The detector geometries may be sorted into four categories: monolithic, annular segmented, pixel arrays, and fully pixellated. An illustration of the three former may be seen in Figure 2.3. The denomination “pixel array” is used for an $n \times n$ detector array with n a small integer² to distinguish it from fully pixellated detectors with $n \gtrsim 60$.

Traditionally, the most common types of detectors in STEM are BF and ADF detectors. The former is placed inside the bright field disk, where it detects electrons that have been weakly scattered or not scattered at all by the sample. On the other hand, an ADF detector an annulus detector which covers an angular range in the dark field. An ADF detector detects only the electrons that were scattered to the angular range of the detector, which for typical HAADF imaging is in the

²Exactly where a “pixel array” detector becomes a “pixellated” detector is debatable, but what’s in a name?

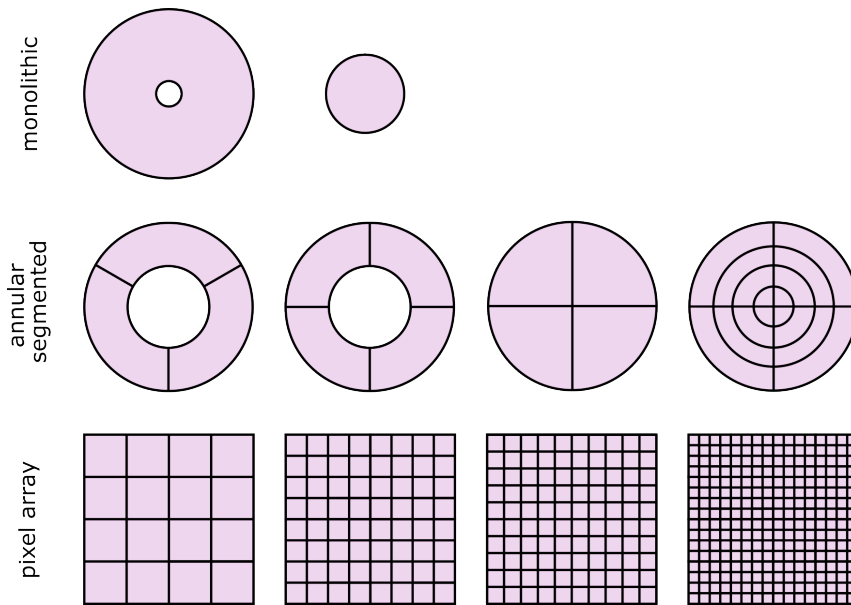


Figure 2.3: Illustration of various detector geometries, grouped into the types of detectors: monolithic (ADF, BF), annular segmented, and pixel array detectors. Pixel array detectors slowly become fully pixellated detectors when the number of array elements, or pixels, is increased.

range 50 – 200 mrad — depending on the semi-convergence angle used [1]. These traditional detectors are single-segment, or monolithic, detectors. This means that they do not distinguish between which direction electrons were scattered in — they simply detect the total intensity of unscattered or scattered electrons respectively. An illustration of their simple geometry may be seen in the top row of Figure 2.3.

While simple BF and ADF detectors offer easily interpretable images, they suffer from some important drawbacks. Since the proportion of electrons from the beam that is scattered out of the BF disk is typically small, the BF images often suffer from low contrast. This is exacerbated at lower electron doses, where the signal-disturbing noise is higher, and thus a high beam current is typically needed to get interpretable contrast, especially for low scattering materials. In addition, the atomic positions can be easily shifted due to misalignment of the beam on the sample, for example via sample mis-tilt [89–92]. Since BF detectors collect the unscattered electrons, atoms appear dark on a bright background in the images.

On the other hand, HAADF images feature bright atoms on a dark background, with the intensity of the bright atoms being proportional to $Z^{1.7}$ [93]. This makes heavy elements very bright and lighter elements difficult to see. Additionally, the proportion of electrons in the electron beam that are scattered to higher angles such that an ADF detector may detect them is small. The intensity that an annular detector may detect in STEM decreases exponentially with increasing inner collection angle of the detector [59, 94], thus the detection efficiency of HAADF is typically around 10% [1]. For low-dose applications, this is far too low to get interpretable

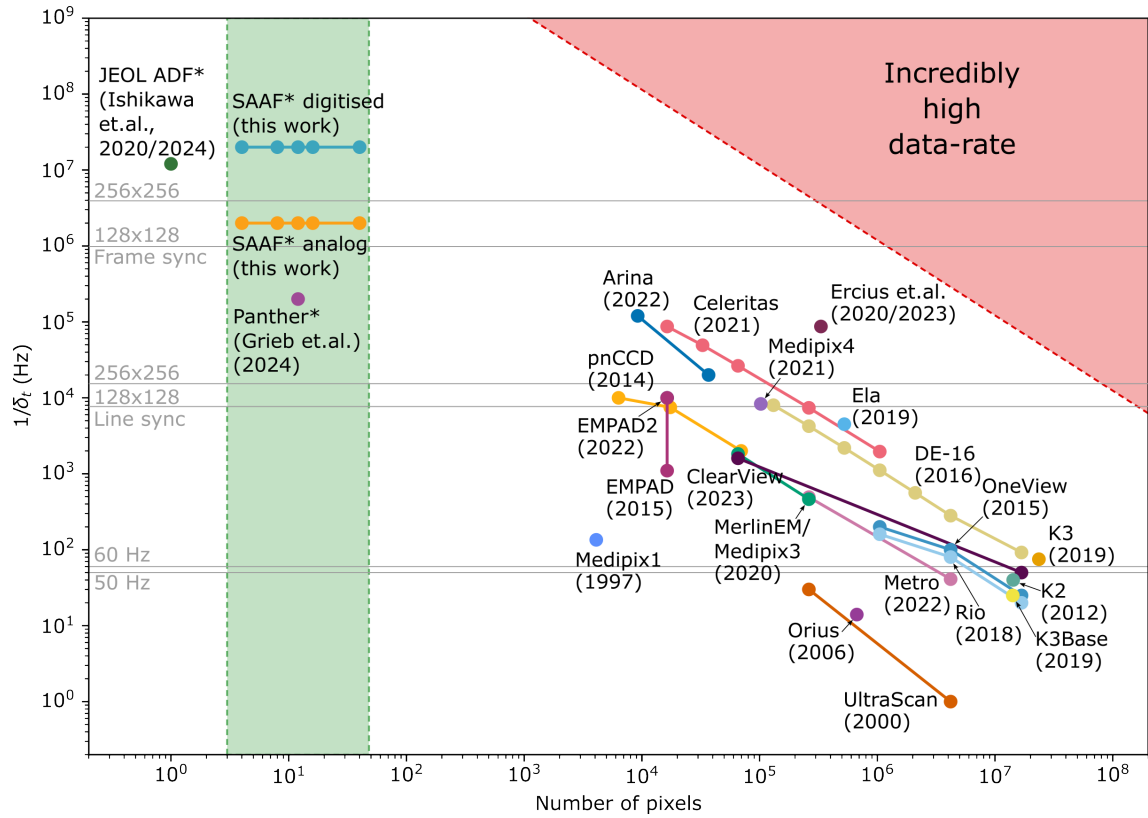


Figure 2.4: Overview of the number of pixels and maximum frame rate (as inverse minimum dwell time) of some commercially available cameras and 4D-STEM detectors. Figure from [95].

information about the sample before sample damage becomes problematic. When the electron dose is lowered significantly, detecting as many of the electrons from the beam as possible is crucial to maximise the information-to-dose ratio. A solution to maximising the information per dose could be to image the whole CBED pattern at every scan position since the CBED pattern contains a vast amount of information about the sample.

Pixellated detectors that capture an image of the CBED pattern at every scan position have become increasingly popular in the last decade. This is with good reason, since the extreme detail captured by such detectors may be used for highly sophisticated characterisation of materials. Thus, this offers amazing new characterisation capabilities for all areas of materials science [96–99]. However, reading out a full image of the CBED pattern at every scan step is time-consuming, which limits the pixel dwell time that may be used, as can be seen in Figure 2.4. This figure shows how the fastest commercially available³ 4D-STEM detector today is the Dectris Arina, which may be operated with $\delta_t = 10 \mu\text{s}$.

³Note that “the 4D camera” never made it to become a commercial product. It was developed by C. Ophus and P. Ercius at the National Center for Electron Microscopy at Lawrence Berkeley National Laboratory, and became a successful research project [100] without commercial adaptation.

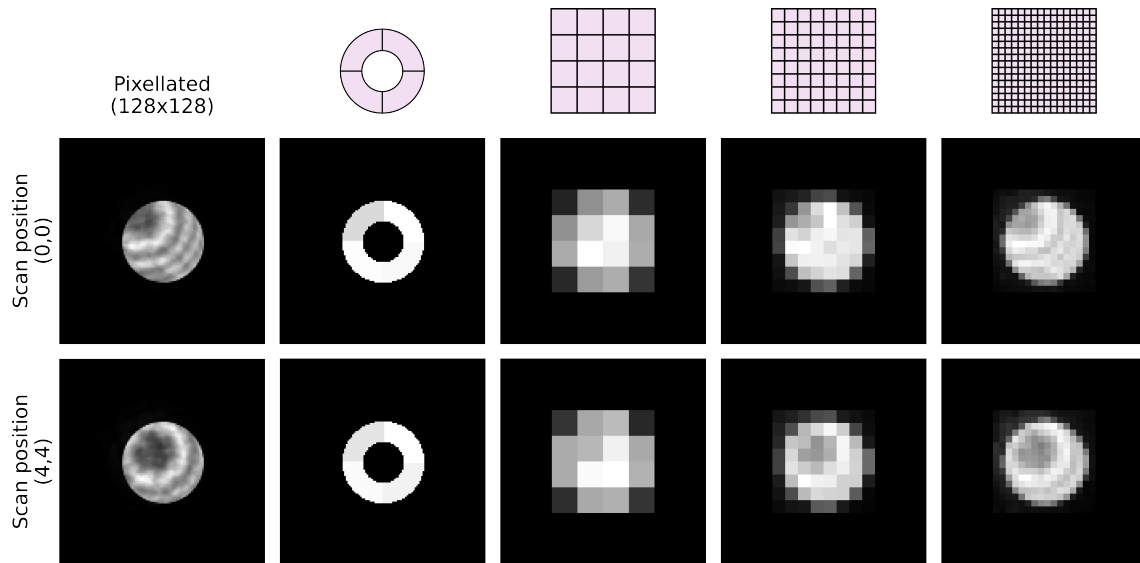


Figure 2.5: Example simulated defocused ($\Delta f = 128 \text{ \AA}$) diffraction patterns as seen by various different detectors in the BF region.

Not only do these detectors limit the dwell time that may be used for STEM characterisation, but the datasets that are produced when a 256 by 256 pixel image is acquired for every pixel in a 256 by 256 pixel image quickly reach several tens of GB, depending on the bit-depth that is used for the data. Some detectors allow the operator to choose their desired bit-depth, from as low as 1-bit to 32-bit, which helps to control the dataset size and the read-out overhead since lower bit-depth data may be read out from the detector faster [88, 101–103]. Albeit, this comes at the risk of clipping data due to the reduced dynamic range available with lower bit-depths. Finally, some detectors encode the intensity value in each detector pixel in more efficient ways than recording the intensity directly. An example is the EMPAD G2, which encodes a gain value and number of charge dumps among the data in its 32-bit read-out. This allows for a higher dynamic range at faster read-out speed [103, 104].

Throughout this work, many different geometries of detectors have been used both in simulation and experimentally either as physical detectors or as virtual detectors on an experimental 4D-STEM dataset. When investigating⁴ properties that only relate to signal detection and detector geometry, no re-simulation of the probe-object interaction is required, which means that virtual detectors may be imposed on a pre-simulated 4D-STEM simulation. This is significantly more efficient than re-simulating for every detector geometry, since the simulation can be computationally expensive and therefore time-consuming.

Virtual detectors are imposed on the simulated CBED patterns by binning pixels in each CBED pattern in the detector shape, see the example diffraction patterns for two close scan positions in Figure 2.5. The geometry of the detector plays a crucial

⁴in simulation

role in the resolution achievable and the information retrievable. Additionally, the experimental parameters chosen, such as the semi-convergence angle and the camera length, profoundly change the signals detected and thereby also the resolution and fidelity of the COM signals. As a rule of thumb, the higher the number of segments on a detector, the higher the achievable resolution [56].

In chapter 3, the optimal camera length for COM imaging with various detector geometries will be investigated. Additionally, how the geometry of the detector impacts the contrast transfer function (CTF) of iCOM will be discussed.

2.1.2 Addition of Dose in Simulations

Emission of electrons from the electron gun, and therefore also detection of electrons in STEM, follows a Poisson distribution, where the probability of $X = k$ electrons being emitted in a finite time interval t , given a rate parameter λ , is

$$P(X = k) = \frac{(\lambda t)^k e^{-\lambda t}}{k!}. \quad (2.1)$$

The rate of emission is given by the beam current, I , as

$$\lambda = \frac{I}{e} \quad (2.2)$$

where $e = 1.6 \cdot 10^{-19}$ C is the absolute value of the electron charge, see the resulting probability distribution for a time interval of 500 ns in Figure 2.6. Using the knowledge of the dwell time per probe position, the beam current used, and the

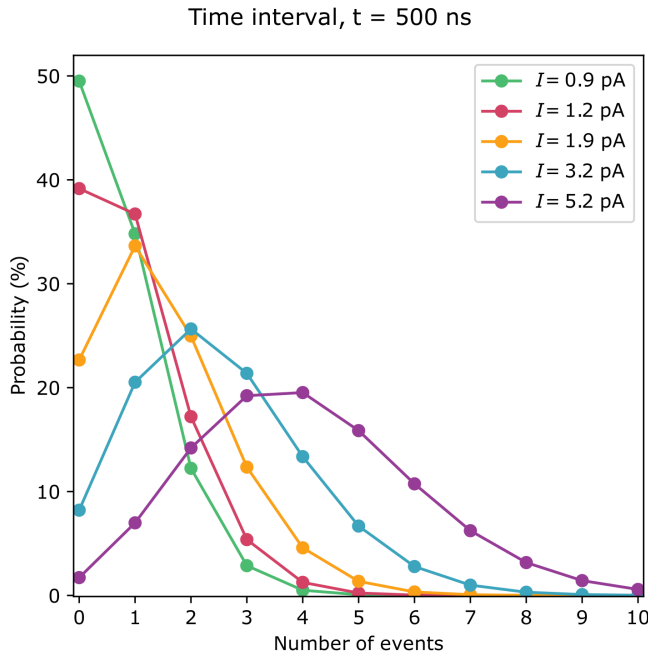


Figure 2.6: Poisson distribution for different beam currents within a finite time interval of $t = 500$ ns.

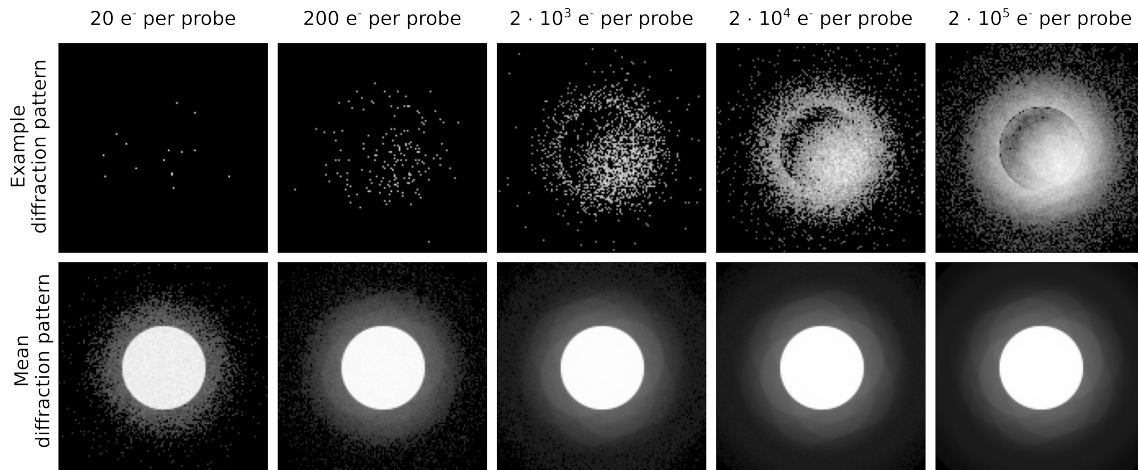


Figure 2.7: Example in-focus diffraction patterns and mean diffraction patterns at various low-dose conditions.

probability of detecting an electron in each pixel, a finite dose may be applied to a 4D-STEM simulation by performing a Poisson draw at each image pixel. This simulates the best-case scenario, low-dose image, where only Poisson noise is present. A few examples of diffraction patterns at various low, and not so low, doses can be seen in Figure 2.7.

In reality, there are many more sources of noise in a STEM image including, but not limited to, electromagnetic and vibrational noise from environmental surroundings, Gaussian noise from the image recording hardware, and sample drift due to the finite time it takes to capture an image. Additionally, the temporal response function of the detector itself plays a key role, especially for images acquired in very fast scans, which will be extensively discussed later. In chapter 4, the addition of dose to the image, including the temporal transfer function (TTF) [105] of the detector and other external noise factors, will be introduced.

2.2 Detector Working Principles and Signal Digitisation

Electron microscopy relies on detectors that can convert electrons hitting the detector to a meaningful signal for the human operator. There are many ways of achieving this, perhaps the most classic one being the fluorescent screen used to view the beam directly at the microscope. Most modern TEMs do not have fluorescent screens anymore⁵ and instead rely on charge-coupled device (CCD) cameras or complementary metal-oxide-semiconductor (CMOS) based detectors to view the TEM image or CBED pattern.

⁵Sadly, but the ones that do typically have screens that fluoresce green, which is the reason why electrons are considered to be green in the electron microscopy community.

A commonly used technology, both for CCD cameras and monolithic and segmented detectors, is to convert electrons to light using a scintillator. The incoming electrons hit a scintillating material, which excites photons that are transported to a photo-multiplier tube (PMT) using light-guides or mirror-lens systems [106–109]. These light-guides typically guide the signal out of the vacuum of the microscope and may be fiber-optics [110] or solid quartz rods [108], depending on the detector design. In the PMT, the light is converted into a current via the photoelectric effect and amplified such that the signal reaches a level that may be read out as a voltage, which is then sent to the computer and displayed to the operator as an intensity value on the screen [111]. Importantly, since the signal is modified along the way by adjusting brightness (constant applied bias) and contrast (applied gain) levels, the correct adjustment of these values for each specific experimental setup is vital for the integrity of the resulting images. In section 4.2 the factors that need to be considered experimentally for monolithic and segmented scintillator-based detectors will be discussed further.

CCD cameras are semiconductor-based cameras that convert electrons to current in each pixel, either directly or via a scintillator, which is then read out in a row-by-row or column-by-column fashion. Some early CCD cameras also relied on scintillators to convert electrons to light for detection [112]. These detectors marked a significant improvement to the photographic film that was standard to image acquisition at the time [113, 114] when they were introduced in the 1990s, since they enabled digital detection of images and thereby significantly increased the data-rate practicable [60, 115, 116]. CCD cameras relying on scintillators offer a good dynamic range, but suffer from low read-out speed and blurring due to neighbouring pixels scintillating when a pixel is hit. Additionally, the scintillating material may keep scintillating after being hit by many electrons, even when the electron exposure is removed, and further, their response to electron events often degrades over time. This may lead to “burn spots” or weak response areas on the detector, particularly in areas that are frequently struck by intense electron exposure frequently⁶. Despite their limitations, scintillator-based CCD cameras are still in use today, but scintillator-based ADF detectors or segmented detectors are far more common.

Scintillator-based CCD cameras may, in theory, be used as 4D-STEM detectors, and some are therefore included in Figure 2.4, but their high pixellation often makes this impractical. Some never CCD cameras transform electrons to read-out current directly, instead of relying on a scintillator to convert the electrons to light first. Regardless of whether a scintillator is used or not, the pixels in a CCD camera are read out in a row-by-row or column-by-column fashion. The serial read-out makes CCD cameras rather slow, and since the entire row is read out before the signal may

⁶Which is another aspect making BF imaging challenging.

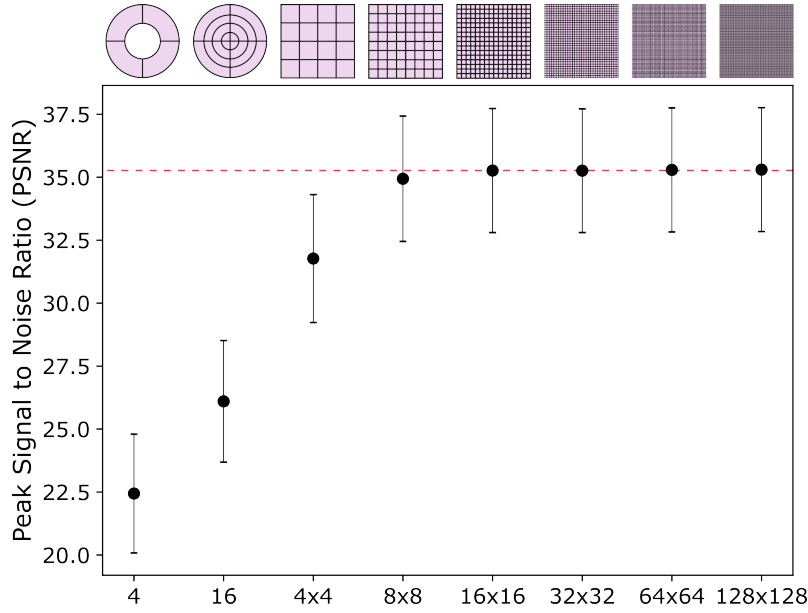


Figure 2.8: Peak signal-to-noise ratio (PSNR) in reconstructed iCOM images when using various simple detector geometries to estimate the COM signal by applying virtual detectors on a 4D-STEM simulation. The dose applied was 10 C/cm^2 or $6241 \text{ e}^-/\text{\AA}^2$. Figure from [95]

be amplified, they have poorer dynamic range than detectors that read out each pixel individually. Some CMOS-based detectors have a separate read-out circuitry for each pixel, which enables individual amplification and improves dynamic range at the cost of higher noise [117].

Some newer CCD and CMOS detectors are direct electron detectors that convert each electron hitting the detector surface to electron-hole pairs directly. The detector layer is biased such that either the holes or the electrons from the generated electron-hole pairs are read out as the signal. Importantly though, a single electron event may generate many electron-hole pairs in the detector, and as such, even direct electron detectors typically do not reach the perfect binary electron detection. Modern pixellated detectors are separated into two categories: monolithic active pixel sensor (MAPS) and (hybrid) pixel array detector (PAD) [109]. Both are direct electron detectors, offering higher efficiency than CCD cameras, but there are some key differences. MAPS detectors are radiation-hardened CMOS detectors that have a very thin sensitive layer, padded on a thicker substrate [117–119]. The thin sensitive layer allows for the individual pixels to be very small, typically around $6.5 \times 6.5 \text{ }\mu\text{m}$. This allows for these detectors to have many pixels, however, the dynamic range is typically limited since the pixels saturate easily. MAPS detectors also typically read out the pixel values in a row-by-row or column-by-column fashion, which is more time-consuming and thus limits their frame rate. On the other hand, PAD detectors feature larger pixel sizes of around $150 \times 150 \text{ }\mu\text{m}$. The active layer is typically in the range $200 - 500 \text{ }\mu\text{m}$, which is enough to fully absorb each

detected electron giving a higher dynamic range [103, 104]. Additionally, each of the pixels in a PAD is typically bonded individually to separate read-out circuits, which means all the pixels may be read out in parallel. As a result, these detectors have more complicated read-out electronics, and the number of pixels they can include is limited, but the benefit is that they have a higher read-out rate and larger dynamic range.

Even though pixellated detectors and 4D-STEM have become very popular, they do come with the drawback of limiting the scan speed, since they require a higher minimum pixel dwell time. Additionally, fully pixellated detectors may not be necessary for all phase retrieval methods. In their 2015 paper, Yang et. al. showed that 8×8 detector pixels seem to be enough for SSB phase retrieval [56]. A similar comparison of achievable PSNR in iCOM images can be seen in Figure 2.8. Since segmented detectors seem to estimate the COM signal very well and produce high-quality iCOM reconstructions, whilst achieving a high real-space frame rate, they are the main focus of this thesis.

Whether scintillator- or solid-state-based, all detectors have a finite response time to an electron hitting them, which will be described in the following.

2.2.1 Single Electron Events

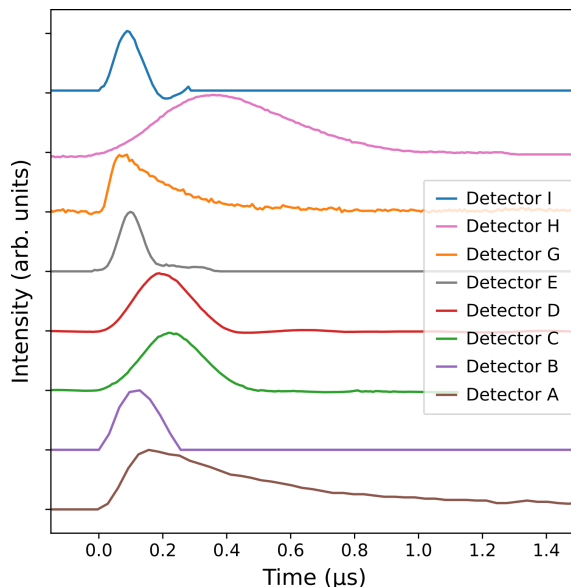


Figure 2.9: Single electron events acquired experimentally on multiple different detectors. The actual intensity of the pulses vary with offset and brightness settings at the specific detectors, so their intensities have been normalised to facilitate direct comparison of their shapes. Figure adapted from [120].

Electrons are, for all intents and purposes here, indivisible and quantised entities. There is no such thing as a weaker or a stronger electron⁷, and as such, electron detection is intrinsically a binary process; an electron is either detected or it is not. However, because electron detection events must be converted to an interpretable

⁷Ignoring any energy difference between electrons in the beam. The energy difference is typically small when compared with the typical beam energy in the range 80 – 300 keV.

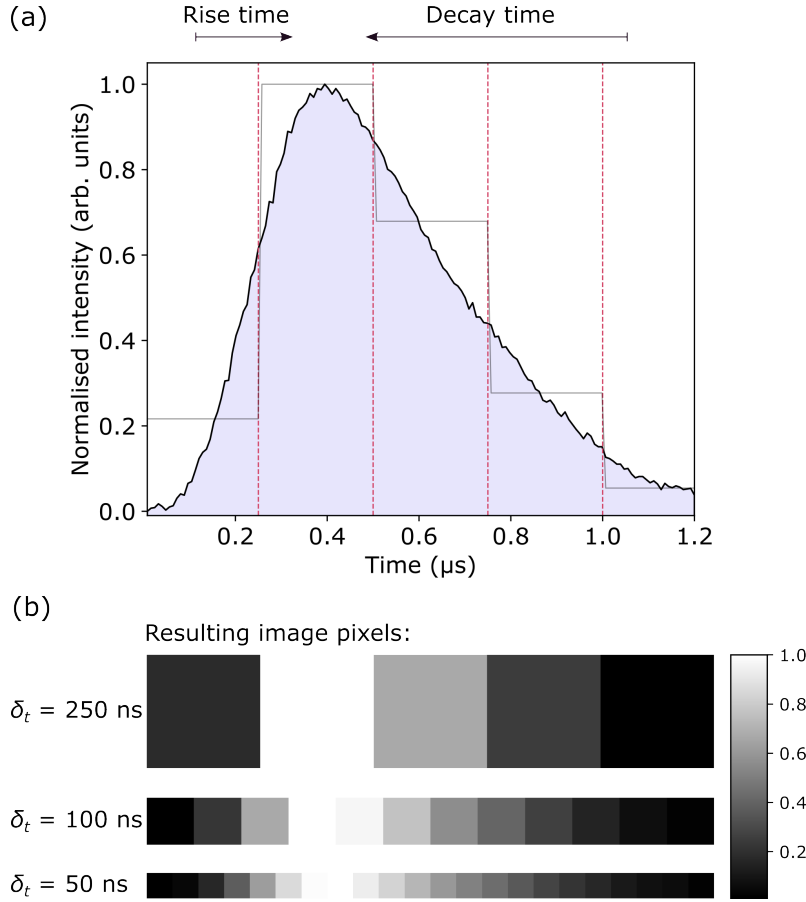


Figure 2.10: (a) A single electron pulse with the rise and decay times indicated. The dashed lines are the pixel borders using a dwell time of $\delta_t = 250$ ns, and the resulting streaking in the image when using three different dwell times can be seen in (b). Figure from [95].

signal, there are physical limitations that have to be accounted for experimentally. Several attempts at single electron counting in-software and characterisation of the shape of a single electron detection event have been made by others [111, 121–125]. Both scintillator-based and solid-state detectors produce a signal with a finite duration > 100 ns, and typically significantly longer, see some experimental pulses in Figure 2.9.

As previously described, for scintillator-based detectors, electrons are not detected directly, but via a light signal. In this case, the signal received by the computer is a convolution between the electron arrivals, the scintillator temporal response, and the response function of the PMT [105], as well as any noise unavoidably present in the detector [117, 126, 127]. The combination of the scintillator and PMT responses is often referred to as the detector response function, and is different for individual detectors. Figure 2.9 shows several different detector responses to a single electron event. Note how the elongated decay time in some of these reaches as far as 1 μs. A more detailed plot of the segmented annular all-field (SAAF) response may be seen in Figure 2.10(a). This elongated decay time can cause streaking artefacts in

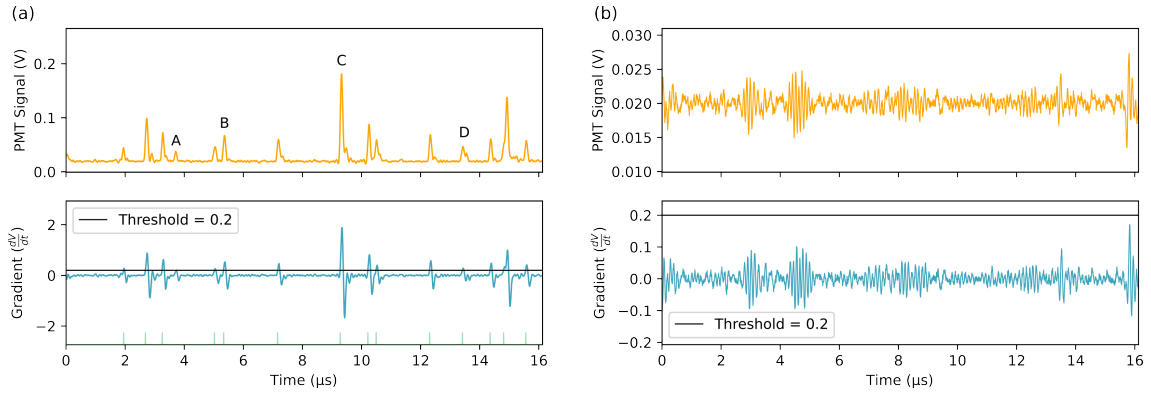


Figure 2.11: (a) Experimental pulse stream (top) and gradient of pulse stream (bottom) from a monolithic ADF detector. The digitised events when threshold $\Gamma = 0.2$ is applied are shown as green ticks at the bottom of the gradient plot. Electron events highlighted are: A, very weak signal — not digitised with the applied threshold. B, weak signal, but strong enough to be digitised correctly. C, very strong signal. D, two events too close to be separated. (b) Experimental noise stream from the same detector acquired with the beam blanked. Figure from [130].

the image [80, 125, 128], especially for lower dwell times, see the resulting image pixels in Figure 2.10(b). Additionally, the detector response function often varies across the detector surface, with some parts of the detector producing a weaker or stronger signal [125, 129]. This inhomogeneity in the detector response function further complicates the reliability of analog electron detection. Digitisation of the signal from scintillator-based detectors improves the speed and homogeneity of these detectors [80].

2.2.2 Digitisation

Digitising an analog signal typically requires applying a threshold for what is to be considered a signal event. An example of a detector signal stream may be seen in the top panel of Figure 2.11(a), and applying a threshold to this signal stream directly is a potential route to digitising the signal. However, this is highly prone to pulse pile-up, or coincidence loss, where fast-arriving electron events are impossible to differentiate [131]. The fast onset of coincidence loss with increasing electron arrival rate is one of the reasons why electron counting efforts by signal thresholding have been abandoned in the past [111, 121–123]. However, using the gradient of the detector signal for the digitisation pushes the onset of coincidence loss to a higher arrival rate, and makes the signal easier to threshold, because individual electron event peaks are more prominent and have shorter duration in gradient space, see Figure 2.11(a).

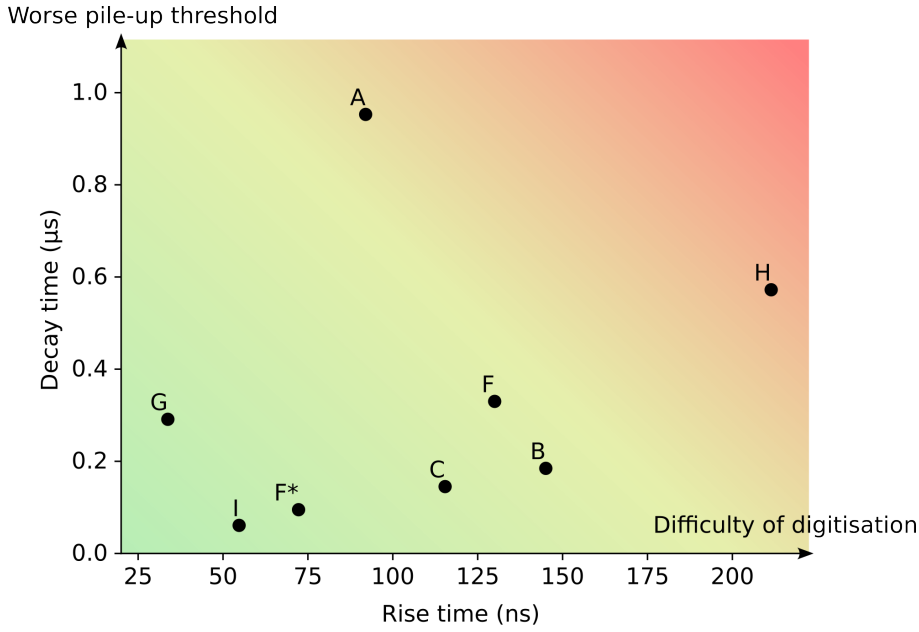


Figure 2.12: Comparison of the pulse rise time vs decay time for several different detectors. Figure adapted from [120].

The ease of digitisation, but also the need for digitisation, varies with the shape and duration of the single electron event at the detector. A scintillator and PMT combination with a longer decay time for each electron event will give streaking artefacts in the image at slower dwell times than a detector system with a shorter decay time, and as such will benefit more from digitisation of the PMT signal. However, the broadness of the electron events in the PMT voltage signal will also result in event count loss due to event pile-up at a lower arrival rate. Additionally, scintillators with a slower rise time have a lower gradient and, as such, are more difficult to digitise compared with scintillators with a sharp onset. An overview of the rise vs decay times for various different detectors can be seen in Figure 2.12.

With electron pulses being sharper and shorter in duration, electrons may arrive closer together in time and still be digitised correctly, which opens up the use of digitisation for practical beam currents. An experimental pulse stream from a fast-scintillator ADF-detector can be seen in Figure 2.11, where the events marked by D are too close to be separated in the digitisation causing coincidence loss. The gradient signal is calculated from the definition of the derivative,

$$f'(t) = \lim_{h \rightarrow 0} \frac{f(t+h) - f(t)}{h}, \quad (2.3)$$

by subtracting the signal from h ns ago from the current signal. Ultimately, the digitisation of single-electron detection events requires the individual events to be sufficiently spaced out in time such that they can be separated from one another by thresholding the gradient of the PMT voltage signal. Using hardware to do this in

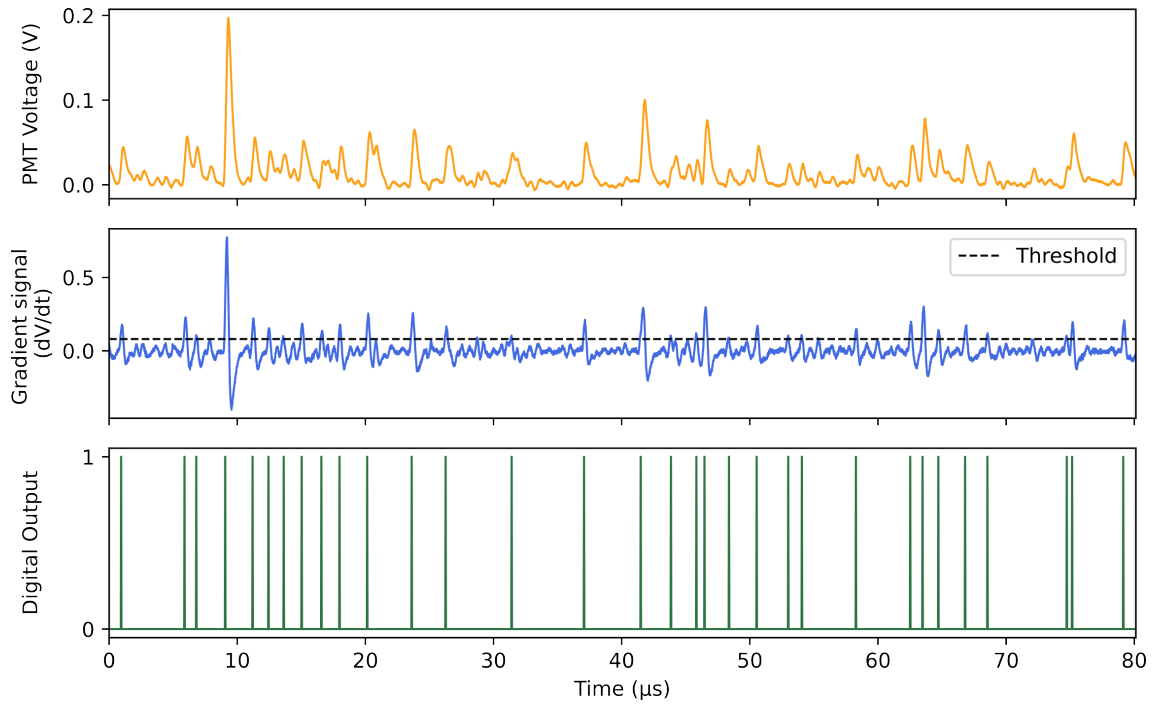


Figure 2.13: The top graph shows the voltage from the PMT as recorded with the digitisation hardware. In the middle is the gradient of the voltage signal and the bottom graph shows the digitised signal output from the digitisation hardware.

real-time, significant improvements in the signal-to-noise ratio for ADF images have been demonstrated [80, 124, 125].

In code the gradient signal is given by

```
pulse_gradient = (pulse_stream[h:] - pulse_stream[:-h]) / h,
```

where h is as small as possible in order to take the gradient as close to real-time as possible, yet large enough to provide a decently low noise level in the gradient signal. Due to noise unavoidably present in the detector [117, 126, 127], pulse streams are typically very noisy and taking the gradient with a small h yields mostly the gradient of the noise. An example of the noise present in a monolithic detector, and the gradient of this noise signal, may be seen in Figure 2.11(b). If the step h is allowed to be too large, the digitisation will lag behind the real-time signal, and with even larger steps it may skip pulses entirely. As a result, the h needs to be high enough to ignore the most high-frequency noise, yet low enough to properly calculate the gradient of weak electron pulses. An experimental pulse stream with 1.2 pA beam current, as well as the gradient with the threshold and the resulting digitised signal, can be seen in Figure 2.13.

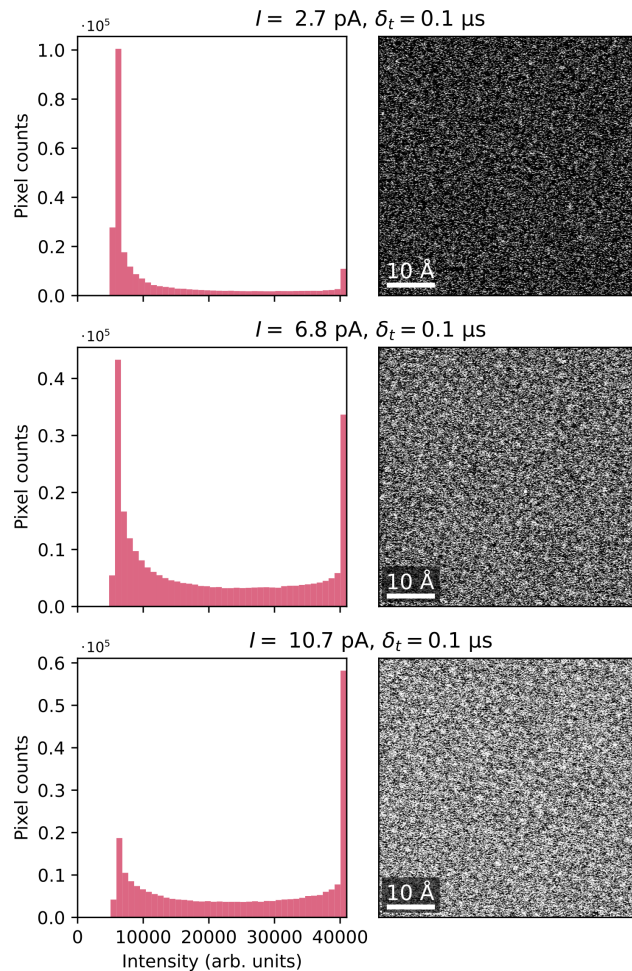


Figure 2.14: Example analog single-frame ADF images acquired with detector offset and gain values set too high for the experiment. As a result, the images are saturated at high values and the image histogram shows an elevated number of image pixels at the highest intensity values.

2.2.3 Setting Gain and Offset Appropriately

Changing the gain and offset of a detector is available to the microscope operator, but care should be taken to set these values correctly, such that no information about the sample is lost in the imaging process. If the offset and gain are set too high, the highest image intensities may be lost due to being clipped in the image capturing software, see the examples shown in Figure 2.14. Conversely, if the offset and gain are set too low, information may be lost due to the lowest image intensities being insufficiently amplified and thus not being adequately recorded in the image capturing software. In other words, the lowest image intensities are clipped, see two examples of this in Figure 2.15. Here, the gain value is set too low for the beam current of 3 pA that is used, which is visible in the histogram of a single frame taken with $\delta_t = 0.1 \mu\text{s}$, but is even more apparent when increasing the dwell time to $1 \mu\text{s}$. Clipping the image intensities, either low or high, causes loss of information in the image and can lead to incorrect image analysis, like artefacts in the fast Fourier transform (FFT)[1]. Images and histograms of image intensities for appropriate gain and offset levels may be seen in Figure 2.16.

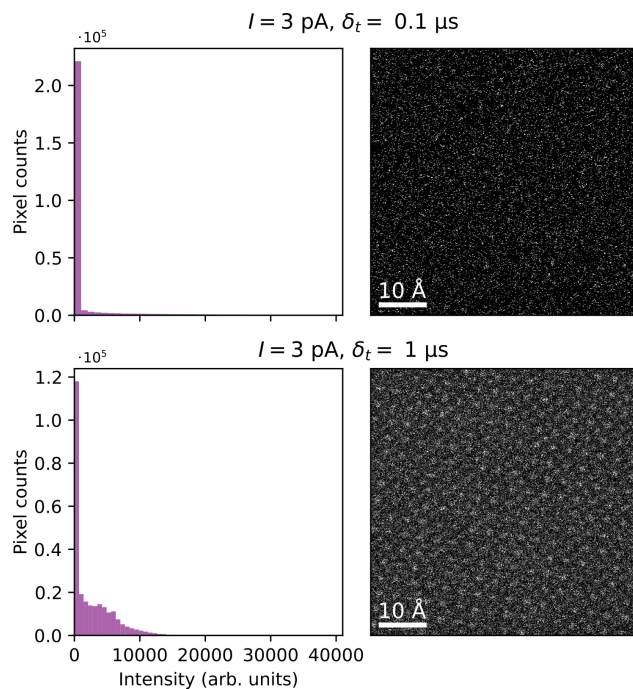


Figure 2.15: Example of the effect of setting the detector offset and gain too low for the experimental parameters used. The lowest image values are clipped, resulting in an elevated count at low image intensities.

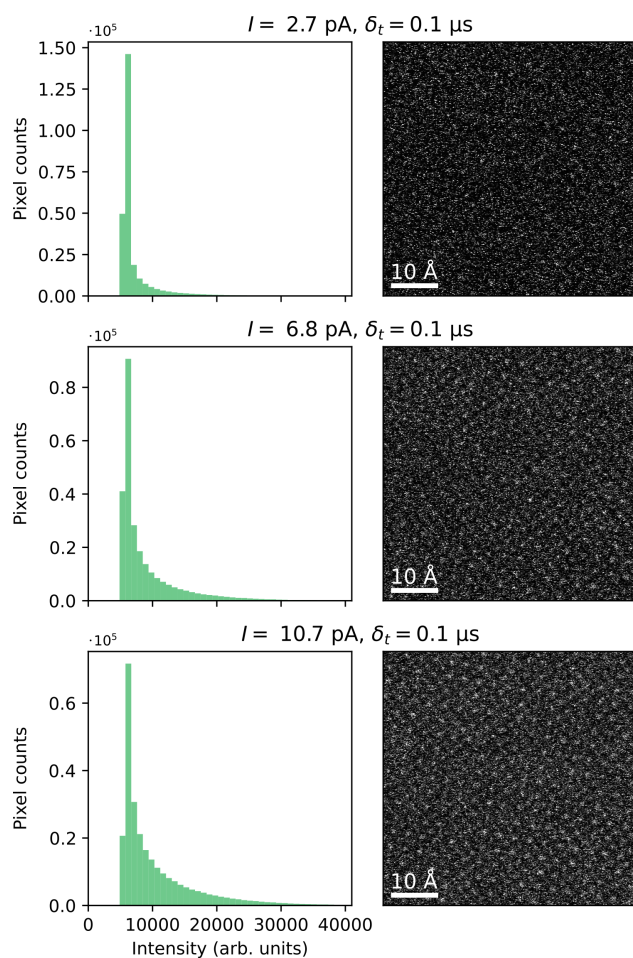


Figure 2.16: Single-frame analog ADF images captured with appropriate gain and offset values for the detector.

When digitising the signal from the detector, the signal read by the digitisation hardware must be taken into account in addition to the signal received by the image capturing software. The digitisation hardware itself can amplify the signal it receives; however, if the gain and offset are set too low for the detector the individual electron events will be weak, and amplifying the signal in the digitisation hardware will also amplify noise significantly. On the other hand, if the gain and offset for the detector are set too high for the digitisation hardware to receive it properly the electron event signals may be clipped, resulting in artefacts in the digitisation, like an event being counted two or more times. For a monolithic detector like the ADF, the gain and offset values can be verified by viewing the histogram of the image in the image capturing software, whilst simultaneously viewing the PMT voltage as received by the digitisation hardware. In this case, the gain and offset can be adjusted until the histogram contains all image values, and no event peaks are clipped in the PMT voltage stream.

For the segmented detector, setting appropriate gain and offset values for the digitisation hardware becomes more convoluted since the gain and offset of individual segments should not be changed separately. This is because it is vital, if the COM signal is to be correct, that the segments are standardised, meaning that the gains and offsets of the segments are set such that all four segments detect electrons equally. In other words, all four segments should detect the same signal intensity when uniformly illuminated. Standardisation of the segmented detector segments is achieved by iteratively changing the gain and offset values until the equalisation requirement is satisfied and the signal intensity level is within the predefined limits. In the SAAF detector software provided by JEOL, this is done with the auto-contrast and brightness scheme which iterates through the segments while sequentially blanking and unblanking the beam to set gains and offsets for the segments according to a goal interval of acceptable intensity values. Subsequently, the image histogram may be checked in DigitalMicrograph[™] and the PMT voltage as received by the digitisation hardware to ensure that all values are within acceptable ranges. The internal gain and offset in the digitisation hardware may then be set such that the signal peaks are as prominent as possible without being clipped, such that the gradient signal becomes as easy as possible to threshold.

2.2.4 Dealing with Digital Signals Experimentally

As a final note on dealing with digital signals experimentally, the fact that digital pulses are square waves needs to be considered. When sending square waves through a coaxial cable, it is important that the terminal port of the cable matches the impedance of the cable itself. If there is a mismatch between the impedance of

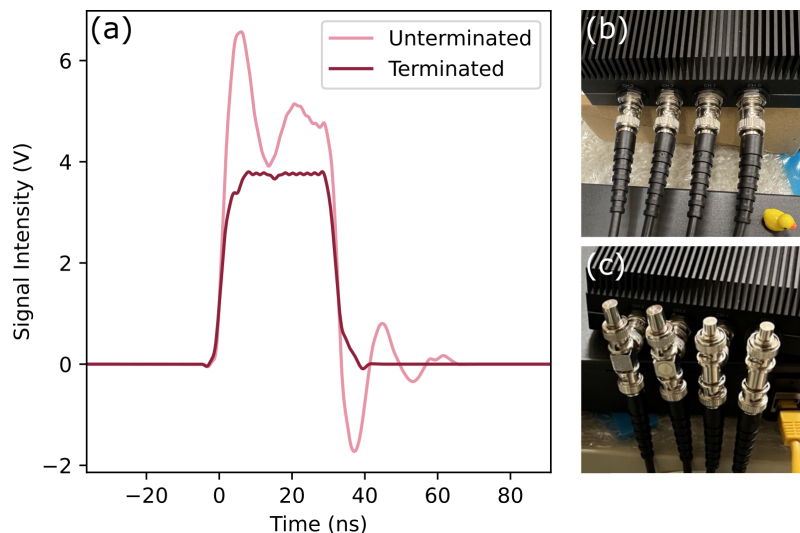


Figure 2.17: (a) Digital square pulse signal in coaxial cable with and without $50\ \Omega$ termination to match the cable impedance. (b,c) Image of coaxial cables connected to the direct digital controller (DDC) without and with a resistor terminating the cable respectively.

the cable and the connection, the signal may be reflected back down the cable and interfere with the incoming signal. The result of this is signal ringing, an example of which may be seen in Figure 2.17. Such ringing of the signal can lead to misinterpretation, and especially difficulty separating fast arriving signals. Therefore it is crucial that the coaxial cables have the correct termination to ensure impedance matching. Since only a few scan generators accept digital signals as inputs, the square digital pulses must be sent into the scan generator as analog signals and re-digitised in software. This exacerbates the need for the correct termination of the coaxial cables to maintain signal integrity.

2.3 Dose Budgets and Achieving Ultra-Low Dose

The high energy electrons that make up the electron beam in STEM may interact with the sample in ways which cause damage. The two main mechanisms for sample damage were introduced in section 1.5, from which it is known that sample damage occurs for all types of samples [69, 70, 72, 73]. However, the severity of the beam damage is highly beam energy- and sample-dependent. Some samples, typically crystalline conductive samples, can withstand electron irradiation on the order of $10^6\ \text{e}^-/\text{\AA}^2$ without showing signs of sample damage. Other samples can start to damage even before a full image of them has been acquired at doses as low as 100s of $\text{e}^-/\text{\AA}^2$. Typically, lighter elements such as lithium are more volatile, which makes battery materials so difficult to image [92]. Similarly, samples with a high content of oxygen, like a metal organic framework (MOF) or biological material, are very sensitive to electron beam irradiation. The general maximum dose that a range of different materials may be exposed to before they undergo irreversible damage, the so-called dose budgets, are summarised in Table 2.1.

Table 2.1: Typical dose budgets for different classes of materials covering a range of beam sensitivity levels [69, 94, 97, 128, 132–138].

Type of material	Typical Dose Budget ($\text{e}^-/\text{\AA}^2$)
Metal nanoparticles (e.g. Au, GaAs)	$10^5 - 10^8$
Perovskites (e.g. SrTiO_3 , PbZrO_3)	$10^4 - 10^6$
2D materials (e.g. graphene, hBN, PtSe_2)	$10^2 - 10^5$
MOFs and zeolites	$10 - 10^3$
Biological materials (e.g. viruses, proteins)	$0.1 - 30$
Liquid cell bio. materials	$0.01 - 5$

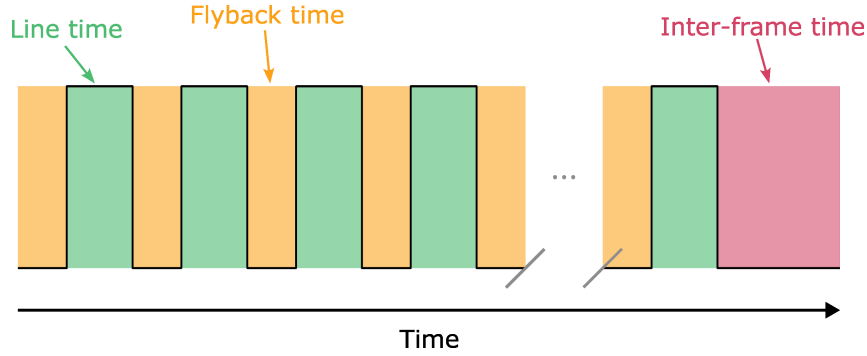


Figure 2.18: Illustration of the times in a multi-frame STEM acquisition. The line time is the only time contributing to the actual imaging. Figure adapted from [139].

Electron dose is defined as the number of electrons incident on the sample per unit area,

$$\text{Dose} = \frac{I \cdot \delta_t \cdot C}{(dx)^2} \cdot F_s \quad (2.4)$$

where I is the beam current, δ_t is the dwell time, C is the Coulomb number, and dx is the pixel size [139]. F_s denotes the fraction of pixels hit if a compressed sensing routine is employed or a subset of the pixels in an image is scanned in another way, like interlaced scanning. Considering Equation 2.4, there are a few routes to lowering the electron dose in STEM, namely reducing the beam current, reducing the dwell time, increasing the pixel size and reducing the number of pixels scanned. The first two are the primary routes taken most often due to being readily available to anyone with a STEM. Increasing the pixel size significantly reduces the resolution, and considering the above limitations of the STEM probe and its limited capability of detecting the projected electric potential, the beam size and thus the pixel size has to be reduced as low as possible. The last possibility is availing of compressed sensing, meaning skipping certain pixels during scanning, can be employed in several different ways, including scanning random pixels from several lines at a time or simply by random position sampling, although these are not necessarily all practical options.

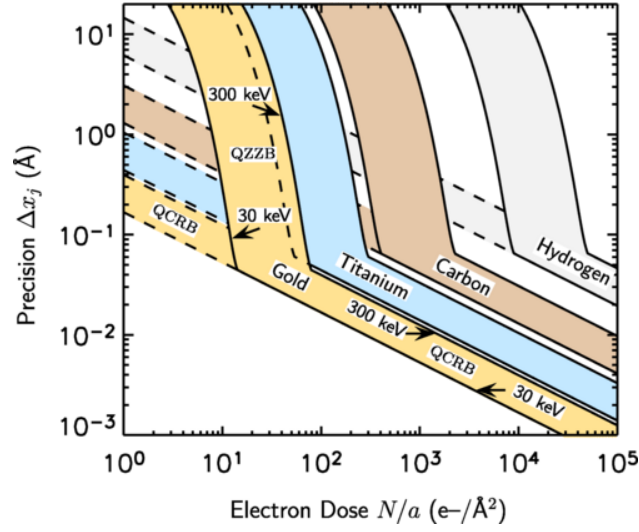


Figure 2.19: Quantum mechanical precision limit for atomic positions for single atom scatterers as a function of electron dose. The outer solid lines of the coloured areas indicate 30 and 300 keV beam energies, respectively, showing that lower beam energies could achieve higher precision provided that all other factors stay the same. QZZB and QCRB denote the quantum Ziv-Zakai bound and the quantum Cramér-Rao bound, which are two different expressions for the quantum mechanical precision bound that are valid in different regimes, see the paper by Christian Dwyer [141].

Importantly, STEM images can be acquired with a range of different scan patterns, with the most common one being raster scanning where all lines are acquired with the beam travelling in the same direction. This means that the beam has to travel back to the start of the next line at the end of each line, and this time is referred to as the line flyback time, T_{LFB} . Since the scan coils used to deflect the beam to the scan points are electromagnetic, the change of direction is always accompanied by hysteresis in the coils. For this reason, the beam typically flies back to a position a significant distance before the scan area to reach a constant velocity before the acquisition of the new line starts. Reducing line flyback time can result in distortions at the start of each line and as a result, line flyback times as long as 500 μs are often used. For multi-frame image acquisition, there is also a flyback time, T_{FFB} , associated with the end of the frame to get back to the starting position before the start of the next frame, see illustration in Figure 2.18. The total frame time may therefore be expressed as

$$\text{Frame time} = \delta_t n_p n_L + T_{\text{LFB}} n_L + T_{\text{FFB}} \quad (2.5)$$

where n_p is the number of pixels in the scan and n_L is the number of scan lines [139]. Efforts are being made to correct for the scan coil hysteresis by post-processing correction, and by developing high-speed scan coils with reduced hysteresis effects [139, 140].

It is also worth noting some consequences that come with ultra-low doses. Previously mentioned was the fact that detector surfaces tend to be quite inhomogeneous [122, 129, 142–146]. At high electron doses, the inhomogeneities will matter less since they will be averaged out by the sheer number of electrons hitting all over the detector for each scan position. However, when the dose is lowered and only a few electrons are the source of signal intensity for each scan position, the possibility of pixels appearing artificially dark or bright simply due to detector effects is increasingly present. Yet, using the digitisation technology described in this chapter, this is a solved problem, as every electron detected is a digital 1 in the digitised image, giving an immediately quantitative image. What is, however, not a solved (or even solvable) problem is the quantum mechanical precision limit. As the dose is severely reduced, the precision with which the sample parameters may be determined is bound by the Fisher information limit [141]. For highly scattering materials, the precision limit does not suffer greatly until doses on the order of $100 \text{ e}^-/\text{\AA}^2$ are reached, as can be seen in Figure 2.19 produced by Christian Dwyer in 2023 [141]. Now, multiple scattering will alter the limits, and for most practical applications the precision achievable is limited well above the quantum limit by other factors. Still, it is worth noting that for low atomic number elements like carbon, the quantum limit is reached far before the typical dose limit at which a biological sample suffers noticeable damage.

2.4 Basic COM and iCOM Data Analysis

In experiments with segmented detectors with four segments, the COM is typically estimated by subtracting opposing detector segments, as shown in Equations 1.22 and 1.23. If the inner and outer collection angle of the segmented detector is known, the center of mass of each segment may be calculated. This allows for quantification of the COM signal through multiplication with the normalised measured intensity from each segment, following Equation 1.22. Normalisation of each detector segment signal in post-processing also corrects any inequalities between the applied brightness and contrast of individual segments, assuming that the full image mean signal level should be similar in each segment, such that normalisation is sensible. Note that this may or may not be the case, depending on the sample and field of view. For example, for atomic resolution images that are on-zone, this is a good assumption.

For low-dose images with low beam current and short dwell times, multiple frames must be acquired to accumulate enough signal to get interpretable images. Such acquisition of multiple subsequent frames inherently gives a time-resolved dataset, as each frame represents (ideally) one static frame in time. This will be discussed further in chapter 5. Depending on the number of frames, a multi-frame dataset can

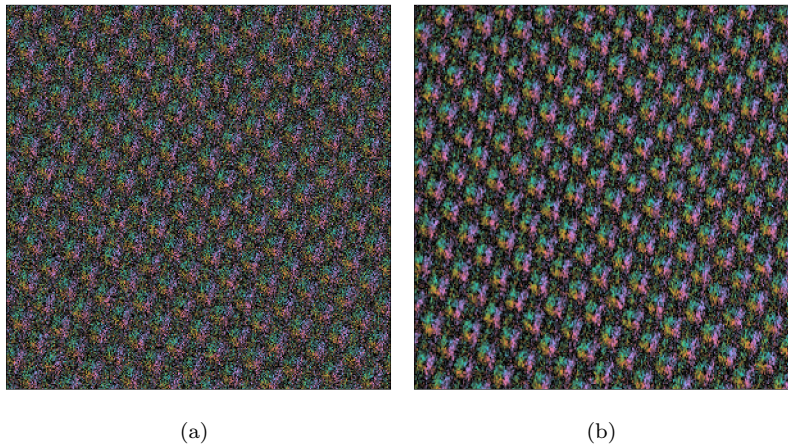


Figure 2.20: (a) Rigid aligned using S_1 , (b) non-rigid aligned using iCOM of single-frame images.

take anywhere from a few seconds up to a minute or more to capture. To correct for sample drift during multi-frame acquisition, the dataset should be drift-corrected in post-processing, and the software SmartAlign is used for drift-correction in this work [4]. Now, for a simple HAADF image stack, drift correction is straightforward since there is only one detector segment and therefore only one image stack. Thus, drift correction is done on the only image stack, typically before any other image processing is applied. On the other hand, for a multi-frame stack from a segmented detector, the order of operations becomes more complicated. Of various different options, the tested methods were:

1. Sum all segments and drift correct the resulting ABF signal.
2. Drift correct the multi-frame stack of a single segment, and then apply the same drift-correction to the three other frames.
3. Calculate the iCOM for each single frame and drift correct the iCOM signals.

For digitised signals from a segmented detector, the resulting ABF signal is too noisy to give a usable drift correction. Thus, rigidly aligning a stack of 200 frames captured at $\delta_t = 200$ ns using the sum of the segments is very difficult, even though the sum of the segments should have a higher signal than a single segment itself. The two other options, however, both provide adequate drift corrections, although drift correcting a single segment before further processing is far simpler and more practical in reality. Resulting COM images from a multi-frame stack, when aligned using a single segment and the iCOM signal, may be seen in Figure 2.20.

2.4.1 Gaussian Blurring and High-pass Filtering

When integrating the COM signal to retrieve the more easily interpretable and phase-proportional iCOM signal, it is very common that the integration introduces low spatial frequency artefacts in the resulting iCOM image. This becomes a bigger

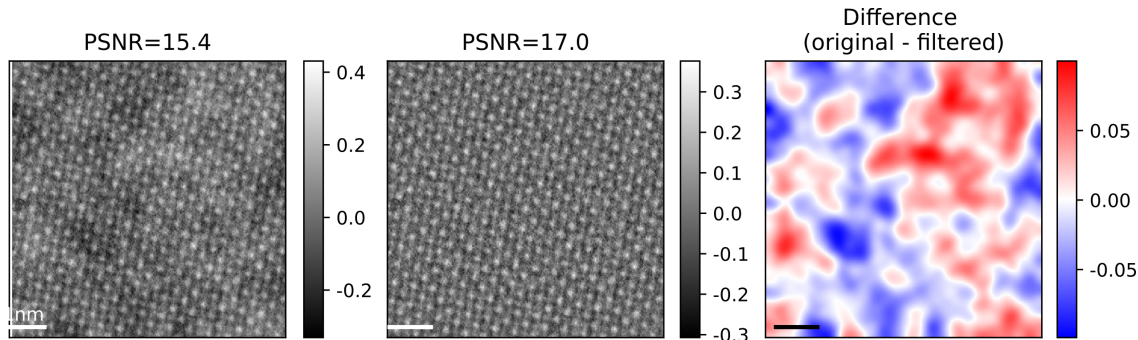


Figure 2.21: Unfiltered and high-pass filtered iCOM images as well as their difference (the low-pass).

problem the lower the dose is because the higher noise introduces more uncertainty, inhibiting the accurate integration of the COM signal. Typically, low signal images, whether analog or digital, are denoised to make them easier to interpret. The simplest form of denoising is Gaussian filtering, which convolves the noisy image with a Gaussian kernel of a given standard deviation, σ . A larger σ yields a broader Gaussian kernel, which produces a smoother image, eventually at the cost of the information limit of the image. Conversely, a smaller σ produces a sharper Gaussian kernel and thus less denoising, such that a balance has to be found where the denoising is useful without compromising image information content. A typical rule-of-thumb is to avoid using a σ that is higher than the minimum feature size the image is resolving.

For monolithic detector signals like HAADF images, the application of the filter can easily be done directly on the image. Translating this operation to the segmented detector scenario, each segment image is here denoised before doing any calculations to get the COM and iCOM signals. In theory, the denoising step could be performed on the COMx and COMy signals, or even on the vector COM signal directly, or on the final iCOM signal. It is worth noting that the subtraction which is performed to get the COMx and COMy signals is a noise-boosting operation, and as such the benefit of denoising is assumed to be highest before subtracting the segments. Oppositely, the integration to obtain the iCOM signal is noise-suppressing. Yet, the more noisy the COM signal is, the worse the noise suppression of the integration works and the more low spatial frequency artefacts are introduced, as will be more extensively discussed in chapter 5. As a result, applying the standard Gaussian filter to an iCOM reconstruction is more likely to exacerbate the low spatial frequency artefact and obscure information, rather than helping interpretability — and these iCOM images are typically smooth enough regardless.

The more natural post-processing of iCOM images is the removal of the low spatial frequency artefacts. This is typically done by applying a high-pass filter, which is equivalent to subtracting a low-pass filtered version of the image. The low-

pass filtered image can be obtained by applying a Gaussian filter with a high σ . An example of iCOM images with and without a high-pass filter, as well as the difference between them, can be seen in Figure 2.21. A more general way of filtering an image is to apply a Butterworth filter in Fourier space. This filter provides a smooth cut-off bandpass for the spatial frequencies in the image, for which the steepness of the cut-off can be user-defined. The higher the Butterworth order, the sharper the cut-off at the specified spatial frequency. Conversely, a Butterworth filter with order 2 for low-pass has a similar behaviour to a Gaussian filter. When applying a high-pass filter to remove low spatial frequency artefacts in iCOM reconstructions, it is important to avoid removing useful information from the image. This can be ensured by investigating the difference between the unfiltered and filtered image, to check that the difference does not contain atomic scale or periodic information, see Figure 2.21.

2.5 Conclusions

This chapter has introduced important background from the literature upon which the current work is built. Primarily, the concept of STEM image simulation and how realistic detector geometries and electron doses may be applied was explained. The investigations show that virtual detectors on 4D-STEM simulations may be used to simulate the COM signal from any detector geometry. When using detectors with few enough segments such that the read-out overhead from the detector is not limiting the scan speed, the minimum dwell time that may be used is limited only by the scan coils of the microscope and the temporal transfer function of the detector [105]. Importantly, a pixel-array detector of 8×8 detector elements seems to be sufficient for iCOM imaging.

Further, the method of signal digitisation using the gradient of the signal from the detector was introduced, and important practicalities to consider when dealing with digital signals in experiment were mentioned. Crucially, the appropriate setting of gain and offset, as well as cable termination, is vital for proper signal acquisition whether it is analog or digitised.

In the following chapter, simulation-informed optimal parameters for COM imaging with segmented detectors will be discussed, as well as the impact of the detector geometry on the contrast transfer of iCOM.

Chapter 3

Optimisation of Experimental Parameters for Phase Imaging

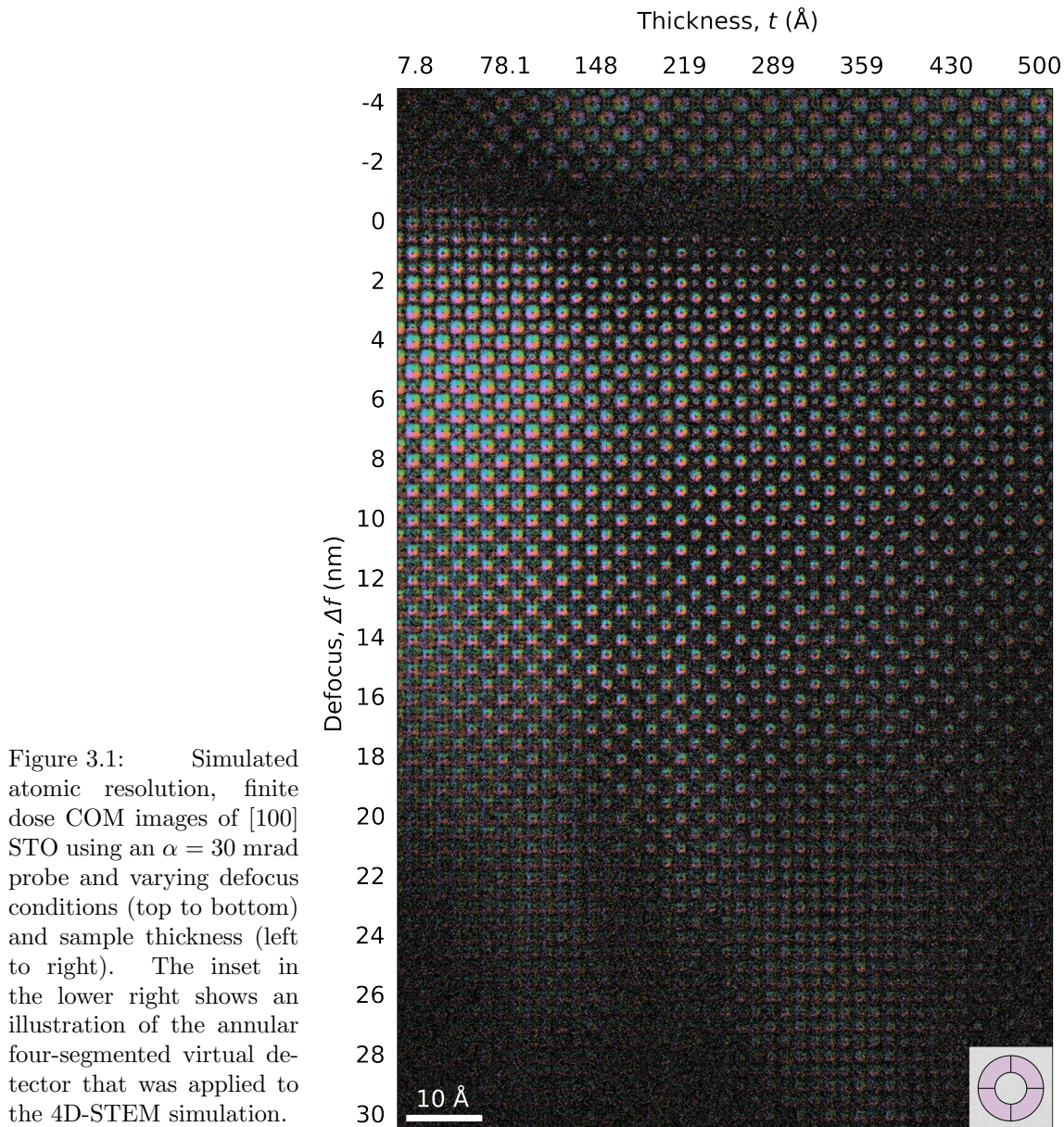
It is known from the literature that phase retrieval imaging is very sensitive to the experimental parameters chosen. Specifically important is the zone-axis of the sample because mis-tilt of the sample may be falsely interpreted as a long-range field [89, 91]. Additionally, the centring of the beam on a segmented detector, the defocus chosen, and the thickness of the sample itself all heavily impact the COM signal [92, 135, 147–149].

This chapter introduces simulations that were done to find the optimal defocus condition and sample thickness for COM imaging. Then, the effects of misalignment of the detector relative to the beam are investigated, and procedures for centring the beam on the detector are explained. Furthermore, the optimal collection angles for annular segmented detectors that yield the optimal signal intensity and COM signal integrity are found. Finally, a framework to analytically and numerically investigate the information transfer for any phase retrieval method, also beyond iCOM, is introduced and used to confirm the results of the aforementioned investigations. The framework is also used to compare the phase retrieval techniques iCOM and iterative ptychography.

3.1 Probe Defocus and Sample Thickness

For thin, non-magnetic samples, the iCOM signal is proportional to the projected electrostatic potential of the sample through the phase shift imposed on the electron wavefunction [150], as was introduced in subsection 1.4.1. Crucially, the phase shift acquired by the electron beam from traversing the sample is dependent on the thickness, t , of the sample, see Equation 1.16, where the projected electrostatic potential is given as

$$V_{proj}(\vec{r}) = \int_{z=0}^{z=t} V(x, y, z) \, dz. \quad (3.1)$$



Naturally, a thicker sample will have a stronger projected potential and thus apply a stronger phase shift on the beam than a thinner sample.

Importantly, the aberration surface of the electron beam also contributes to its phase, and it is crucial for COM signal integrity that the optimal defocus condition is found. Furthermore, as sample thickness is increased, the probability increases for electrons scattering off atoms in the sample multiple times, or interacting inelastically with the sample. Typically, the beam electrons have a tendency to travel down the atomic columns, which is a phenomenon known as channelling [143, 151]. Therefore, it is specifically important that the zone-axis of the sample is aligned with the optic axis, as channelling of electrons down the atomic columns contributes to the offset of the COM of the CBED pattern, and can therefore be falsely interpreted as a long-range electric field [89, 91]. As a result, the beam deflection that is the

basis of COM measurements may be lost due to obfuscating signals from multiple scattering and inelastic scattering events in thicker samples.

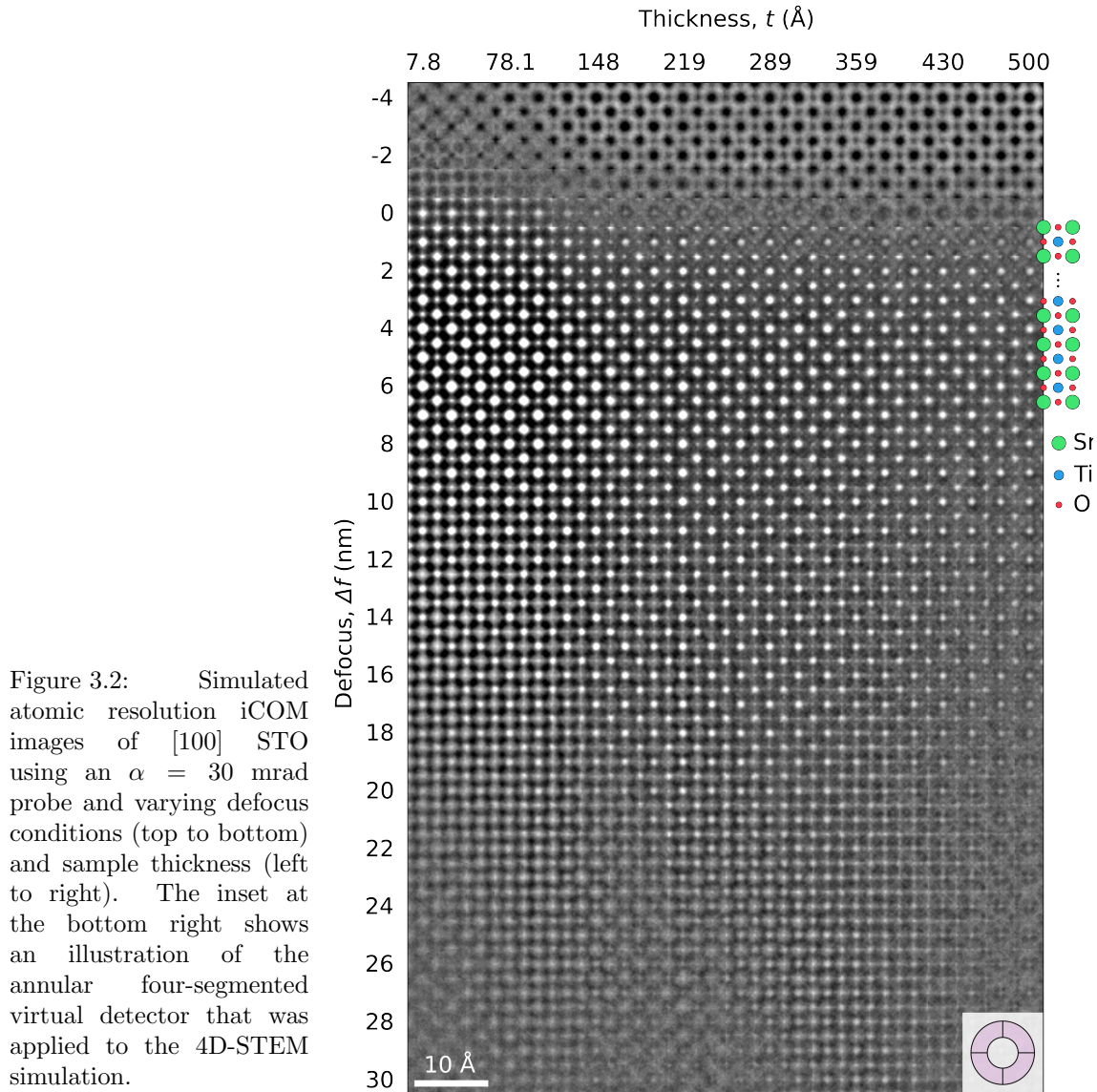
To investigate the optimal defocus and thickness for COM experiments, a series of simulations with varying defocus and sample thickness was done using the abTEM simulation software [16]. A 300 keV beam energy was used, and the semi-convergence angle was $\alpha = 30$ mrad. The collection angle for the annular four-segmented detector was $15 - 30$ mrad. To simulate more realistic conditions, frozen phonons have been included in the simulation¹. Figure 3.1 and Figure 3.2 show the COM images and iCOM reconstructions, respectively, for [100] STO with varying defocus and sample thickness at a finite, but relatively high, dose of $2 \cdot 10^4 \text{ e}^-/\text{\AA}^2$. Here, the sample entrance surface is taken to be the reference point where $\Delta f = 0$ nm, and over-focus ($\Delta f < 0$) denotes the beam cross-over being before the sample, see illustration in Figure 1.4. Importantly, probe defocus is the only probe aberration considered in these simulations.

The simulated results in Figure 3.1 highlight the importance of the sample being thin to get interpretable COM signals. Additionally, both Figure 3.1 and Figure 3.2 show that the cross-over of the beam should be inside the sample, or even slightly after the exit surface of the sample for the thinnest samples. This is in agreement with similar results found by others in the literature, and also in strong contrast to the optimal defocus condition for HAADF, which tends to be close to the entrance surface of the sample [89, 149, 152, 153]. For thicker samples, the optimal defocus range narrows and is found closer to the entrance surface than the middle.

Importantly, for all sample thicknesses, the four-fold symmetry of the detector is visible for most of these simulated COM and iCOM signals. This suggests that the detector geometry plays a key role in the contrast transfer, which will be investigated further later in this chapter. In Figure 3.2 it is also shown how the iCOM image contrast is reversed for negative defocus, when the beam cross-over is before the sample, similar to the contrast reversal that is observed for high-resolution transmission electron microscopy (HRTEM) imaging [149, 154]. While this effect is small for very thin samples, it becomes prominent for practical sample thicknesses, > 10 nm for STO.

In conclusion, the sample should be thin enough for the WPOA to be a good approximation. In practice, that means thin enough to avoid significant presence of dynamical interactions, in other words, thinner than the inelastic mean free path [1]. Additionally, a slight underfocus should be applied such that the beam cross-over is around the middle of the sample for thin samples and just beyond the entrance surface for thicker samples.

¹However, progressively fewer frozen phonon configurations were used as the sample thickness was increased since each new layer of the STO sample adds to the effective number of configurations.



3.2 Centring the Beam on a Segmented Detector

When acquiring data for iCOM phase reconstruction, the beam must be centred on the segmented detector for the COM signal to be accurate [147]. A miscentring of the beam can give features in the images that may lead to misinterpretation of sample properties, like obscuring sample tilt [89]. Furthermore, if final alignments of the probe, like correction of astigmatism, are done based on the live COM image at the microscope, these alignments may be flawed if performed on an unreliable COM image. Ideally, the segmented detector is aligned with the optic axis of the TEM, such that proper alignment of the microscope itself should ensure proper centring of the electron beam. However, this is not necessarily the case, and even if it is, the centring of the beam on the segmented detector should always be double-checked before an experiment.

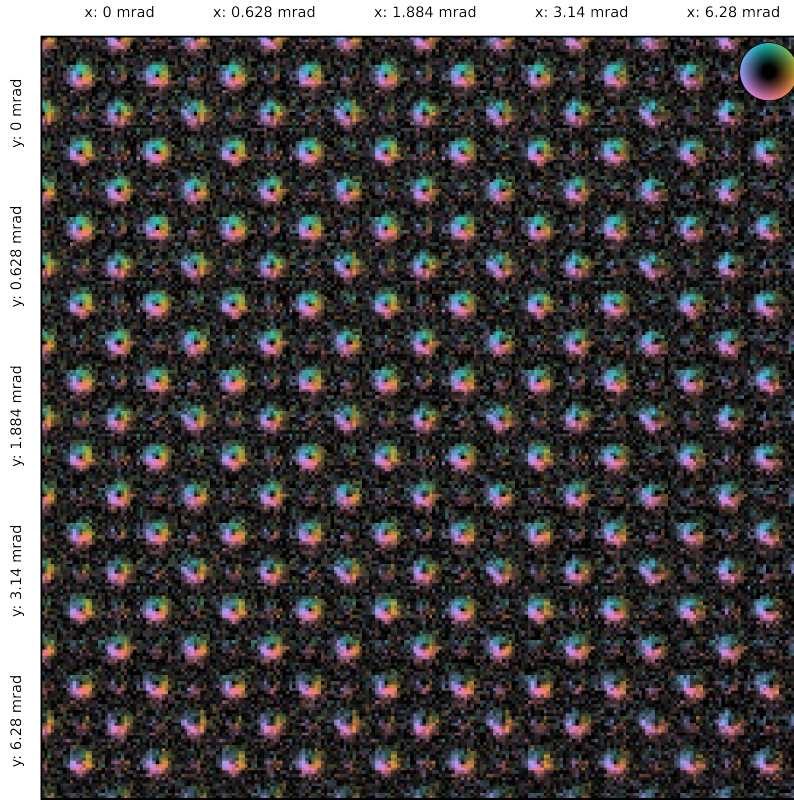


Figure 3.3: Simulation of COM images of two unit-cells of [100] STO, using an $\alpha = 30$ mrad beam, for various detector miscentring in both x - and y -direction. The collection angles of the detector were $\frac{\alpha}{2} - \alpha$, and the finite dose of $2000 \text{ e}^-/\text{\AA}^2$ was applied.

The effects of detector miscentring were investigated in simulation by applying a miscentered virtual detector to a 4D-STEM simulation of ten unit-cell thick STO, simulated with 50 frozen phonon configurations. A semi-convergence angle of $\alpha = 30$ mrad and a beam energy of 300 keV were used for the simulation, and an eight-by-eight unit-cell area was simulated to achieve the necessary resolution in reciprocal space. The resulting COM images for a two-by-two unit-cell area for each detector miscentring configuration is shown in Figure 3.3, where a finite dose of $2000 \text{ e}^-/\text{\AA}^2$ has been applied. This figure shows how a miscentring of the beam up to $\sim 10\%$ has minor visible effects on the finite dose COM images. When approaching $\sim 20\%$ miscentering, the effect becomes more apparent in the COM images, but is still hard to see in the iCOM reconstruction, see Figure 3.4. In conclusion, the effects of beam centring on the segmented detector are much smaller than other parameters like zone axis condition for atomic resolution imaging [89, 147].

There are multiple ways of centring the beam on a segmented detector. Importantly, the detector first needs to be normalised, such that all channels in each ring respond equally when uniformly illuminated. Then the beam may be centered by using the projector lenses to deflect the beam post-specimen. Using the desired semi-convergence angle with the optimal camera length, as will be defined in the following section, a vacuum beam may be parked in the centre pixel and shifted until all detector segments output the same signal level. It is important that the beam does not traverse a sample while the beam is being centered in this way, as that will

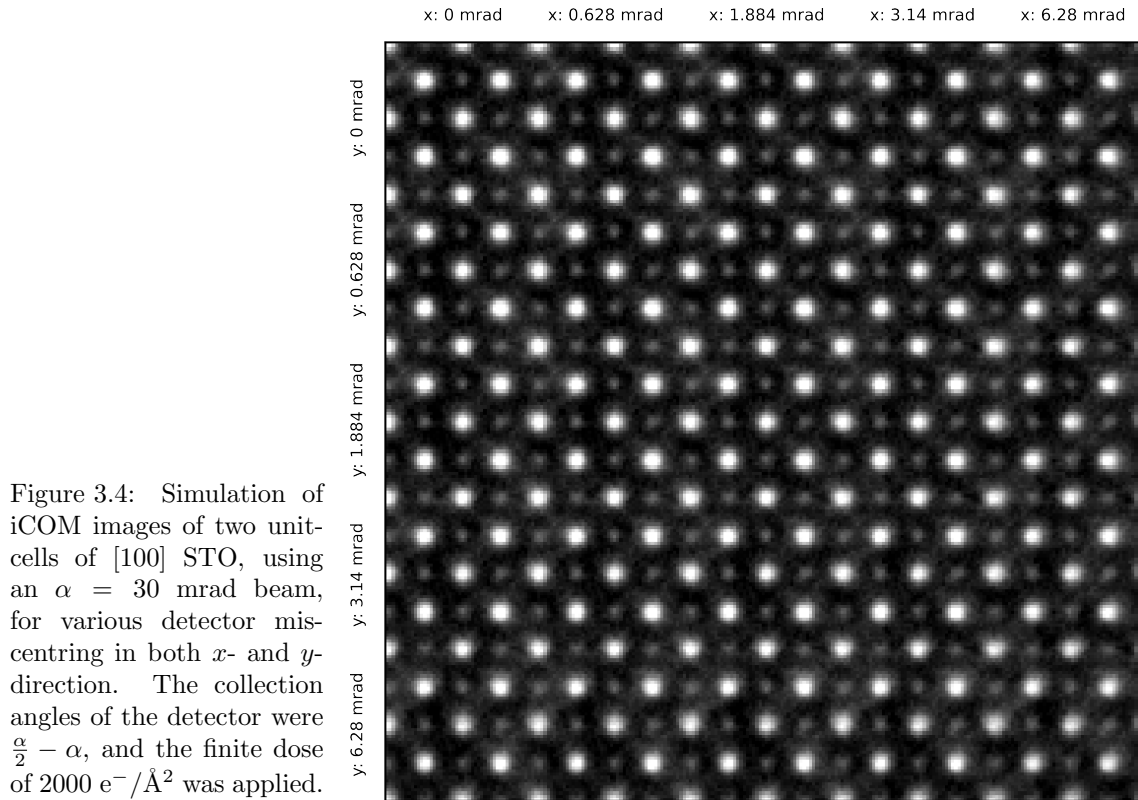


Figure 3.4: Simulation of iCOM images of two unit-cells of [100] STO, using an $\alpha = 30$ mrad beam, for various detector miscentring in both x - and y -direction. The collection angles of the detector were $\frac{\alpha}{2} - \alpha$, and the finite dose of $2000 \text{ e}^-/\text{\AA}^2$ was applied.

give a non-uniform beam. Similarly, the beam may be scanning through vacuum or a part of the sample, if there is no long-range field, in which case the mean signal level in each segment may be equalised. Equivalently, the mean COM deflection over the field of view (FOV) should be as close to zero as possible and the beam may be centered with this goal in mind. These approaches all rely on the operator of the microscope, which in and of itself is not a problem². Yet, for mundane tasks like these, where the goal for the optimisation is clearly defined, it is fair to assume that a computer would likely do a better job than any human. The goal may be defined as “make all these four segment signals have the same mean intensity” or “minimise mean COM deflection”, and the means of getting there requires adjusting two independent parameters: projector lens deflection in x and y .

Since the goal of centring an electron beam is so well-defined, a retrofittable hardware and control code to allow the computer to do the centring was developed and tested. A pair of stepper motors, connected to the projector lens deflectors by rubber bands around the multi-function knobs at a TFS Titan 80-300kV TEM, were controlled using an Arduino, see Figure 3.5. Acquiring a detector map allows for the determination of the centre of the detector via the COM of intensity in the detector map, which can be used as an initial guess for the centre position. A more accurate centre position of the detector can be found by repeating the procedure. Naturally, this procedure assumes precise control of the beam position using the

²It does require a lot of skill to drive a TEM after all.

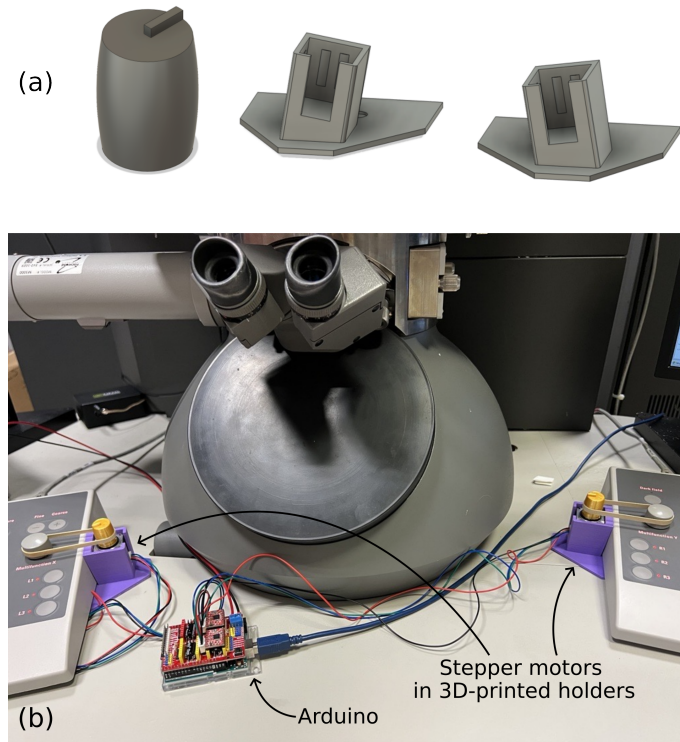


Figure 3.5: (a) Models for the 3D-printed cap and holders for the stepper motors. (b) Image of the prototype Arduino-based auto-centring device operating the multi-function knobs of the TFS Titan 80-300.

multi-function knobs. Though multiple attempts were made to achieve a better result using the retrofitted hardware than a human could with direct feedback of the intensity difference between opposing segments, satisfactory results could not be achieved. When considering that most microscopes may be controlled by code, either through a manufacturer-provided application programming interface (API) or by otherwise accessing the microscope-controlling code on older TEMs, it is unlikely that such retrofittable hardware will prove very useful.

3.3 Optimal Camera Length

It is well-known that the intensity that may be detected by an annular detector in STEM decreases exponentially with increasing inner collection angle, α_{det}^i , of the detector [59, 94]. This is the main reason why phase retrieval techniques have become a popular alternative to the more traditional HAADF imaging for beam-sensitive materials characterisation in STEM. A higher collection efficiency means that a higher proportion of the beam electrons are detected, such that the electron dose applied to the sample may be reduced, which results in less sample damage. Using pixellated detectors to capture the entire CBED pattern at every scan position provides the operator with a massive amount of data, but comes with the downside of limiting the practicable scan speed, as seen summarised in Figure 2.4. Annular segmented detectors offer more flexibility with regard to scan speed, but require a choice of the optimal outer collection angle to achieve the highest possible COM

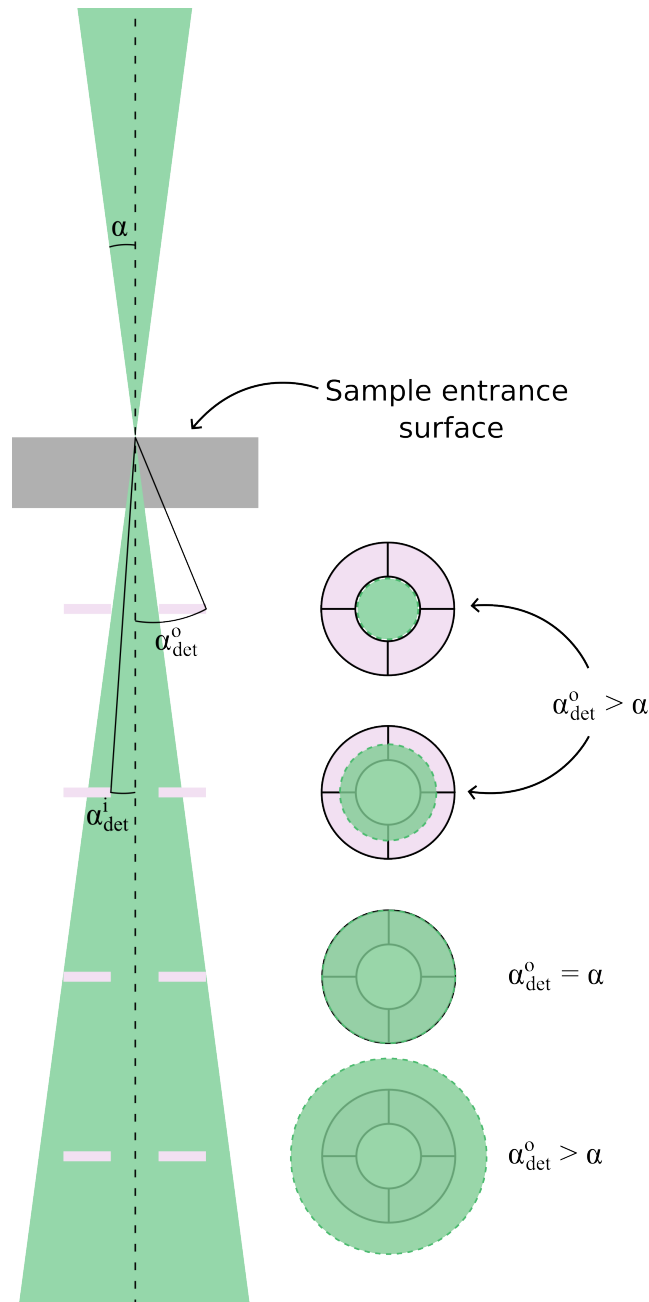


Figure 3.6: A simplified illustration of the effect of how placing the detector at different camera lengths adjusts the collection angles of the detector. This is true for any detector, whether segmented, monolithic, or pixellated.

signal integrity. Note that while it is, in principle, possible to optimise the whole collection range — both inner and outer collection angle — this is typically already limited by the physical geometry of the detector. In this section, the annular segmented detectors all have inner collection angle equal to half the outer collection angle; modelled after the SAAF detector [110].

Adjusting the outer collection angle of an annular segmented detector may be done by adjusting the camera length of the microscope, which essentially magnifies or demagnifies the CBED pattern on the detector by changing the effective distance between the sample and the detector, see illustration in Figure 3.6. The collection angles, for a fixed camera length, may also be altered by the semi-convergence angle (α) of the STEM probe, but α is typically determined by other experimental

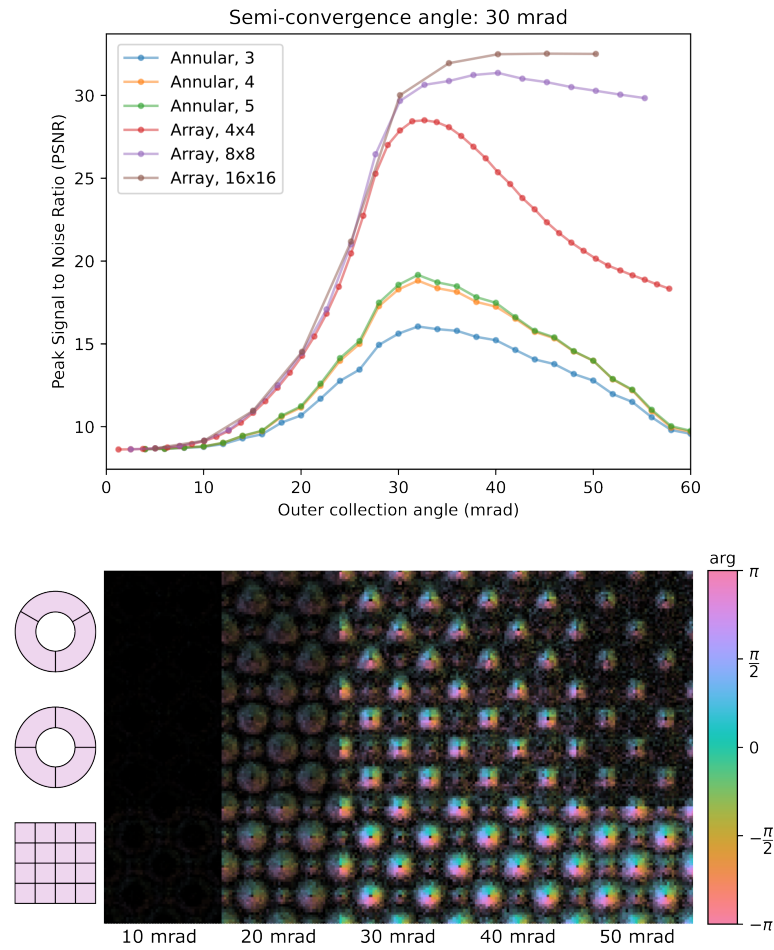


Figure 3.7: PSNR of COM images at finite dose 10 C/cm^2 ($6241 \text{ e}^-/\text{\AA}^2$) for various different detector geometries as a function of outer collection angle of the detector. The reference image used is the finite dose COM as calculated from the full-field 4D-STEM data with maximum collection angle 2α . Example images are the resulting images from the detector geometries indicated by the illustrations on the left-hand side, at the given outer collection angles.

constraints, such as the aberrations of the microscope or the sensitivity needed for the specific sample. As such, the collection angles of the detector are most easily changed by altering the effective camera length. Experimentally, this is done by changing the strength of the projector lenses post-sample, meaning that the dose delivered to the sample is not altered by changing the camera length. In simulation, changing the camera length may be done by varying the size of the virtual detectors applied on a 4D-STEM simulation.

Figure 3.7 shows the PSNR of COM images acquired with various detector geometries as a function of outer collection angles. From this, it is evident that the optimal outer collection angle for annular segmented detectors, both with three, four, and five annular segments, is slightly above the semi-convergence angle. The sample used for these simulations was a 30 unit-cells thick [100] STO, imaged using a 300 keV beam with $\alpha = 30 \text{ mrad}$, where the maximum collection angle of the

4D-STEM simulation was 2α . The COM image from the full 4D-STEM simulation was used as the reference image for the PSNR calculation. An outer collection angle of $\sim 1.1 \cdot \alpha$ ensures that the optimal amount of signal from the CBED pattern and the intensity variation within it are detected by the annular detector. Interestingly, the PSNR of COM images from a 4×4 array detector also peaks around 1.1α , alluding to the importance of discerning the details within the CBED pattern for COM image fidelity.

For collection angles larger than $\beta_{det} = 2\alpha$, there is no phase signal to be detected [56]. This is evident from how the PSNR of the annular segmented detectors with a central hole drops as the outer collection angle approaches 60 mrad, or 2α , since the inner collection angle then approaches $\alpha = 30$ mrad. The lower panel of Figure 3.7 also shows how the intensity of the finite dose COM images from the detector geometries with a hole drops for both very low outer collection angle, where the detector is simply too small to detect much signal, and high outer collection angle where there is little signal left to detect. Another reason why the PSNR drops for the higher outer collection angles is that the COM signal tends to be overestimated, since the COM of the segments lie outside where most of the intensity is detected. The following section will discuss this more rigorously.

In conclusion, the overall optimal camera length is taken from this investigation to be one where the detector outer collection angle is 1–3 mrad higher than the semi-convergence angle. Using this optimal condition, the error in the calculated COM_x and COM_y signals as compared to the fully pixellated COM may be estimated to give an insight into the effect of the detector geometry on the CTF of COM imaging. This will be further investigated in the following.

3.4 Contrast Transfer Function of Phase Retrieval Techniques

The fact that the detector geometry impacts the phase reconstruction achievable with iCOM has been extensively reported [56, 57, 155]. In section 2.2, the effects of detector geometry on the PSNR of iCOM images of an STO sample were shown. Similarly, the error in the COM_x and COM_y signals when using a segmented detector versus a pixellated detector may be calculated to visualise how segmented detectors introduce errors with the same foldedness as their geometry. That means that a four-segmented detector introduces a four-fold symmetry in the COM image.

Interestingly, this remains true for any detector with a four-fold symmetry, even when the detector has 16 segments spread over four rings with the same orientation. The COM_x and COM_y errors for five different detector geometries may be seen in Figure 3.8. Here, it is worth noting how the error decreases and the roundness of the

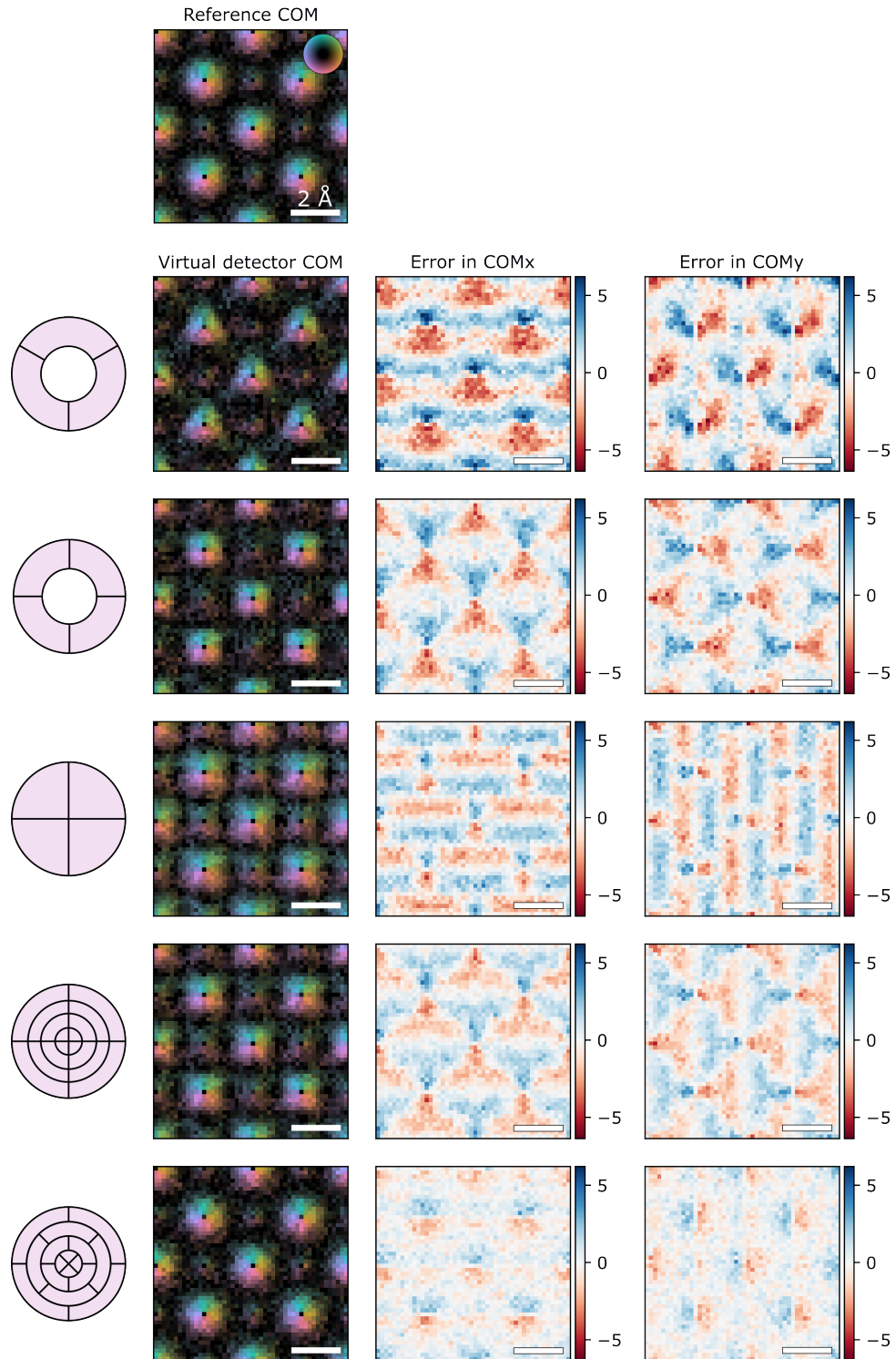


Figure 3.8: Virtual detectors and their resulting COM images, as well as the errors in COMx and COMy. All COM images are limited to the radial resolution of the detector. Note that the 16-segmented annular detector does not produce significantly better COM images than the four-segment ones, unless half of the segments are rotated by 45° .

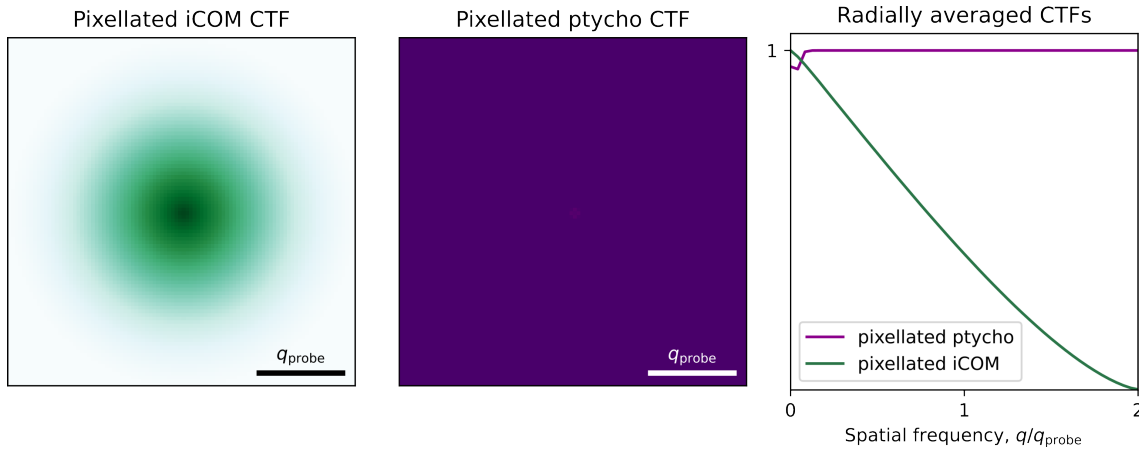


Figure 3.9: Upper limits of CTF for iCOM and iterative Ptychography showing that iCOM is limited by the aperture auto-correlation and iterative Ptychography can reach unity CTF across all spatial frequencies.

atoms appears a lot better for the 16-segment detector where half of the detector rings are rotated 45° . This is attributed to its enhanced ability to resolve information azimuthally in addition to radially, like the standard 16-segment detector directly above it in Figure 3.8. A similar effect for a 4×4 pixel array detector may be seen in the images in the lower row of Figure 3.7.

These figures are anecdotally investigating the impact of the detector geometry on the resulting image. In order to investigate the effects of using a segmented detector for phase retrieval, and compare with other phase retrieval methods in a fair way, the sample needs to be taken out of the equation. Then, when only evaluating the two-dimensional CTF, the effects of the detector geometry on the method may be fully and fairly investigated.

To be able to evaluate the impact of the detector geometry on the CTF, it is useful to first become familiar with the upper limits. An ideal phase retrieval method would be able to retrieve information at all spatial frequencies equally — that means that the two-dimensional CTF would be unity everywhere. For STEM measurements, the CTF is given mathematically as [156]

$$\text{CTF}_j(\vec{q}) = \frac{i}{2} \int A(\vec{k}) D_j(\vec{k}) \left\{ A(\vec{q} - \vec{k}) e^{-i[\chi(\vec{q} - \vec{k}) - \chi(\vec{k})]} - A(\vec{q} + \vec{k}) e^{i[\chi(\vec{q} + \vec{k}) - \chi(\vec{k})]} \right\} d\vec{k}, \quad (3.2)$$

where $A(\vec{k})$ is the probe forming aperture, as seen in Equation 1.8, and $\chi(\vec{k})$ is the aberration function given in Equation 1.10. In the ideal case, where the probe is aberration-free ($\chi(\vec{k}) = 0$) and a circular probe-forming aperture is used, the CTF in Equation 3.2 reduces to the aperture auto-correlation function,

$$\text{CTF}^{\text{ideal}} = [A \star A](\vec{k}) = \text{Re} \left\{ \mathcal{F}_{\vec{r} \rightarrow \vec{k}}^{-1} \left[|\mathcal{F}_{\vec{k} \rightarrow \vec{r}}[A(\vec{k})]|^2 \right] \right\}, \quad (3.3)$$

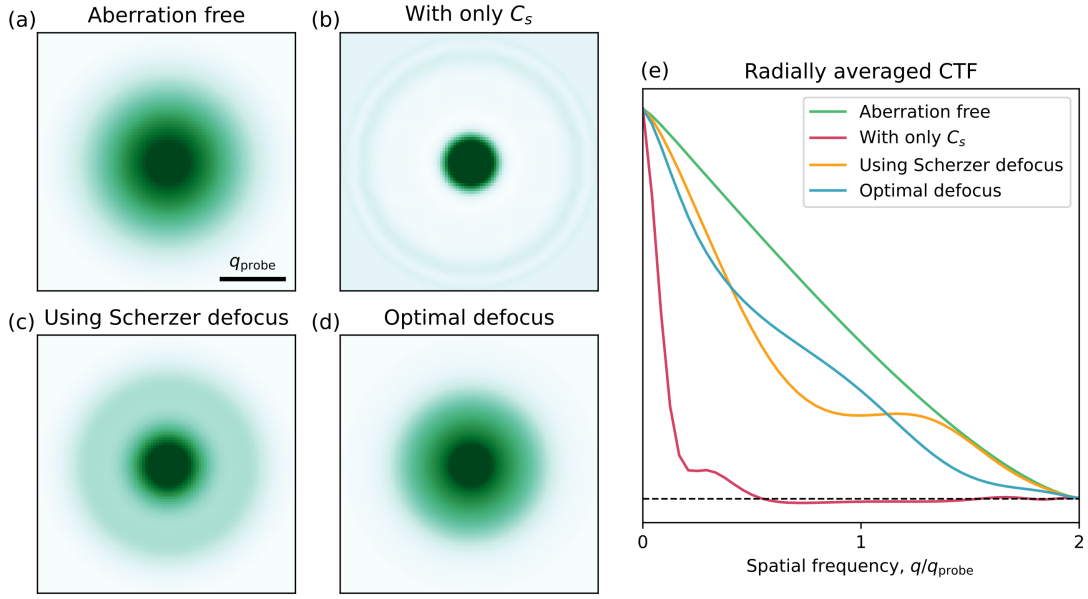


Figure 3.10: CTF for iCOM with an (a) aberration-free probe, and (b) probe with spherical aberration, $C_s = 50 \mu\text{m}$. When Scherzer defocus, here $\Delta f = 121.5 \text{ \AA}$, is applied to the aberrated probe, this gives the CTF in (c) which is slightly sub-optimal. (d) The optimal defocus for iCOM with a spherically aberrated probe is slightly less underfocused than Scherzer defocus, here $\Delta f = 71.5 \text{ \AA}$. (e) The radially averaged CTFs, showing how the optimal defocus condition for the aberrated probe yields the most linear CTF which also most closely matches the aberration-free one.

which may be interpreted as the overlapping region between the aperture and a shifted aperture centered at $\vec{k}$. iCOM is limited by this aperture auto-correlation, see the ideal case iCOM CTF in Figure 3.9, since all the directly obtainable phase information is contained in the double overlap regions — meaning where the direct beam overlaps with a diffracted beam. In other words, there is no phase information to be collected beyond 2α using direct phase retrieval methods like iCOM [157]. Contrastingly, iterative ptychography may reach the ideal limit of unity phase retrieval for all spatial frequencies, given practically infinite dose, a pixellated detector, and enough iterations, see Figure 3.9 [158].

Allowing the aberration function to be non-zero, the aperture auto-correlation function is replaced by the complex probe auto-correlation, which is

$$\text{CTF} = [\psi \star \psi](\vec{k}), \quad (3.4)$$

where $\psi(\vec{k}) = A(\vec{k})e^{-i\chi(\vec{k})}$ is the complex-valued electron wavefunction at the detector. Using this analytical probe auto-correlation formalism, the optimal defocus condition for an aberrated probe may be investigated. For HRTEM the optimal defocus condition to counteract the effect of spherical aberration is Scherzer defocus [17], as defined in Equation 1.12. However, when this defocus is applied for iCOM phase retrieval, the resulting CTF is sub-optimal, delivering non-linear con-

trast transfer across spatial frequencies, see Figure 3.10. A slightly less defocused probe delivers a more linear contrast transfer, and this is also the CTF that most closely matches the aberration-free probe CTF.

Finally, the CTF for segmented detector iCOM may also be found analytically, using the complex-probe auto-correlation with each of the segments [158]. A detector segment may be given mathematically as

$$D_j(\vec{k}) = \begin{cases} 1, & \text{for } \vec{k} \text{ inside detector segment } j \\ 0, & \text{elsewhere.} \end{cases} \quad (3.5)$$

For a segmented detector with detector segments estimating the COM shift in two orthogonal directions, the vectorial detector function may be expressed as

$$\vec{D}(\vec{k}) = \sum_j k_x(\vec{k}) D_j(\vec{k}) \hat{k}_x + \sum_j k_y(\vec{k}) D_j(\vec{k}) \hat{k}_y. \quad (3.6)$$

The analytical COM CTFs in x - and y -directions may then be obtained by

$$\begin{aligned} \text{CTF}_x^{\text{COM}}(\mathbf{q}) &= [\psi \star \psi D_x](\mathbf{q}) - [\psi D_x \star \psi](\mathbf{q}) \\ \text{CTF}_y^{\text{COM}}(\mathbf{q}) &= [\psi \star \psi D_y](\mathbf{q}) - [\psi D_y \star \psi](\mathbf{q}), \end{aligned} \quad (3.7)$$

which means the CTF is now a vector since it consists of a set of two CTFs for two orthogonal directions. The iCOM CTF may be found by the same Fourier integration as presented in Equation 1.24 [159].

Unfortunately, this analytical framework does not lend itself well to the iterative nature of iterative ptychography, and thus it is more productive to proceed numerically. In the following, a numerical framework for finding the CTF for both iCOM and iterative ptychography, using any detector geometry, is introduced.

3.4.1 White Noise Object

So far, the fidelity of the COM images and the iCOM reconstructions with segmented detectors has mostly been investigated anecdotally in simulated atomic resolution images. While this gives valuable insight into the quality of phase retrieval achievable with iCOM, it has one important flaw namely the symmetry of the sample. To investigate the effects of only the detector geometry on the achievable phase retrieval, and to be able to compare different phase retrieval methods directly in a fair way, the sample needs to be isotropic. Ideally, this isotropic sample should be a weak phase object, as defined in Equation 1.16, with a potential $V(\vec{r})$ that is real-valued in real space and has a constant amplitude and random phase in Fourier space. The

Fourier space potential may be given as

$$\tilde{V}(\vec{k}) = Ae^{2\pi i\varphi(\vec{k})} \quad (3.8)$$

where A is a constant and $\varphi(\vec{k})$ is the phase. In the implementation of the isotropic weak-phase object to use for the CTF investigations, the amplitude A is set to unity and a random phase $\varphi(\vec{k})$ is drawn from a standard normal distribution. This yields an isotropic “white noise” object with a random distribution of spatial frequencies to resolve in all directions.

Since the white noise object is a weak phase object by definition, it is sufficient to perform a single-slice 4D-STEM simulation. This is most easily done by multiplying the object with a shifted probe for every scan position, and taking the absolute square of the Fourier transform of the result to get the detectable 4D-STEM dataset. Then, once the 4D-STEM dataset is simulated, the virtual detectors may be applied and the phase reconstructed using iCOM or a ptychographic phase reconstruction method. Finally, the Fourier transform of the reconstructed image is the CTF of the phase retrieval method used with the applied detector geometry. This numerical method of obtaining the CTF is useful because of its universal applicability to any phase retrieval method, with any detector geometry, without needing to work out analytic expressions. Since the random distribution of phase factors yields a random distribution of spatial frequencies to resolve in all directions, the final Fourier transform gives an easily interpretable two-dimensional CTF which clearly indicates the directions in which a detector geometry performs better or worse, as can be seen in Figure 3.11.

3.4.2 iCOM Contrast Transfer

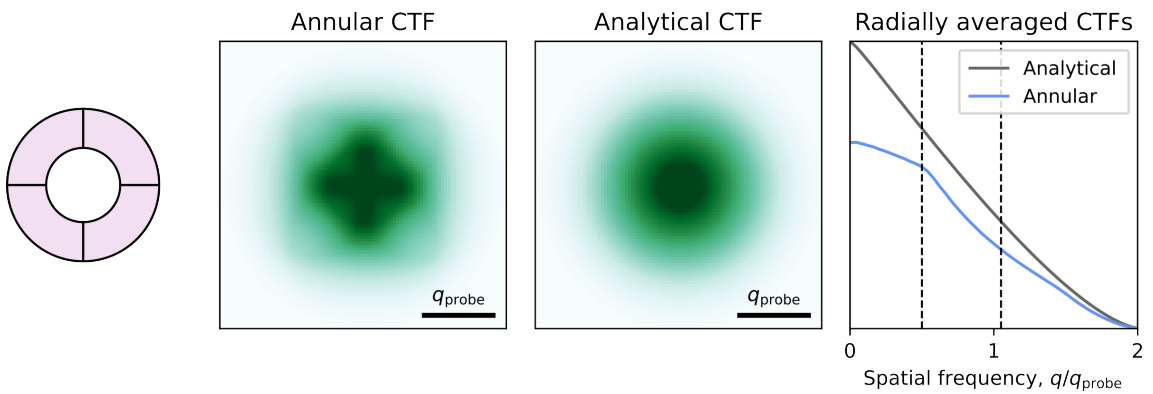


Figure 3.11: Comparison of the CTF of iCOM with the segmented detector geometry shown and the analytical iCOM CTF for an in-focus probe. The radially averaged CTFs clearly show how segmented iCOM falls short of the analytical limit. The horizontal lines mark the collection angles of the detector.

For a pixellated detector and a perfect probe, the iCOM CTF decreases roughly linearly with increasing spatial frequency, following the probe auto-correlation seen previously. In Figure 3.10 it was also shown how the presence of spherical aberration degrades the contrast transfer for iCOM, even when counteracting the C_s with defocus. Since spherical aberration is an isotropic aberration, the degradation of the contrast transfer is also isotropic, and a radial average adequately represents the CTF without information loss. Using segmented detectors also limits the information retrievable from the dataset, as can be seen in Figure 3.11, however, now the effect is anisotropic. The detector geometry is reflected in the CTF, resulting in information along certain directions being better transferred than others. Note that while taking the radial average over the two-dimensional CTF shows how the segmented detector CTF falls short of the ideal probe auto-correlation limit, the anisotropic nature of the 2D CTF gets hidden by the radial average.

The reflection of the detector geometry in the CTFs for all the detector geometries shown in Figure 3.8 can be seen in Figure 3.12. Here, it is worth noting that the three-segmented annular detector CTF has a six-fold symmetry due to the Friedel symmetry, namely that the square amplitude of a Fourier transform is centrosymmetric; $|\mathcal{F}(\vec{k})|^2 = |\mathcal{F}(-\vec{k})|^2$. For the three middle detectors, which all have a four-fold symmetry, the CTF also exhibits a four-fold symmetry. In fact, the rotation of the detectors is directly reflected in the corresponding CTF, demonstrated in Figure 3.12 for rotations 20° and 45° . This helps elucidate why certain detector rotations yield better iCOM reconstructions for specific samples because it directly shows that the contrast transfer is better along the directions through the middle of the segments [160]. For samples with a four-fold symmetry, like STO, where the $\langle 100 \rangle$ -directions exhibit atomic columns at a higher spatial frequency than the $\langle 110 \rangle$ -directions, a four-fold detector may be rotated such that the $\langle 100 \rangle$ -directions align with the segments to exploit the enhanced contrast transfer. This illustrates the power and usefulness of an isotropic “white noise” object to directly inform us of the impact that the detector geometry has on the CTF.

Optimisation problems discussed previously in this chapter may also be investigated with this CTF formalism. For example, that iCOM data should always be taken with in-focus conditions is easily observed by the degradation of the CTF with increasing defocus in Figure 3.13. These results are in agreement with other findings in the literature [91, 149, 152, 153]. A diverging colour-map was used for the 2D CTFs in Figure 3.13 to show the contrast reversal that occurs when the defocus is too high. Additionally, the radial averages of the CTFs highlight how the contrast transfer is negative for certain spatial frequencies at $\Delta f \geq 80 \text{ \AA}$, which explains the contrast reversal seen in COM images at high defocus.

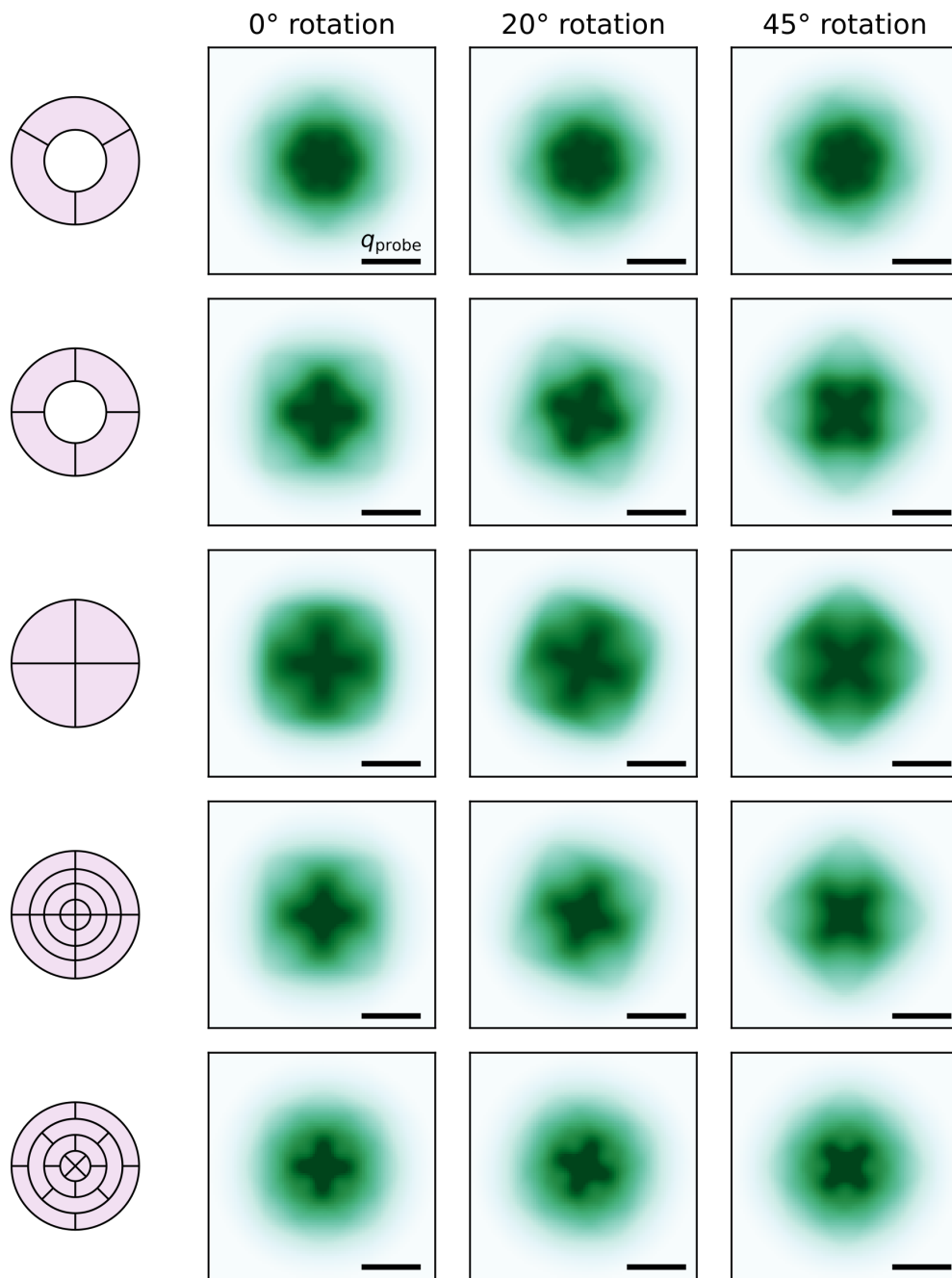


Figure 3.12: Segmented detector two-dimensional CTF for iCOM phase retrieval for all the detector geometries for which the COM images of STO can be seen in Figure 3.8.

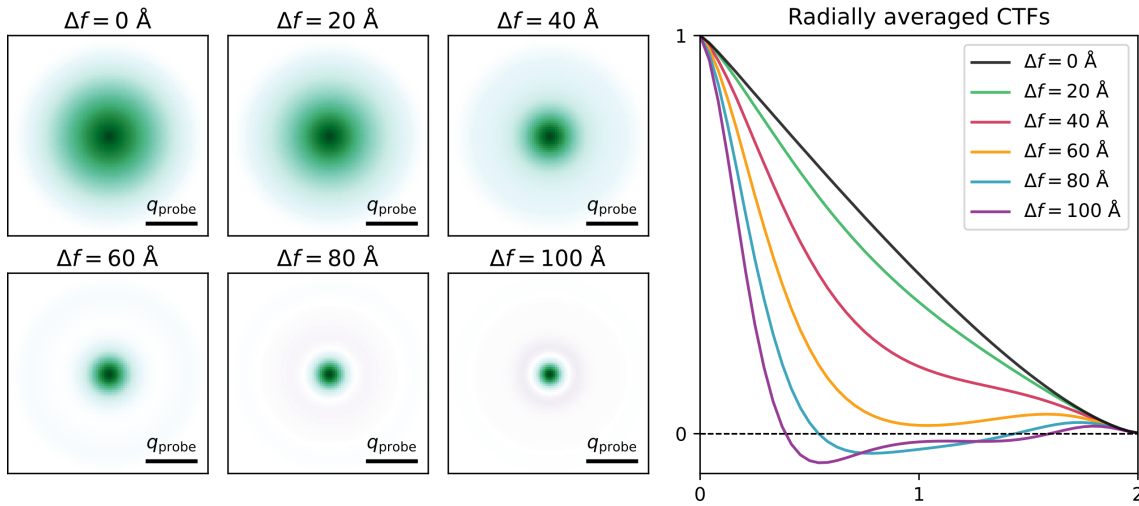


Figure 3.13: Analytical iCOM CTF for increasing defocus values in an $\alpha = 20$ mrad beam. Both the 2D CTFs and the radial averages show that iCOM requires in-focus data for optimal contrast transfer. Note that a diverging colour-map has been used to highlight that the CTF becomes negative for high defocus values.

Naturally, achieving in-focus conditions is much easier with a single-slice object than with a real sample of finite thickness. Additionally, the effect of positive and negative probe defocus is symmetric for the white noise object since it has effectively 0 thickness. The situation is much more complicated for a real sample with non-negligible thickness — that is, samples for which a single-slice simulation is insufficient. When the sample has a finite thickness, the beam effectively “sees” different parts of the sample at different defoci, necessitating investigations like the defocus-thickness simulations in section 3.1. Still, an important takeaway is that iCOM does not gain anything from being performed on a dataset with deliberate defocus, unlike iterative ptychography³ — which will be elaborated on in section 5.4.

Other optimisation problems discussed in this chapter lend themselves better to being investigated through the CTF framework. Firstly, the optimal camera length — or more generally detector collection angles — that optimise the contrast transfer may be found. An example for a detector where the inner collection angle is always half of the outer collection angle can be seen in Figure 3.14. This figure also shows how a four-segment detector with outer collection angle larger than α overestimates the COM. The reason for this is that the COM of the segments badly represents the COM of the signal detected in them and, as a result, the COM signal is overestimated and the CTF exceeds unity. From this, the conclusion is that an outer collection angle closely matching, or slightly larger than, the semi-convergence angle is optimal for annular detectors [149, 158]. This agrees with the findings from previously using atomic resolution STO simulations, and findings in the literature [148, 149]. Adding more radial rings to the detector can help reduce the effects of overestimation caused

³And some other ptychographic methods not covered in this thesis.

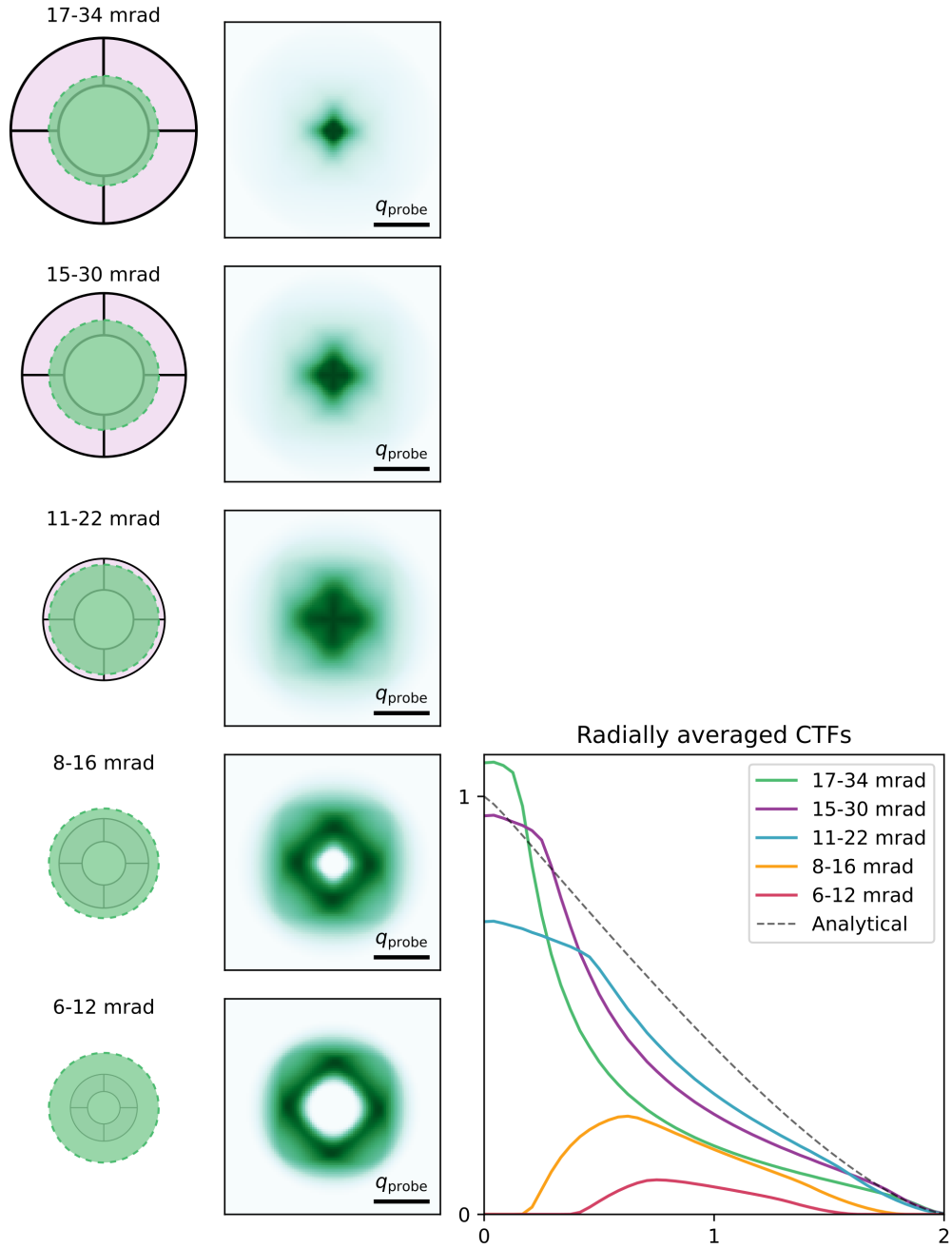


Figure 3.14: White noise object numerical estimation of CTF for iCOM with an $\alpha = 20$ mrad beam, using various effective camera lengths which alter the collection angles of the annular segmented detector. Both the 2D CTFs and the radial averages show that an outer collection angle close to α is optimal.

(0.0, 0.0) mrad (2.5, 2.5) mrad (4.9, 4.9) mrad (7.4, 7.4) mrad

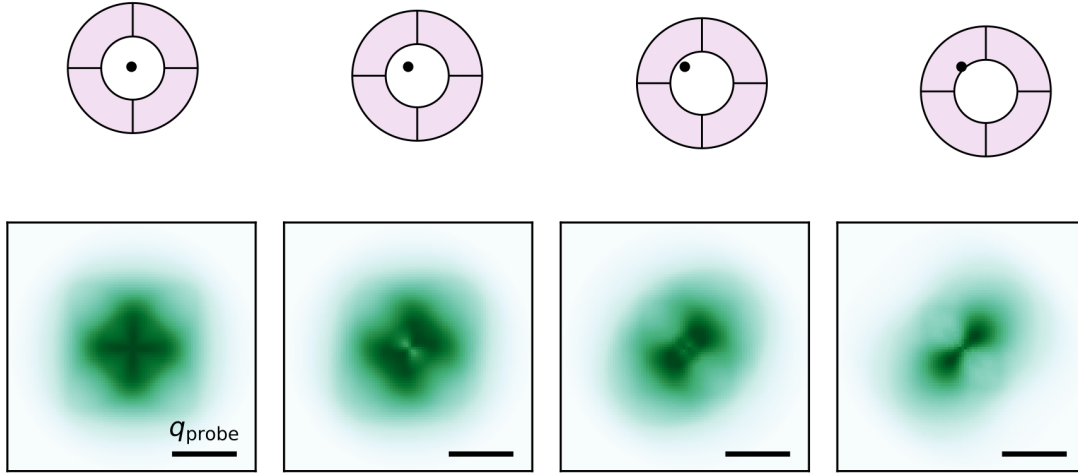


Figure 3.15: Effect of diagonal detector miscentering on the CTF of iCOM. The miscentering in x - and y -direction is given in the tuple above each panel and illustrated by the detector sketch where the black dot signifies the optic axis. The semi-convergence angle is $\alpha = 20$ mrad.

by using sub-optimal detector collection angles because the COM of each segment is more accurately determined for a smaller segment. This is seen in Figure 3.12 when comparing the four-segmented detector without a central hole and the 16-segmented detector to the analytical CTF in Figure 3.11. Adding a 45° rotation to half of the rings in the 16-segmented detector yields a somewhat rounder, and more isotropic contrast transfer.

Furthermore, the effects of detector miscentering, which were discussed in section 3.2, may be understood more directly through assessing the CTFs. The effect on the iCOM CTF from a diagonal shift of the segmented detector centre for a 20 mrad beam may be seen in Figure 3.15. Note how the CTF becomes increasingly anisotropic with increasing detector miscentering. A full tableau of miscentering in x - and y -direction can be seen in Figure 3.16. Here, the radially averaged CTFs in the bottom row show how the magnitude of the contrast transfer suffers with increasing detector miscentering. Note that, again, the radial average of the CTFs obscure the anisotropy in the 2D CTF, hindering the full interpretation of how detector miscentering impacts the iCOM images. Figure 3.15 shows that miscentering up to $\sim 10\%$ has minimal effect on the iCOM contrast transfer, similar to the result from section 3.2. Finally, miscentering exceeding 20% degrades the CTF significantly, further confirming the conclusions from the previous results.

Similar to how the 16-segment detector with half the rings rotated 45° produces a rounder CTF, because it is capable of resolving information better azimuthally, a

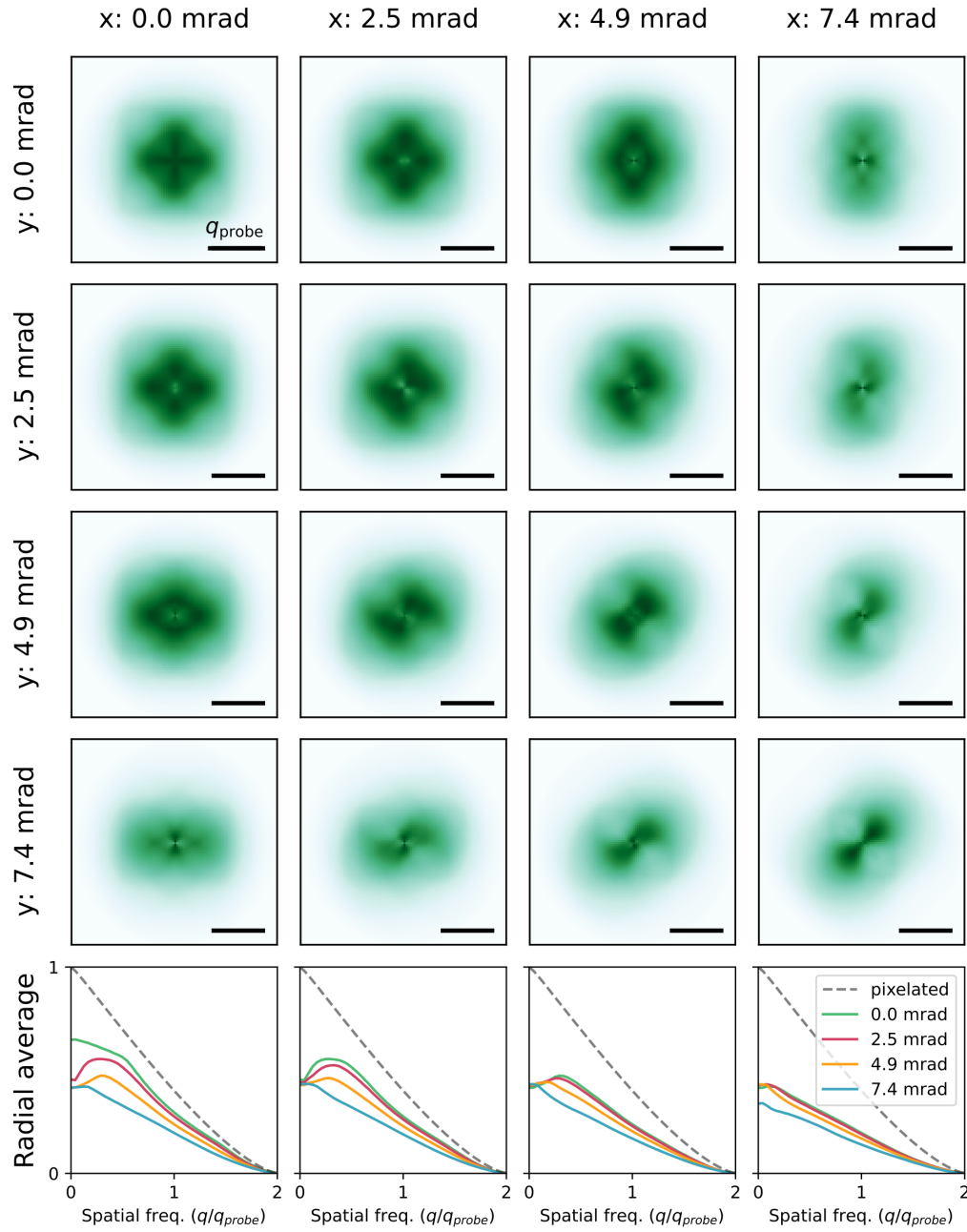


Figure 3.16: Miscentering of the detector causes the CTF to become anisotropic, and the magnitude of the information transfer is hampered — as shown by the lighter 2D CTFs and the lower valued radially averaged CTFs. The semi-convergence angle is $\alpha = 20$ mrad.

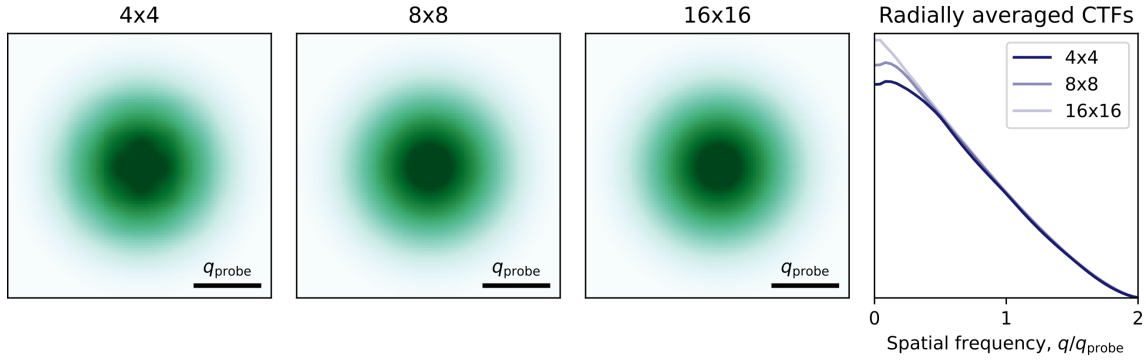


Figure 3.17: iCOM CTF when using $n \times n$ pixel array detectors for three different values of n . Note that already at 16×16 detector elements the CTF closely resembles the analytical CTF for iCOM.

pixel array detector also produces rounder CTFs than traditional annular 4-segment detectors. The iCOM CTF when using 4×4 , 8×8 , and 16×16 pixel array detectors may be seen in Figure 3.17. These show that array detectors perform better than annular detectors for iCOM phase retrieval, and the 16×16 array detector CTF lies very close to the analytic iCOM CTF, which reinforces the findings from Figure 2.8. This is attributed to the detector array elements having a better spread of the COM of each detector element compared with a simple annular segmented detector, thus being capable of resolving the CBED more isotropically, and as a result, the CTF is better. To see this more visually, revisit Figure 2.5 which shows CBED patterns binned using various detector geometries. As a rule of thumb, more detector elements will give a more precise estimate of the COM, however, Figure 3.17 illustrates how higher detector pixellation beyond 16×16 yields diminishing returns for iCOM phase retrieval since there is minimal information left to be gained before reaching the upper limit given by the probe auto-correlation.

Before proceeding, it is worth noting that the CTF does not tell the full story, which is apparent from that it tends to claim that iCOM recovers the mean phase exactly, shown by the CTF approaching unity as spatial frequency approaches 0. This is a shortcoming of CTF, since it does not take into account the error in the measurement at the various spatial frequencies. For iCOM, the noise scales linearly with inverse spatial frequency, meaning that lower spatial frequencies are noisier and harder to resolve. The CTF, being the maximum possible signal, divided by the noise, gives the signal-to-noise ratio (SNR). Figure 3.18 shows the SNR for the same detector collection angles as shown in Figure 3.14. When taking the noise into account, it becomes more clearly evident that the optimal outer collection angle is one that closely matches α . Note that an analytical expression for the noise of iterative ptychography does not exist, and therefore the CTFs of iCOM will be used in the following when comparing the two methods since this provides a more fair comparison of upper limits.

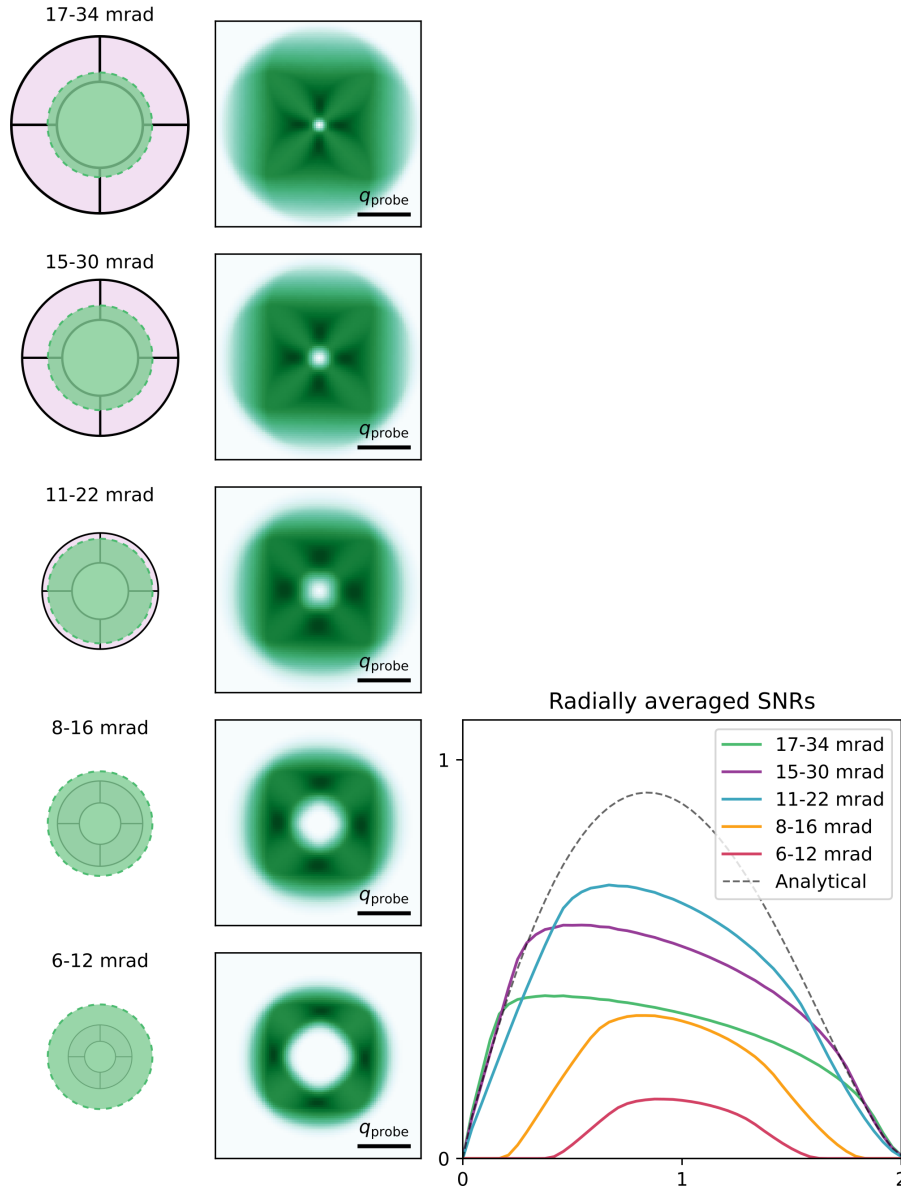


Figure 3.18: Camera length impact on the signal-to-noise ratio (SNR) of iCOM reconstructions for an $\alpha = 20$ mrad beam using a four-segmented detector. This figure shows the same collection angles as Figure 3.14, and shows more clearly how the outer collection angle close to α provides the optimal signal.

3.4.3 Comparison of Phase Retrieval Methods

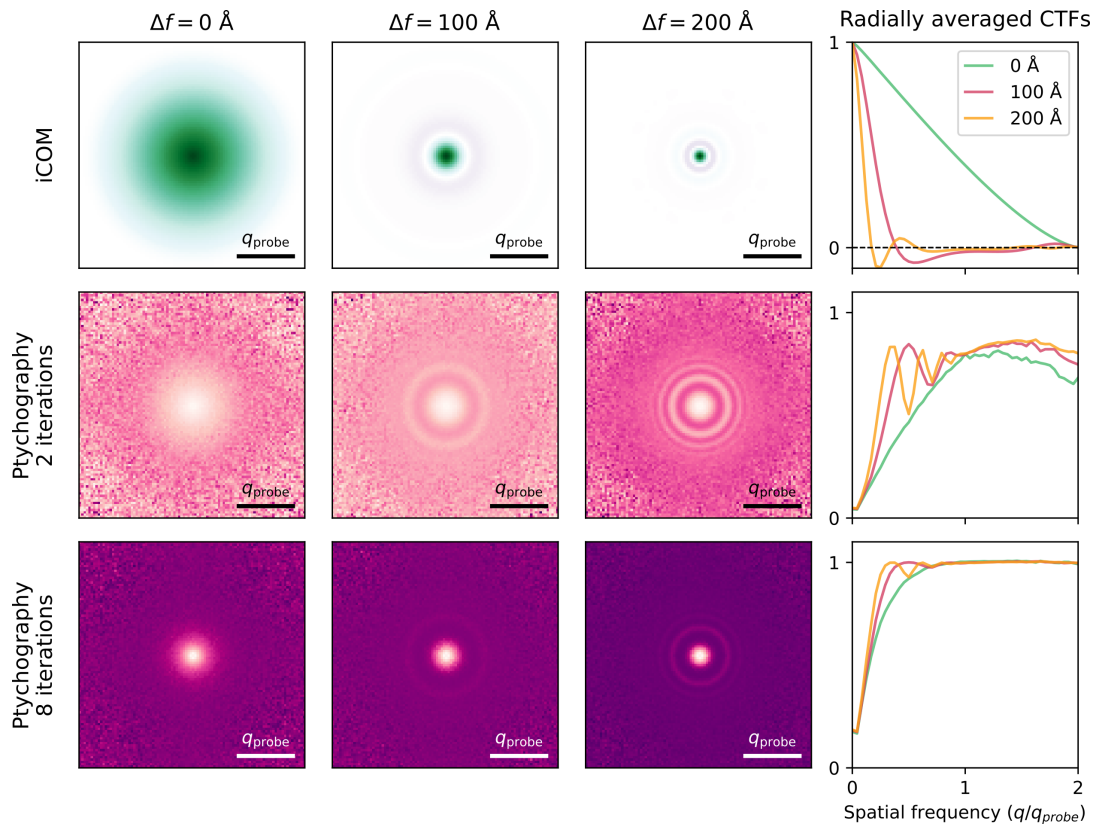


Figure 3.19: Comparison of CTF for iCOM and iterative ptychography with pixellated detectors, with increasing probe defocus, highlighting that increased defocus can help the iterative ptychographic reconstruction to reach unity information transfer faster.

Evaluating the CTF also yields a fair platform on which to directly compare different phase retrieval methods. This section will compare iCOM and iterative ptychography. Figure 3.13 shows that the CTF for iCOM is degraded at high defocus values, which means that added defocus hinders iCOM contrast transfer. Iterative ptychography, on the other hand, benefits from probe defocus because defocused probes have more overlap in real space in addition to the overlapping disks in reciprocal space. This introduces more redundancy in the dataset, which causes the algorithm to converge to a solution faster, see Figure 3.19. However, when the defocus is increased, the features in the CBED pattern become smaller and therefore more pixels in the detector is required to resolve them such that the algorithm may successfully deconvolve the object and the probe. As a result, segmented annular and few-element pixel array detectors typically do not yield satisfactory reconstruction for heavily defocused datasets. Still, iterative ptychography with any detector yields better contrast transfer with defocused probes when compared with iCOM. Practical implications of a balance between defocus and detector pixellation will be investigated in chapter 5.

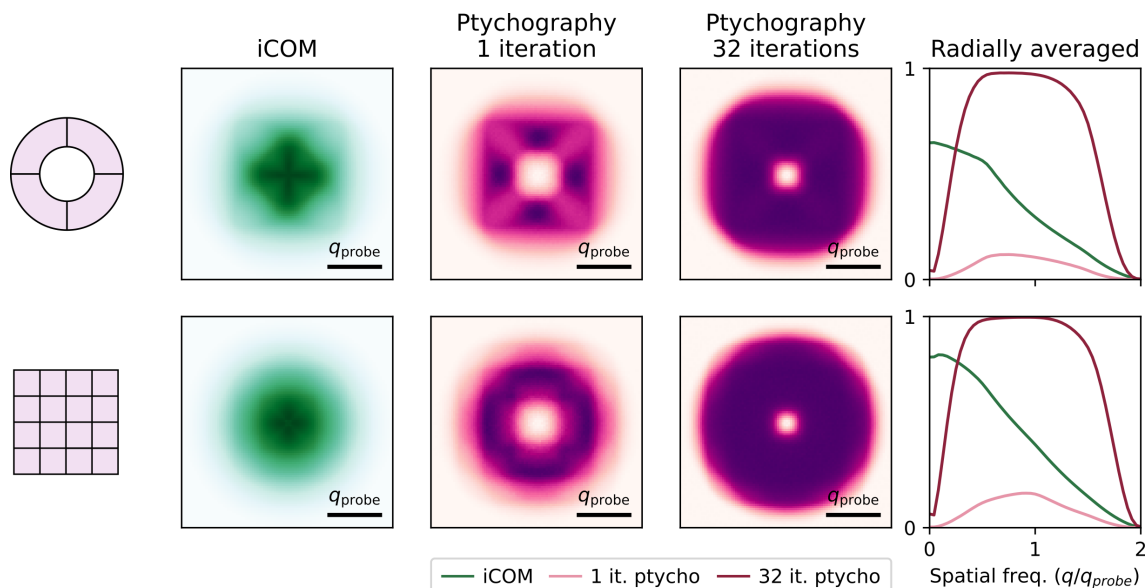


Figure 3.20: Comparison of iCOM and iterative ptychography CTFs for annular four-segmented and 4×4 array detectors with an $\alpha = 20$ mrad beam. Note how the CTFs for the 4×4 pixel array detector are more isotropic for both iCOM and iterative ptychography, and that the latter reaches unity for mid-range spatial frequencies already at 32 iterations.

For in-focus datasets, iterative ptychography works well with both annular segmented detectors and pixel array detectors, as may be seen from the comparison in Figure 3.20. Here it is clear how the geometry of the detector is reflected in the CTF for both phase retrieval techniques, and that the first iteration of iterative ptychography provides a worse contrast transfer than iCOM. However, this is quickly remedied by more iterations, and already after 32 iterations, the anisotropy imposed on the CTF from the detector geometry is mostly removed. The radially averaged CTFs show how the 4×4 array detector provides faster convergence by showing that it reaches unity for mid-range spatial frequencies already at 32 iterations. Further iterations will help fill in the lower and higher spatial frequencies, but these detectors are unlikely to be able to provide super-resolution⁴. Since the outer collection angle is chosen to match the optimum found previously, which is slightly larger than α , these phase retrievals are effectively limited by 2α . Iterative ptychography has been shown to yield super resolution both with pixellated and annular segmented detectors [98, 161], but this requires sampling of a significant amount of dark-field electrons. Acquiring dark-field electrons as well as adequately sampling the direct beam requires multiple rings in annular detectors or an $n \times n$ pixel array detector with $n > 4$.

The early iterations of ptychographic reconstruction with an annular four segment detector have a four-fold symmetry in their CTF, as can be seen in both Figure 3.20 and Figure 3.21. With an increasing number of iterations, this improves and

⁴Meaning resolution beyond the diffraction limit given by the numerical aperture of the microscope.

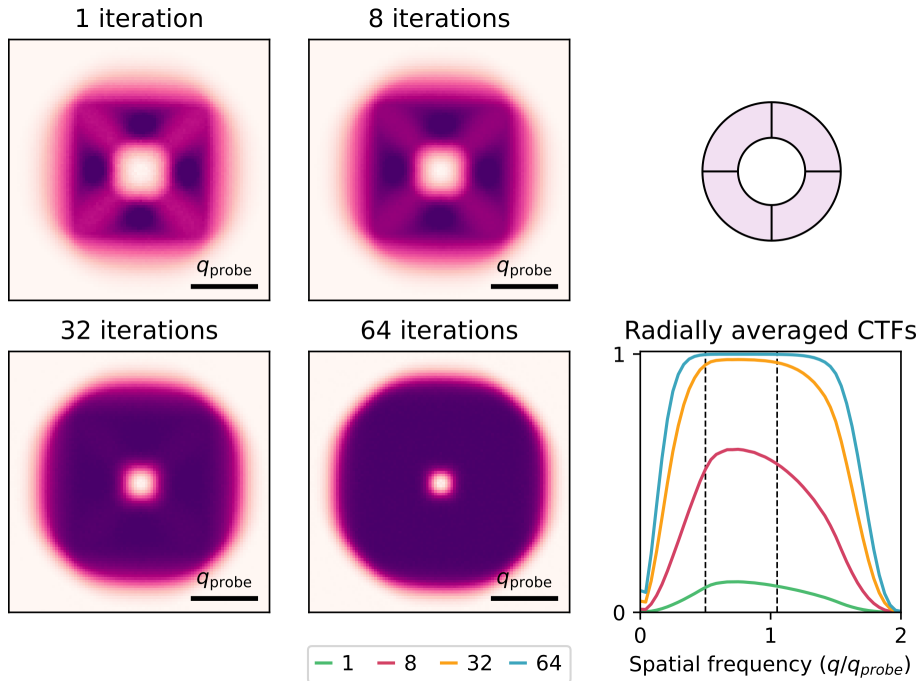


Figure 3.21: Information transfer for in-focus iterative ptychography at different iterations, showing that the detector geometry is reflected in the CTF anisotropy in early iterations. With an increasing number of iterations, the CTF becomes more and more isotropic.

the CTF is significantly better after 8 iterations. Again, the radially averaged CTFs obscure the anisotropy, but provide a clear picture of the increased magnitude of the contrast transfer. Importantly, at 64 iterations the CTF reaches the ideal unity for mid-range spatial frequencies, highlighting the usefulness of segmented detectors for iterative ptychography.

3.5 Conclusions

This chapter has introduced two ways of optimising experimental parameters for phase retrieval experiments. Firstly, the optimal defocus condition and sample thickness for a [100] STO sample were investigated specifically. The later investigation of defocus condition using the CTF framework confirmed that iCOM does not gain anything from being performed on a defocused dataset. However, the CTF framework as presented here is insufficient for evaluating the appropriate sample thickness for such experiments due to the single-slice nature of the “white noise” object used for numerical investigations and the sample-less nature of the analytical auto-correlation framework. Further, the effects of misalignment of the beam on the detector were elucidated. While centring the beam on the detector is important for COM signal integrity, it is empirically evident that human alignment based on the mean signal intensity is sufficiently accurate. This conclusion is also based on the minor effect that a few mrad misalignment of the beam has on the

Table 3.1: Summary of the optimal experimental collection values for iCOM.

Parameter	Optimum	Comments
Sample thickness	2 – 40 nm	Approximately weak phase object
Defocus	1 – 6 nm	Below sample entrance surface
Beam miscentering	< 10%	Manual centering sufficient
Optimal collection angles		
Annular segmented detectors:		
Inner collection angle, α_{det}^i	$0 - \frac{\alpha}{2}$	Too large will cause over-estimation of COM
Outer collection angle, α_{det}^o	$\alpha - 1.2\alpha$	
Array detectors:		
Outer collection angle, α_{det}^o	$1.1\alpha - 2\alpha$	
Detector pixellation	< 16×16	Negligible benefit beyond this

Table 3.2: Summary of optimal experimental collection values for iterative ptychography.

Parameter	Optimum	Comments
Sample thickness	2 – 40 nm	Approximately weak phase object ^a
Defocus	> 0 nm	If detector allows ^b
Optimal collection angles		
Annular segmented detectors:		
Inner collection angle, α_{det}^i	$0 - \frac{\alpha}{2}$	Can go beyond if multiple rings ^c
Outer collection angle, α_{det}^o	$\alpha - 1.2\alpha$	
Array detectors:		
Outer collection angle, α_{det}^o	$1.1\alpha - 4\alpha$	Extend if more detector pixels ^d
Detector pixellation	3 – 256×256	More pixels $\rightarrow$ more flexibility ^d

^a For single-slice reconstruction. Thicker samples may be reconstructed with multi-slice ptychography [162–165].

^b The fewer segments/pixels the detector has, the less defocus can be supported. For fully 4D data, defocus of 200 – 300 nm is reconstructable with an $\alpha = 30$ mrad beam.

^c Outer rings collecting dark-field electrons can help reach super-resolution with segmented detectors [161].

^d Few segments/pixels work for in-focus experiments, more segments/pixels allow for higher defocus and more sampling of dark-field electrons yielding super-resolution.

CTF for iCOM. Additionally, the optimal outer collection angle for annular segmented detectors is found to be where the outer collection angle is slightly larger than the semi-convergence angle. This is confirmed by investigations with the iCOM CTF framework, and made more obvious by taking into account the noise of iCOM measurements.

Finally, the information transfer of phase retrieval methods iCOM and iterative ptychography was compared using a numerical CTF framework. This shows that the detector geometry, which strongly impacts the iCOM CTF, only impacts the CTF of iterative ptychography in early iterations. With more than 32 iterations, the CTF anisotropy from the detector geometry disappears, and the CTF becomes increasingly isotropic with more iterations. Importantly, iterative ptychography is also limited by 2α when using an annular segmented detector due to the detector's limited ability to resolve information beyond 2α .

Chapter 4

Digitisation of Segmented Detector Signal

Traditionally, the intensity detected by an electron detector is summed for the duration of a pixel. This is the analog and simplest way of recording a signal from a detector. It assumes a few crucial things; firstly that the intensity delivered from the scintillator and PMT system scales linearly with the number of electrons hitting it, and secondly that the scintillator response to an electron hitting it does not vary significantly depending on where on the detector the electron hit. More importantly, it assumes the duration of an electron detection event is shorter than the pixel dwell time¹. This last assumption becomes more and more problematic the more the dwell time is reduced — in other words, as the beam is scanned faster across the sample. There are multiple motivations for scanning faster, for example, to space out the sample exposure to the beam by acquiring multiple subsequent fast frames to delay the onset of sample damage. While the effects of this temporal transfer function of the detector have been well explained and studied by others [105, 124], the effects on segmented detectors and phase retrieval are less studied.

In this chapter, the effects of detector streaking on COM and iCOM signals will be introduced to motivate why the digitisation hardware can be useful also in phase imaging. Furthermore, experimental results comparing analog and digitised COM and iCOM images will be presented. Finally, experimental considerations of coincidence loss and the optimal setting of the threshold for the digitisation setup will be discussed in more detail to aid future work in the digitisation of signal from segmented detectors in the BF region.

¹ideally much shorter, at least $< \frac{\delta_t}{2}$

4.1 Simulations Including Detector Temporal Response

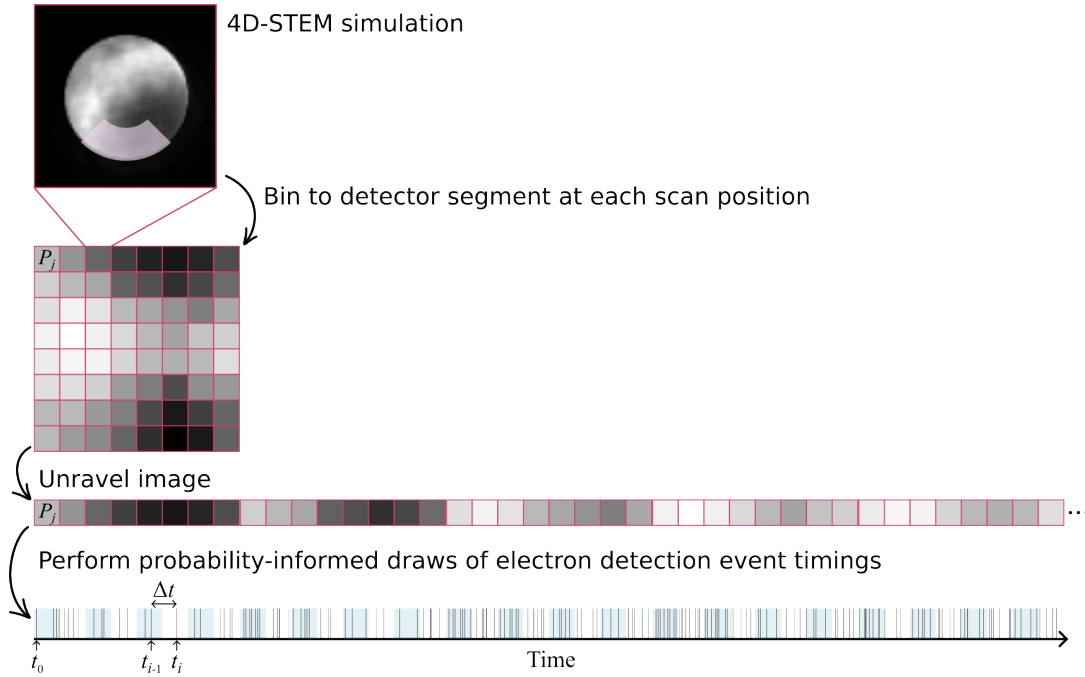


Figure 4.1: Process of simulating a time stream of electron events for a detector segment image calculated from a 4D-STEM simulation. A dummy image of eight by eight scan positions is used for illustration purposes.

Most simulated low-dose STEM images in literature have applied a finite dose by applying a representative Poisson noise, which arises due to the Poisson distribution of the electron emission from the electron gun. This is the absolute lowest limit of noise in experimental images. All experimental datasets have other sources of noise in addition to the Poisson noise. These other sources of noise include electromagnetic influence from outside wires and other detectors, 50 or 60 Hz interference from power sources, electrical interference from noisy ground, sample drift, and scan coils instability, to mention a few. For segmented detectors, there may also be crosstalk between the segments, causing signal from one segment to disturb the other segment signals. The latter, and the potential effects on the digitised data, will be discussed further in subsection 4.1.2.

In addition to all the sources of noise, the finite duration of the detector response to single electrons can cause information loss, especially at high scan speeds. Importantly, at significantly lowered electron doses, where every single electron counts, the margin for loss of information is minimal. To accurately evaluate the experimental images, it is common practice to compare them with simulations. However, comparison of experimental images with real noise levels with simulated images with only Poisson noise can, in the worst case, lead to misinterpretation of experimental data. In short, this necessitates the addition of real noise sources in simulations.

As introduced in section 2.1, the STEM image simulation softwares output probability images. That means that each pixel in the simulated CBED pattern contains the probability of detecting an electron in that pixel, and hence, a real-space image made from summing all or parts of the CBED pattern at each scan position is the probability of detecting an electron in that scan position with the applied detector geometry. Then, the expected number of electrons in each pixel, $\langle N_e \rangle$, is the rate parameter λ multiplied by the dwell time and the detection probability of the specific pixel. The rate parameter, λ , is given from Equation 2.1 as the beam current divided by the electron charge.

In order to simulate images including temporal noise effects, it is necessary to simulate a time-stream of the electron events arriving at the detector such that the detector signal may be simulated in the time-domain². In practice, the time-stream of events arriving at a detector segment is calculated by first binning the diffraction pattern to the detector segment for each scan position to obtain the probability of the segment detecting an electron at each position. The process is illustrated in Figure 4.1, where the probability of detecting an electron in the j^{th} position is given by P_j . Then, the probability of detecting an electron in each scan position is used to calculate the rate of arrival of electrons, λ_j , to the detector segment for each scan position independently. This is then used to draw Poisson distributed time stamps for each electron detection event. Note how the darker pixels in the example image in Figure 4.1 result in fewer electron events in the event stream because dark pixels represent a lower probability of electrons being detected.

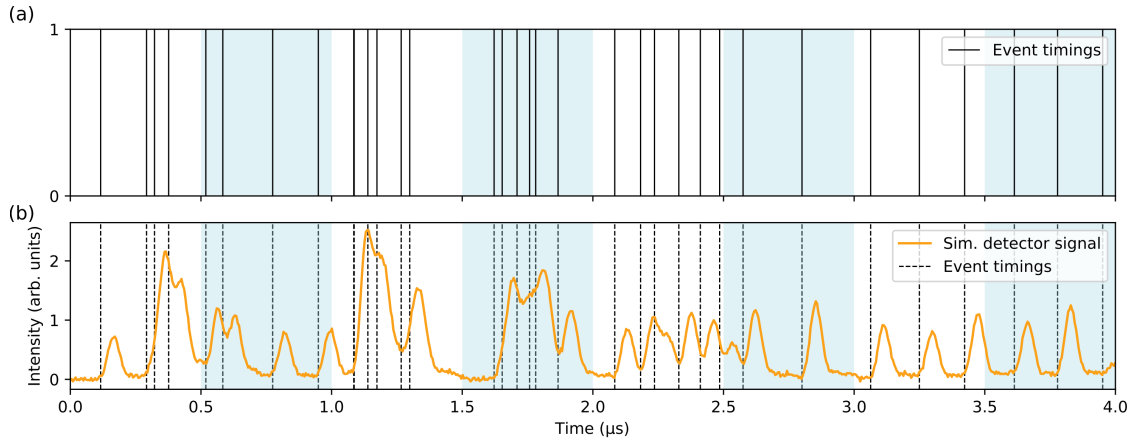


Figure 4.2: Illustration of (a) generated electron event stream, and (b) pulse stream through applying a detector response function to each electron event time. The shaded areas indicate the pixel boundaries for a dwell time of 500 ns. An experimental detector pulse shape from Figure 2.11

²Practically, the signal from electron detectors in experiment is a function of time before it is binned to each pixel.

The Poisson distributed time stamps for detection events are generated using the fact that the time between events in a Poisson process follows an exponential distribution. Therefore, if t_i denotes the time of the i^{th} electron detection event, then the time difference $\Delta t = t_i - t_{i-1}$ follows the exponential distribution with probability distribution function (PDF)

$$f(\Delta t) = \lambda_j e^{-\lambda_j \Delta t}, \quad \Delta t \geq 0, \quad (4.1)$$

where $\lambda_j = \langle N_e \rangle / \delta_t$ is the time-normalised rate parameter giving the expected number of events per unit time for pixel j . Generation of these inter-event times is done by drawing a random number, $U \in (0, 1]$, from a uniform distribution and calculating the time difference

$$\Delta t = -\frac{1}{\lambda_j} \ln(1 - U). \quad (4.2)$$

This is added to the global time variable, t , which starts at 0 and is only incremented with each valid Δt . When $t_i = t_{i-1} + \Delta t > \delta_t$ for the j^{th} pixel the event is discarded, and the rate parameter is updated with the detection probability of the following pixel. The process is repeated for every pixel in the $n \times n$ image such that the result is an event stream of Poisson-distributed time stamps over the interval $[0, n^2 \delta_t]$, which is weighted by the detection probability of each individual pixel. This is effectively a more computationally intensive way of applying Poisson noise to a simulated two-dimensional STEM image, if the event stream is now binned into image pixels.

Having simulated the electron detection events in time allows for addition of temporal effects to the event stream. Firstly, the detector response function can be applied to each detection event time, simulating the effect of the finite response of the detector on the image. The detector response function may be either a model or an experimental pulse shape, like the one seen in Figure 2.10. Since the detector response function is applied to each detection event time separately, the intensity of each pulse may also be varied to simulate detector inhomogeneity. An example event and pulse stream can be seen in Figure 4.2. Note how the signal from events occurring in one pixel sometimes pours over into the next pixel, which in more extreme cases causes streaking artefacts in the image [124]. Additionally, the inevitable background noise in analog detectors may be added by adding Gaussian noise to the pulse stream.

For monolithic detectors, a single event stream is sufficient, but for segmented detectors that have multiple detector channels it is necessary to simulate multiple event- and pulse streams. The simplest way of achieving this is to simulate each low-dose segment image separately. Since each segment image is a probability image,

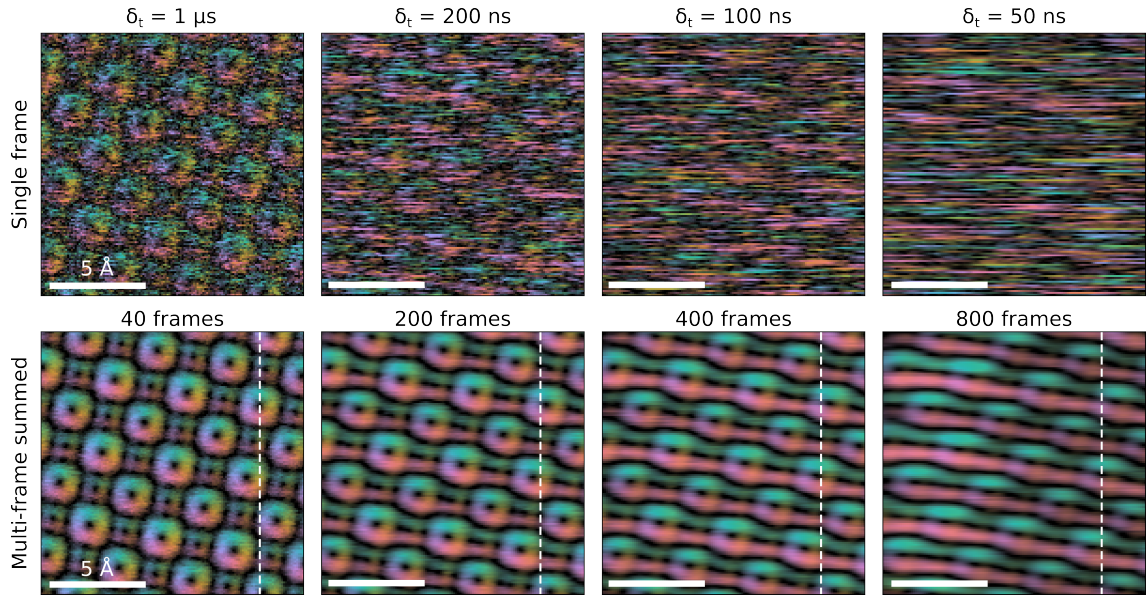


Figure 4.3: Simulated analog, single-frame (top) and multi-frame (bottom) COM images of [100] STO at increasingly fast scan speeds. The number of frames simulated was increased with decreasing dwell time in order to keep the total beam exposure time the same, such that the images may be directly compared.

accounting for the probability of detecting an electron in that specific segment, independent low-dose image simulations are possible. The mathematical validity of this in terms of the Poisson statistics is outlined later, in subsection 4.1.4. However, in cases where it is advisable to include effects that are dependent on how the segments interact with one another, for example crosstalk, simultaneous processing of the segments is required. This can be achieved by simulating an event stream for the full beam current and then subsequently performing a probability-informed choice of detection status for each electron event individually. Then, for a four-segmented detector, there exists five possible outcomes: the electron is detected by one of the four segments or it is not detected at all.

4.1.1 Streaking Effects on Phase Images

In order to investigate how beneficial digitisation of the signal from a segmented detector could be, it is informative to first study the effects streaking has on COM images and iCOM phase reconstructions. Since streaking is a detector response effect, it will directly affect the acquired images from individual segments of a segmented detector. This means that the probability images from each detector segment must be simulated, and then these can subsequently be used to simulate streaking in each individual segment.

The images in Figure 4.3 are the resulting COM images from simulating streaking artefacts in segment images at increasing scan speeds. Note that the underlying STEM simulation providing the probability images is the same for all of these

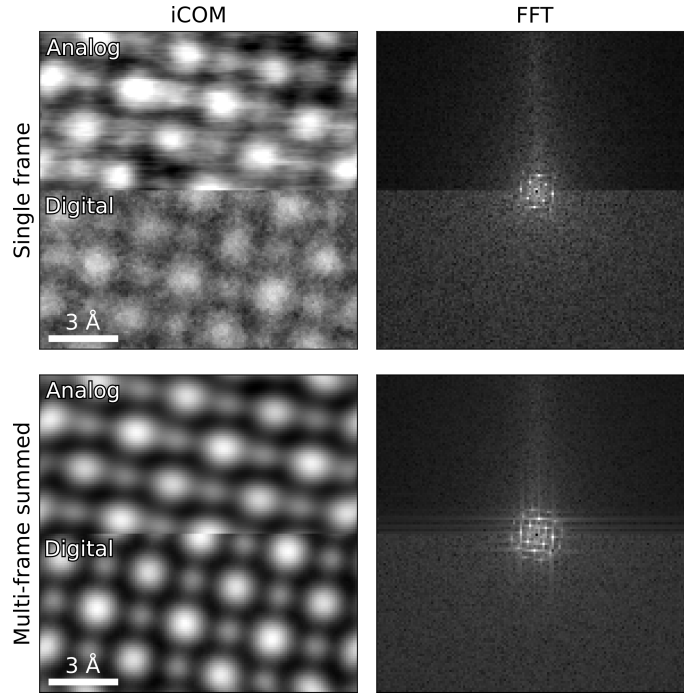


Figure 4.4: Simulated analog and digital iCOM phase retrieval images from using a four-segmented detector at $\delta_t = 200$ ns and $I = 5$ pA.

streaking simulations. Thus, it is possible to see directly how the streaking causes the atomic column positions to shift towards the right — essentially downstream in *time*. A vertical line has been added to the multi-frame COM images in Figure 4.3 to help see this shift in column position. Calculating the iCOM images from these COM signals shows how the streaking in the segment images affects the final, phase-retrieved images, an example of which can be seen in Figure 4.4. These images clearly show how the detector response function is expected to hamper the transfer of information at high spatial frequencies in the fast scan direction. The bottom row of Figure 4.4 shows that multi-framing may help even out some of the low spatial frequency artefacts of the iCOM reconstructions, but the FFT confirms that the information transfer at high spatial frequencies is still limited by the detector response function at the dwell time $\delta_t = 200$ ns.

Calculating the iCOM images also allows for the use of atomic column position recognition software typically used on HAADF images. Since this is a simulation, the ground truth position of the atomic columns is known, and thus the error in the estimated position from iCOM reconstructions from the exact column positions may be found. This error increases significantly for increasing scan speed in analog images because of the streaking artefacts, see Figure 4.5. Unsurprisingly, the error also increases for the single-frame digital iCOM images as the dwell time is decreased due to the lowered signal level. When the signal level is recovered by acquisition of many frames, such that the total exposure time is the same across all dwell times, the error of atomic column positions in the digital iCOM images does not

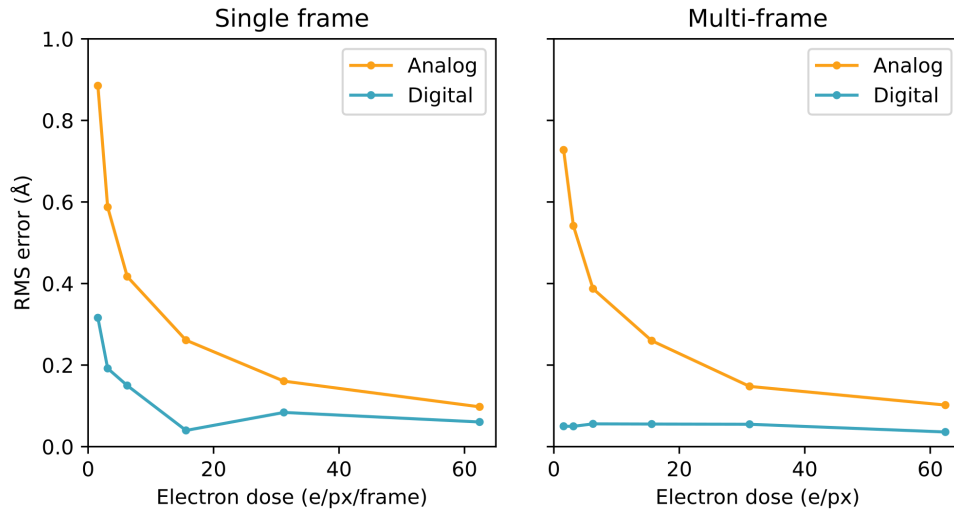


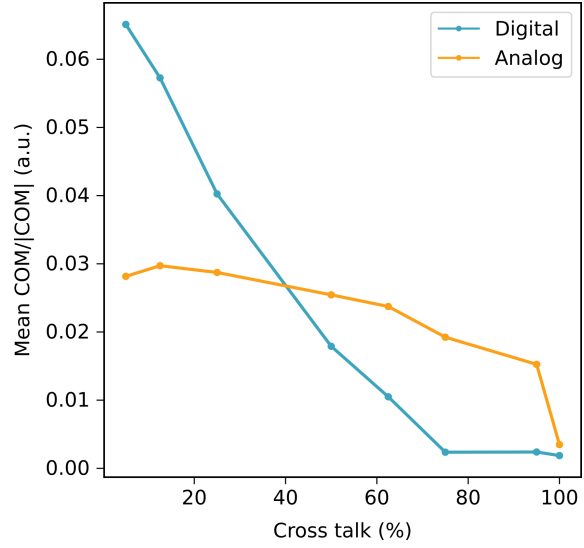
Figure 4.5: Error in position of the atomic columns from the ground truth iCOM images from simulated analog and digital low-dose segment images. The beam current used was 5 pA and the dwell time was 200 ns.

increase with decreasing dwell time. Crucially, the error in the estimated position of atomic columns from the analog images increases for reduced dwell times, also for the multi-frame iCOM images.

4.1.2 Crosstalk Between Detector Segments

For segmented detectors, there exists a probability that electron events from one segment also show up in one or more of the other segments. This disturbance is less likely to show up in an ADF detector because it is monolithic. Only if a monolithic detector is somehow connected with another detector or source of signal disturbance could this effect be present. However, when using a segmented detector, which inherently requires the readout of several signals simultaneously, there is always a finite probability that an electron is detected in more than one segment at a time. This can be because of a shared event between two segments, i.e. an electron hitting the border between segments and thus being recorded in two or more segments simultaneously. Furthermore, the detector segments might not be sufficiently insulated, leading to light or other signal leakage from one segment into another, which has previously been reported for x-ray and CT detectors [166, 167]. Additionally, there are other sources of both noise and potential crosstalk in the wiring of the detector, the grounding of components if the ground is noisy, and in connecting pieces that might be less electrically insulated and thus more susceptible to electromagnetic influence from outside sources. The shared-events type crosstalk can only be realised for bordering segments and thus, for the 2nd ring on the SAAF, this type of crosstalk can not occur between segments S1 and S3, and S2 and S4, see Figure 2.3 for detector illustration.

Figure 4.6: Absolute value of the average phase information in multi-frame analog and digital images for different cross-talk probabilities for shared events in four segments. The beam current used for the simulation is 3.2pA and the dwell time is 100 ns.



Shared events type cross-talk has been implemented in the simulation software discussed previously. An additional event stream for each segment is defined such that for every simulated electron detection event in a given segment, there is a certain probability that the event occurs in one or more of the other segments as well. Using this scheme, segment images with varying cross-talking probability have been simulated. The mean phase information, $\text{COM}/|\text{COM}|$, versus cross-talking probability for analog and digitised signals can be seen in Figure 4.6. Here, it is evident that the digital signals suffer more drastically from cross-talk. This is to be expected since the subtraction of the segment signals, as given by Equations 1.22 and 1.23, for analog signals with cross-talk does not necessarily lose all information when cross-talk events occur. For digital signals, on the other hand, if both the event and the cross-talk event in the opposite segment are picked up and digitised, that can result in $\text{COM}_x = 1 - 1 = 0$ for that pixel. However, Figure 4.6 also indicates that at lower cross-talk probabilities, less than about 40%, digitised COM images retain more of the phase information in fast scans.

4.1.3 Addition of noise

To make sure that any outside periodic noise is added to the image correctly, the line fly-back time (T_{LFB}) must be taken into account. A natural solution would be to add the corresponding number of pixels to the end of the image, such that the dwell time for each pixel would in itself account for the fly-back time. However, this adds a significant area to the simulation and thus slows it down significantly. The area the beam would have covered during T_{LFB} if it had spent that time scanning with dwell time $\delta_t \in (50, 500)$ ns is much larger than the image size itself. Therefore, another solution is devised, where the actual calculation of the periodic noise is performed for the duration of each line only, but the time variable used for each line takes the

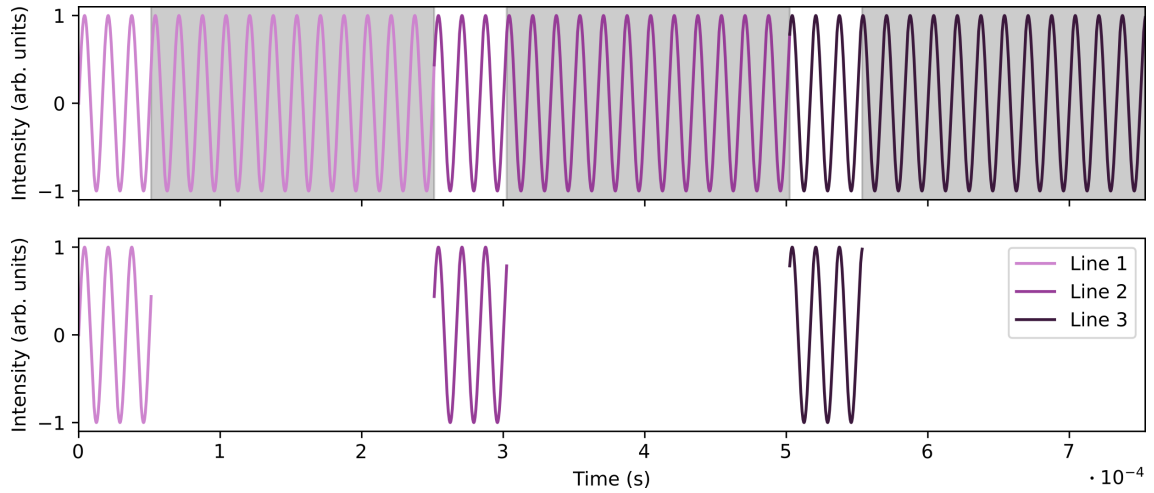


Figure 4.7: Illustration of a 60 kHz periodic noise during three lines of a STEM scan with 512 pixels and $\delta_t = 100$ ns. The grey boxes indicate the time spent while the beam is flying back to start the next line. The top graph shows the entire duration of each line, including the line fly-back time. The bottom graph shows only the parts of interest; the noise during the scan.

corresponding fly-back time into account. In practice this is done by concatenating several time-arrays

```

1   t0 = np.linspace(0, T_line, npix*steps)
2   t1 = np.linspace(dur, dur+T_line, npix*steps)
3   .
4   .
5   .
6   tn = np.linspace(n*dur, n*dur+T_line, npix*steps)

```

where n is the number of lines, **steps** is the sample points per single electron event pulse, and

$$\text{dur} = \delta_t \cdot n_{px} + T_{\text{LFB}}$$

$$T_{\text{line}} = \delta_t \cdot n_{px}.$$

This essentially picks out only the noise that occurs during active scanning, as illustrated in Figure 4.7, which allows for the simple addition of the noise into the simulated pulse streams.

The reason why it is useful to simulate this is that a common trick to avoid disturbances from e.g. the power supply frequency (50 or 60 Hz) is to sync the start of each pixel, line or even frame to the power supply frequency. When scanning with $\delta_t \in (50, 200)$ ns, it is possible to sync the start of each frame to mains frequency if the frame size is low enough, typically 128^2 to 256^2 pixels. Given this, it is unsurprising that the mains frequency varies at a much lower rate than the line acquisition frequency and as such the noise is fairly uniform throughout a single line. Keeping this in mind, the noise seen in experimental images captured with $\delta_t = 100$ ns, as can be seen in Figure 4.8, must have a much higher frequency than

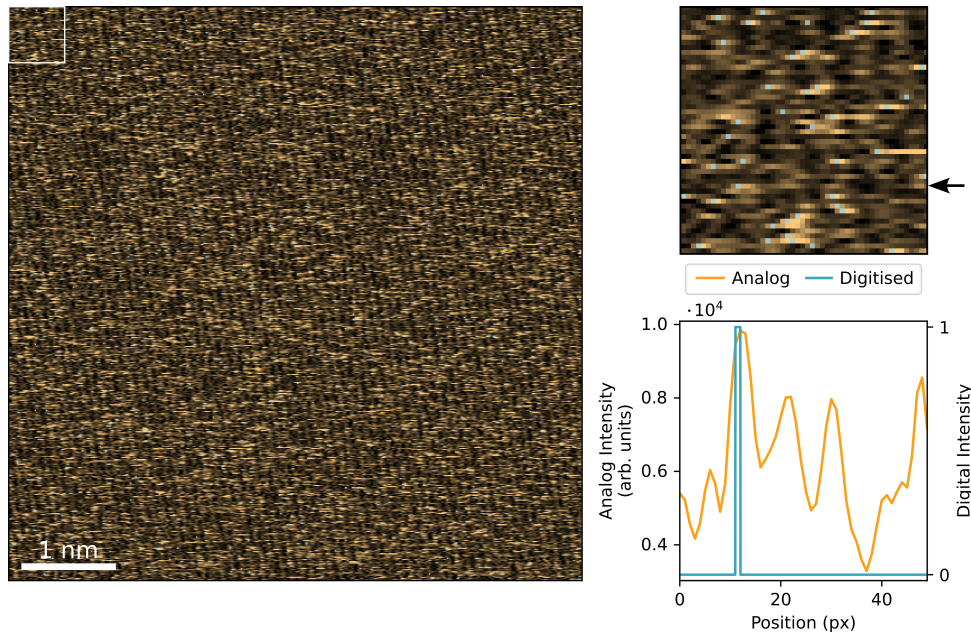


Figure 4.8: Experimental analog (orange) and digitised (blue) single-frame (512×512 pixels) image from the SAAF detector. The zoomed in image is the 50×50 pixels in the top left corner, marked with a white box, and the line profiles plotted are along the line marked with an arrow.

50/60 Hz and therefore likely comes from a different source of noise than the power supply. For this reason, the example frequency used in Figure 4.7 is 60 kHz.

4.1.4 Simultaneous Poisson Draws

A question that quickly arises when trying to simulate segmented detector signals at ultra-low doses is whether the segment images may be simulated separately or not. As mentioned in subsection 2.1.2, the electron emission from the gun in a TEM is a Poisson-distributed process. Even though scattering probabilities are deterministic and alter the distribution of the Poisson-distributed electrons in the beam in space, the electrons stay Poisson-distributed in time. As a result, the arrival of electrons to any detector, segmented or not, is still Poisson-distributed. Due to the presence of the sample and the finite size of the detector in STEM, the arrival rate of electrons to the detector will be lower than the rate of electron emission from the gun. The sample may backscatter or absorb some of the beam electrons, lowering the number of electrons transmitted through to the detector. Additionally, the collection angles of the detector determine how many of the scattered and unscattered electrons are detected by the detector. For segmented detectors in the ABF region, mostly unscattered electrons are detected. Still, a proportion of the beam electrons will have been scattered to higher angles, thus lowering the rate of electron arrivals to the detector.

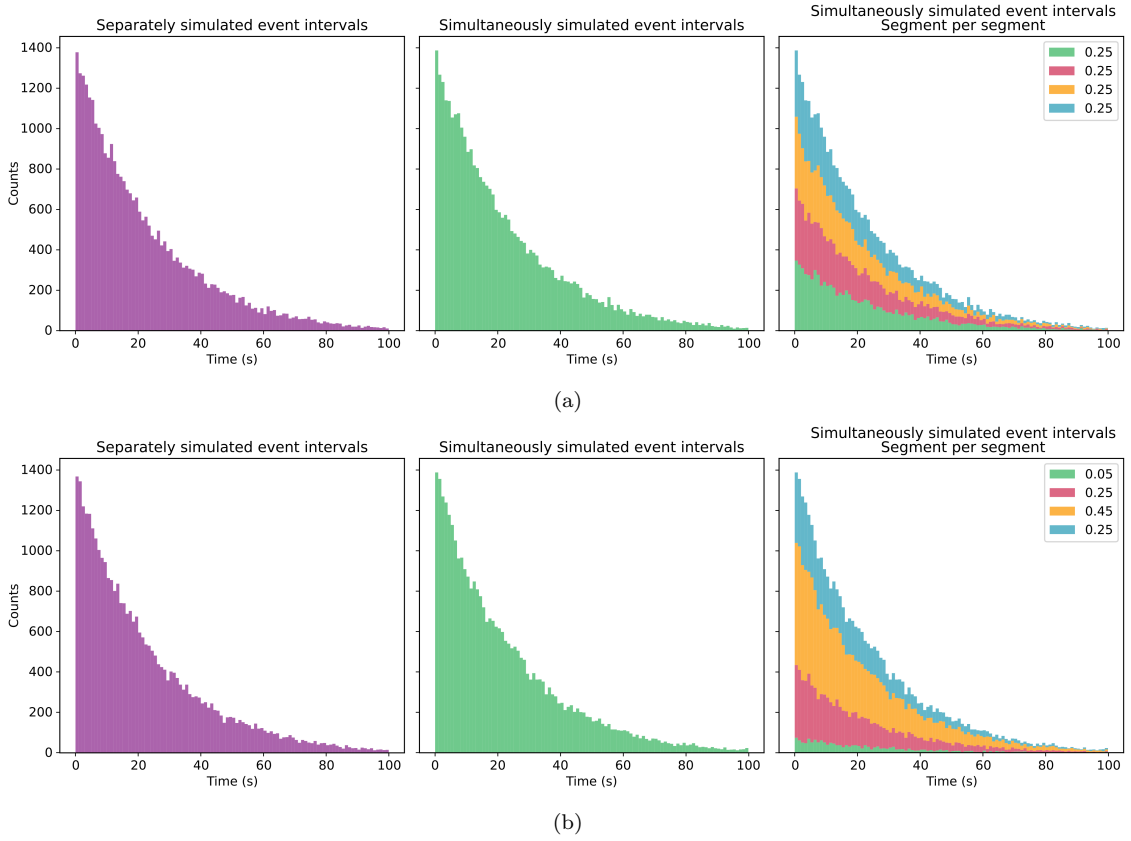


Figure 4.9: The histograms of time between consecutive events in a Poisson process with (a) equal probability of electron detection in each segment and (b) unequal probabilities. These visualise how simulating the four segments separately and simultaneously give the same statistics overall.

For simplicity, let I denote the beam current arriving at a segmented detector, such that the rate parameter follows directly from Equation 2.2. The probability distribution for the time between two consecutive events arriving at the whole detector follows the exponential distribution

$$dP_{\text{detector}}(\lambda, t) = \lambda e^{-\lambda t} \quad (4.3)$$

for which the cumulative distribution function (CDF) is given by the integral

$$P_{\text{detector}}(t) = \int_0^t dP(t) = 1 - e^{-\lambda t}. \quad (4.4)$$

Dividing the beam into n equal segments divides I equally such that the rate parameter of electron arrivals for the i^{th} segment becomes

$$\lambda_i = \frac{I}{n \cdot e} = \frac{\lambda}{n}. \quad (4.5)$$

Now the probability distribution for the time between two consecutive events in all of the segments together may be found by considering the probability that no event

occurs in any of them. The inverse of this probability distribution yields the PDF for consecutive events in all the segments. Let us first consider the probability that no events occur in the i^{th} segment,

$$P'_i(t) = 1 - P_i(t) = e^{-\lambda_i t}. \quad (4.6)$$

Now the probability distribution for consecutive events in any of the segments is unity minus the probability that no events occur in any of the n segments. Mathematically that is

$$\begin{aligned} P_{\text{segmented}}(t) &= 1 - \prod_{i=1}^n P'_i(t) \\ &= 1 - \left(e^{-\frac{\lambda t}{n}}\right)^n \\ &= 1 - e^{-\lambda t} \\ &= P_{\text{detector}}(t), \end{aligned}$$

and it shows that it is irrelevant whether the low-dose segment images are simulated together or separately. This result is shown visually by the histograms in Figure 4.9(a), and holds for the case where the detector segments are equal in geometry and simply divide the total detected current equally between them.

The assumption that the current arriving to each segment of a segmented detector is simply the total divided by the number of segments is oversimplified, and not a fair assumption realistically³. Instead, the total current arriving at the detector, I , may be spread arbitrarily across the n detector segments such that I_i is the current arriving in segment i and $\sum_{i=1}^n I_i = I$. Then the arrival rate λ_i , from Equation 4.5, may be redefined as

$$\lambda_i = \frac{I_i}{e},$$

such that

$$\sum_{i=1}^n \lambda_i = \frac{1}{e} \sum_{i=1}^n I_i = \frac{I}{e} = \lambda.$$

From here, the mathematical argument follows the same structure as above. Again, we make use of the probability that an event *does not occur* in any of the segments, given by Equation 4.6, to find the PDF for the time between events in *any* of the segments. Now, the probability that no event occurs in any of the segments is the

³If you think about it, COM imaging would be meaningless if there was never any difference between the intensity detected in the segments.

product of P'_i for all n segments, which gives

$$\begin{aligned}
 P_{\text{segmented}}(t) &= 1 - \prod_{i=1}^n e^{-\lambda_i t} \\
 &= 1 - e^{-t \sum_{i=1}^n \lambda_i} \\
 &= 1 - e^{-\lambda t} \\
 &= P_{\text{detector}}(t)
 \end{aligned}$$

This is visualised with the histograms in Figure 4.9(b). Thus, the conclusion from above, namely that it is irrelevant whether the event streams for each segment image are calculated simultaneously or separately, also holds with an unequal spread of the total detected current between the detector segments. This covers both the use of segmented detectors of arbitrary geometries, where the segments are not necessarily of equal geometry, but more importantly, any arbitrary scattering probability to the segments. Importantly, even if this allow for simulation of the event stream for each segment separately, some effects like cross-talk still require simultaneously simulating event streams for all the segments because they rely on the events occurring in the other event streams as well.

Finally, the fact that the Poisson-distributed events for low-dose segment images may be simulated separately is backed up by Raikov's theorem [168], which states that if two random variables X_1 and X_2 are independent and Poisson-distributed, then their sum $X = X_1 + X_2$ is also Poisson-distributed⁴.

4.2 Experimental Digitisation

So far, this chapter has introduced the simulation of real-life effects on COM and iCOM images and unique challenges that arise for segmented detectors. The simulations of the detector streaking effects on COM images in Figure 4.3 suggest that digitisation of the detector signal will significantly improve the signal quality achievable. This section will elaborate on the experimental results from digitisation of a segmented detector and comparisons with the analog versions. Additionally, some limitations of the digitisation and practicalities of optimising the acquisition parameters are explained.

⁴The converse is also true.

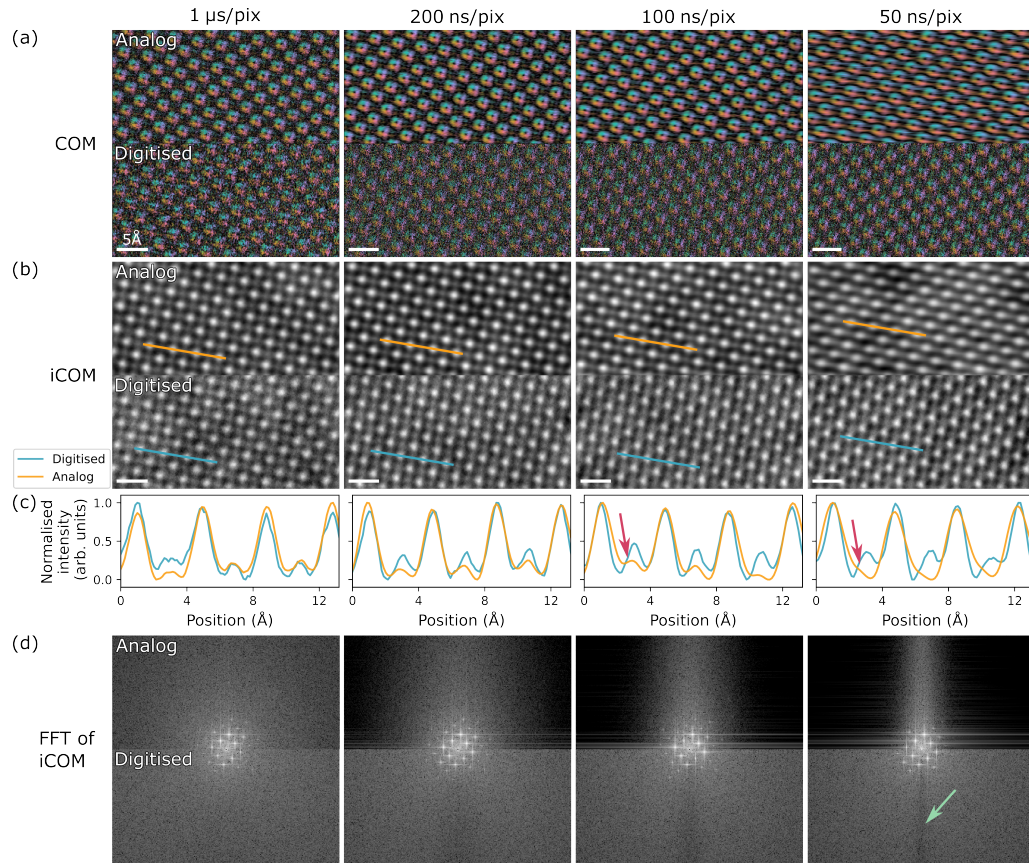


Figure 4.10: (a) Analog (top) and digitised (bottom) atomic resolution COM images of STO acquired with beam current $I = 5.2$ pA and increasingly fast dwell times. (c) Resulting iCOM images from the above COM without high-pass filter. Note how the resolution is severely reduced in the horizontal (fast scan) direction in the analog images due to image streaking. (d) Line profiles from the lines indicated in (c) showing clearly how the oxygen columns become increasingly unresolvable when the dwell time is reduced to increase the scan speed. In the analog iCOM images the oxygen columns become completely unresolvable at the Figure from [95].

Digitisation of the detector signal may be the solution to a few problems, but that is not to say that it does not create some other problems instead. Primarily, there is always a probability of two electrons arriving too close together to be separated into two events in the digitisation process, which leads to coincidence loss. Secondly, the digitisation process used here requires the setting of a threshold for what is to be counted as an electron event, which introduces human bias to the experiment when the operator has to choose the threshold that they think gives the best digitised signal. This section will elaborate on these two problems by providing some detailed insight into where coincidence loss comes from, and the effects both in theory and in experiment.

Experimental stacks of analog and digitised images at increasingly fast scan speeds were acquired on the second ring of a SAAF detector at the JEOL GrandARM at the University of Tokyo. The resulting COM and iCOM images of [100] STO, acquired with 300 keV beam energy and semi-convergence angle $\alpha = 30$ mrad, can

be seen in Figure 4.10. For all dwell times, the total image capture time⁵ used was 40 μs , meaning that 40 frames were acquired at $\delta_t = 1 \mu\text{s}$, whereas 800 frames were acquired at $\delta_t = 50 \text{ ns}$. Consequently, all images have approximately the same accumulated electron dose, which allows for direct comparison between them. From Figure 4.10, it is evident that the resolution in the fast scan direction is severely reduced for the fastest scan speeds due to image streaking. This is particularly clear from the line profiles in Figure 4.10(c) which highlights that the oxygen columns disappear in the analog images acquired with the fastest scan speed. The digitised COM images, on the other hand, retain the atomic positions even at $\delta_t = 50 \text{ ns}$ and yield a more isotropically reliable image.

The loss of information in the fast scan direction for the analog images is illustrated by the attenuation of higher spatial frequencies in the FFT. On the other hand, the FFTs of the digitised iCOM images have much flatter backgrounds when compared with the analog equivalents, which confirms that the digitised images are more reliable and less prone to losing high-frequency information in the fast scan direction. However, the FFT of the digitised iCOM images does reveal a small dip in information transfer in the fast scan direction for increasingly low frequencies as the scan speed is increased. This dip is partially due to coincidence loss, and as scan speed is increased, the average spacing of loss of counts in space becomes higher since the average spacing in time is constant. As a result, when the scan speed is increased, the increased spatial separation between coincidence loss events causes information loss at lower frequencies.

4.2.1 Coincidence Loss

Digitisation of an analog electron detector signal inherently involves choosing a criterion for what signal level is to be counted as an electron event. As mentioned in subsection 2.2.2, several methods of digitising the signal from the detector directly have been tested previously by others [111, 121–123]. Using the detector signal directly for the digitisation process easily leads to severe coincidence loss because events arrive too close to be separated with a threshold. However, a common observation is that the onset of a single electron event is sharp, while the decay is typically longer [105]. As a result, the gradient of the detector signal will have sharp peaks at the onset of each electron detection event. Thus, the threshold is here applied to the gradient signal rather than the direct analog signal from the detector — the same digitisation procedure as was introduced in subsection 2.2.2. This has the benefit that each electron event has a sharper peak, such that electrons arriving in fast succession may be more easily differentiated — thus coincidence loss

⁵not including fly-back times

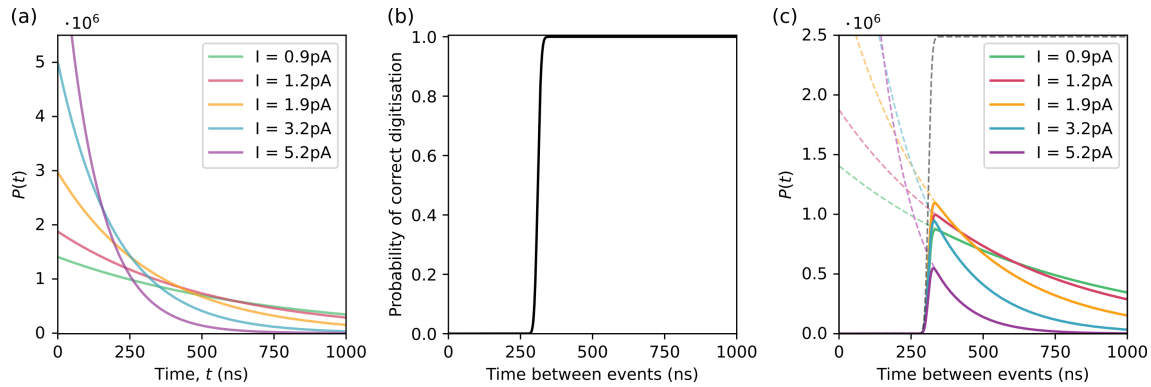


Figure 4.11: (a) PDF of time between events in a Poisson distribution, which follows an exponential distribution. These PDFs are adapted to a segmented detector with four segments covering the BF disk by assuming a rate based on $\frac{1}{4}$ of the beam current in the label. (b) The CDF of correctly digitised electrons for the SAAF detector as a function of the separation between electron detection events. (c) Combination of (a) and (b) gives the total PDF of time between correctly digitised electron events.

is minimised [80, 124]. The applied threshold is a crucial factor which defines the quality of the digitised signal, and this will be discussed in the next section. For now, the severity of coincidence loss will be characterised and avenues to minimise it investigated.

The emission of electrons from the electron gun is Poisson-distributed, and thus the arrival of electrons to a detector is also Poisson-distributed. Coincidence loss occurs when electrons arrive at the detector in too close succession to be successfully separated in the digitisation process, and this occurs even the gradient of the detector signal is used. One of the main reasons why phase retrieval methods are promising for ultra-low-dose characterisation is the high collection efficiency of using the bright field region of the electron beam. However, the placement of a detector in the BF or ABF region, giving the high collection efficiency, also means that the detector sees a very high arrival rate of electrons. This leads to a higher probability of coincidence loss when digitising the signal from the detector. Loss of electron events to coincidence loss causes degradation of the signal recorded and ultimately loss of valuable information, especially at low dose when the signal itself is already very low. Therefore, the arrival rate of electrons should be kept at a level where electrons are statistically likely to arrive at the detector one at a time. However, this is not necessarily easy to achieve in practice, which is why both the statistical models for Poisson-distributed events and the practical aspects of the arrival of electrons to various detectors in STEM are explored here. This will help shed light on the severity of coincidence loss for different experimental conditions.

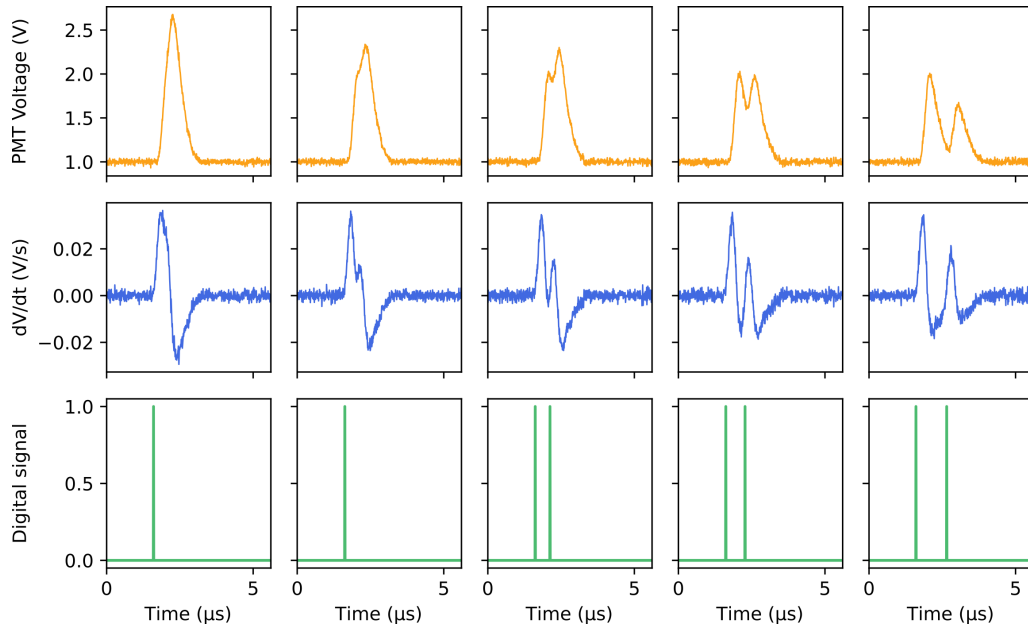


Figure 4.12: Simulation of coincidence loss by simulating two pulses with increasing separation eventually being digitised as two event rather than just one.

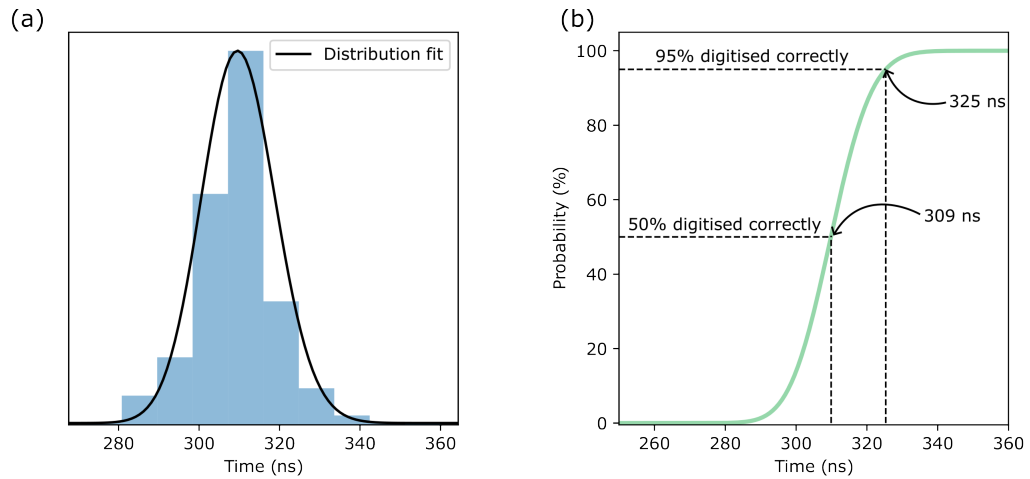


Figure 4.13: (a) Histogram of event separations at which two SAAF pulses are digitised correctly. 10000 repetitions of the simulation were run. A skew normal probability distribution has been fit to the histogram data. (b) Skew normal CDF using the fitted parameters to evaluate the chances of consecutive events being digitised correctly as a function of event separation.

For a Poisson distribution, the time between consecutive events follows an exponential distribution, see Figure 4.11(a). In order to determine at what event separation coincidence loss is likely to occur for a specific electron event shape, digitisation of two events may be tested when the separation between them is continually increased. This simulation scheme is illustrated in Figure 4.12. By repeating this simulation multiple times, statistics on what separation correctly yields two digitised events is gathered. Naturally, this will vary based on the relative signal strength of the two events. This is incorporated in the simulation by multiplying the normalised event signal with a strength factor with unity mean and standard deviation $\sigma = 0.1$, based on the average flatness of analysed detectors from Mullarkey *et. al.* [125].

Using the experimental electron event shape from the SAAF detector, see Figure 2.10, and running 10000 repetitions of the simulation yields the distribution shown as a histogram in Figure 4.13(a) for the event separation at which the two events are correctly digitised. Assuming this histogram well represents the probability distribution function (PDF) of correctly digitising the electron events, a skew normal distribution is fitted, see the solid black line in Figure 4.13(a). This leads to the cumulative distribution function (CDF) shown in Figure 4.13(b), from which the event separation at which two events have e.g. a 50% chance of being digitised correctly may be extracted. For this particular experimental electron event shape this separation time turns out to be 309 ns. For two events separated by 325 ns, the probability of them both being digitised correctly is 95%. While this is impossible to ensure in-experiment, it does help inform what beam current is practically usable for the given detector, considering its response function to electron detection events.

Importantly, the detector's position in the microscope will also have to be taken into account, as ADF detectors receive significantly fewer electrons than detectors in the BF region. The arrival rate of electrons to the detector depends not only on the beam current, but also on the scattering probability of the sample. For an ADF detector, a higher scattering probability results in a higher arrival rate of electrons. Conversely, for detectors in the BF and ABF region, a higher scattering probability lowers the arrival rate of electrons to the detector because of the positioning of the detector. This is because the intensity detected by a detector decreases exponentially with increasing inner collection angle [59], which means that the electron arrival rate is much higher in the BF region than in the ADF region. Segmentation does somewhat improve this, since each segment only detects a maximum of $1/n$ proportion of the total beam current. Therefore, using a detector with more segments could not only improve contrast transfer as was seen in section 3.4, but also help combat coincidence loss provided that signal disturbance like cross-talk between segments could be avoided. Although, importantly, the digitisation happens in-hardware and is performed on a per-segment basis. This means that the number of digitisation

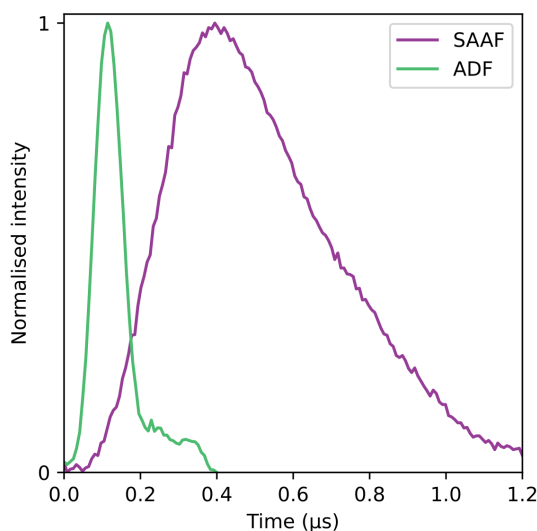


Figure 4.14: Single electron event shapes for segmented SAAF detector and monolithic ADF detector, showing how much faster the detector response function is for the latter.

channels on the digitisation hardware scales linearly with the number of detector segments that are to be digitised, and anything beyond 16 - 32 segments⁶ becomes impractical.

Using the CDF from Figure 4.13 and assuming a segment that detects $1/4$ of the total beam current, the total PDF for several different beam currents becomes the graphs shown in Figure 4.11(c). This reveals that when using a beam current of 5.2 pA, up to 92% of the electrons hitting the detector may be lost to coincidence loss if the entire beam current is sent directly to the detector. Scattering of electrons to higher angles outside the detector will lower the effective arriving current to the detector, and thus meaningful results are still possible with $I = 5.2$ pA experimentally. Yet, lowering the beam current is the primary route to reducing coincidence loss, and around 1.2 pA the proportion of electrons lost to coincidence loss for the full-beam case is reduced to 44%.

Using a Faster Scintillator

The severity of coincidence loss will also depend heavily on the single-electron event shape — that is, the detector response function. A sharper scintillator rise time and a shorter decay time give a single-electron event that is easier to digitise, and also a detector whose analog signal is less prone to producing streaking artefacts in the images. Figure 4.14 shows the single electron event shape for two very different scintillator detectors: a JEOL SAAF and a JEOL ADF detector. The latter has been specifically designed to support fast scanning and therefore has a much sharper detector response function. Performing the same analysis of simulating what event separation is required to correctly digitise two events for this faster detector shows

⁶Depending on how many four-channel signal digitisers you wish to have in your lab.

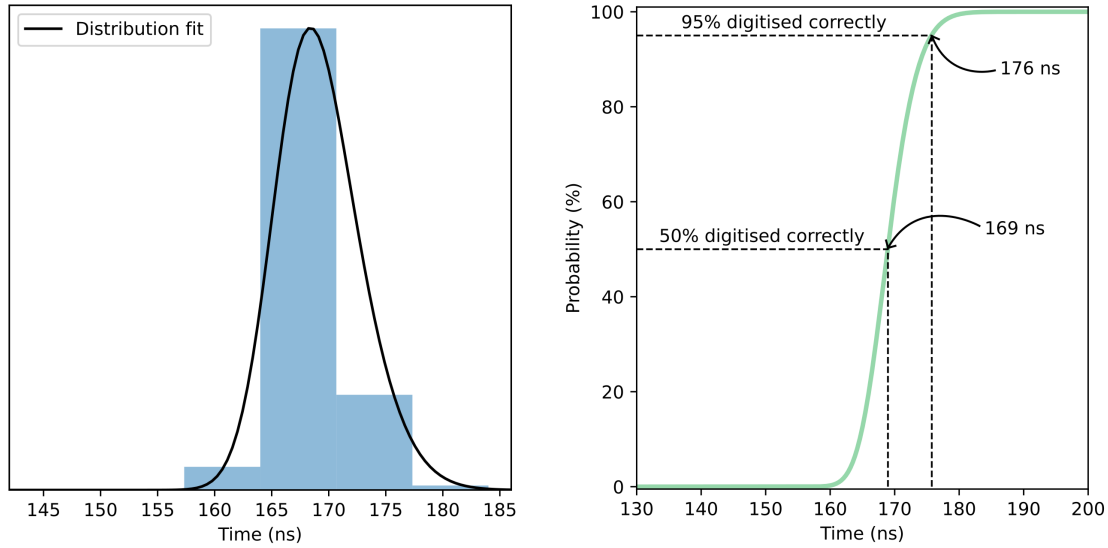


Figure 4.15: Histogram of event separation yielding correct digitisation of two electron events with the experimental detector response function of a monolithic ADF detector. A skew normal distribution has been fitted to the histogram and its corresponding CDF shows the probabilities of two events being digitised correctly as a function of their separation.

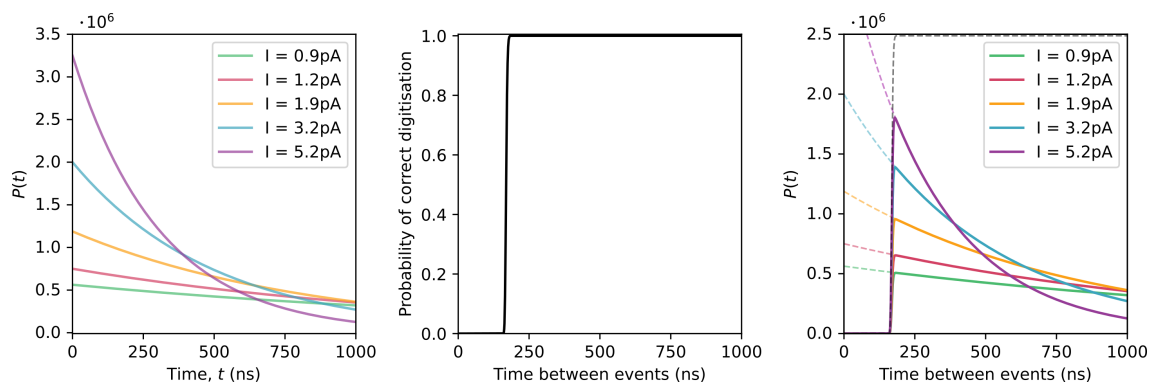


Figure 4.16: PDF of time between events arriving at a detector assuming 10% of the beam current indicated is scattered to the detector. Combination of the PDF with the CDF from the histogram fit yields the total PDF of correct digitisation.

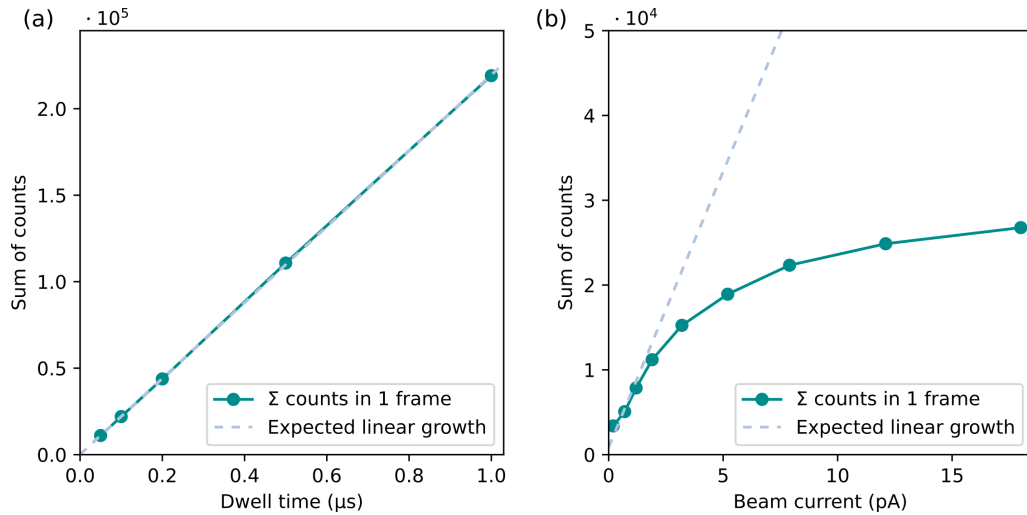


Figure 4.17: Sum of counted electrons in digitised images when increasing dose with (a) increasing the dwell time and (b) increasing the beam current, showing the expected linear increase in number of detected electrons is not observed when increasing the beam current due to coincidence loss.

that this detector can support much higher electron arrival rates than the SAAF detector. Since this is an ADF detector, a scattering probability of 10% is assumed for the PDFs in Figure 4.16, such that the beam current listed is the total beam current. This allows for a more straightforward comparison with the same analysis for the segmented detector shown in Figure 4.11. For this setup, over 50% of electrons are expected to be correctly digitised for a beam current of 5.2 pA — which is significantly higher than the meagre 8% for the segmented detector.

Experimental Observations

So far, the analyses of coincidence loss have all been from simulations, even though experimental detector response functions were used. The number of electrons detected in an image is expected to scale linearly with increasing pixel dwell time and increasing beam current, following Equation 2.4. Thus, the severity of coincidence loss may be investigated by looking at the total count of electrons detected per frame when the dwell time and beam current are increased, respectively.

The linear expectation is shown as the dotted lines in Figure 4.17, where the graph for increasing dwell time follows the expected linear growth in the number of electrons detected. In contrast, the number of electrons detected when the beam current is increased does not follow the expected linear growth due to the increased occurrence of coincidence loss. Figure 4.17 shows that for the SAAF detector used in the experiment, the number of electron counts diverges from the expected value already around a beam current of $I = 1.9$ pA, which is consistent with what was found by simulation for this detector.

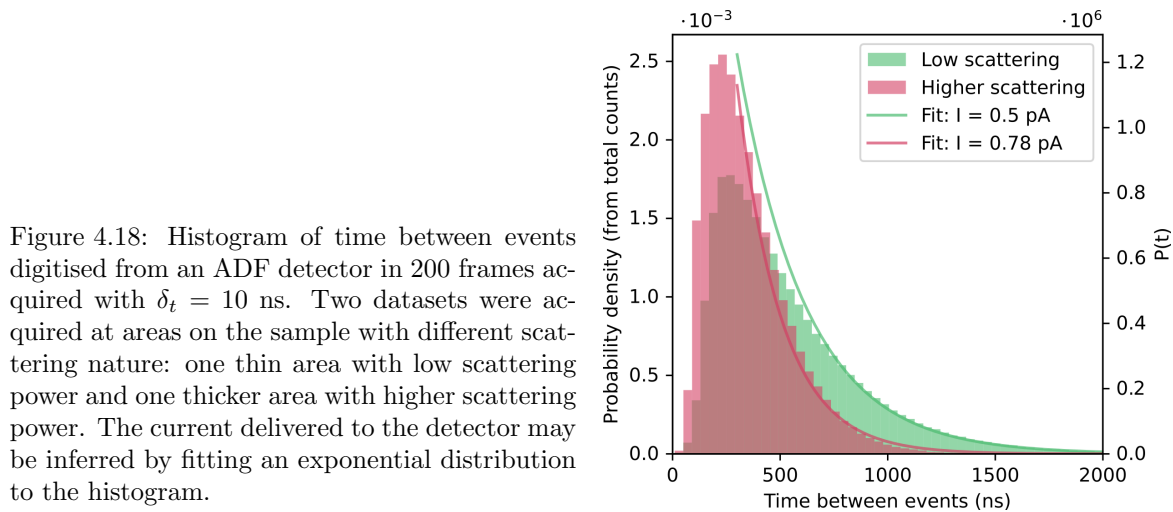


Figure 4.18: Histogram of time between events digitised from an ADF detector in 200 frames acquired with $\delta_t = 10$ ns. Two datasets were acquired at areas on the sample with different scattering nature: one thin area with low scattering power and one thicker area with higher scattering power. The current delivered to the detector may be inferred by fitting an exponential distribution to the histogram.

Finally, the coincidence loss may be investigated more directly by analysing the time between events detected directly in a digitised multi-frame series with very low dwell time. 200 frames were acquired with a dwell time of $\delta_t = 10$ ns using an ADF detector⁷ and two different areas on a sample. The two different areas on the sample had different scattering power, which yields different probability distributions for the time between events delivered to the detector, even though the electron beam current was kept constant. From the histograms in Figure 4.18, the current delivered to the ADF detector may be inferred by fitting an exponential distribution. Although, importantly, this experiment has the significant flaw that the time between events in experiment is only able to record the time between *detected* events. The time of arrival for any electron events lost to coincidence loss will always be lost, and as such, the time between events will inevitably ignore some lost electron events. Still, these histograms adequately follow the predicted shape from the analysis that gave Figure 4.11 and Figure 4.16 — that is, following an exponential distribution up until short time-between-events on the order of 100s of nanoseconds. Furthermore, the higher scattering power region exhibits a sharper peak with fewer events occurring with long separation times compared with the lower scattering region, which has more events occurring with higher temporal separation. Crucially, both histograms peak around 250 ns and significantly decline for event separations lower than that.

4.2.2 Auto-thresholding

As introduced previously, the digitisation scheme used here applies a threshold to the gradient of the signal stream from a detector or detector segment. Electron events are counted only when this gradient signal is higher than the threshold for more than a defined time-over-threshold. The use of such a time-over-threshold limit reduces the chance of accidentally counting noise as events. Crucially, the accurate

⁷A different ADF detector than the one investigated previously in this section.

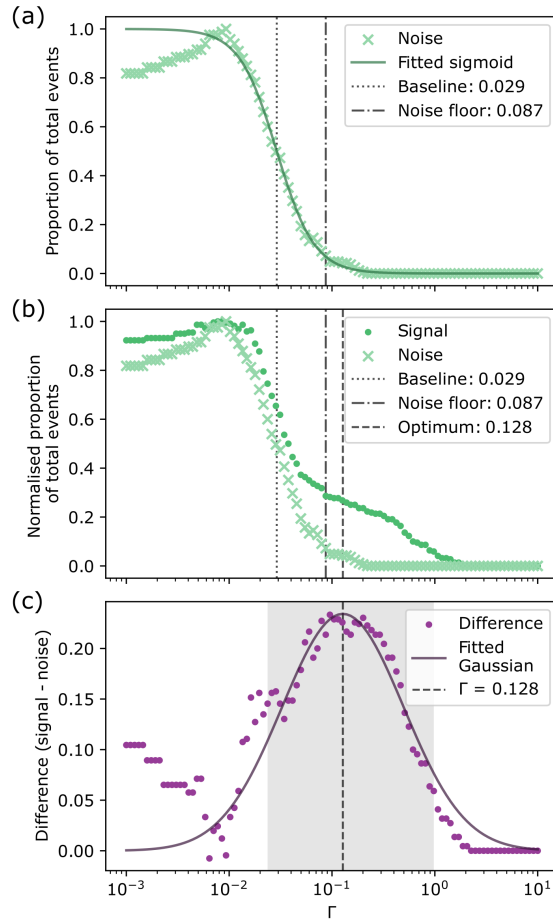


Figure 4.19: Plots used for optimisation of the threshold for a monolithic ADF detector. (a) The normalised number of events counted versus Γ for a noise stream acquired with the beam blanked. (b) Normalised number of events counted versus Γ for noise stream (x) and signal stream (dots). (c) Difference between the number of counts in signal and noise streams versus Γ .

digitisation of analog detector signals relies on choosing the optimal threshold. A threshold that is set too low risks counting noise as events, giving false positives, even with the time-over-threshold limit. Conversely, a threshold that is set too high risks missing signal events, especially those that give a weaker scintillation signal, yielding false negatives. In other words, the threshold must be set low enough to pick up less pronounced electron detection events, but high enough to avoid picking up noise and potential cross-talk events in segmented detectors. Allowing the microscope operator to set this threshold manually introduces human bias in deciding what threshold yields the highest quality digitised image. Thus, an automated and unbiased method of determining the optimal threshold to use for the digitisation process is necessary for achieving unbiased, reproducible results with digitised STEM imaging.

The goal when optimising the threshold is to minimise the number of false positives and the number of false negatives simultaneously. In other words, the threshold that allows correct digitisation of the highest proportion of actual electron events, whilst noise being accidentally counted as events happens as little as possible. This requires two data streams from the detector: one of the intrinsic noise in the detector with the beam blanked — a noise stream — and one with the beam on so that there are real electron events — a signal stream. To find the optimal threshold,

100 test thresholds evenly spaced logarithmically between 10^{-3} and 10^1 are applied to both the noise stream and the signal stream. The streams are independently digitised using each test threshold, and the number of electrons counted in the stream is recorded, see Figure 4.19(b), where the dots and the crosses represent the normalised counts from the signal and noise stream respectively. Then, these counts from the noise stream and the signal stream are compared by subtracting the normalised noise counts from the normalised signal counts, which produces the difference graph in Figure 4.19(c). The peak of this difference graph yields the threshold that maximises the difference between the signal counts and the noise counts, giving the lowest possible amount of noise counted as electron events and the highest amount of electron events counted correctly.

To ensure that the algorithm reliably finds the peak of the difference, a Gaussian function is fitted to the 30 points surrounding the maximum of the difference curve, and its peak is taken to be the central point of the shaded region in Figure 4.19(c). The fitting of a Gaussian is then repeated on the 40 points surrounding the point closest to the maximum of the first Gaussian fit. Such a double Gaussian fit scheme gives a robust and reliable way of finding the peak of the difference graph, which yields the optimal threshold for the detector⁸.

When automatically determining the optimal threshold by using this approach, it is important to set reliable limits for how low the threshold is allowed to be set. Ideally, there would be no accidental electrons in the noise stream, in which case the threshold could simply be set right above where noise starts being falsely counted as events. Unfortunately, it is impossible to know exactly how many accidental electron events appear in a given experimental noise stream. Therefore, a baseline lower limit for the threshold and a user-defined acceptable noise floor must be determined. The baseline is defined by fitting a Sigmoid function to the graph of the number of events counted in the noise stream versus the threshold in log-space, see Figure 4.19(a). The inflection point of this Sigmoid function is taken to be the baseline lower limit for the threshold, which is the absolute lowest limit for the threshold. Additionally, the operator will need to define a noise floor for what an acceptable number of accidental electrons per time is when the beam is blanked. Typically, this should be on the order of 10 electrons per 80 μs of noise stream, which is roughly 1 false event counted per 1000 sampling points or just over one electron per 10 μs . Importantly, the acceptable number of events should be above zero because, even when the beam is blanked, accidental electrons can occur and allowing no counts may therefore set the noise floor artificially high. The baseline, noise floor, and optimum threshold may all be seen as the vertical lines in Figure 4.19.

⁸For the current setup, particularly offset and gain settings.

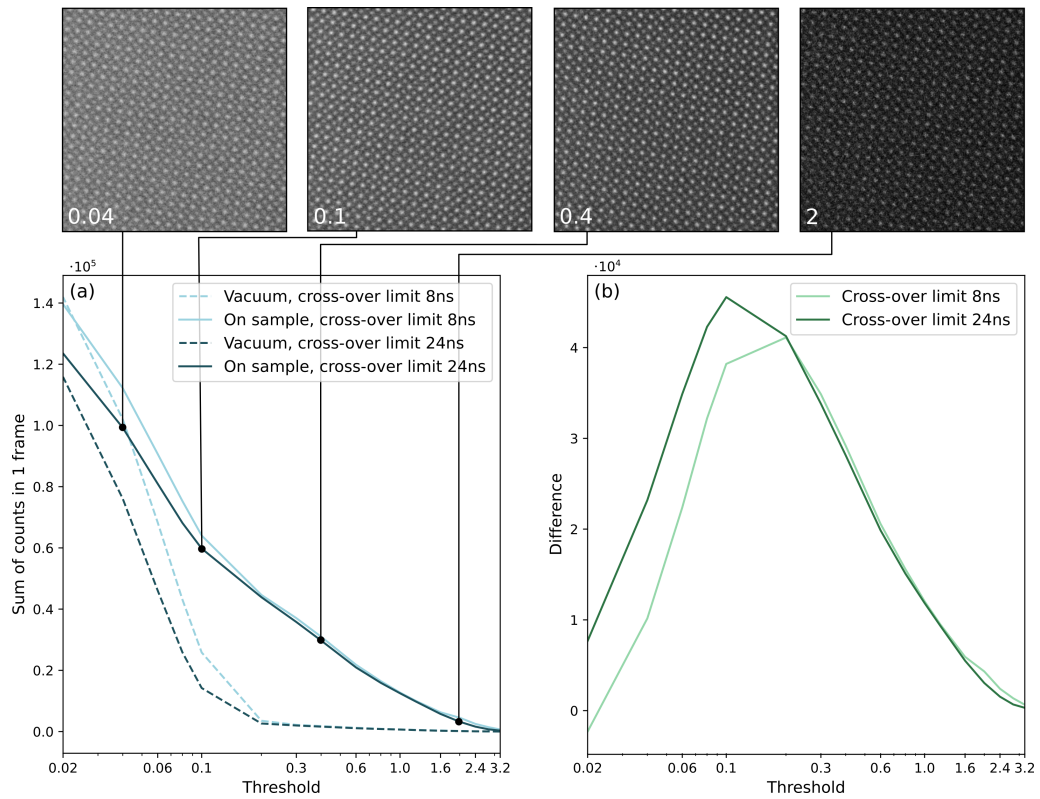


Figure 4.20: Experimental threshold series with the ADF detector showing (a) the sum of electron counts in a single frame digitised image of a [100] STO sample (lines) and of vacuum (dotted lines), for two different cross-over limits. (b) shows the difference between the number of counts in the on-sample images and the vacuum images for the two different cross-over limits, which gives an indication of the optimal threshold for this experimental setup. The images on the top are the resulting experimental, digitised images using four different thresholds as indicated by the markers on the plot.

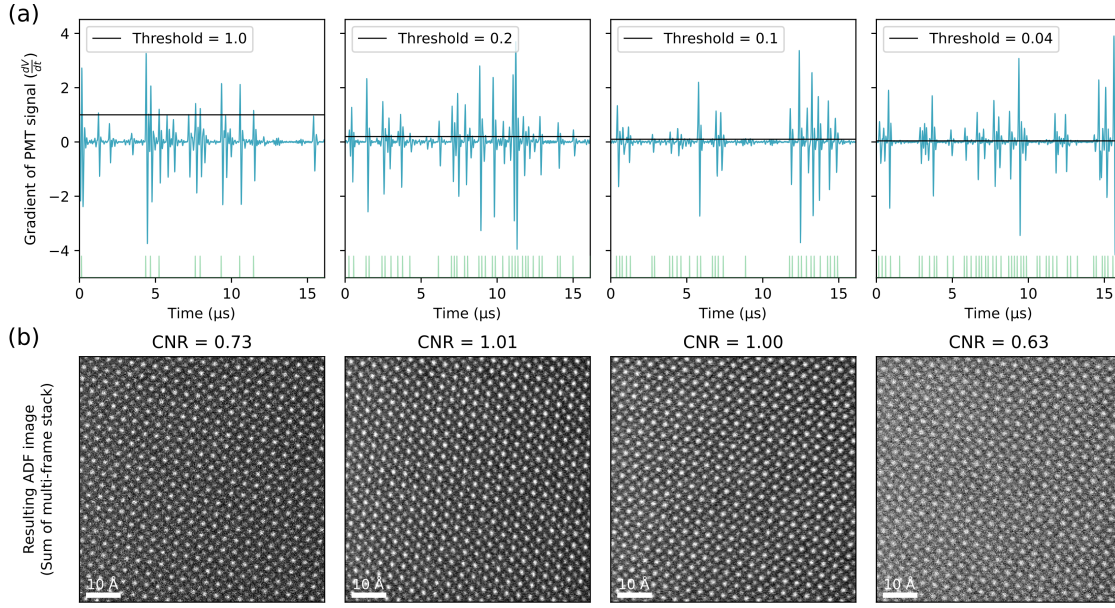


Figure 4.21: Experimental gradient of pulse streams and images with varying thresholds. (a) Detector signal gradient with the threshold indicated by the horizontal black line. The green ticks at the bottom show the digitised events with the relevant threshold. (b) Resulting digitised ADF images of STO when the relevant threshold was applied. These images are the sum of 100 frames acquired with 100 ns dwell time, beam current 8 pA, and semi-convergence angle $\alpha = 30$ mrad. The contrast-to-noise ratio (CNR) value was calculated by comparing the Sr and O columns.

It is also important to keep in mind that this procedure should be done after the gain and offset are set for the detector, and making sure they are set appropriately, see subsection 2.2.3. This is because changing the gain and offset changes the signal output from the detector, which may impact the optimal parameters for digitisation. Crucially, the output signal from the detector should always be within the limits of what the digitisation hardware can record: $-10 - 10$ V. Any signal beyond this will be clipped and result in incorrect digitisation.

Experimental Observations

To investigate the effects of varying the threshold experimentally, a threshold series was performed with a [100] STO sample. A total of 100 frames were acquired at 100 ns dwell time and 8.3 pA beam current and compared with a similar multi-frame dataset of vacuum. The resulting sum of counts in a single frame image, along with some multi-frame images at chosen thresholds, can be seen in Figure 4.20. In the vacuum scans, minimal counts are expected because the ADF detector inner collection angle is higher than the semi-convergence angle, meaning that counts in the digitised images are either stray electrons or noise being counted as electrons. Subtracting the number of counts in a vacuum image from the number of counts in an STO image for each applied threshold yields a similar difference graph to that

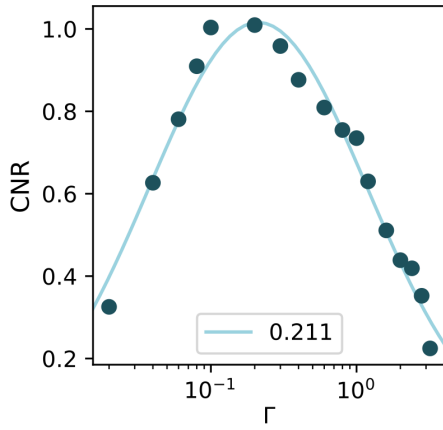


Figure 4.22: CNR between highly scattering Sr columns and weakly scattering O columns in experimental, digitised, multi-frame ADF images as a function of the threshold used for digitisation.

achieved when applying the test thresholds to the noise and signal streams in the automated threshold optimising algorithm, see Figure 4.20(b) and Figure 4.19. To investigate if a threshold around the estimated optimum around 0.1 – 0.2 indeed yields the highest quality image, the CNR in the experimental images from the threshold series has been evaluated, see the results in Figure 4.21. Here, CNR, defined as

$$\text{CNR} = \frac{|\mu_1 - \mu_2|}{\sqrt{\sigma_1^2 + \sigma_2^2}}, \quad (4.7)$$

is chosen rather than SNR since the former gives a much better idea of the quality of the final image. This is because it is challenging to clearly define “useful signal” in an experimental dataset without a ground truth. The parameters of Equation 4.7 are here μ_1 and σ_1 the mean and standard deviation of Sr columns and μ_2 and σ_2 the mean and standard deviation of O columns in the ADF images [169, 170]. Analysing the CNR of all the images from the experimental threshold series yields the points in Figure 4.22. Signal gradient streams and images from a subset of these points can be seen in Figure 4.21, where the green ticks in (a) indicate the digitised events with the applied threshold. Fitting a Gaussian to the CNR points in Figure 4.22 suggests that the optimal threshold to use in order to maximise CNR of these ADF images is around 0.211. Considering that the threshold optimisation algorithm found the optimal threshold to be 0.128, it is deemed to yield a successful estimate.

As alluded to at the beginning of this section, the determination of the optimal threshold to use for digitisation becomes significantly harder when using segmented detectors. This is mainly because of the probability of cross-talking, that is, electron detection events occurring in more than one segment. Another factor making digitisation harder with segmented detectors is that the signal level should not be changed in one segment at a time to a level that is acceptable for the signal digitiser. This is because it is still crucial that the segments are standardised, also before digitising the signal from them. After the gain and offset for the segments have been set, upholding the standardisation requirement, and the signal level checked

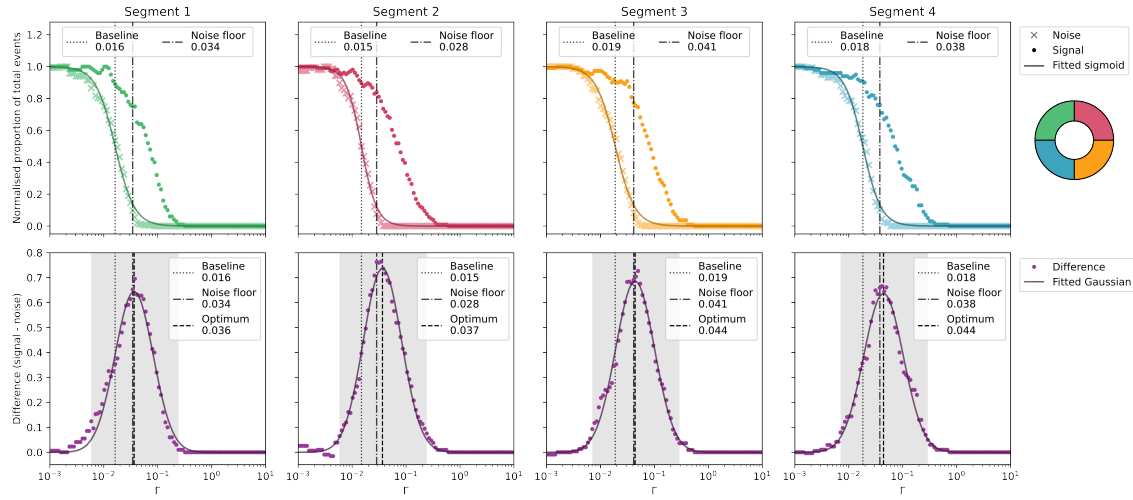


Figure 4.23: Illustration of the automatic finding of the optimal threshold for four segments on the SAAF detector. The noise streams and the signal streams are acquired for each segment separately, such that the optimal threshold is found independently for each segment.

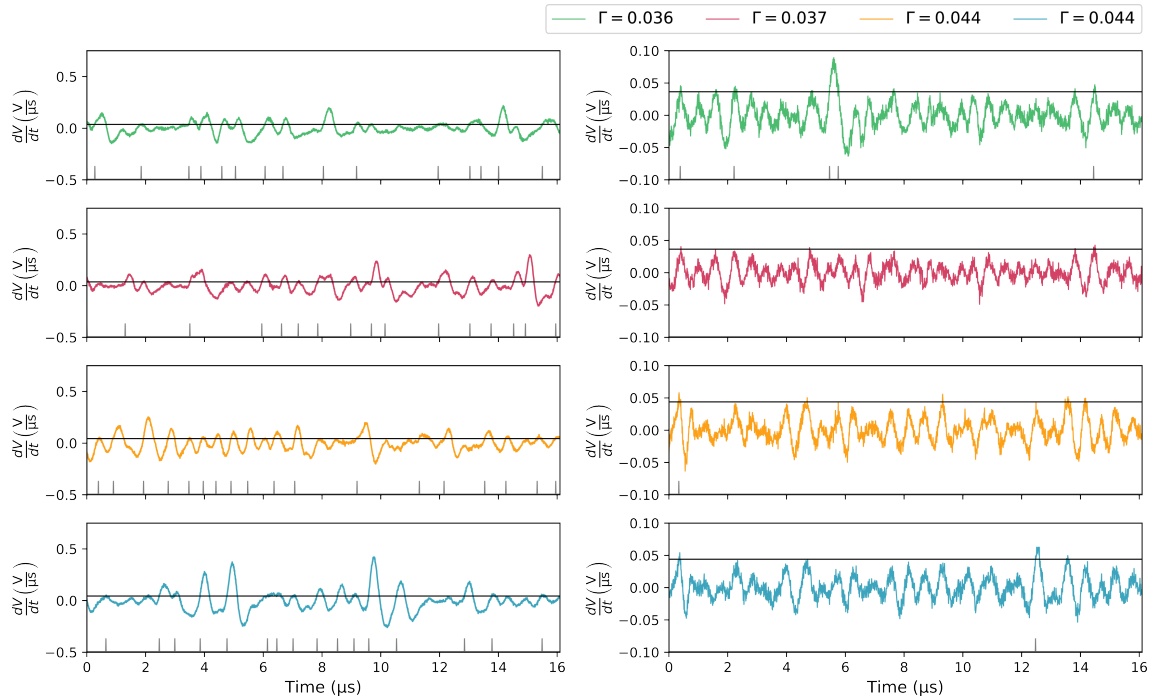


Figure 4.24: Subset of a signal stream (left) and a noise stream (right) from each of the four segments used from the SAAF detector.

in the control software for the digitisation hardware, the optimal threshold may be found similarly to that for the monolithic detector. Importantly, the noise and signal streams must be acquired for each individual segment. Then, the threshold optimisation algorithm may find the optimal threshold for each segment by following the same test thresholding procedure as explained previously. For the SAAF detector, this yields the graphs shown in Figure 4.23, which show that slightly different optimal thresholds are found for the individual segments. It is also worth noting that this detector is much noisier than the monolithic ADF detector, see the noisy single-segment image in Figure 4.8 and gradient streams from four segments in Figure 4.24. Comparing the gradient of the signal and noise streams from Figure 4.24 to the similar streams from the monolithic ADF detector in Figure 2.11 illustrates the difference in the detectors' noise levels and explains why the segmented detector is more challenging to digitise.

4.3 Conclusions

The finite response time of an electron detector impacts the integrity of the signal that may be recorded, not just for monolithic detectors, but also for segmented detectors. This, in turn, affects the quality and integrity of the iCOM phase retrieval practicable with segmented detectors at severely elevated scan speeds through an attenuation of the information transfer at higher spatial frequencies in the fast scan direction. Simulations show that digital images dodge this attenuation of the signal, and therefore provide more isotropically reliable iCOM images and transfer higher mean phase information than their analog equivalent. Similarly, experimental results show severe streaking artefacts in analog COM and iCOM images, with the expected attenuation at higher spatial frequencies in the fast scan direction. Digitisation provides, as expected, more isotropically reliable iCOM images, also at the highest scan speed with dwell time $\delta_t = 50$ ns.

The experimental digitisation results also highlight some key issues that stand in the way of unlocking the full potential of digitisation. Primarily, the positioning of the detector in the BF region means it is prone to coincidence loss — that is, missing electron event counts because electrons arrive too close in time to be separated into separate events. This may be somewhat remedied by dividing the detector into more segments, or spreading the beam over more segments on the detector, but this requires more digitisation channels in-hardware. Secondly, the choice of threshold for the digitisation process significantly impacts the resulting digitised images, and in turn, the phase-retrieved images from the segment images. Therefore, an automated procedure for determining the optimal threshold for a specific detector and experimental setup has been developed. This ensures that the number of false

positives and false negatives in the digitised signal are minimised simultaneously, whilst also removing human bias from the digitisation process by removing the need to set a threshold manually. Using the automatically found optimal threshold makes the digitisation of analog detector signals more reliable and reproducible.

Chapter 5

Low-dose and Temporal Resolution

Achieving truly low-dose conditions in STEM requires a holistic view of the whole microscope and experimental parameters. Many of these parameters and consequences have been introduced in the previous chapters, and possible solutions have been outlined. The primary avenue to reaching very low dose conditions is naturally to lower the beam current, typically by reducing the extraction voltage from the electron gun. There are other mechanisms through which the beam current delivered to the sample may be reduced, including reducing the amount of electrons that pass through a monochromator [171–173] and duty-cycling the electron beam exposure with an ultra-fast electrostatic beam blanker [174] — although these are mentioned with no further discussion here.

Another avenue is spacing out sample exposure to the beam in small bursts by acquiring multiple subsequent frames, as was introduced in section 2.3 and elaborated on in chapter 4 for both simulations and experiments. Not only has this multi-frame technique been shown to significantly delay and reduce sample damage [4], it also introduces a time dimension to the dataset, which may be exploited to characterise new aspects of materials in terms of dynamic development *in situ* either under stimuli or simply resulting from exposure to the beam. Additionally, this may help with characterisation of beam-sensitive materials since the onset of beam damage may be identified in post-processing of the data, and all the frames before the sample is damaged may be used for the analysis.

In this chapter, the temporal resolution is investigated for experimental iCOM reconstructions. Further, the use of denoising to aid interpretability of the iCOM images is investigated. Finally, we also see how iterative ptychography may allow for dose rate reduction via increased defocus and how that may give reduced dose via increased real-space step size.

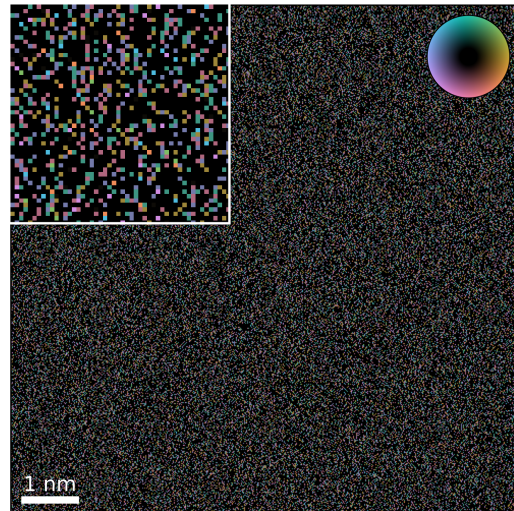


Figure 5.1: Ultra-low-dose single-frame COM image from a dataset acquired with a four-segmented detector, beam current 1.9 pA, and dwell time 200 ns. The inset shows a small part of the image at increased zoom to highlight the noise.

5.1 Data Analysis for Digital Data

Acquiring images with very low beam current and short dwell time means that each frame will have very few electrons; in most pixels, there are no electrons detected at all. An example of a single-segment, single-frame image can be seen in Figure 4.8 in the previous chapter. Even though the detector is placed in the high-fluence ABF region, digitised single-frame, single-segment images typically consist of 0s and 1s only, and there are typically no pixels with more than one event counted. Since the x - and y -components of the COM signals are calculated by subtracting opposing detector segments, as outlined in subsection 1.4.1, and subtraction is a noise-boosting operation, the resulting COM image for a single frame is typically very noisy. An example of such a COM image may be seen in Figure 5.1. As mentioned previously, calculation of the COM signal becomes especially challenging in the presence of cross-talk between detector segments, since a single electron event being accidentally doubly detected means that it gets lost in the subtraction if the two segments that detected it are S1 and S3, or S2 and S4¹.

As seen from the SNR calculations in section 3.4, iCOM struggles to retrieve the information at low spatial frequencies since the noise in iCOM phase retrieval is inversely proportional to spatial frequency. For noisy measurements, this weakness is exacerbated and low-dose iCOM images often suffer from more severe low spatial frequency artefacts than higher dose iCOM images. Thus, the extraction of information from a single frame image at low dose is often difficult. Since increasing the beam current will increase the amount of electron events that are lost to coincidence loss, while also being more damaging to the sample, that is not a viable route to increasing the signal. Acquiring multiple frames in quick succession is an alternative, and relatively easy, way to recover enough signal to get analysable images, which

¹We aptly termed this the $1 - 1 = 0$ -problem.

also helps to delay sample damage [4]. As previously mentioned, this also adds a time dimension to the dataset, which will be elaborated on in the following.

5.2 Achieving Time Resolution

By acquiring multiple fast frames consecutively, it is possible to gain enough signal to obtain a reliable iCOM reconstruction. Naturally, the more frames we sum, the higher the signal, which gives higher a PSNR in the iCOM image. Balancing the PSNR and need for the temporal resolution opens the route for characterisation of dynamic properties in *in situ* experiments with iCOM. Figure 5.2(a) shows the PSNR in iCOM reconstructions as a function of the time it took to acquire enough frames to get an iCOM image with that PSNR. This time takes into account the fly-back times, T_{LFB} and T_{FFB} , which are the time it takes for the beam to return to the start of the line and frame, respectively.

Both line fly-back and frame fly-back are unavoidable constant additions to the time it takes to acquire a multi-frame stack. The total frame time may be calculated by using Equation 2.5, reprinted here for convenience:

$$\text{Frame time} = \delta_t n_p n_L + T_{\text{LFB}} n_L + T_{\text{FFB}}.$$

The lower the dwell time used, the higher the proportion of the frame time that is fly-back time, meaning that the more the dwell time is lowered, the smaller the return in terms of increased temporal resolution is. Because of this, the iCOM

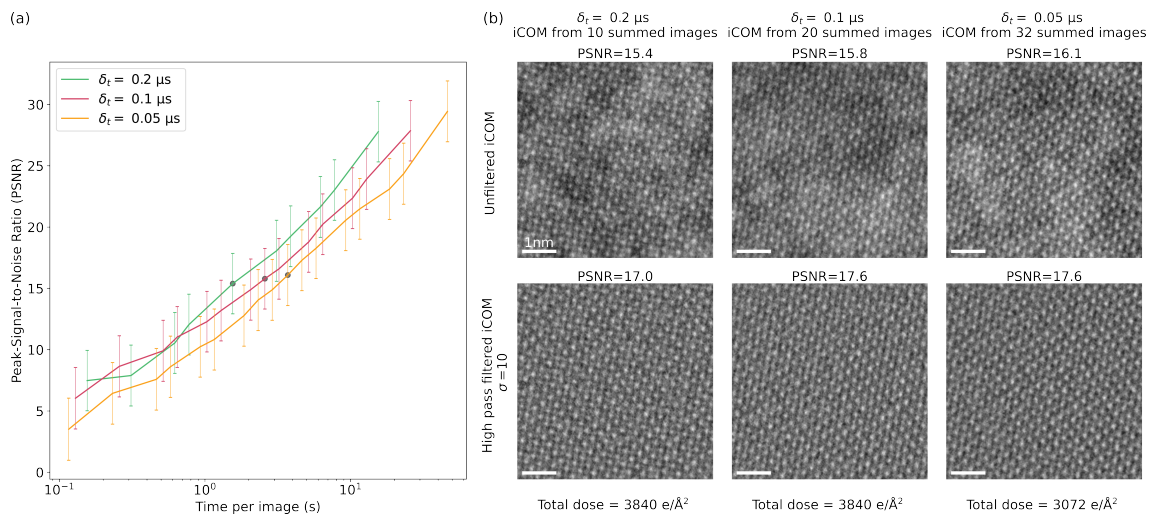


Figure 5.2: Balancing the temporal resolution and the PSNR of the image paves the way for *in situ* characterisation of dynamical processes in STEM. (a) shows the PSNR of experimental images as a function of the “time per image”, meaning the full time it took to acquire all the binned frames, calculated by Equation 2.5. (b) iCOM images from the dots in (a) at the three different dwell times. The beam current used in this experiment was $I = 5.2 \text{ pA}$. Figure adapted from [95].

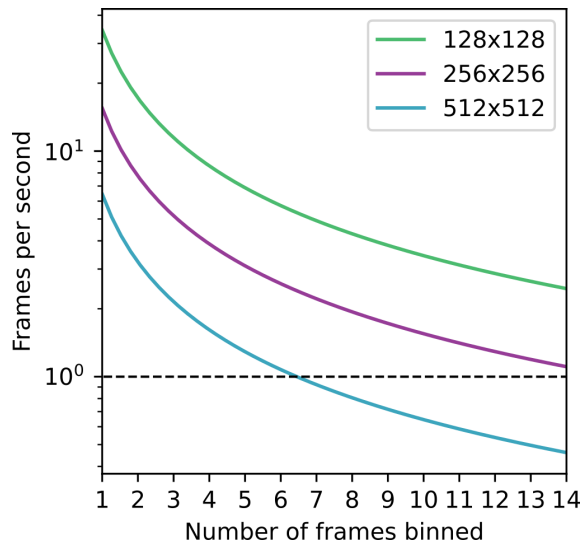


Figure 5.3: Achievable real-space frame rate with $\delta_t = 200$ ns and $T_{\text{LFB}} = T_{\text{FFB}} = 200$ μs as a function of how many frames are summed for three different scan sizes.

reconstructions from images acquired with the higher dwell time, $\delta_t = 200$ ns, in Figure 5.2 counterintuitively provide better temporal resolution — less time required to accumulate enough frames to gain higher PSNRs values — than the faster scans with $\delta_t = 100$ ns and $\delta_t = 50$ ns.

The graph in Figure 5.2(a) suggests that time resolution on the order of a few seconds is achievable for 512×512 -pixel images. At least if the images from 10 summed frames acquired with $\delta_t = 200$ ns from Figure 5.2(b) are acceptable. This means that we can characterise dynamics of samples where the change happens on the order of seconds for a real space frame with size 512×512 pixels [95]. Naturally, this could be significantly improved upon if the frame size is reduced or fewer frames needed to be summed, see the graphs in Figure 5.3.

Acquisition of 14 frames for a 512×512 -pixel image, with $\delta_t = 200$ ns and $T_{\text{LFB}} = T_{\text{FFB}} = 200$ μs , takes just over 2 seconds. Reducing the scan area to 256×256 reduces the acquisition time to less than a second, 0.9 seconds. Consequently, the FOV will be smaller, which is illustrated with the colour-coded boxes in Figure 5.4. As a result of the smaller FOV, it becomes increasingly important that the sample is physically stable to avoid large sample drift because this will reduce the size of the analysable FOV further after drift correction. The images shown in Figure 5.4 were acquired as 512×512 and reduced to 507×507 pixels after drift correction. Note, still, that the single-frame iCOM image, with dose $384 \text{ e}^-/\text{\AA}^2$, does exhibit visible atomic columns.

5.3 Lowering the Dose Further

The loss of electrons to coincidence loss at $I = 5.2$ pA is likely to be significant, as was seen at the end of subsection 4.2.1. Since the results so far seem to indicate that lower doses are achievable, a lower beam current of $I = 1.9$ pA is tested, see

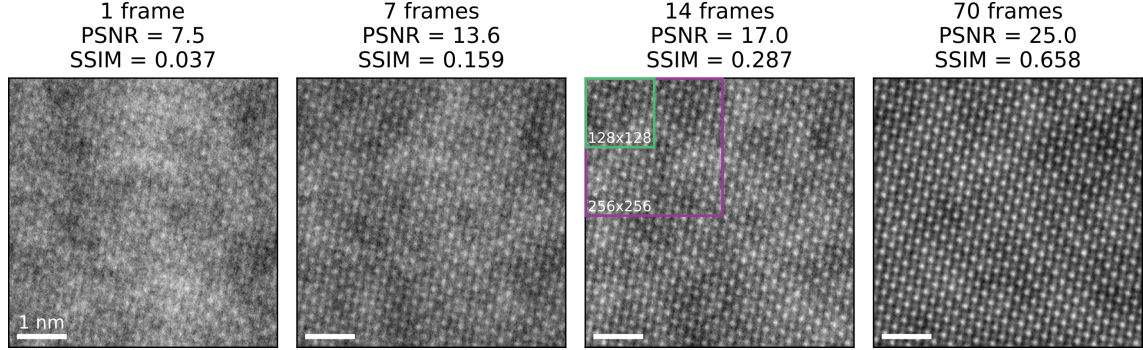


Figure 5.4: iCOM images from binning 1, 7, 14, and 70 frames before phase retrieval of 500×500 -pixel segment images. The data was acquired with 512×512 scan positions, beam current $I = 5.2$ pA and dwell time $\delta_t = 200$ ns, and a pixel size $dx = 0.13$ Å, which yields a dose of $384 \text{ e}^-/\text{Å}^2$ per frame.

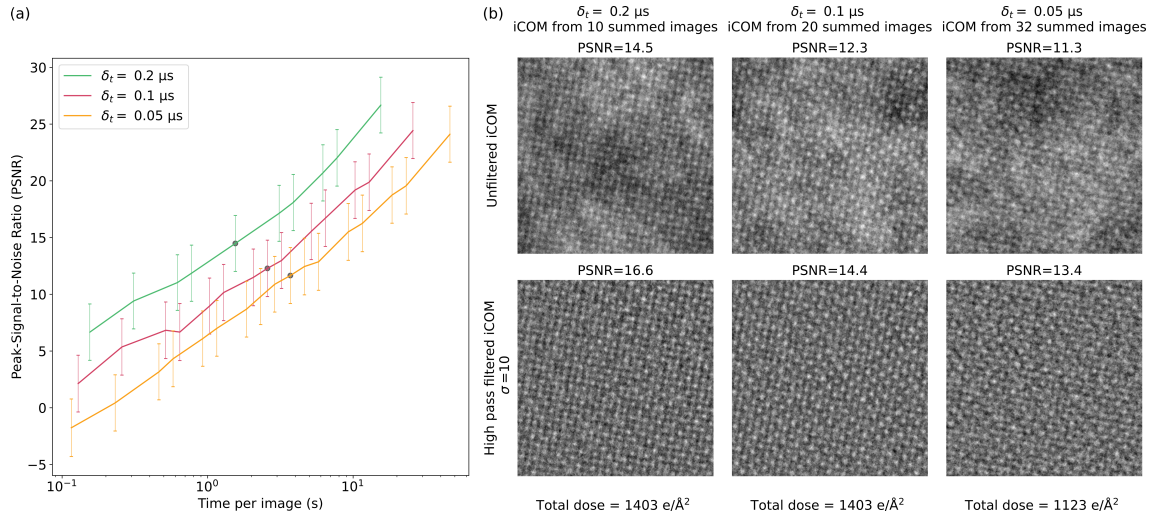


Figure 5.5: (a) Analysis of the time resolution and the achievable PSNR in experimental multi-frame dataset, like Figure 5.2 but with a lower beam current of $I = 1.9$ pA. Example iCOM images from the dots in (a) at the three different dwell times. Note that the number of frames that were summed for these images is the same as for the example images in Figure 5.2 to ease comparison between the two.

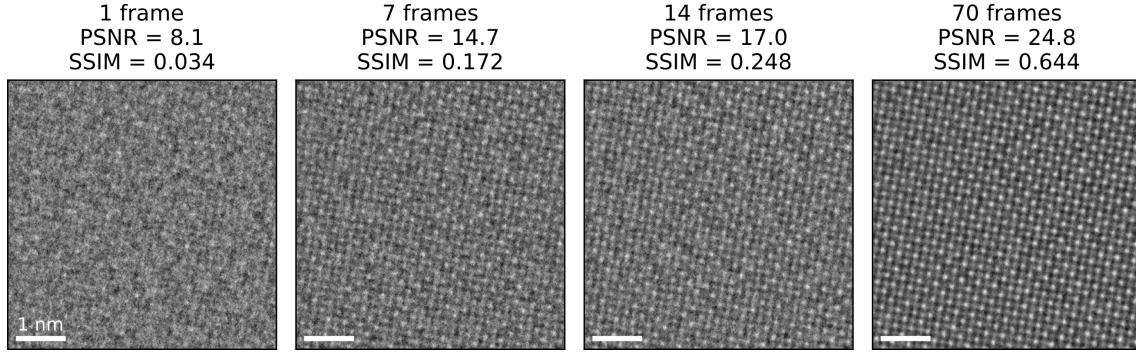


Figure 5.6: iCOM reconstructions from low-dose, digitised, experimental multi-frame dataset. The beam current used was $I = 1.9$ pA and dwell time $\delta_t = 200$ ns, which yields a dose of $140 \text{ e}^-/\text{\AA}^2$ per frame. The reconstructed images have been high-pass filtered with $\sigma = 12$ pixels, which is equivalent to $1.56 \text{ \AA}$ here.

PSNR as a function of time per image in Figure 5.5. Unsurprisingly, the achieved PSNR values are lower, especially for the faster scans with dwell times $\delta_t = 100$ ns and $\delta_t = 50$ ns. However, atomic columns are clearly visible in the iCOM image from the sum of 10 frames for $\delta_t = 200$ ns. This image has a slightly lower PSNR than the equivalent from the $I = 5.2$ pA dataset, but does not appear significantly worse. This is especially promising considering that the dose delivered to the sample is less than half of the dose in Figure 5.2, where the beam current used was $I = 5.2$ pA. The doses in the lower and higher beam current datasets are $1403 \text{ e}^-/\text{\AA}^2$ and $3840 \text{ e}^-/\text{\AA}^2$ respectively.

Note, however, that the lower dose causes the iCOM reconstructions to have more low-spatial frequency artefacts, which are typically removed with a high-pass filter. Such a high-pass filtered version of iCOM images from a sum of 1, 7, 14, and 70 frames may be seen in Figure 5.6. From these, it is evident that the single-frame reconstructed image is unusable, and the reconstruction from 7 summed frames is insufficient. Still, at this dose, summing 14 frames starts yielding analysable images where atomic column positions could be more reliably extracted.

In the following, denoising will be introduced to enhance the interpretability of these low-dose images.

5.3.1 Denoising Digital Data

As was introduced in the previous chapter, the digitisation of signals from detectors reduces the impact of inherent noise in the detectors on the images by removing the background noise and the effects of the finite response time to an electron detection event. This produces digital images with a true-zero noise floor and pure Poisson noise, which contains more information per signal because other sources of noise are removed. However, the resulting images appear very noisy. A single frame image from one segment with both the analog and digitised signal was seen in Figure 4.8,

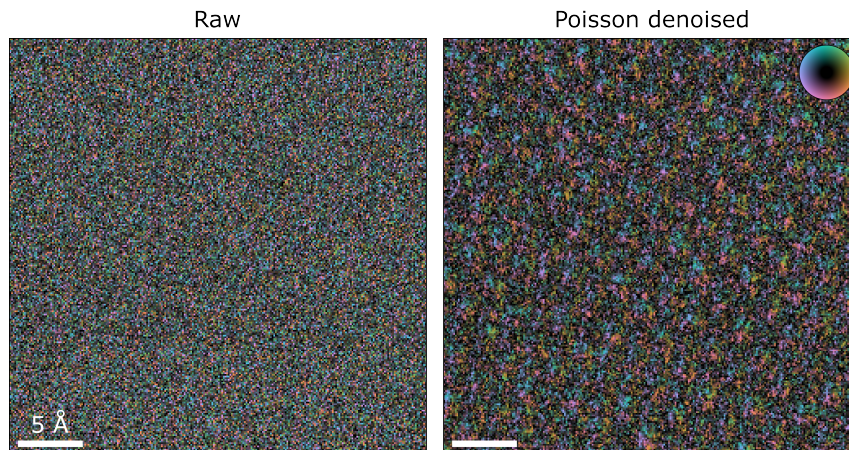


Figure 5.7: Comparison of the COM image from summing 21 frames of raw and denoised segment data before calculating the COM signal. 3 denoised frames from the denoised dataset were summed, because each frame in the denoised dataset is a denoised version of a 7-frame average; $3 \cdot 7 = 21$ frames. Note how the denoising makes the atomic columns more easily visible. The experimental beam current was $I = 1.9$ pA and dwell time $\delta_t = 200$ ns.

which illustrates how much intrinsic noise a detector can have, and also how most of the pixels in the digitised dataset are zeros.

The finite detector response function, see Figure 2.10, acts as a smoothing function in time on the detector signal. As a result, analog images typically appear less noisy and more visually smooth. Still, as seen in previous chapters, the detector response function may also lead to information loss — especially at high scan speeds. Conversely, the pure Poisson noise in the digitised images appears very noisy, but the digitised images may be made more visually pleasing, and thus more easily analysed, by denoising.

Denoising digital data requires summing enough frames for denoising to be sensible, which for the data presented here was deemed to be 7 frames. The denoising algorithm used is developed by researchers at the University of Tokyo, and takes the Poisson distributed nature of the digitised electron events into account [175]. The denoising was applied to a rolling average of 7 drift-corrected frames to produce a denoised dataset. Summing 3 denoised frames, making sure to avoid summing the same frame multiple times by extracting every 7th frame from the denoised dataset, yields the Poisson denoised COM image shown in Figure 5.7. The denoising clearly improves the visibility of the atomic columns from what is seen in the COM image from the raw data, highlighting the power of denoising for making images more visually analysable. When the COM images are less noisy, the iCOM reconstructions also become better. This is observed from comparing the high-pass filtered iCOM reconstructions from raw data to the ones from denoised data, see Figure 5.8 or compare Figure 5.9 to Figure 5.6.

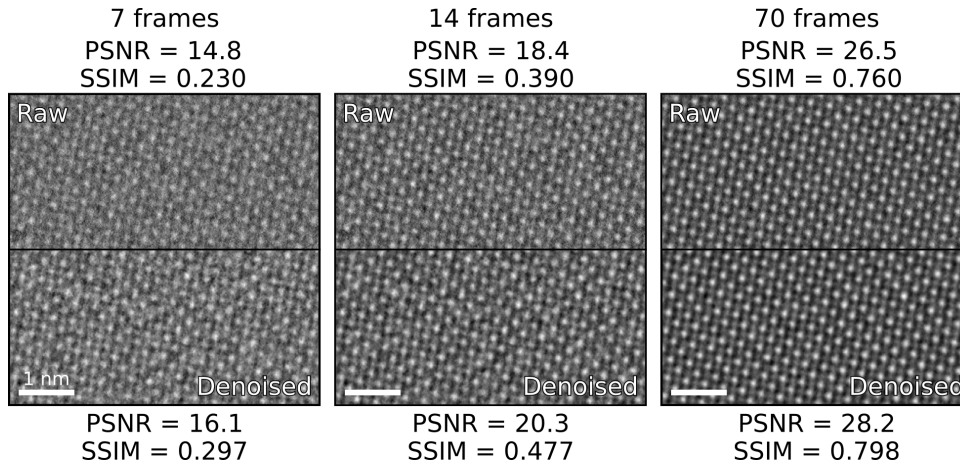


Figure 5.8: Denoised and high-pass filtered ($\sigma = 12$ pixels) iCOM reconstructions from raw and denoised data. Digitised data acquired with beam current $I = 5.2$ pA and dwell time $\delta_t = 200$ ns, which yields a dose of $384 \text{ e}^-/\text{\AA}^2$ per frame.

To ensure that a fair comparison is being made between reconstructions from raw and denoised data, these figures show two image quality metrics: peak signal-to-noise ratio (PSNR) and structural similarity index (SSIM). Where PSNR gives a good indication of how noisy the tested image appears in comparison with the reference image, SSIM provides a more complete comparison of the tested image with the reference image by taking both the image luminance, contrast, and structure into account [176, 177]. This places more emphasis on how close the tested image resembles the reference image in structure than what PSNR does. The SSIM is a number between 0 and 1, where 0 is completely different images and 1 indicates perfectly correlated images — an image will always have $\text{SSIM} = 1$ with itself. As a result of checking both metrics, the quality of the iCOM reconstructions from denoised and raw segment data may be compared more fairly.

In Figure 5.10, the quality metrics of iCOM reconstructions from raw and denoised data, acquired with $I = 5.2$ pA, are plotted as a function of the number of frames summed. From this, it is evident that denoising yields higher quality reconstructions, even when 70 frames are summed before reconstructing. This is interesting because, since integration is a noise-suppressing operation, it might be expected that denoising will have a less pronounced effect when many frames are summed, but this is not seen to be the case. Investigating the resulting iCOM images directly, see Figure 5.8, elucidates that the denoising yields more easily interpretable images through subtly improving the smoothness of the reconstructed image.

For the images with even lower dose, the denoising helps make the lowest dose images, the sum of only 7 frames, significantly more interpretable. This is seen from comparing Figure 5.6 and Figure 5.9. It is, however, important to note that denoising only helps the visual interpretability and analysability of images and does not help bring out any more information.

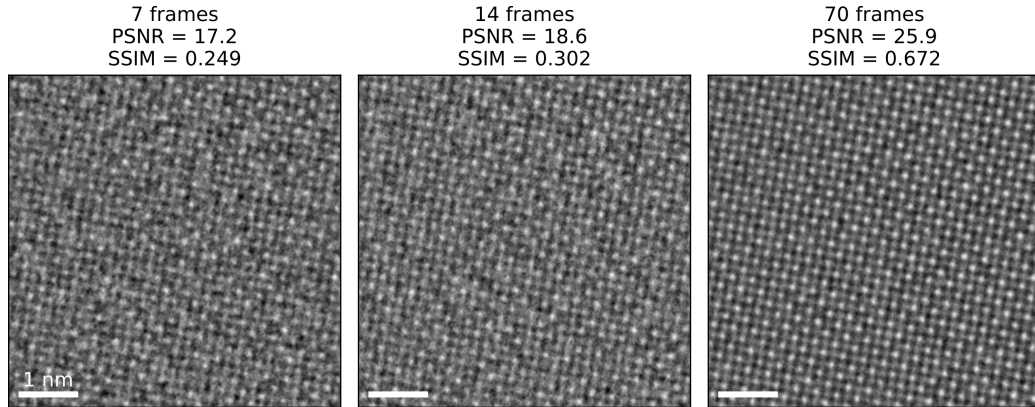


Figure 5.9: High-pass filtered iCOM reconstructions from the denoised dataset. Note how these images are smoother and more easily interpretable than the raw equivalents in Figure 5.6. The experimental data were acquired with beam current $I = 1.9$ pA and dwell time $\delta_t = 200$ ns.

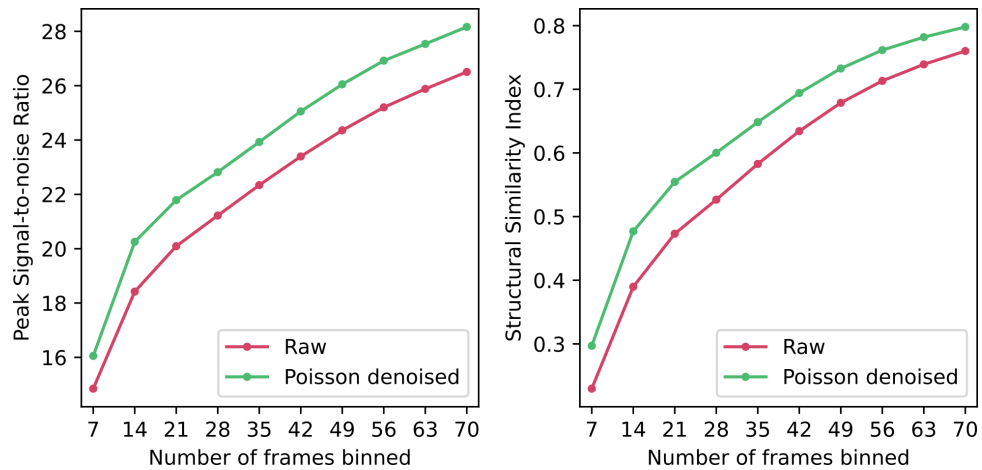


Figure 5.10: Comparing the quality of high-pass filtered iCOM reconstructions from raw and denoised datasets using both metrics, PSNR and SSIM. The denoised images are of better quality overall, and the benefit of denoising is not limited to the lower number of summed frames. Examples of iCOM images may be seen in Figure 5.8.

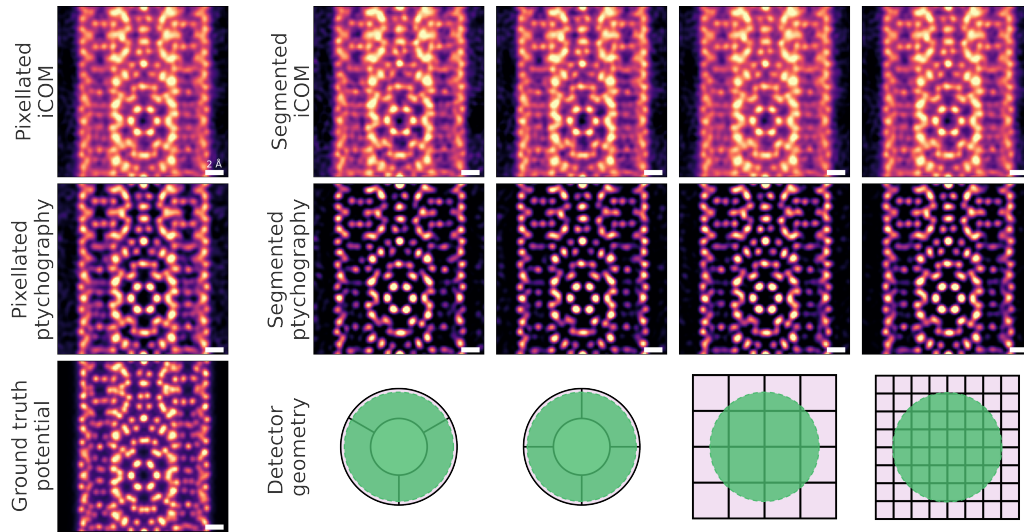


Figure 5.11: In-focus iCOM and ptychographic reconstructions of a double-walled carbon nanotube, at finite dose $5 \cdot 10^4 \text{ e}^-/\text{\AA}^2$ using four different detector geometries. Simulated with 200 keV and $\alpha = 25 \text{ mrad}$. The green circle on the detector illustrations indicate the BF disk to show the collection angles used.

5.4 Ptychography with Simple Detector Geometries

One of the main benefits of iCOM over ptychography is the simple reconstruction algorithm, allowing for live processing at the microscope. Although live iterative ptychography is an active and interesting area of research [67], the strength of ptychography lies in its ability to recover all spatial frequencies equally well in the ideal case [178], giving it super-resolution capabilities for high electron doses [98, 161]. Both in-focus and out-of-focus ptychography are routinely done with pixelated detectors, but the CTF investigations in section 3.4 alluded to the usability of segmented and pixel array detectors for iterative ptychography. This was co-developed by the author and contributed to the open-source analysis software py4DSTEM [65, 178, 179].

Figure 5.11 shows a comparison between the iCOM and iterative ptychographic reconstructions of a simulated double-walled carbon nanotube at a relatively high dose of $5 \cdot 10^4 \text{ e}^-/\text{\AA}^2$ when four different simple detector geometries were used as virtual detectors. These simulated results show that iterative ptychographic reconstructions are possible with very simple detector geometries for in-focus data. This means that the segmented detectors that have previously been thought of as “DPC detectors” can in reality also be used for iterative ptychography — and more broadly for phase characterisation in general [158].

In this section, the performance of iCOM and iterative ptychographic phase retrieval techniques will be investigated for several different experimental parameters and settings. The main focus is on dose efficiency and beam damage reduction.

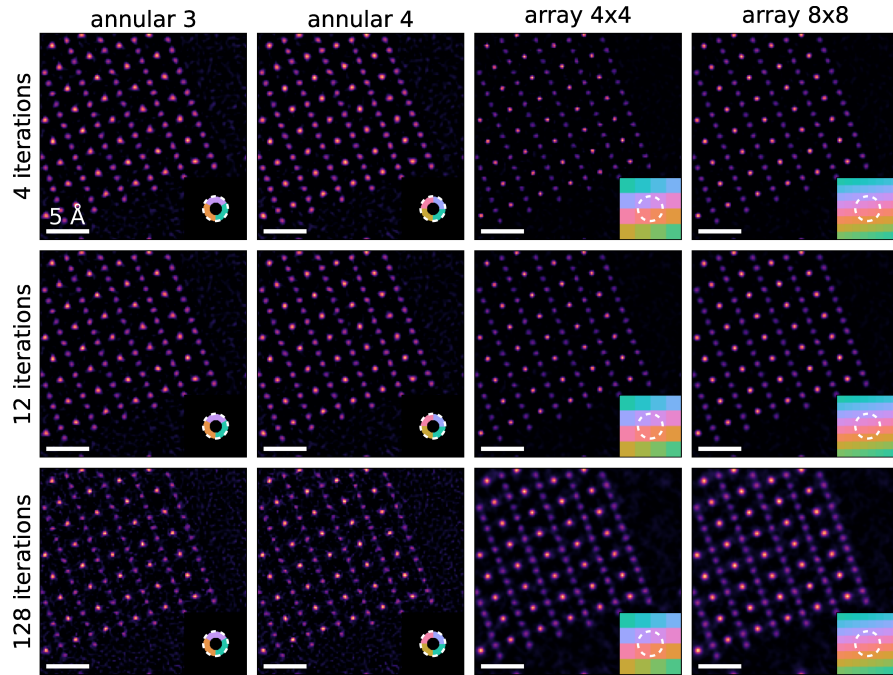


Figure 5.12: Iterative ptychographic reconstructions of $[110]$ STO with simple detector geometries using an increasing number of iterations. Notice how the lower spatial frequencies are the slowest to converge. The inset indicates the detector geometry, and the dotted white circle shows the edge of the BF disk.

5.4.1 Investigations for In-focus Conditions

Iterative ptychography can reach extremely high information transfer [98, 158, 161], but this is dependent on letting the reconstruction converge fully. As a result, ptychographic reconstructions can take hours² to get right and to achieve the highest possible information limit [180]. Even in simulation, a higher number of iterations gives better reconstructions. This is illustrated in Figure 5.12, where the reconstruction of a $[110]$ STO sample at dose $2000 \text{ e}^-/\text{\AA}^2$ is done with 4, 12, and 128 iterations. Notably, the algorithm struggles to reconstruct the object properly with simple detector geometries at this dose, because the gradient steps are fixed such that the algorithm may overshoot and inject noise into the reconstructed object. This is seen as the added noise in the vacuum areas of Figure 5.12. Note also that the array detectors in Figure 5.12 cover a significant amount of area in the dark field region. This is because sampling the dark field has proved to be helpful for reconstructing the lower spatial frequencies. However, it comes at the cost of a loss of resolution within the BF disk since fewer of the detector pixels are then in the BF region.

Naturally, at high scan speeds, the streaking artefacts appearing in segment images will also disturb the ptychographic reconstruction, similar to what was seen for iCOM reconstructions in section 4.1. Preliminary simulations of streaked signal in each of the detector pixels of a 4×4 pixel array detector show that the pty-

²Days if you count the researcher's time to tune the hyper-parameters for the reconstruction.

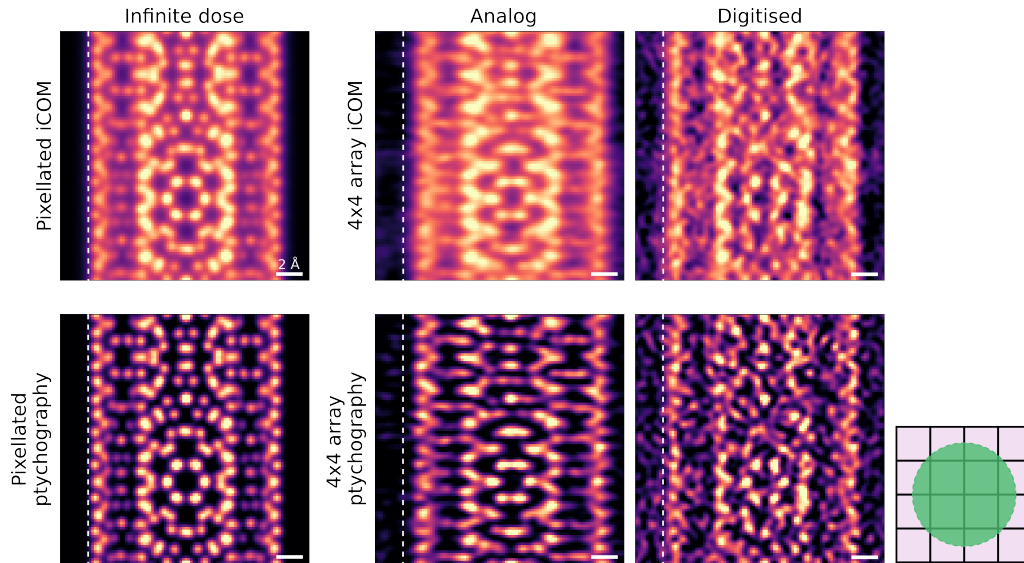


Figure 5.13: Preliminary simulation of the effect of streaking artefacts on iCOM and iterative Ptychographic phase reconstructions from a 4×4 pixel array detector. A beam current of 3.2 pA and dwell time of 100 ns, which yields a decent dose of $\sim 24000 \text{ e}^-/\text{\AA}^2$, were used for the simulation of the detector array signals. The infinite-dose pixellated reconstructions are shown for reference.

chography algorithm can not resolve the streaking artefacts any better than iCOM could, see Figure 5.13. Notably, the same drifting of the atoms downstream in time as was seen earlier in this thesis is seen in the Ptychographic reconstruction from the analog signals. The vertical dotted line is at the same position in all the images of Figure 5.13 to help see the misplacement of the edge of the carbon nanotube in the reconstructions from the simulated analog signal. The reconstructions from the digitised signals suffer significantly from the noise of the segment signals, likely due to severe coincidence loss and a poor choice of threshold, which both exacerbate the noise. Additionally, as mentioned previously, the algorithm used here [179] struggles to reconstruct noisy signals from simple detector geometries and tends to inject the noise from segments into the object. Still, it is promising that the atoms appear at the correct place in the digitised simulation reconstructions, as seen with the dotted white line. Thus, with improvements being made to both the digitisation parameters and the Ptychographic reconstruction algorithm, this holds great promise for the expansion of the usability of iterative Ptychography.

5.4.2 Defocused Probe Reconstructions

One of the main concerns about STEM for scientists working with truly beam-sensitive materials is the high dose rate that comes with focusing the beam to a very fine point. In contrast to HRTEM, where the dose is delivered across the whole FOV continuously, the beam in STEM is focused to a probe such that it delivers all the beam fluence to each scan position sequentially. Naturally, this is cause

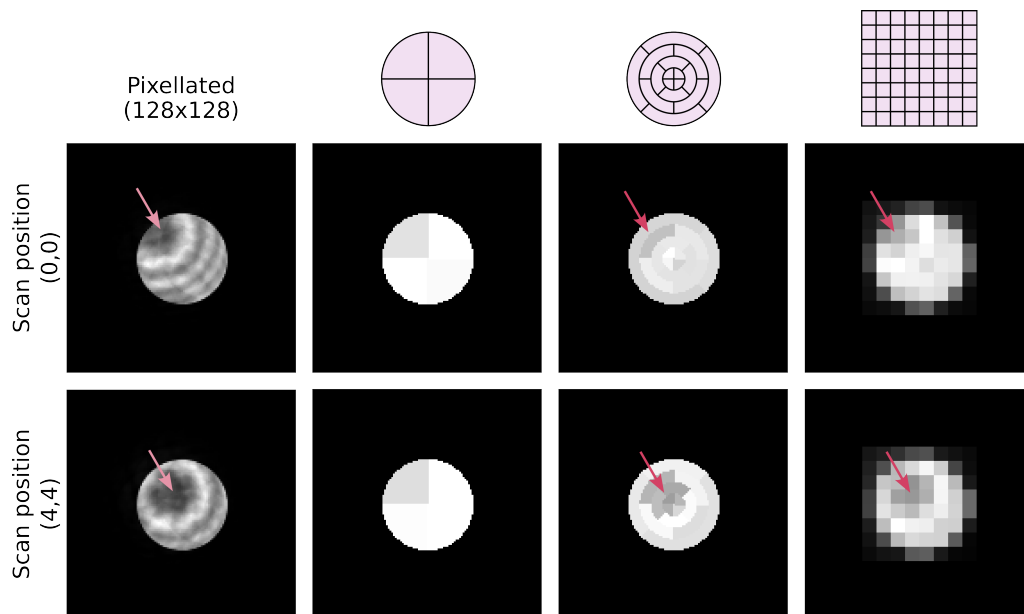


Figure 5.14: Defocused diffraction patterns as seen by a pixellated detector and three different detector geometries. Arrows indicate a change in the position of the feature seen in the pixellated CBED pattern, which is visible in the binned diffraction patterns for the 16-segment and 8×8 pixel array detectors.

for concern for samples that damage at very low doses. Additionally, when using extremely low doses, the detection system and imaging method have to optimise the information extraction from every single electron.

The CTF investigations in section 3.4 showed that ptychography works with, and typically benefits from, a defocused probe — which is also well-known from the literature [178, 181]. With increased defocus, the features in the diffraction patterns become smaller, which requires more detector pixels to properly resolve them. As a rule of thumb, the iterative ptychographic algorithm will be able to successfully reconstruct the object if features are traceable from one scan position to the next by eye in the detected diffraction pattern. In Figure 5.14, the CBED patterns as “seen” by a pixellated and three different simple detector geometries for two close scan positions are shown. This illustrates how it is hard to discern the two scan positions using a four-segmented disk detector. On the other hand, the 8×8 pixel array detector and the 16-segmented detector with rotational offset between the rings are undoubtedly able to track the motion of the indicated feature. This is reflected in the way these detectors with more detector elements can reconstruct the object from datasets with higher defocus, as shown in Figure 5.15. Interestingly, these line profiles also seem to indicate that there is little difference between an annular 16-segment detector with rotational offset between the rings and a 4×4 pixel array detector — all the while they both have 16 total detector elements. For simulations of how segmented and pixel array detectors handle other probe aberrations, like C_S and three-fold astigmatism, see Appendix B.

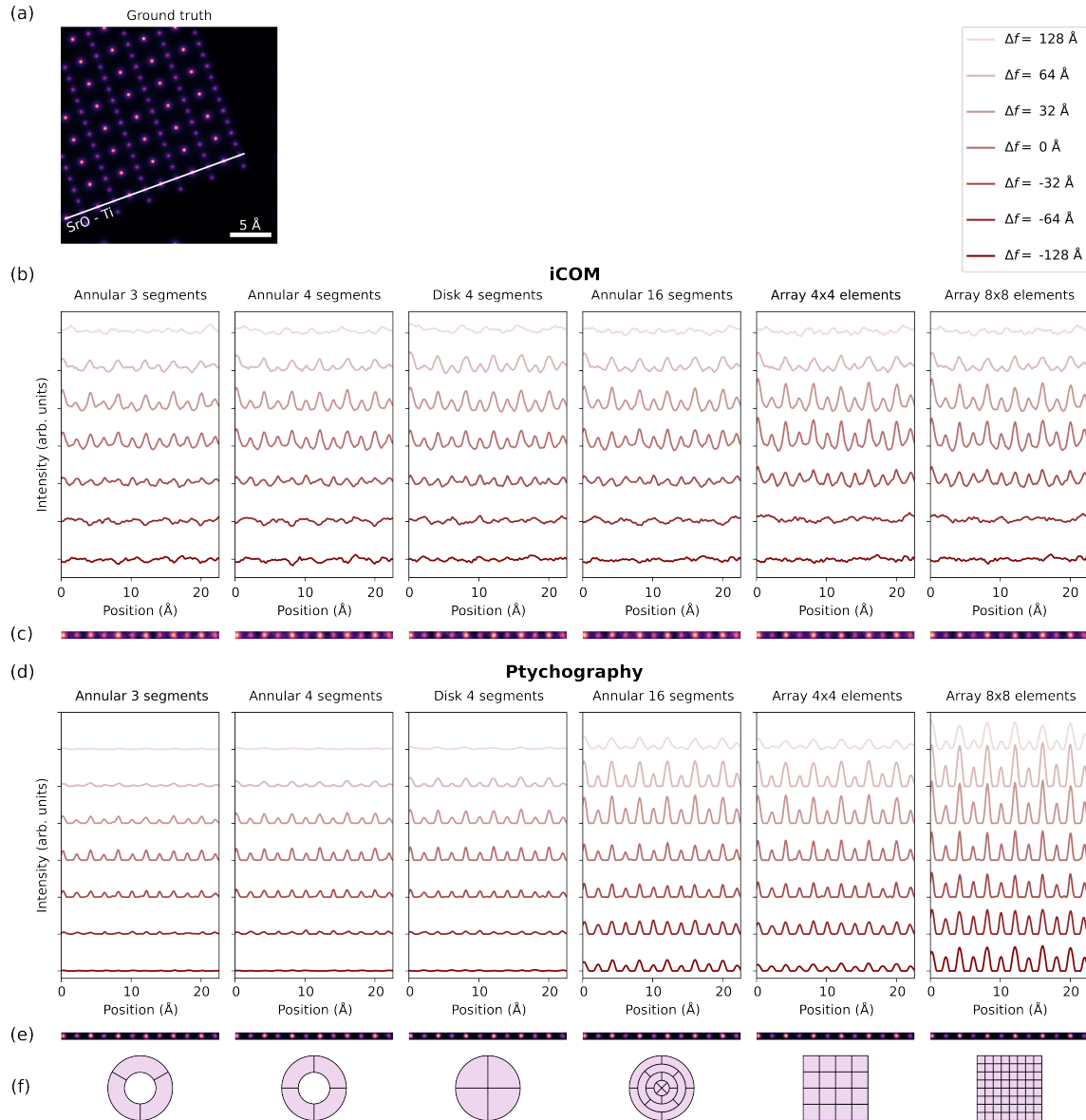


Figure 5.15: Line profiles through SrO–Ti columns in reconstructed phase images from finite dose ($2000 \text{ e}^-/\text{\AA}^2$) simulations with various defoci. The zero defocus point is taken to be the middle of the sample. Defocus helps the reconstructions for ptychography, provided that the detector has enough elements to decipher them.

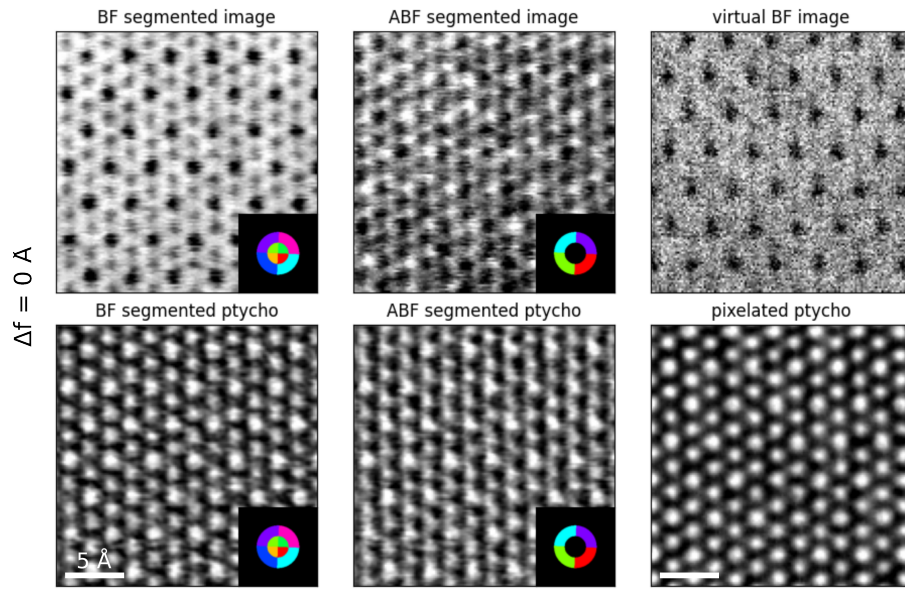


Figure 5.16: Experimental image from data acquired with an in-focus probe, from preliminary experiments. Top row: virtual BF and ABF calculated by summing the segments shown in the inset for the segmented detector, and the full BF disk for the pixelated data. Bottom row: iterative ptychographic reconstructions from segmented detector and pixelated detector as indicated. With no applied defocus, the ptychographic reconstructions are successful for both segmented and pixelated detectors.

Experimental Observations

To confirm the usability of segmented detectors for iterative ptychography, some preliminary experiments were done at the TFS Spectra 300 (S)TEM at Stanford University. Data was collected with eight segments on an annular segmented Panther detector and 192×192 pixels on a pixelated Arina detector. Figure 5.16 shows the successful ptychographic reconstructions of a 2D material sample of mechanically exfoliated PtSe_2 using data from the segmented and pixelated detector at in-focus conditions. The semi-convergence angle was $\alpha = 28$ mrad, and the collection angles for the two rings on the segmented detector were $0 - 15$ mrad and $15 - 30$ mrad, respectively. Note that these data are from preliminary experiments and, as such, were acquired with beam current $I = 45$ pA and dwell time $\delta_t = 50$ μs , which by no means yields a low dose. Still, these reconstructions confirm that iterative ptychography may be done using segmented detectors at in-focus conditions.

A similar reconstruction for a defocused dataset, with $\Delta f = -150$ Å, can be seen in Figure 5.17. This shows that the information in the segmented detector dataset is not sufficient to give a successful ptychographic reconstruction at this defocus. On the other hand, the pixelated data reconstructs well due to the higher sampling of the CBED pattern. Reconstructions from datasets at other defocus values may be found in Appendix B.

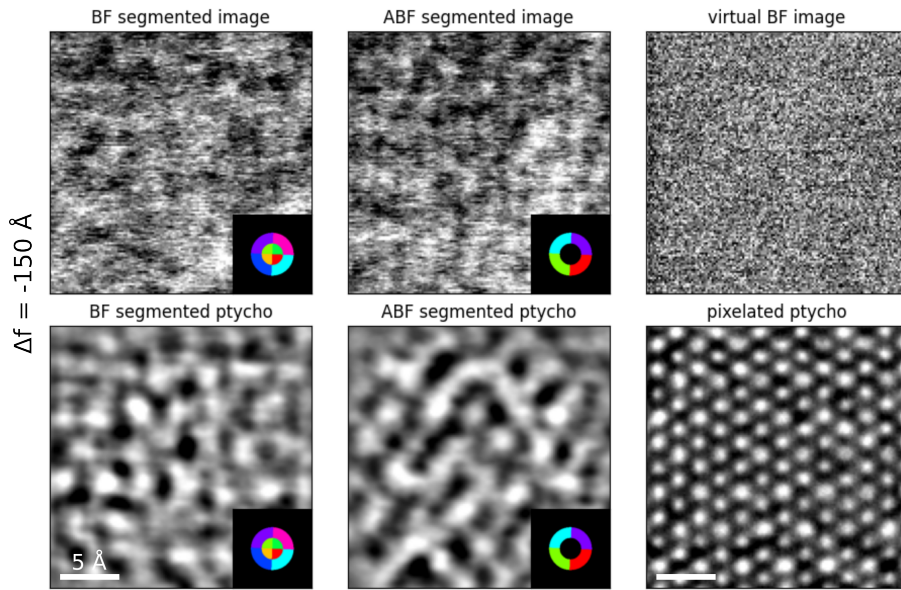


Figure 5.17: BF/ABF image and iterative ptychographic reconstructions from experimental data acquired with defocus $\Delta f = -150$ Å. Note how the ptychographic reconstruction fails for the segmented detector data at this defocus, while the pixelated detector data still reconstructs well.

5.4.3 Low Dose Iterative Ptychography

Combining all the knowledge gained so far, three significant efforts may be made to accommodate the most beam-sensitive samples. Firstly, scanning fast over the region of interest (ROI) and multi-framing, such that the dose is delivered in small flashes with significant breaks. This has been shown to reduce sample damage significantly [4]. Additionally, this allows for the subsequent determination of where in time the sample begins to damage by averaging over a subset of the frames acquired. This requires a non-pixelated detector with minimal read-out overhead.

Secondly, the beam may be defocused, spreading the electron dose over a larger area at each scan step. This requires a detector geometry and a phase reconstruction algorithm capable of decoupling the defocused probe and the object phase. As seen in Figure 5.15, ptychography works significantly better with defocus than iCOM, as was already known from the literature. Defocused probes are typically used for ptychography and help the iterative reconstruction algorithms to converge faster. Importantly, a fully pixelated (128×128) detector is not necessary to utilise this attribute of ptychography: the simulations shown in Figure 5.15 suggest that 16 detector elements may be sufficient. Figure 5.15 further shows that increasing the detector array to 8×8 pixels yields a significant improvement in the reconstructions, especially at higher defocus values. This suggests that there is a middle-ground where you can get the temporal resolution that pixelated detectors are unable to achieve due to the significant read-out overhead and, simultaneously,

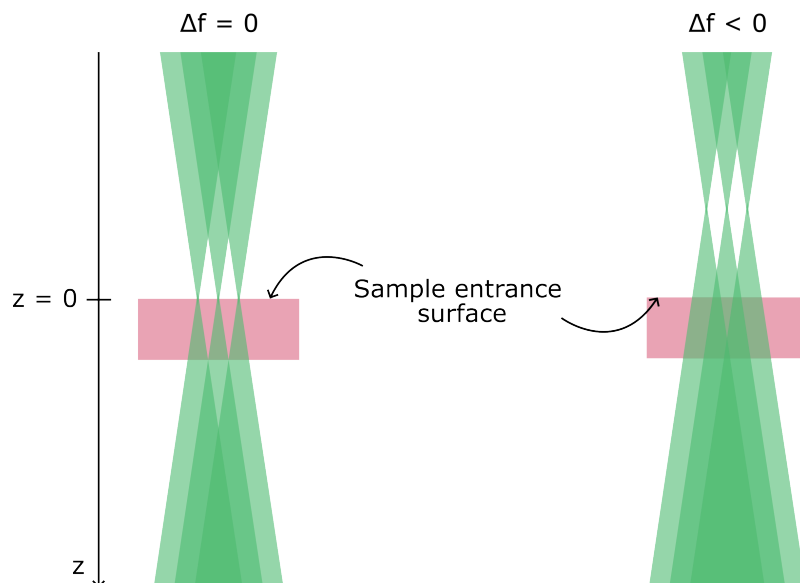


Figure 5.18: Illustration of how a defocused probe allows for a larger step size between scan positions since the size of the probe on the sample surface will overlap, which introduces more redundancy in the dataset.

the successful decoupling of a defocused probe and the phase object, using a detector array. Together, these give beam damage reductions in two ways at once — by multi-framing with fast scanning, and using a defocused probe.

Finally, where iCOM imposes very strict sampling requirements, iterative ptychography is more lenient and may often be able to successfully reconstruct heavily undersampled datasets. As long as there is sufficient information in the dataset for the reconstruction algorithm to deconvolve the probe and object phase information, the object may be reconstructed, even if the scan step frequency is lower than Nyquist sampling. This especially manifests when using a defocused probe, since adjacent scan positions will have overlapping regions of their probes, leading to more redundant information in the dataset, see illustration in Figure 5.18. In short, this allows for the scan step size to be increased, and as a consequence, the dose is reduced further. Combining high defocus with a large step size reduces the fluence of electrons delivered to the sample surface and the total dose delivered because the probe covers more area at a time, and fewer probe positions are required to image the ROI. Ptychographic reconstructions from simulated data with increasing scan step size of a [110] STO sample when three different detector geometries were applied as virtual detectors can be seen in Figure 5.19. These show that the more elements a detector has, the more resilient it is to the loss of information in the dataset with increased step sizes.

These techniques, all combined, show promise for the characterisation of beam-sensitive materials using iterative ptychography. However, low-dose significantly reduces the redundancy of a dataset, which can fast lead to the information content

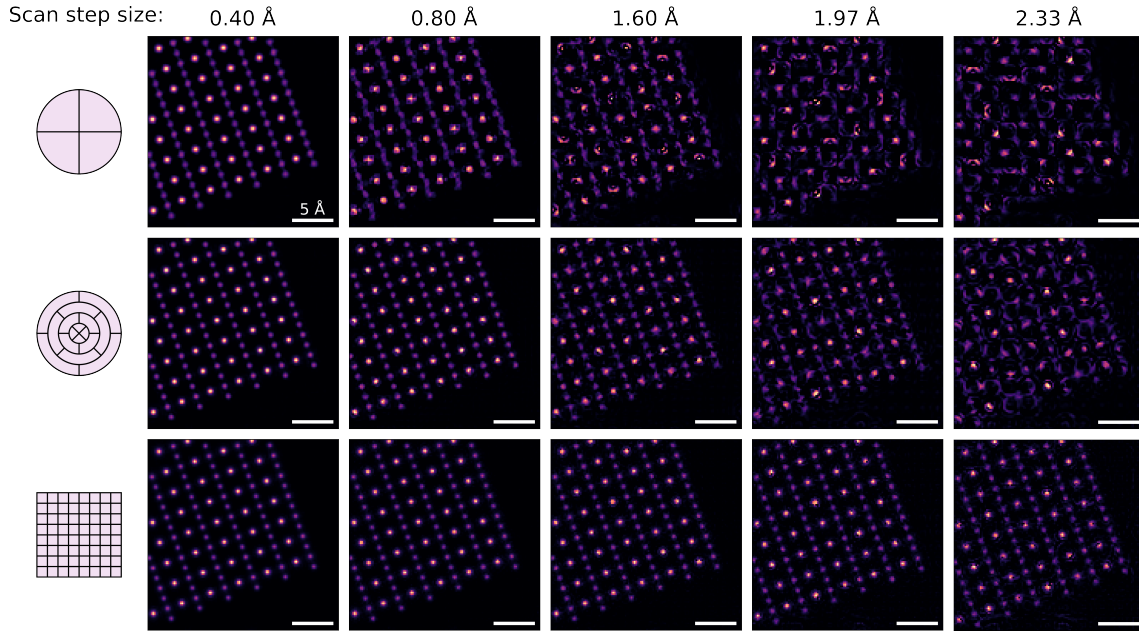


Figure 5.19: Iterative ptychographic reconstructions of a simulated [110] STO sample with three different detector geometries and increasing scan step size. This shows how more elements in the detector is more robust when the redundancy in the dataset is reduced.

in the data being too low for the reconstruction to converge successfully. Using simple detector geometries further reduces the redundancy of the dataset, which adds to this difficulty. While ptychography with simple geometry detectors is showing very promising results so far [158, 161], it suffers significantly with reduced dose. Algorithm development is still ongoing to elucidate issues where the noise is boosted along the detector geometry with an increased number of iterations for low-dose reconstructions.

5.5 Conclusions

Acquiring a stack of many consecutive frames with very high scan speeds allows for the use of low enough beam current to be digitisable whilst recovering a high enough signal level for phase reconstruction. Additionally, this helps delay beam damage on the sample and allows for analysis of frames prior to the onset of beam damage. Furthermore, this adds a time dimension to the dataset, giving the opportunity to sum a subset of the frames in the stack to observe dynamic changes in the sample. In this chapter the achievable temporal resolution with 512×512 real-space scan positions was found to be on the order of seconds, with significant room for improvement via scanning a smaller ROI and/or using a detector with a sharper onset of each electron detection event to ease digitisation and thereby reduce the effects of coincidence loss. Denoising the digitised images was seen to improve the interpretability of reconstructions from summing very few frames.

For ptychography, the usability of segmented detectors was demonstrated for both in-focus and defocused datasets, although the amount of defocus that may be handled by simple detector geometries is limited. The simplified detector geometry reduces the redundancy in the dataset, and combining that with further redundancy reduction via lowering the dose makes successful reconstruction difficult. Still, there is likely an optimal middle ground — perhaps around a 16×16 pixel array — where a balance between temporal resolution through low readout overhead, dose efficiency to accommodate the sample requirements, and information content for ptychographic algorithm convergence may be found. Whether that is low enough to accommodate the most beam-sensitive of samples is still an open question. It may very well seem like iCOM performs better, or at least more reliably, at ultra-low doses, even though it requires that data is acquired in focus. Importantly, there are also other flavours of ptychographic reconstruction — e.g. tilt-corrected BF-STEM and single side-band — which may very well be more suited for the most beam-sensitive samples, but these are outside the scope of the current work.

Chapter 6

Conclusions

This thesis aimed to investigate how segmented detector signals and real experimental effects can be better understood, simulated, and processed to push the boundaries of ultra-low-dose STEM phase retrieval. To achieve this, this thesis has explored how simulations and experimental analysis of both analog and digitised STEM signals can guide the optimisation of phase contrast and retrieval techniques at ultra-low doses. The central goal was understanding how realistic detection conditions impact phase retrieval performance, enabling more dose-efficient workflows for characterising beam-sensitive materials.

Phase retrieval methods in STEM have become increasingly popular in recent decades because they allow for the use of lower electron doses, thus enabling characterisation of materials not traditionally suited for STEM characterisation. This is because phase characterisation techniques offer superior collection efficiency by using the BF region of CBED patterns to extract information about the sample. This helps make STEM characterisation viable for beam-sensitive samples where the electron dose has to be significantly reduced, because detectors used to collect data for phase contrast and retrieval imaging have higher collection efficiency than traditional HAADF characterisation.

This thesis has investigated the use of segmented detectors, and other simple detector geometries, for efficient phase contrast and retrieval imaging in STEM. Throughout this work, it has been demonstrated that simulations incorporating realistic detector parameters, including geometry, temporal response, and coincidence loss, are crucial for understanding the practical limits of phase retrieval techniques at ultra-low-dose conditions. Importantly, simulation-informed parameter optimisation was shown to significantly enhance contrast transfer and reduce artefactual signal loss. Parameters optimised include camera length, from which the optimal outer collection angle for COM imaging was found to be $\sim 1.1\alpha$, probe defocus, and thresholding condition. This work confirmed through CTF investigations that deliberate probe defocus reduces contrast transfer for iCOM, but aids phase retrieval for iterative ptychography; conclusions which are in agreement with literature. The numerical CTF formalism introduced in this work provides an unambiguous platform

on which to compare different phase retrieval techniques and certain experimental parameters. From this, it was found that Scherzer defocus is sub-optimal for iCOM contrast transfer when the probe has significant spherical aberration, and a slightly lower defocus than Scherzer performs better.

The high arrival rate of electrons to the detectors in the BF region proves challenging from a signal digitisation perspective. Since digitisation requires a minimal time-separation between electron detection events, the beam current usable in digitisation experiments is heavily reduced from normal STEM beam currents. This work has presented an algorithm for finding the optimal threshold to use for digitisation which uses a comparison of the number of events in digitised noise- and signal-streams. The algorithm ensures that the number of false positives and false negatives are both minimised. Additionally, a method for determination of the severity of coincidence loss given an experimental detector response function has been presented. This may be used to inform the maximum practicable beam current for a specific detector response and geometry. Naturally, lowered beam currents are necessary for low-dose STEM imaging of beam sensitive materials and thus digitisation of the analog PMT voltage signal from scintillator-based detectors help reduce the effects of noise from other sources than the low-dose Poisson distribution of electrons. Experimental results have confirmed that digitised signal from segmented detectors can provide reliable phase imaging at low doses and ultra-fast scan speeds by retaining precision of electron arrivals to the detector. As a result, image series with practical real-space time-resolution of a few seconds for a 512×512 pixel scan area was achieved via multi-framing. This both delays sample damage and allows for determination of the onset of the sample damaging in post-processing. Importantly, the segmentation of the detector and the high arrival-rate of electron in the BF region makes digitisation challenging, and this thesis has investigated ways to mitigate and minimise the effects of these challenges.

Finally, the use of segmented detectors for iterative ptychographic reconstruction has shown promising preliminary results both for simulations and experimental data. While the benefit of converging faster at higher defocus, which is seen for pixellated iterative ptychography, is lost when using a four-segmented detector, the potential use of a 16×16 pixel array detector could make this possible in the future. Since ptychographic reconstructions are not able to converge properly with the low redundancy provided by few detector elements, more work is needed to find the balance between the necessary resolution in reciprocal space and the scan speed which provides acceptable real-space temporal resolution. The benefits of enabling faster scanning and lower dataset sizes from non-pixellated detectors may be balanced with the need for redundancy in the dataset to find the optimal conditions for low-dose characterisation and necessary time-resolution.

In conclusion, this work contributes important results and reflections on how simple detector geometries and signal digitisation help make high-resolution STEM phase retrieval more accessible, and expands the scope of materials that may be characterised.

Chapter 7

Future work

While this thesis has uncovered and addressed some key challenges in ultra-low-dose STEM phase imaging when digitising the signal from segmented detectors, there are still several aspects that remain open for further exploration. In particular, hardware developments for improved detector design with digitisation in mind, algorithmic advances to improve interpretability and tolerance of low-dose, and more realistic simulation frameworks can all significantly improve the applicability of methods demonstrated in this thesis.

This thesis has demonstrated the use of digitisation to retain the precision of electron detection events from segmented detectors for COM and iCOM imaging when the dwell time is severely reduced. Besides that, it has also demonstrated that array detectors has a more isotropic contrast transfer than four-segmented detectors through simulation. The physical realisation of array detectors for experimental proof that they could provide results near those seen in simulations is ongoing work. Particularly, the design of detectors to minimise occurrence of cross-talk and optimise digitisation capability through a sharper scintillator response will help expand the practical applicability of phase retrieval methods for beam-sensitive materials. Not only does the design of new detectors have the opportunity to optimise the scintillator choice, but a detector with more segments will divide the beam into more digitisation streams in parallel, reducing the effective beam current in each segment which reduces the information lost to coincidence loss. Both of these advances will help increase the fidelity of the digitised signal from the detector, increasing the information contained both in each single frame and in a multi-frame stack as a whole.

Simulation of real, signal-degrading effects allows for more realistic comparisons between experiments and simulations. Additionally, the ability to simulate periodic noises may help elucidate their sources and help identify and potentially remove the source of the noise. Further improvements to the streaking simulation code presented in this thesis will allow such investigations and may be highly beneficial to further refine and improve digitisation parameters.

While the iterative ptychographic algorithms used in this work have provided adequate preliminary results, their robustness at low doses can be improved. Specifically, the Poisson noise of the measurements is currently the source of algorithmically injected artefacts at ultra-low doses, since the algorithm is solving for a smoothly varying object. Allowing the noise to be separated from the reconstructed probe and object through, for example, multi-object or multi-probe solutions could help provide a more accurate reconstruction. Additionally, implementations that provide faster reconstructions, potentially approaching real-time, live phase retrieval, would significantly improve the feasibility of ptychography for dose-limited conditions.

Combining digitisation for segmented detectors and phase retrieval techniques with sub-sampling of the ROI ($F_C < 1$), could significantly reduce the applied dose on the sample and the real-space frame rate. This may be realised through various forms of sub-sampling or compressed sensing, for example via interlaced scanning. Another potential sub-sampling route is mid-pixel blanking of the beam, which has been shown to provide image contrast similar to ADF via calculation of the rate of electron arrivals rather than full-pixel counting of detected electrons. What implications this has for COM estimation with a segmented detector and what criterion to be set for the beam to be blanked, are both open research questions. A potential solution is requiring a total number of electrons for all the segments combined and calculate the rate of electron arrival to each individual segment, similar to what has been done with ADF, and then subsequently calculate COM signal from that. Testing of various ways of solving this for experiments is left for future work.

To summarise, these future directions reflect the dynamic and ever-changing landscape of STEM imaging, especially phase contrast and retrieval methods. These techniques are powerful, but rely on hardware, simulation, and algorithm development for their full potential to be realised. Crucially, improvements in these areas are tightly coupled, and continued research is needed to make low-dose, high-resolution imaging more accessible and robust across a wide range of materials and use cases.

Advances in highly sensitive, low-dose characterisation techniques will aid research in many important areas of science for a healthier, cleaner, and safer world. For example, higher sensitivity phase retrieval methods will help with characterisation of materials for battery technology, which typically contain low- Z number elements like lithium and oxygen. Understanding of the relation between the structure and functional properties of anode and cathode materials help scientists develop higher capacity batteries that degrade less from repeated charging and discharging [92, 182–185]. Additionally, these sensitive characterisation methods will aid characterisation of MOFs and zeolites, which are typically affected by both knock-on damage and radiolysis [136, 186]. These materials play a crucial role in development of efficient carbon capture and micro-plastics absorption techno-

logy [187–192]. Using characterisation techniques with high spatial resolution aids understanding of the morphological changes these materials undergo upon absorption of carbon-containing molecules, which underlines why low-dose STEM characterisation techniques are beneficial for such studies. Further, the fast, low-dose multi-frame technique presented in this thesis enables identification of sample damage in post-processing of each data set, and frames prior to the onset of sample damage may be used for analysis.

This work also paves the way for time-resolved and accurate characterisation of beam-sensitive materials and other low- Z number elements as they change under beam illumination or changing of the local environment *in-situ*. Traditionally, samples have been studied pre- and post-exposure to understand how the exposure alters the materials. However, this typically fails to address how materials change during the exposure [193]. The temporal resolution advances presented in this thesis help address this by bringing time-resolution on the order of seconds to phase retrieval imaging in STEM. As a result, dynamical changes in materials during *in-situ* experiments may be observed, such as changes in anode and cathode materials during applied bias. This means that not only the material atomic structure may be observed, but also the material function during conditions close to operational conditions. Methodological advances, like those presented in this thesis, are important in building characterisation techniques that enable the materials discoveries and scientific advancements for the future.

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Appendix A

Aberration notation

In this thesis multiple types of aberrations have been mentioned by name. Maybe most prominently defocus, which incidentally is the most confusing one of them all. In Table A.1, the aberration coefficients and their notation is summarised. The notation used is the Krivanek notation [14], as is it the common notation used for aberration coefficients in STEM and also the notation used by abTEM.

Table A.1: Overview of aberration names and the Krivanek [14] notation for them. Table adapted from [194].

Aberration alias	Krivanek notation	Order in $\vec{k}$ -space	Azimuthal symmetry
Two-fold astigmatism	$C_{1,2}$	2	2
Defocus	$C_{1,0}$	2	—
Three-fold astigmatism	$C_{2,3}$	3	3
Axial coma	$C_{2,1}$	3	1
Four-fold astigmatism	$C_{3,4}$	4	4
Axial star aberration	$C_{3,2}$	4	2
Spherical aberration	$C_{3,0}$	4	—
Five-fold astigmatism	$C_{4,5}$	5	5
Fourth order axial coma	$C_{4,1}$	5	1
Three lobe aberration	$C_{4,3}$	5	3
Six-fold astigmatism	$C_{5,6}$	6	6
Fifth order rosette aberration	$C_{5,4}$	6	4
Fifth order axial star aberration	$C_{5,2}$	6	2
Fifth order spherical aberration	$C_{5,0}$	6	—
Seven-fold astigmatism	$C_{6,7}$	7	7
Sixth order axial coma	$C_{6,1}$	7	1
Sixth order three lobe aberration	$C_{6,3}$	7	3
Sixth order pentacle aberration	$C_{6,5}$	7	5
Eight-fold astigmatism	$C_{7,8}$	8	8

Appendix B

Ptychographic Reconstructions with More Aberrations

In section 5.4, some preliminary simulations and experimental results with segmented and pixel array detectors for iterative ptychography were presented. As mentioned, one of the strengths of iterative ptychography is its ability to resolve, and correct for, aberrations in the probe. In particular, defocused probes are often used when acquiring datasets for ptychographic reconstruction because it helps the algorithm converge faster by introducing more redundancy in the dataset. Similarly, other aberrations can help “phase” the probe wavefront, but is not as easily controlled as the defocus. The success of a ptychographic reconstruction typically requires a decent guess for the incoming probe wavefront. Tools from other phase reconstruction techniques, like SSB and tilt-corrected BF-STEM, are customarily employed to estimate the values for common aberrations to initialise the iterative ptychographic reconstruction. Therefore, investigating how the algorithm performs with aberrated probes when simple detector geometries are used instead of pixellated detectors is important to solidify its usefulness with alternative detectors.

First, the usefulness of iterative ptychography with simple detector geometries for probes with substantial spherical aberration was studied with simulations. In these simulations, Scherzer defocus was applied to counteract the impact of the high spherical aberration coefficient of $C_S = 1.2$ mm, which represents a typical value for an uncorrected STEM instrument. For this value, the Scherzer defocus is roughly $\Delta f = 595$ Å, and for an uncorrected STEM this would roughly be the defocus at which the sample looks to be in focus¹. The resulting iCOM images and ptychographic reconstructions for annular segmented and pixel array detectors are shown in Figure B.1 and Figure B.2, respectively. Based on these, it seems that ptychography can provide better phase retrieved images than iCOM for uncorrected microscopes, and that even simple detector geometries can be sufficient to get successful reconstructions.

¹Although we later found that Scherzer defocus is not the optimal defocus value for optimal iCOM contrast transfer, see section 3.4.

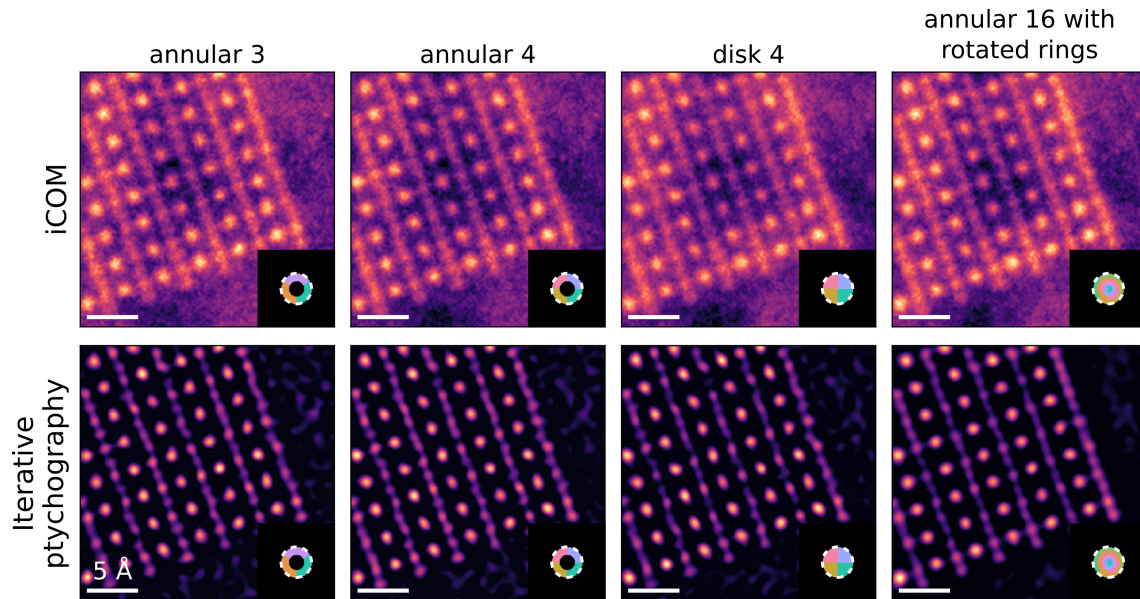


Figure B.1: iCOM and ptychographic reconstructions from simulated low dose ($2000 \text{ e}^-/\text{\AA}^2$) data with various detector geometries. Simulated with $C_S = 1.2 \text{ mm}$ and Scherzer defocus ($\sim 595 \text{ \AA}$), to investigate how well segmented ptycho can work for a non-probe-corrected STEM instrument.

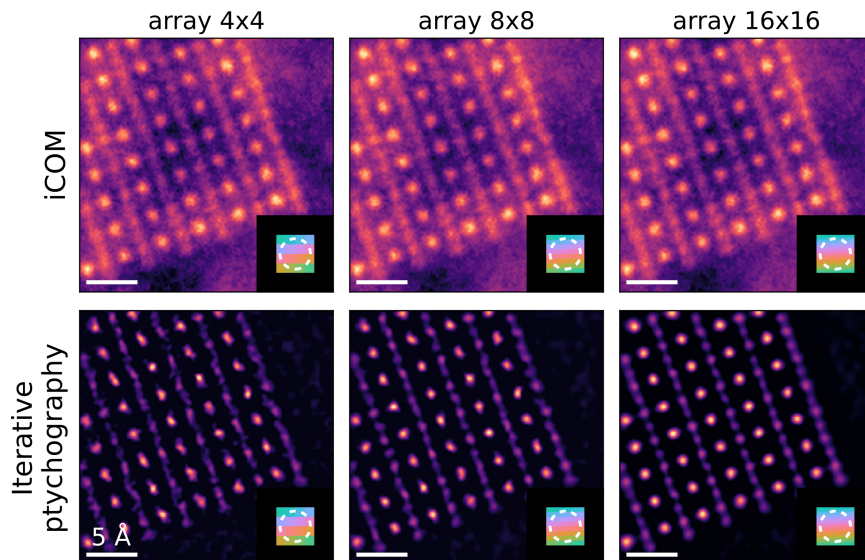


Figure B.2: iCOM and ptychographic reconstructions from simulated low dose ($2000 \text{ e}^-/\text{\AA}^2$) data with various detector geometries. Simulated with $C_S = 1.2 \text{ mm}$ and Scherzer defocus ($\sim 595 \text{ \AA}$), to investigate how well segmented ptycho can work for a non-probe-corrected STEM instrument.

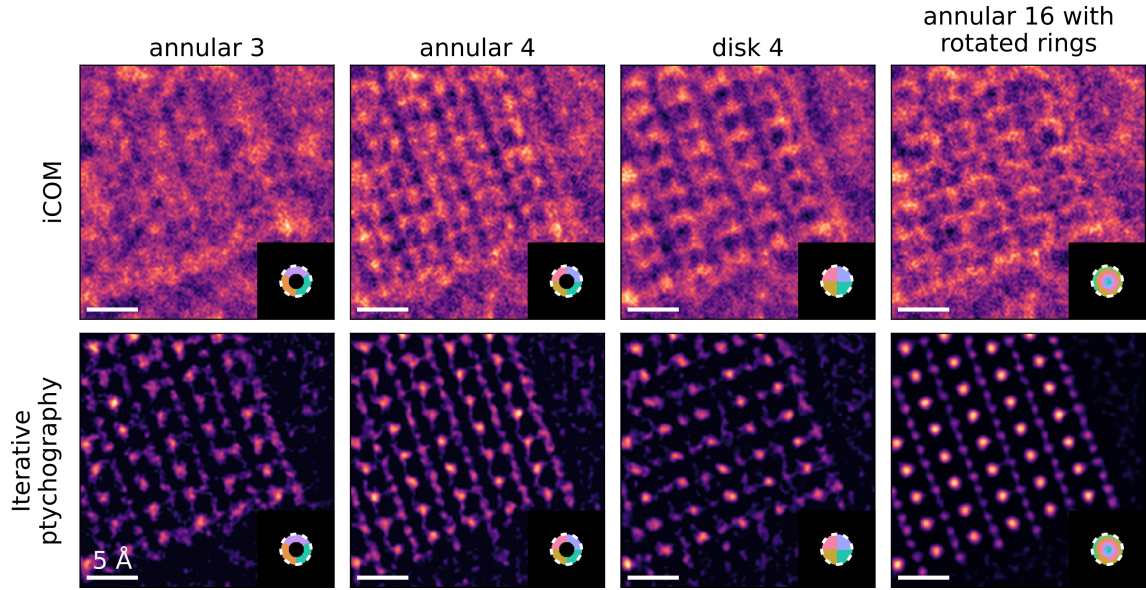


Figure B.3: iCOM and ptychographic reconstructions from simulated low dose ($2000 \text{ e}^-/\text{\AA}^2$) data with annular detectors with 3, 4, and 16 segments. Simulated with $C_{2,3} = 5 \cdot 10^3 \text{ \AA}$ and $\Delta f = 128 \text{ \AA}$, to investigate how well segmented detectors work for the iterative ptychographic algorithm to decode heavily aberrated probes, and if detector geometry matters for how well the probe aberrations may be de-tangled from the object.

Another aspect that was worthwhile exploring was whether simple geometry detectors would provide enough data for the ptychographic reconstruction to correct non-spherical aberrations. Test simulations with three-fold astigmatism $C_{2,3} = 5 \cdot 10^3 \text{ \AA}$ and defocus $\Delta f = 128 \text{ \AA}$ was made and binned to simple detector geometries for the ptychographic algorithm to reconstruct. The results, shown in Figure B.3 and Figure B.4, show that the simplest annular detectors are unable to provide enough information for the algorithm to successfully reconstruct the object, but the detectors with 16 and more detector elements perform better.

Importantly, simulation have a crucial benefit: that the aberration coefficients are known exactly. This yields a luxury of a very precise guess for the initial probe, which again helps the algorithm disentangle the probe and object successfully. In experiment this is typically not the case, and estimates for the aberration coefficients must be found in other ways. A typical experiment includes a pixellated capture of the vacuum probe acquired right after the experiment is done. Assuming that higher-order aberrations drift slowly, and aberration correction was optimised at the start of a session, that typically leaves only defocus and astigmatism to be estimated from the data.

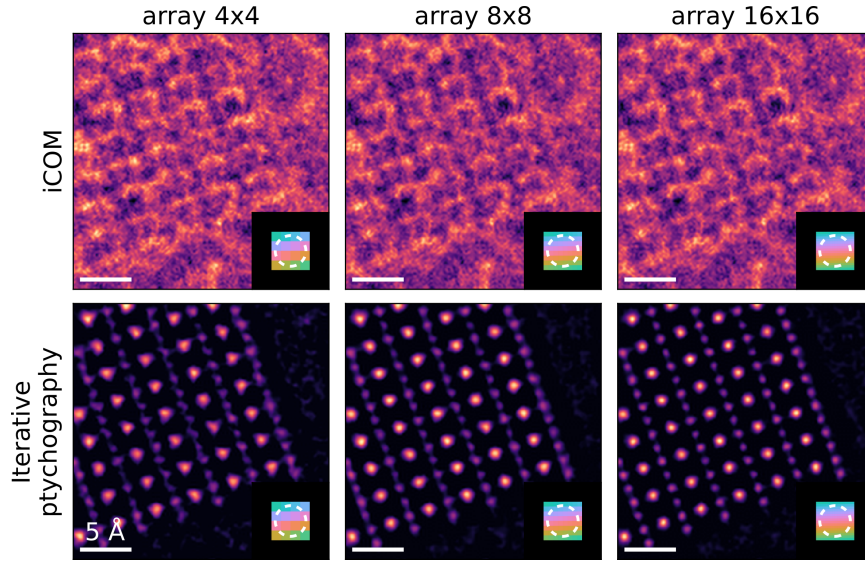


Figure B.4: iCOM and ptychographic reconstructions from simulated low dose ($2000 \text{ e}^-/\text{\AA}^2$) data with various pixel array detector geometries. Simulated with $C_{2,3} = 5 \cdot 10^3 \text{ \AA}$ and $\Delta f = 128 \text{ \AA}$, to investigate if array detectors better allow the iterative ptychographic algorithm to de-tangle the probe aberrations from the object.

Experimental Observations

The following figures show ptychographic reconstructions for data acquired with a segmented eight-segment Panther detector and pixellated Arina detector on a TFS Spectra 300 using increasing defocus values. The experimental parameters were $E = 300 \text{ keV}$, $I = 45 \text{ pA}$, and $\delta_t = 50 \text{ }\mu\text{s}$. These preliminary results confirm what was found in simulations, see chapter 5, which is that the usable defocus when using a segmented detector is heavily limited. Future work will determine if array detectors can expand the defocus range from which successful reconstructions can be achieved, and also if this is a useful route to fast frame-rate, low-dose ptychography.

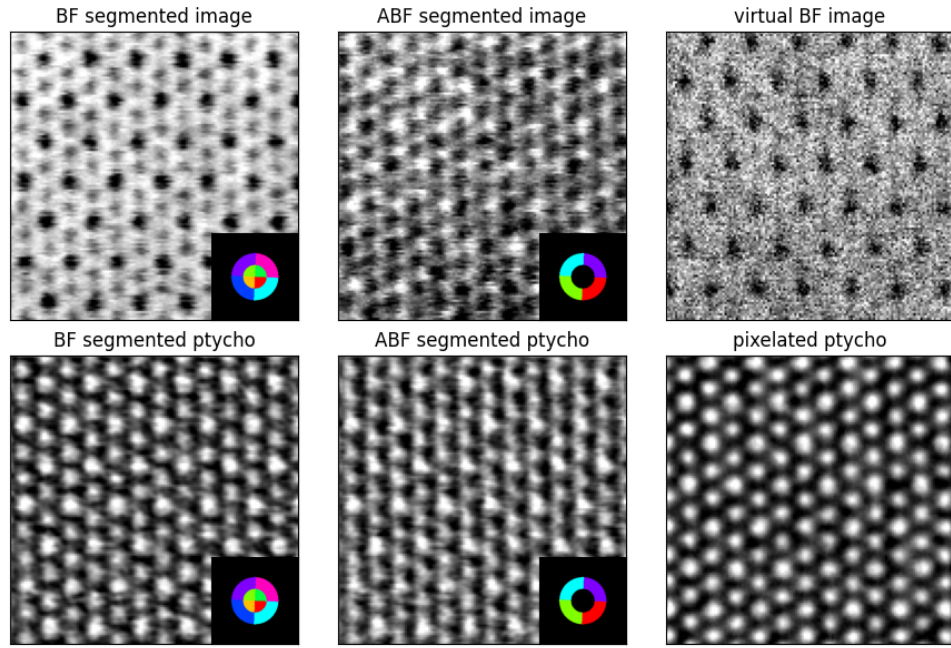


Figure B.5: Ptychographic reconstructions from experimental data (preliminary experiments) with in-focus probe.

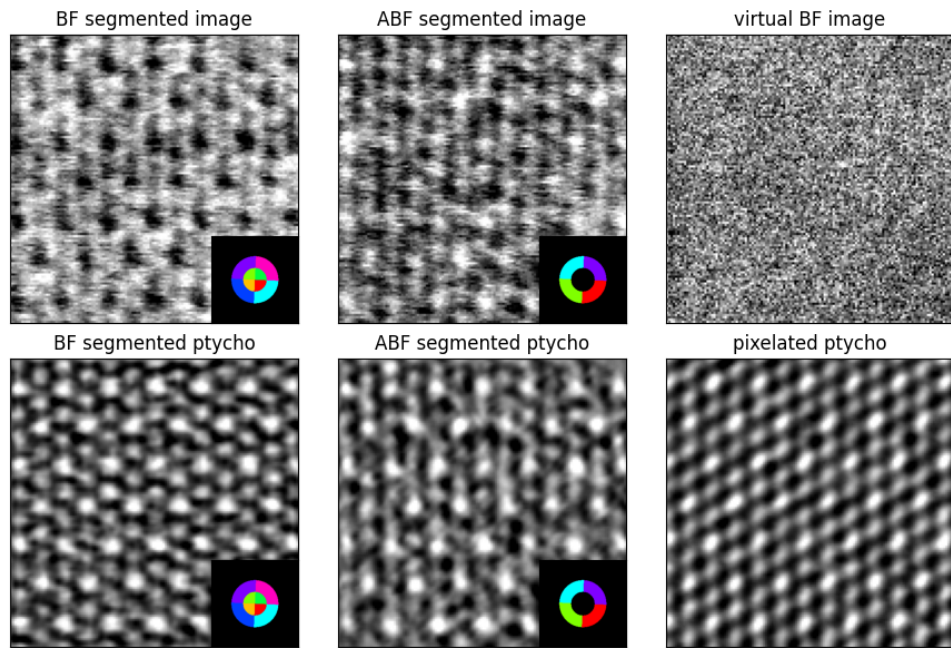


Figure B.6: Ptychographic reconstruction from preliminary experiments. The insets show the detector geometry for the segmented detector reconstructions. $\Delta f = -50$ Å.

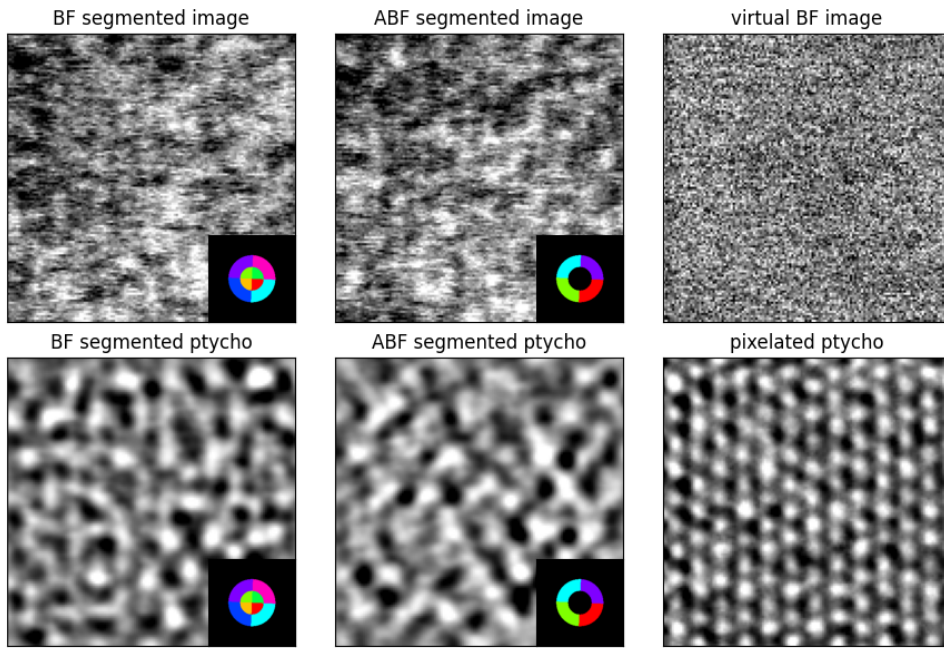


Figure B.7: Ptychographic reconstruction from preliminary experiments. The insets show the detector geometry for the segmented detector reconstructions. $\Delta f = -100$ Å.

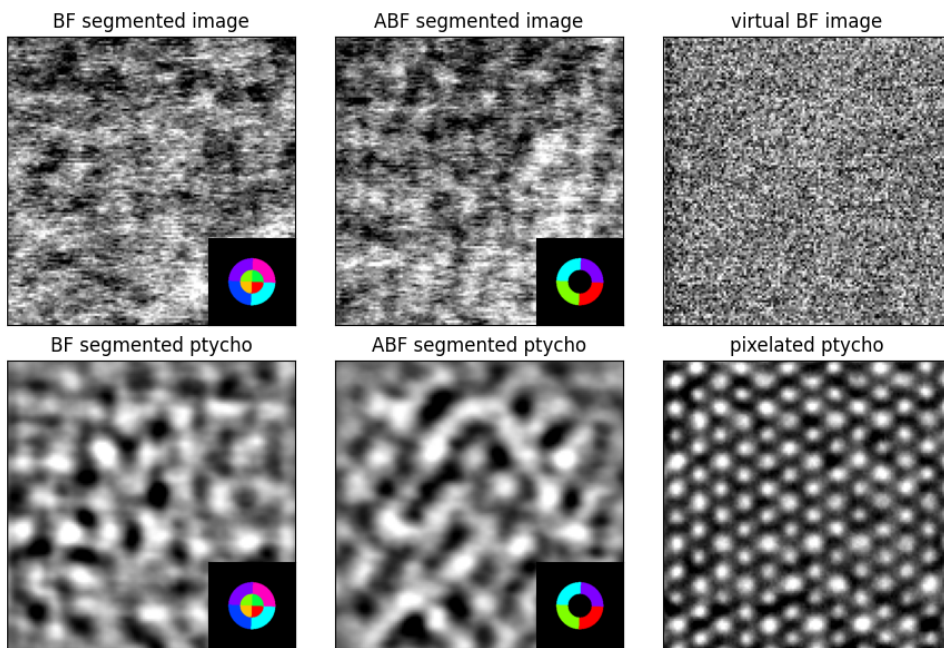


Figure B.8: Ptychographic reconstruction from preliminary experiments. The insets show the detector geometry for the segmented detector reconstructions. $\Delta f = -150$ Å.

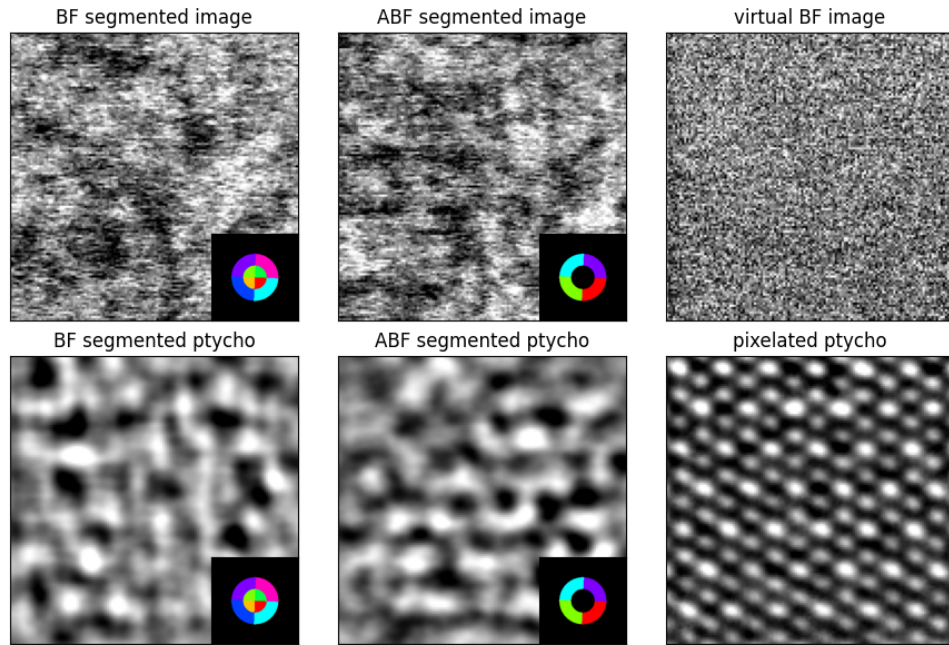


Figure B.9: Ptychographic reconstruction from preliminary experiments. The insets show the detector geometry for the segmented detector reconstructions. $\Delta f = -200 \text{ \AA}$.

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All the best, Julie Marie Bekkevold

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