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Report on phase manipulation with Electrostatic Phase Plates for depth of focus increase in STEM

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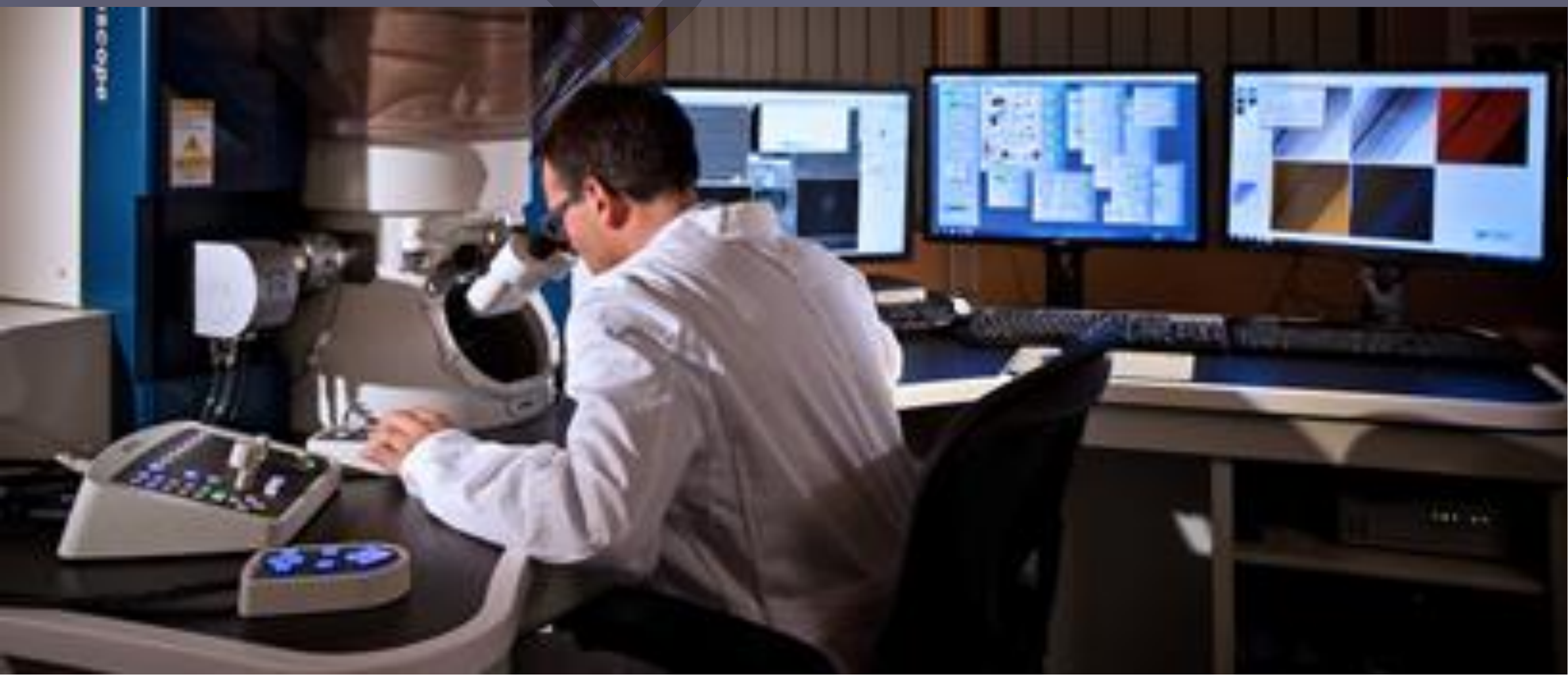

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Revision history log

Version number	Date of release	Author	Summary of changes
V1	26/04/2022	Jo Verbeeck, Francisco Vega Ibañez	Final report
V1.1	27/04/2022	Peter van Aken	Approval of report
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1. Introduction

Transmission electron microscopy provides a view of the structure of materials down to the atomic scale. As the electrons are transmitted through the sample, this is often regarded as a projection technique, where the sample is imaged as a flat projection. Just like in optical microscopy, however, this point of view breaks down at higher magnification, especially in cases where focused (STEM) probes are used.

Indeed, in an attempt to increase the spatial lateral resolution, the focused beam needs to converge, and an inverse relationship between opening angle and spatial resolution emerges. In practice, this opening angle cannot be increased too much as higher-order lens aberration will quickly deteriorate the spatial lateral resolution. Besides this in-plane spatial resolution, one can also expect different image properties at different depths as the beam is only focused in a specific plane and quickly defocuses away from the plane. This effect is even more prominent, when the beam opening angle is larger. A trade-off thus appears between spatial in-plane resolution and out of plane z-resolution.

Depending on the task at hand, this can prove to be either advantageous or problematic. Indeed this limited depth of field could be used to provide depth information about the sample by deliberately changing the focus and probing different sample layers. In optical confocal microscopy, this is exploited to gain complete 3D information of, e.g., life science samples. Implementation of this concept in TEM has been attempted and discussed but suffers from the limited opening angle. As the z-resolution can be demonstrated to scale with $1/\alpha^2$, we get that the resolution in the z-direction is a factor of alpha lower than the xy resolution. If we consider a modern instrument, alpha is typically limited to [30, 50] mrad. As it is unexpected to get to a significant increase in the near future, we can only hope for depth resolution of a few 100 times that of the xy resolution.

In other applications such as tomography, it is necessary to have all features throughout the thickness of a sample to remain focused, obtaining an accurate geometric projection required for the tomographic reconstruction. In this case, the depth of field needs to be increased as much as possible, which often means choosing between lower in-plane resolution or working with very thin samples if high resolution is required.

All the above arguments rely on the free-space propagation of the electron beam. However, the interaction with the sample makes the situation far more complicated and results in sample-dependent scattering and channelling conditions that can severely complicate the experimental interpretation.

In this report, we will investigate if the emerging capability of shaping the coherent wavefront of an electron beam with programmable phase plates would alleviate the difficulties mentioned above. Indeed, progress in optics shows that, e.g., adaptive tuning of laser beams can maintain accurate, focused probes inside thick and heavily scattering media if adaptive optics is applied. On a theoretical level, Bessel beams are receiving attention for their ability to 'self-heal' and their increased depth of focus. These arguments would hint at the fact that also in electron microscopy, similar techniques could be applicable. We present here a series of initial simulations to test the feasibility of such methods that could become a reality with the advent of programmable phase plates.

2. Definition of Depth of Field and Depth Resolution

Depth of field can be defined in many ways. However, taking incoherent light optics as a reference, we can define that depth of field corresponds to the range of defocus without a significant change of contrast in the image. A more quantitative assessment of what “significant” means in this context can be taken from Born and Wolf’s definition, where a 20% variation from the maximum intensity gives a reasonable focal tolerance.

If we have negligible aberrations in the instrument, we can express this loss of intensity in the following way:

$$I(z) = \left[\frac{\sin\left(\pi \frac{\alpha^2}{2\lambda} z\right)}{\pi \frac{\alpha^2}{2\lambda} z} \right]^2 I_0$$

From this expression, we can extract that the value for either z-resolution or depth of focus will be proportional to the inverse of the opening angle squared:

$$\Delta z \sim \frac{\lambda}{\alpha^2}$$

Having a large Δz means that we will keep in-focus more features throughout the thickness of the sample (while having to compromise xy resolution). On the other hand, we can also aim for the highest possible z-resolution (getting to the smallest possible value of Δz). With this in mind, the resolution along the z-direction will be the smallest possible distance, at which we can distinguish between two points (Rayleigh’s criterion), thus giving:

$$\Delta z = 2\frac{\lambda}{\alpha^2}$$

To put this into perspective, we will require an aperture angle greater than *100 mrad* to start reaching an atomic resolution of barely 4 Å.

In a general manner, we can look at the intensity of the probe propagating in free space (or inside a sample) as an ellipsoid. The sharpest, most intense small volume will yield the highest z-resolution, whereas having the largest intense cigar-shaped ellipsoid will give an equally elongated depth of focus. The xy resolution of this ellipse is determined by the Full Width at Half Maximum (FWHM) of the probe for a given z value of propagation.

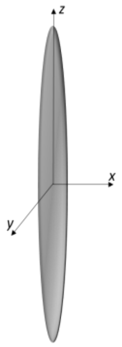

Figure 1 Sketch of the ellipse formed by the intensity of the probe propagating in free space

3. Bessel and related beams

Bessel beams are exact solutions to the wave equation that do not experience spreading in the transverse direction (diffraction). The zeroth-order Bessel beam, which is used for most applications in the field of optics, can be expressed as:

$$\psi^B(r,z) \sim exp(ik_z z)J_0(k_\perp r)$$

This Bessel function consists of two parts: one pure phase propagation, which depends only on z, and an in-plane distribution that depends only on r (thus non-diffracting). Moreover, if we take the Fourier transform of this Bessel function, we get:

$$\tilde{\psi}^B(p_\perp, \varphi, p_z) \sim \delta(p_\perp - \hbar k_\perp)\delta(p_z - \hbar k_z)$$

where δ is the Dirac delta function. This Fourier Transform shows how the Bessel beam is an eigenstate of transverse momentum, which can be taken as a conical superposition of plane waves. Knowing this, we can see that imprinting a conical phase to a plane wave (axicon) via a spatial light modulator, or by creating a hologram, can create approximate Bessel beams. Another property that this superposition of plane waves yields is self-healing: if the central lobe of the beam is blocked, the converging plane waves will recreate it after some propagation length proportional to the size of the obstruction:

$$z_{rec} \approx \frac{ak_\perp}{2k_z}$$

with a the size of the obstruction [1].

However, due to the deviation from the ideal Bessel beam, these properties are only maintained to a certain extent. If the finite energy beam is generated by an aperture consisting of a ring, with $\Delta k_\perp$ the thickness of this ring, we obtain an approximated Bessel beam with a waist inversely proportional to the ring width. Such experiments have been documented by us and others, but the practical gain in depth of field turned out to be minor [2]–[4].

3.1. Can we change the propagation of a beam with beam shaping?

We now would like to evaluate if we can obtain the aforementioned benefits (z-resolution increase or enlarged depth of field) with the aid of an electrostatic programmable phase plate.

The strategy relies on creating an array of phase shifting elements in the aperture plane, thus allowing to gain some degree of freedom over the phase of the electron wavefront [5].

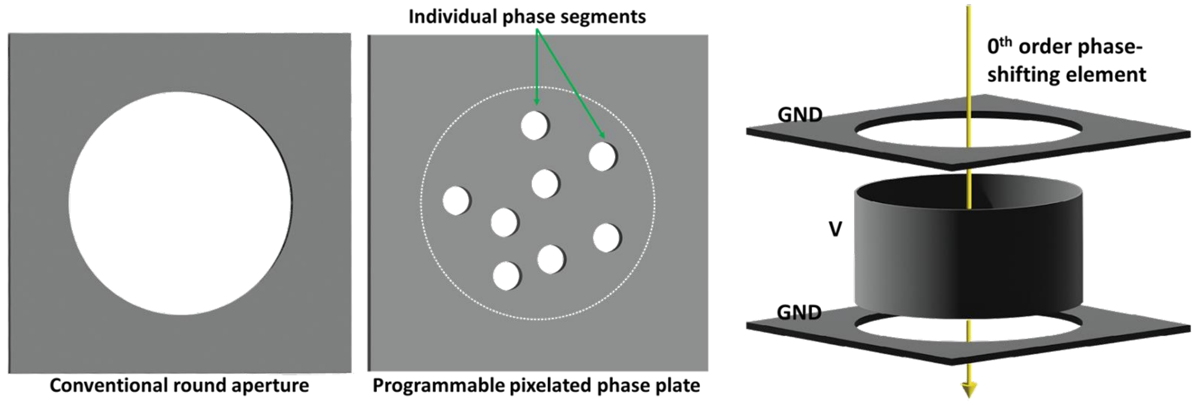

Figure 2 Sketch of a round aperture, programmable phase plate, and individual phase segment.

The design of these pixelated apertures, in the context of z-resolution and depth of field, can be seen from either perspective as follows:

a. **Obtain the best possible z-resolution:** for this, since z-resolution goes as $1/\alpha^2$, the design should be aiming to correct for higher-order aberrations, which prevent a regular microscope from increasing the opening angle further. Not only this, but also blocking as little of the electron beam as possible in the process.

b. **Enlarge the propagation length of the beam:** To enlarge the beam propagation length (Bessel-like beam), annular-like features are necessary in the phase plate.

3.2. Can we increase depth resolution with beam shaping?

As mentioned earlier, the depth resolution is inversely proportional to the square of the aperture angle. With this in mind, we can now attempt to design a phase plate that corrects for the aberrations at higher aperture angles (i.e., $\alpha > 21$mrad). Only considering rotationally symmetric aberrations (defocus and C_3), the design concentrates on sampling the radial coordinate as well as possible. However, corrected instruments at higher convergence angles often come with non-rotationally symmetric aberrations (see Figure 3(a)), which further complicates the potential phase plate design due to the need for increased azimuthal coordinate sampling.

With this in mind, we want to propose a theoretical design for a phase plate to correct aberrations and allow to reach higher aperture angles in a corrected instrument, which is shown in Figure 3(b). We estimated the aberrations as follow in our calculations: C_1= -16.2 Å, C_3 = 1µm, B_2 = 20 nm, A_2 = 60 nm, S_3 = 500 nm, A_3 = 1µm, A_4 = 20µm.

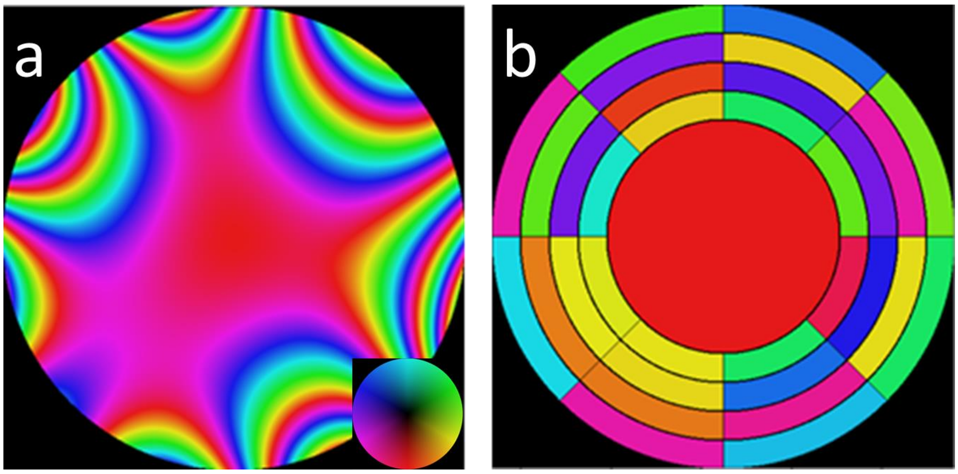

Figure 3 (a) Aberrations at a beam energy of 300kV and a convergence angle of 50 mrad for a round aperture. (b) Phase plate design correcting for this aberrated profile over the same opening angle. The inset on (a) gives the scale with amplitude represented by intensity and phase by hue.

With this segmented design for the phase plate, we calculate the integrated FWHM (d_{50}) at different opening angles and show the result in Figure 4. Our proposed theoretical design can clearly correct for aberrations, allowing us to get to higher aperture angles with reduced probe size.

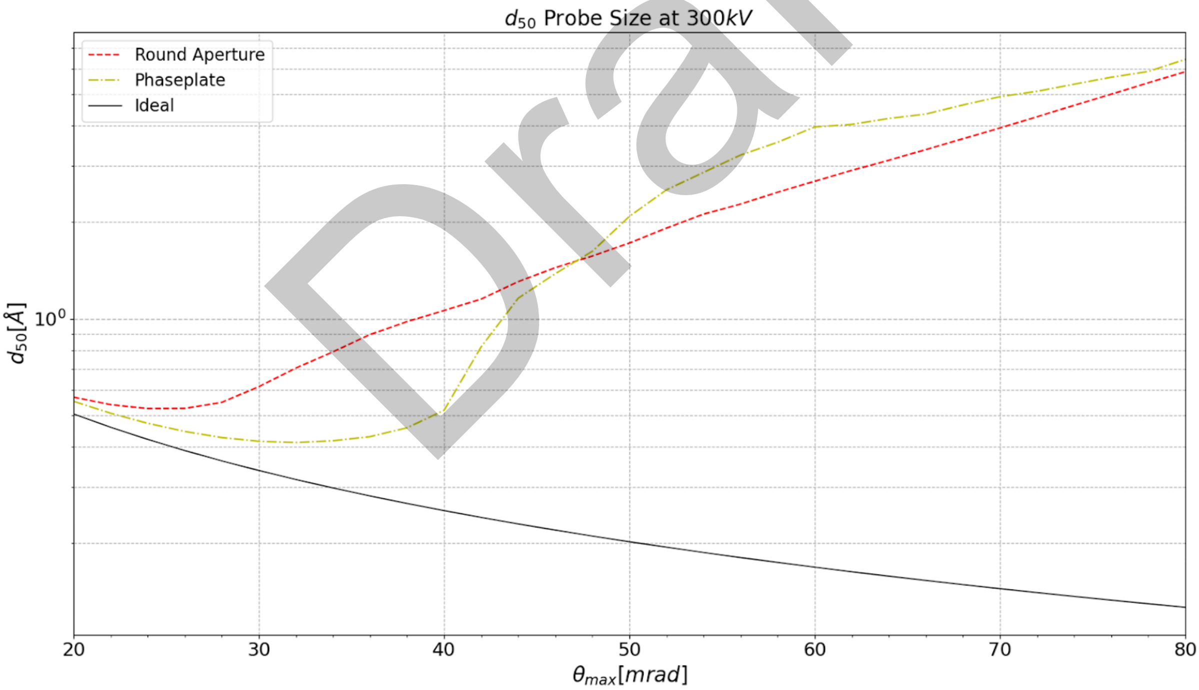

Figure 4 Simulated probe size d_{50} assuming a 300 keV electron beam going through either a phase plate or a round aperture. The black continuous line shows the diffraction limit.

Furthermore, we also replicate the measurement for probe size (d_{50}) at different propagation lengths, for different apertures, and for different opening angles (Figure 5). To obtain the best z-resolution we aim to get the sharpest possible lowest point when coming into focus (z≈0). This added to a small d_{50} implies that the ellipsoid volume forming the probe is the smallest.

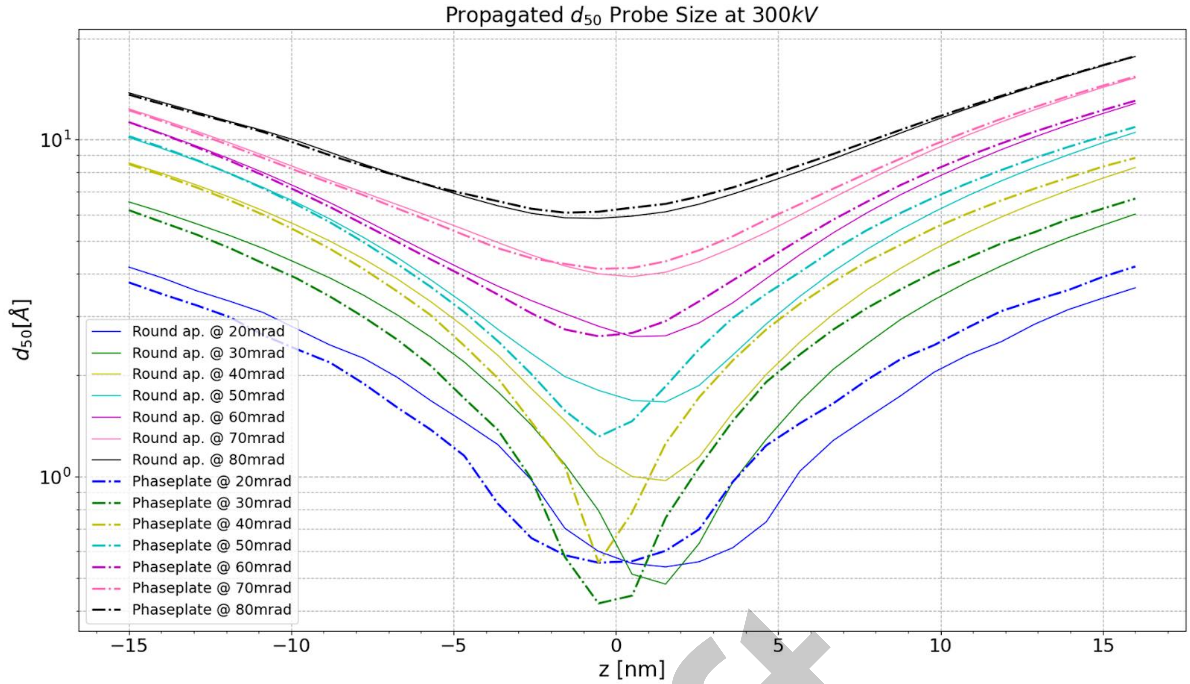

Figure 5 Change of d_{50} over different propagation lengths at different opening angles. The value on the y axis gives the xy resolution of the probe, whereas the 'steepness' of the curve is proportional to the z-resolution.

From Figure 5, we can also extract the propagation distance within which the xy resolution remains under 50% of the minimum probe size. This calculation yields, for the phase plate, to an increase in z-resolution of ~1.8x compared to a standard round aperture (see Figure 6).

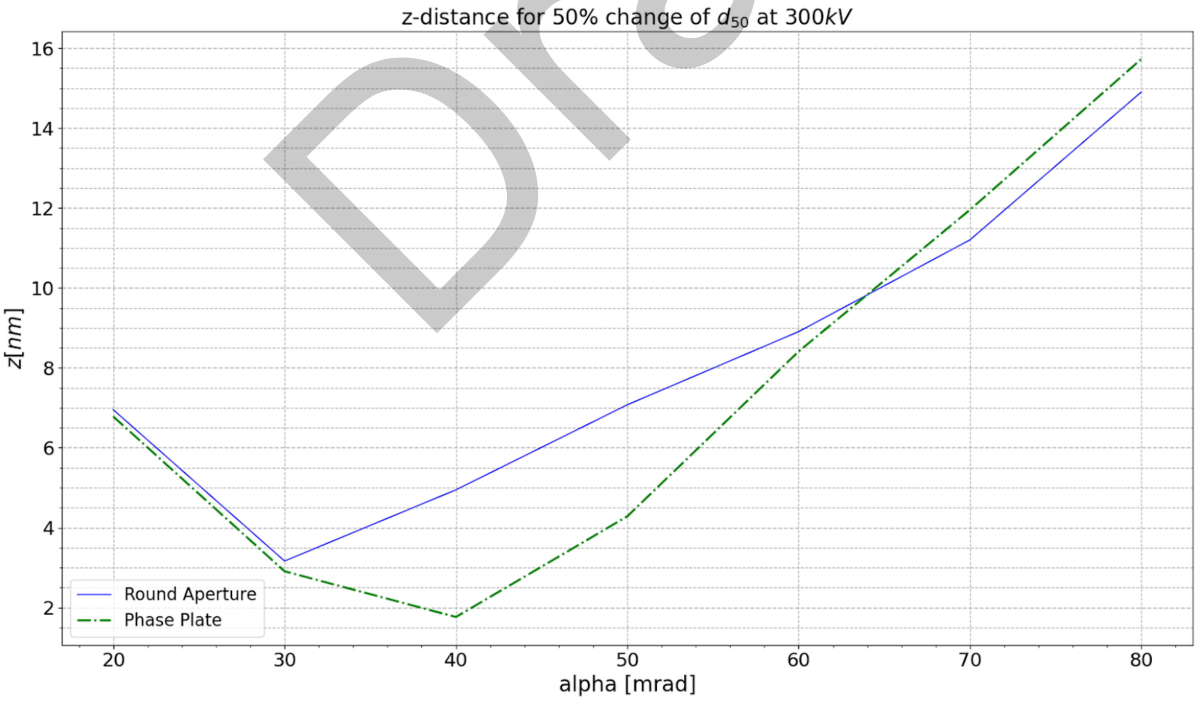

Figure 6 Propagation distance at which the probe gets 50% larger than its minimum size for different opening angles.

3.3. Can we increase depth of field with beam shaping?

Opposite to what we discussed in the previous section, if we aim to enlarge the beam, we will gain depth of field, thus keeping more of the sample in focus along the z-direction. As the theory mentioned earlier shows, we want to create ring-like features as thin as possible, yielding an enlarged cigar-shaped beam, which unfortunately comes at the expense of some loss of intensity. We can see the effect of the ring thickness on the depth of field in Figure 7, where we gradually increase the width of the ring aperture and calculate the integrated intensity (up to 50%) as it propagates in free space.

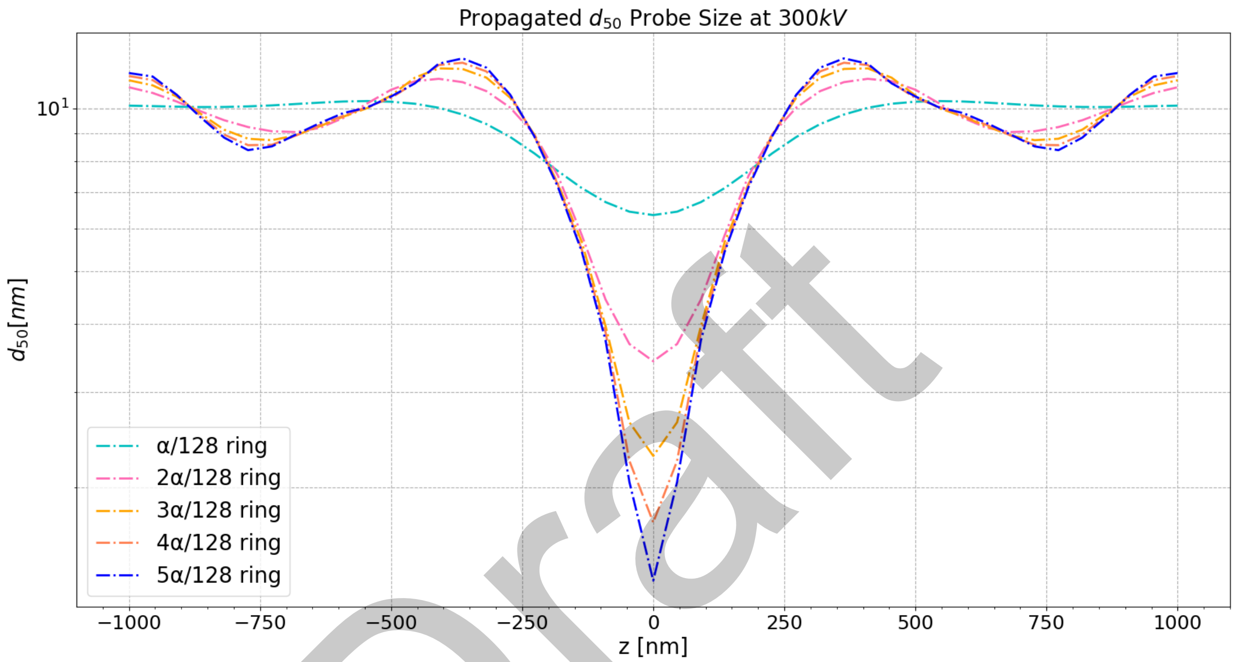

Figure 7 Change of d_{50} over different propagation lengths with different ring widths. As we make the width of the ring aperture smaller, the probe remains focused for a longer propagation length (at the expense of xy resolution).

Now, to evaluate what a programmable phase plate can bring to increase the depth of focus, we want to propose a phase plate design that has been already demonstrated [6]. This design consists of 48 pixels arranged in 12 petals with 4 segments each (see Figure 8).

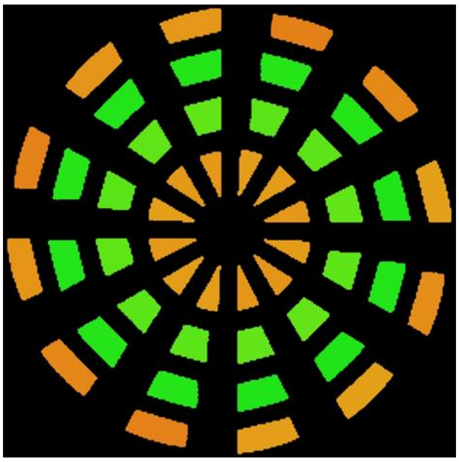

Figure 8 'Segmented' phase plate design

We can see the pixels arrangement as four concentric annular segments, so the probe coming from any of these annulus will have some Bessel-like behavior.

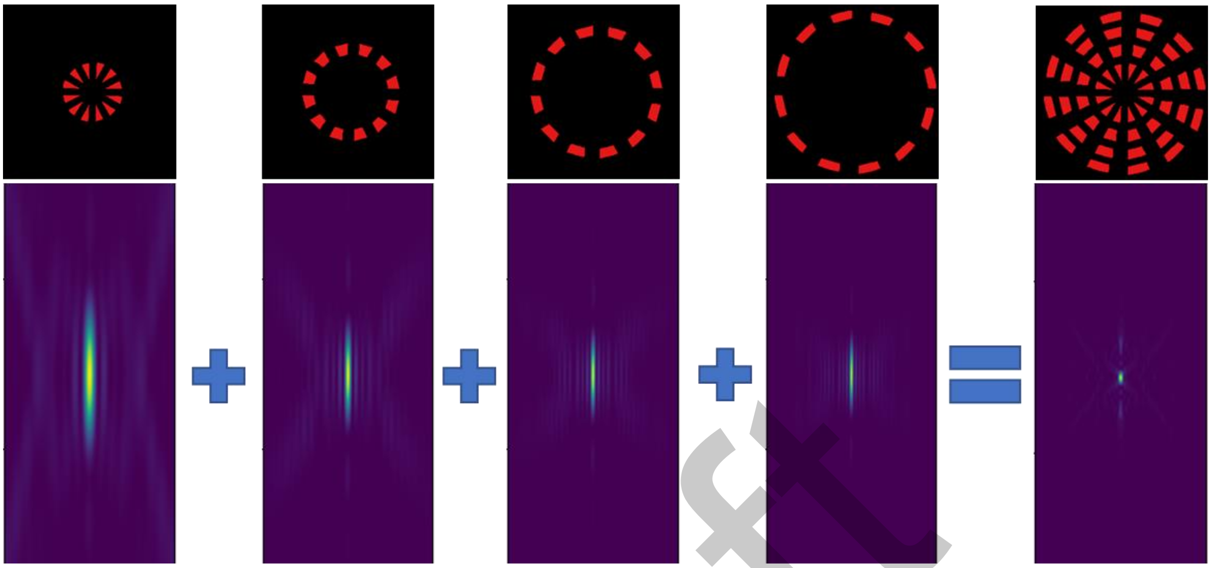

Figure 9 Propagated probe coming from all the annular-like segments of the phase plate and the resulting probe from the whole phase plate.

From Figure 9 alone, we see that adding all the ring segments with the plate off (no phase in the segment) does not yield to an elongated cigar-shaped probe, as the resulting phase plate comes closer to a standard round aperture. Fortunately, the versatility and adaptability of a programmable phase plate allows rapidly changing the phase of each pixel segment. If we incoherently add up all the combinations of zero and π phase shift on each ring segment faster than the dwell time, we obtain the equivalent of an enlarged cigar-shaped probe from the phase plate (see Figures 10 and 11).

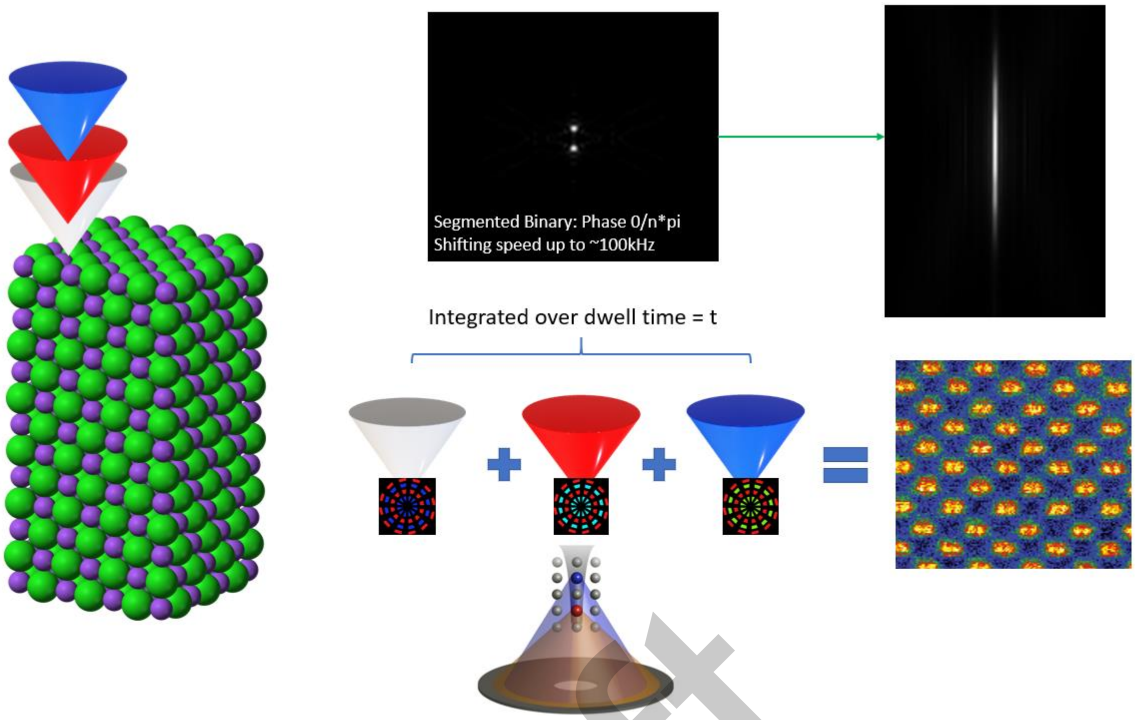

Figure 10 Sketch of the proposed alternative scanning scheme using a phase plate. The phase on the pixels can be rapidly shifted and added up to get signal from different probes integrated over the dwell time.

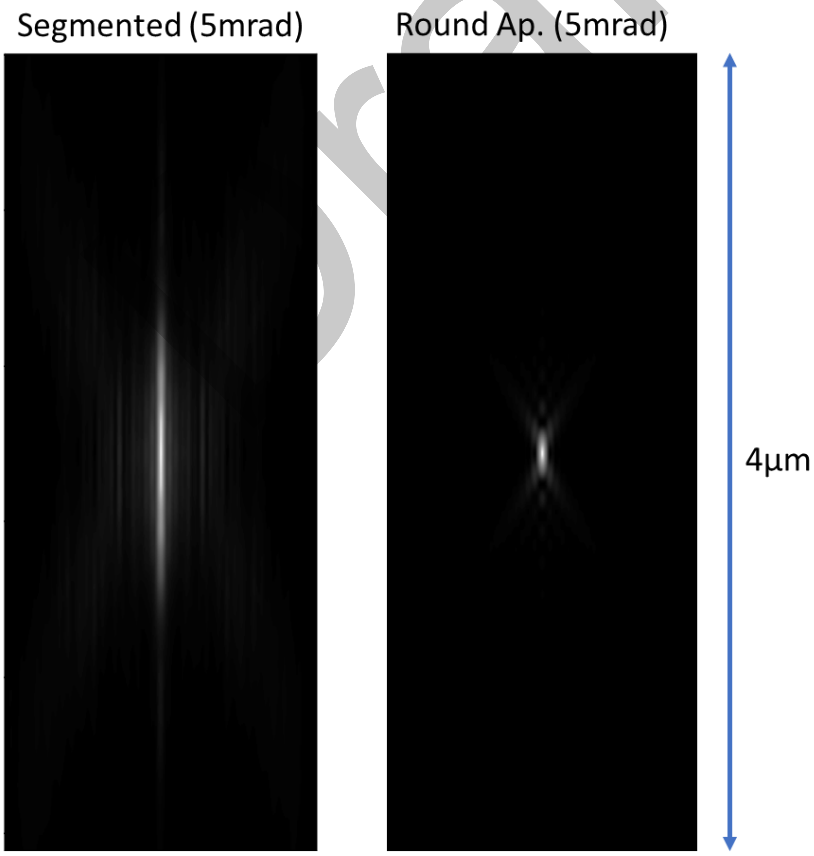

Figure 11 Propagated probe adding up the 16 combinations of zero and π phase inside the annular segments of the plate (left) next to a propagated beam coming from a round aperture (right). The box is ~10_nm wide. Despite showing an enlarged propagation length, the beam coming from the phase plate has an intensity spread out to higher angles.

3.4. Noise and other considerations

Several parameters need to be considered when designing an electrostatic phase plate. Firstly, adding material to make up for the plate segments leads to the concept of the fill factor (ζ), the amount of optically transparent vs. non-transparent parts in the phase plate. This modulation of the local amplitude of the electron beam will inevitably lead to a broadening of the probe as high spatial frequency tails are introduced. Having a low ζ leads to image artefacts and loss of xy resolution, so having ζ as close as one as possible is highly desired.

Another important issue, especially when dealing with z-resolution, is the possibility of having electronic noise coming from the phase plate controller itself. This is evaluated and shown in Figure 12, which displays how phase noise up to π/8 does not affect the probe size significantly. As standard off-the-shelf electronic components provide much better noise properties, such effect is not expected to have strong experimental influences.

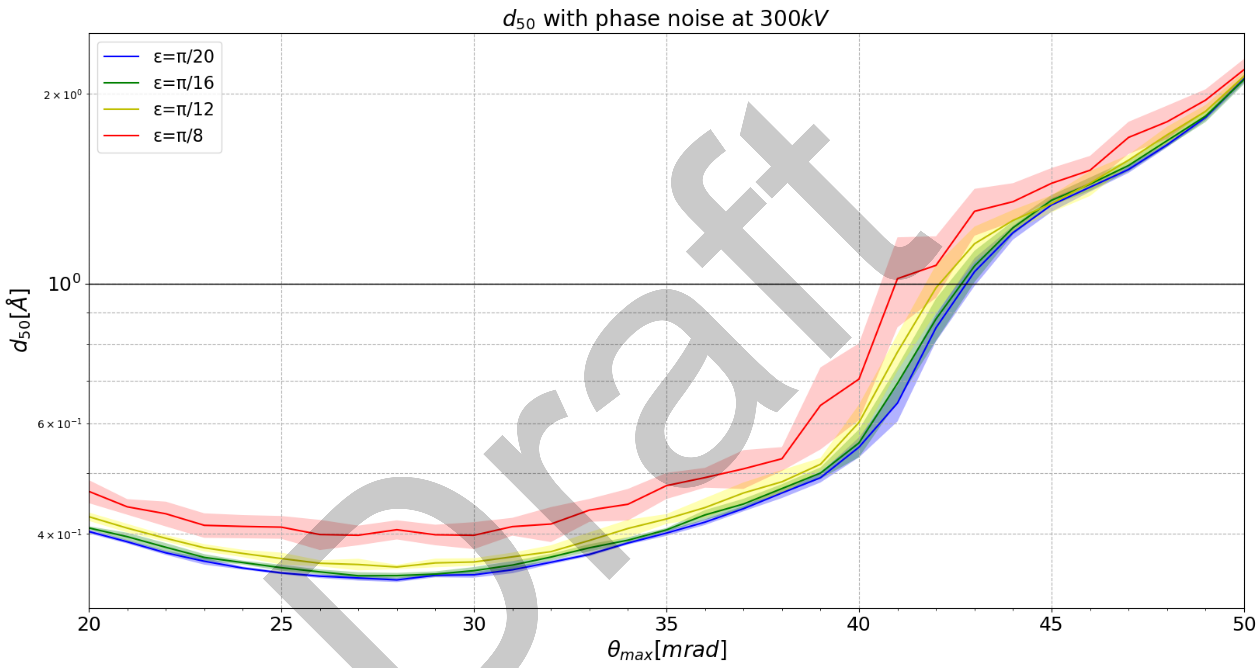

Figure 12 Effect of electronic noise on the phase pixel. For each data point in each plot, we average ten values for d_{50} applying Gaussian noise with std. deviation epsilon.

4. Future development

Future development will focus on the use of adaptive algorithms for optimization of the phase profile in each of the segments forming the phase plate. With the support of multi-slice simulations, we can reconstruct the optimal phase profile on the aperture with any goal function in mind, from there we can enclose regions to make up for the optimal placement and size of the phase-shifting elements in our aperture.

5. Conclusion

In conclusion, we have demonstrated that the newly gained degree of freedom brought by programmable phase plates does provide the ability to both increase the depth of field, as would be desirable for tomography or for imaging thick samples and increase the depth resolution as it would be attractive to look inside the third dimension of the sample without the requirement of tilting the sample.

For increased depth of field, we found that fast dynamic control of the phase plate allows to incoherently sum different coherent beams in the course of the dwell time of a STEM recording. In principle, similar methods could also be performed with a defocusing lens, but it is unlikely that such a method would be fast enough given the high inductance (and thus slow response) of any magnetic lens with some focusing capability.

Through focus atomic depth resolution remains highly attractive for rapid 3D information e.g., in in-situ liquid or gas experiments. We demonstrated that, in principle, a significant increase in depth resolution is possible with a programmable phase plate (factor ~2). However, the atomic scale seems to require a level of detail in the phase plate (number and size of programmable pixels) that is well beyond possible at this moment. 48 segments is the current limit and higher is imaginable, but lithographic constraints in pixel dimension and in getting interconnects towards a dense stacking of such pixels seems to be highly challenging. On the other hand, if making such higher detail phase plates becomes possible, it would also solve the issue of higher-order aberrations, and could further increase the spatial resolution of EM, reaching a numerical aperture that is closer to what is possible in light optics.

We hinted at adaptive algorithms, which are undoubtedly promising, but future simulation work would be needed to explore this idea further. Such algorithms could likely improve the imaging of thick samples, which might be especially interesting for applications such as, e.g., life science imaging of viruses and cells.

We conclude by stating that depth of field and depth resolution are just one aspect where phase plates show promise, and many other applications and methods are expected to be developed in a very near future.

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