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**Aberration-corrected ADF–STEM depth sectioning and prospects for reliable 3D imaging in S/TEM**

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**Abstract**

The short depth of focus of aberration-corrected scanning transmission electron microscopes (STEMs) could potentially enable 3D reconstruction of nanomaterials through acquisition of a through-focal series. However, the contrast transfer function of annular dark-field (ADF)-STEM depth sectioning has a missing-cone problem similar to that of tilt-series tomography. The elongation as a function of the probe-forming angle is found to be \( \sqrt{\frac{3}{2}} \frac{1}{\phi_{\text{max}}} \). For existing aberration-corrected STEMs operated at optimal imaging conditions, the elongation factor for depth sectioning is larger than 30. This large elongation factor results in highly distorted shapes of 3D objects and unexpected artifacts due to the loss of information. Depth-sectioning experiments using a 33-mrad 100 keV C₃-corrected aberration-corrected STEM demonstrate the elongation effect and the missing-cone problem in real and reciprocal space. The performance limits of different S/TEM-based imaging modes are compared. There is a missing cone of information for bright-field S/TEM, ADF-STEM, hollow-cone ADF-STEM and coherent scanning confocal electron microscopy (SCEM). Only incoherent SCEM fills the missing cone.

**Keywords**

depth sectioning, scanning transmission electron microscope, aberration-corrected STEM, scanning confocal electron microscope

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**Introduction**

Tilt-series electron tomography has grown increasingly popular as a method for determining the structure of materials at the nanometer scale [1–12]. With annular dark-field (ADF) and incoherent bright-field (BF) scanning transmission electron microscopy (STEM), diffraction artifacts can be minimized [13] and thick samples of strongly scattering materials [14] can be reliably reconstructed. However, tilt-series tomography is still primarily limited by the precision of image registration at different tilting angles and the time required to tilt the sample and record the data.

Traditionally, the depth of focus (DoF) of uncorrected STEM is comparable to or larger than the sample thickness generally studied by STEM. In the absence of channeling, ADF-STEM images provide an approximately linear projection view of the mass contrast of the specimen. However, with the development of aberration correctors, the ability to open the illumination angle greatly improves the depth resolution: a 3-fold improvement in the illumination angle would result in a 9-fold improvement in DoF [15]. The short DoF of aberration-corrected STEM (∼3–6 nm) potentially could enable 3D reconstruction of nano-objects by recording through-focal series in a similar fashion to optical microscopy depth sectioning [16–18]. Without the need to mechanically tilt the sample, this technique can potentially record data much more rapidly than tilt-series tomography, and with less sample movement depth sectioning should enable more precise image registration than would be possible by tilting the sample.

In amorphous materials, through-focal series are well described by the convolution of the three-dimensional point spread function (PSF) with the underlying 3D structure [18]. Intuitively, we might expect depth-sectioning reconstructions to be the original objects blurred out by ∼3–6 nm in the z direction. However, the 3D contrast transfer function (CTF) of STEM depth sectioning has a missing cone in the CTF [19,20] which results in almost all features being proportionally elongated by the same factor in depth-sectioning.
reconstructions. As the opening angle of the missing cone is equal to the probe convergence semi-angle \( (\alpha_{\text{max}}) \) \([19,20]\), the elongation factor scales inversely with \( \alpha_{\text{max}} \). For current aberration-corrected STEMs, the probe-forming aperture can only open up to 40 mrad in the best case, which results in an elongation factor of roughly 30 times. More typically an aperture of 20–25 mrad is used, yielding elongation factors of \(~50\) or more. This would mean that a 10-nm-diameter nanoparticle would be stretched over 500 nm. Such large elongation factors distort the shapes of the reconstructed objects and can produce unexpected artifacts. This is potentially dangerous if one interprets the reconstructions without a priori knowledge of the underlying artifacts. To demonstrate the elongation effect and the missing-cone problem, we report experimental depth-sectioning results taken on a 100 keV \( C_3 \)-corrected Nion UltraSTEM with a 33-mrad aperture in this paper.

We also examine the performance limits of other S/TEM-based imaging modes. The analytic boundaries of the 3D CTFs of BF-STEM/TEM, hollow-cone illuminated ADF-STEM, coherent BF-scanning confocal electron microscopy (SCEM) and incoherent SCEM are derived. It is found that BF-STEM/TEM only transfers information on two parabolic surfaces, and both hollow-cone illuminated STEM and coherent BF-SCEM have the same missing-cone problem as ADF-STEM. In contrast, incoherent SCEM is the only mode of those investigated which does not display a missing-cone artifact.

The rest of the article is organized as follows. In Methods we discuss ADF-STEM depth sectioning. Theory is presented – including an explanation and derivation of the elongation factor as a function of the probe-forming aperture size. In Results, we first present experimental evidence for the elongation effect and the missing-cone problem. Then we discuss the elongation artifacts for extended structures and the special properties of imaging point-like/1D/2D objects. The Discussion section examines the reliability of alternative imaging modes in depth sectioning.

**Methods**

The depth sectioning of amorphous material can be approximated by a convolution of the incoherent 3D PSF with the underlying material \([18]\):

\[
I(x, y, z) = \iiint O(x', y', z') \times \text{PSF}
\]

\[
(\pi \lambda(k_{df}) \chi(k; df = df_0 + z) - 2\pi |\mathbf{k} \cdot \mathbf{r}| d^2\mathbf{k})^2
\]

where \( A \) is a normalization factor which only depends on \( \alpha_{\text{max}}/\lambda \), \( df \) is defocus, \( df_0 \) is the reference defocus where the center of the PSF is defined, \( H \) is the aperture function which is 1 for \( k \leq \alpha_{\text{max}}/\lambda \) and 0 for any \( k > \alpha_{\text{max}}/\lambda \) and \( \chi \) is the phase error due to geometrical aberrations and defocus. If we neglect non-spherical aberrations and assume a monochromatic source, the expression for \( \chi \) up to the seventh order is

\[
\chi(k; df) = \frac{\pi df \lambda^2 k^2}{\lambda} \alpha \left[ C_3 \lambda^4 k^4 + C_5 \lambda^6 k^6/6 + C_7 \lambda^8 k^8/8 + O(k^{10}) \right] - \pi df \lambda^2 k^2
\]

where \( C_3, C_5 \) and \( C_7 \) are the third-, fifth- and seventh-order spherical aberration coefficients, respectively. The finite energy spread of the source can lead to a chromatically limited probe when the major geometric aberrations are corrected, in which case an additional integral over chromatic defocus spread is also required in Eq. (2) \([19,25,26]\).

Because a Fourier transform translates between convolution and multiplication of functions, the physical formation of depth sectioning can be understood more transparently in reciprocal space as

\[
\tilde{I}(k) = \mathcal{F} [I(x, y, z)] = \mathcal{F} [O(x, y, z) \times \text{PSF}(x, y, z)] = \tilde{O}(k) \cdot \tilde{\text{PSF}}(k) = \tilde{\tilde{O}}(k) \cdot \text{CTF}(k)
\]

where \( \mathcal{F} \) denotes 3D Fourier transform and CTF is the 3D contrast transfer function, which is the 3D Fourier transform of the PSF defined in Eq. (2). Depth sectioning in reciprocal space is a simple point-wise product of the 3D CTF and the Fourier transform of the 3D sample, and thus the CTF describes how the final depth-sectioning reconstruction responds to the original object at different spatial frequencies. In particular, if any part of the CTF is zero, the information at the corresponding spatial frequency is not transferred to the final image.

The 3D CTF of ADF-STEM depth sectioning is only non-zero between the two parabolas in \( k_r \) and \( k_z \) described by the relation

\[
\frac{1}{2} \lambda k_{\text{max}}^2 \left[ 1 - \left( \frac{k_r}{k_{\text{max}}} \right)^2 \right] < k_z < \frac{1}{2} \lambda k_{\text{max}}^2 \left[ 1 - \left( \frac{k_r}{k_{\text{max}}} \right)^2 \right]
\]

where \( k_{\text{max}} = \alpha_{\text{max}}/\lambda \) \([19,20]\). Figure 1b shows the numerically simulated 3D CTF of a 100 keV 33-mrad aberration-corrected STEM, demonstrating that information outside the two parabolas is not transferred to the final depth-sectioning reconstruction.

At low spatial frequencies,

\[
\frac{dk_z}{dk_r} \bigg|_{k_r=0} = \lambda k_{\text{max}} = \alpha_{\text{max}}.
\]

the opening angle of the CTF is exactly equal to the convergence semi-angle of the microscope. Therefore, taking a depth-sectioning reconstruction is essentially the same as
versely proportional to the convergence angle. As shown in Eq. (8), the elongation factor is roughly in-
horizontal spatial frequency, resulting in the proportionality frequency for the vertical information is proportional to the
this wedge (or cone) shape in reciprocal space, the cut-off
information in the CTF, the main lobe of the ver-
i.e. $A$ had corresponding Fourier information in the missing cone. (Fig. 2a). However, due to the
loss of features that would have
of the missing-cone problem. Under some circumstances, a
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new field of view in electron microscopy
conventional tilting tomography with a max tilt angle equal to the convergence angle of the microscope (Fig. 1c and d). In tilt-series tomography, a tilt range smaller than $\pm 90^\circ$ results in an elongation of the reconstructed object in the direction of the missing information [27,28]. The elongation factor [28] for conical tilting tomography [27] is given by

$$\epsilon_{xx} = \epsilon_{yy} = \sqrt{\frac{3 - \sin^2 \alpha_{\text{max}}}{2 \sin^2 \alpha_{\text{max}}}} \tag{7}$$

where $\alpha_{\text{max}}$ is the max tilt angle in tomography. The CTF of the tilting tomography and STEM depth sectioning takes a similar form; we can replace the max tilt angle in Eq. (7) by the convergence angle to estimate the elongation factor for STEM depth sectioning. Because the STEM convergence angle is much smaller than 1, we can take a limit as $\alpha_{\text{max}}$ goes to 0:

$$\epsilon_{xy} = \epsilon_{xz} = \lim_{\alpha_{\text{max}} \to 0} \sqrt{\frac{3 - \sin^2 \alpha_{\text{max}}}{2 \sin^2 \alpha_{\text{max}}}} = \sqrt{\frac{3}{2} \frac{1}{\alpha_{\text{max}}}}. \tag{8}$$

As shown in Eq. (8), the elongation factor is roughly inversely proportional to the convergence angle.

The elongation effects of both tilting tomography and STEM depth sectioning result from the missing wedge (or missing-cone) problem. If there is missing information in this wedge (or cone) shape in reciprocal space, the cut-off frequency for the vertical information is proportional to the horizontal spatial frequency, resulting in the proportionality

For example, consider a 3D square box with a width of $L$. In reciprocal space, the spatial information approximately extends to $k = 1/L$ because the Fourier transform of a solid square box is a product of three sinc functions, i.e. $A \sin(nk_{xL}) \sin(nk_{yL}) \sin(nk_{zL})$ (Fig. 2a). However, due to the missing-cone problem in the CTF, the main lobe of the vertical information is cut off outside $k_z = [\alpha_{\text{max}}/L]$ (Fig. 2b). Using the reciprocity, this is a feature of length $L/\alpha_{\text{max}}$ in real space in a first-order approximation (Fig. 2c); therefore, the elongation factor is approximately $1/\alpha_{\text{max}}$.

However, this simple analysis misses the leading $\sqrt{3}/2$ factor from the missing cone in Eq. (8). It is worth pointing out that the leading factor from the missing wedge of single-axis tilt-series tomography is $\sqrt{3}$ (lim $n \to \infty \sqrt{\frac{\sin(n\pi)}{\sin(n\pi/3)}} = \sqrt{3}$ $\times \sqrt{3}$ [13,28]). By reducing the wedge-shaped missing trench to a missing cone, a $\sqrt{2}$-fold improvement in reducing the elongation is obtained.

The elongation effect is only a first-order approximation of the missing-cone problem. Under some circumstances, a more serious problem is the loss of features that would have had corresponding Fourier information in the missing cone. This effect can be potentially more damaging than the blur, because it is strongly specimen dependent and thus more difficult to analyze and predict.

For existing aberration-corrected STEM, the probe-forming aperture can only open up to $20–40$ mrad. As explained above, in theory, this leads to a $177.7–178.9^\circ$ missing-cone problem and an approximate elongation factor of $30–60$ in depth sectioning. However, STEM depth sectioning has attracted considerable interest due to the availability

Fig. 1. (a) The central cross section of the 3D PSF of a 100 keV 33-mrad STEM ($C_1 = -0.0749$ mm, $C_3 = 68.79$ mm); (b) the corresponding 3D CTF shown in log scale; (c) an illustration of the boundary of the 3D CTF of ADF-STEM depth sectioning and (d) an illustration of the sampling of the Fourier space of tilt-series tomography.

Fig. 2. A simple explanation of the elongation effect: (a) the main lobe of the Fourier transform of a solid box with a width of $L$; (b) central cross section of the cone-missing CTF and (c) the remaining information transferred.
Fig. 3. Nine selected images of a 101-frame STEM through-series of a 6-nm gold particle (100 keV, $\alpha_{\text{max}} = 33$ mrad, FoV = 13.9 nm, NoP = 512 x 512, dwell time = 20 $\mu$s, ADF collection angle = 80–240 mrad, probe current = 60 pA): (a–f) defocus = −192, −88, −8, −4, 0, 4, 8, 88 and 192 nm, respectively.

of aberration-corrected STEMs. While most studies largely focus on the advantages of the short DoF of aberration-corrected STEM, less attention has been paid to the inherent problems of this technique. In the next section, we will explore both the elongation effect and the missing-cone problem in experiments.

Results

In order to understand the elongation effect and the missing-cone problem experimentally, a 33-mrad 100 keV C$_5$-corrected Nion UltraSTEM was used to take STEM through-focal series. The sample used in the experiments is a standard combined test specimen with gold particles on a carbon support. The defocus of the microscope is controlled by changing the high-tension voltage. The defocus was calibrated by measuring the aberration coefficients using Ronchigrams of the combined test specimen [29] for a series of high-voltage settings. The calibration coefficient was measured to be 11.1 nm V$^{-1}$, which is very close to that expected from the chromatic aberration of the optics. The approximate DoF of this operating setting is 5.8 nm [19,30]. The defocus spread due to the energy spread of the gun is approximately an additional 3 nm. The ADF collection angle is 80–240 mrad. Through-focal image acquisition was automated in Digital Micrograph.

The elongation effect

Figure 3 shows typical STEM images of a 6-nm gold particle at nine different defocus settings taken from a larger through-focal series of a total of 101 images acquired at 4 nm defocus intervals. The field of view (FoV) is 13.9 nm, the number of pixels (NoP) is 512 x 512, the dwell time is 20 $\mu$s and the probe current is $\sim$60 pA. The DoF and the chromatic defocus blur are 5.8 nm and 3 nm, respectively; thus, a 4-nm step size is neither undersampling nor oversampling. The resulting images are also close enough for automated post-imaging registration for removing drifts and shifts during acquisitions.

The reconstruction is built by stacking the 101 images together along the $z$ direction (the systematic shift in the series is removed by least-squares minimization). Figure 4b shows the central slice of the reconstruction in square pixels (i.e. equal number of nanometers per pixel along the $x$ and $z$ axes) and (c) the $z$ axis is compressed 15-fold (i.e. nanometers per pixel of the $z$ axis is 15-fold bigger than that of the $x$ axis); (d) the simulated reconstruction of a 6.3 nm solid sphere (the $z$ axis is compressed 15-fold; the black dashed line outlines the original solid sphere) and (e) the comparison of the central vertical line profile.

Fig. 4. Depth-sectioning reconstruction of a 6-nm gold particle: (a) central slice of the gold particle. The white dashed-line labels where the central cross section is taken; (b–c) the central cross section of the reconstruction; (b) plotted in square pixels (i.e. equal number of nanometers per pixel along the $x$ and $z$ axes) and (c) the $z$ axis is compressed 15-fold (i.e. nanometers per pixel of the $z$ axis is 15-fold bigger than that of the $x$ axis); (d) the simulated reconstruction of a 6.3 nm solid sphere (the $z$ axis is compressed 15-fold; the black dashed line outlines the original solid sphere) and (e) the comparison of the central vertical line profile.
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The missing-cone problem

The elongation effect shown in the last section is the first-order consequence of the missing-cone problem. Missing information outside the cone is potentially more problematic than an elongation blur, especially if a $>170$° cone of information is missing. In order to show the missing cone from experimental data, we need to apply a 3D discrete Fourier transform to the reconstruction. Pixel size in reciprocal space is reciprocal to the full dimension in real space. In order to obtain a 3D diffractogram with sufficient detail, a through-focus series needs to cover a very wide defocus range. Figure 5 shows a through-focus series of a 5.6 nm gold particle that goes $\pm 500$ nm out of focus. The detail of the missing-cone problem is clearly demonstrated in the 3D diffractogram (Fig. 5c). The opening angle of the missing cone is measured to be the same as the convergence angle ($\theta_{\text{max}} = 33$ mrad). The diffractogram fringes are a 3D analog of the single-slit diffraction pattern. Moreover, as the gold particle is approximately round, the fringe envelope can be approximated by the 3D Fourier transform of a solid sphere as shown in (Fig. 5d.).

Extended objects versus point/1D/2D objects

In previous sections, we have shown the elongation effect and the missing-cone problem. The experimental results agree well with the theoretical predictions. While existing aberration correctors can correct geometrical aberrations up to the fifth order [31], allowing us to open the aperture up to 50 mrad, chromatic aberrations remain as the limiting factor in existing machines. In order to balance the defocus blur due to the energy spread of the gun and the instability of the lens current, the aperture size needs to be lowered to 20–35 mrad to achieve optimal image resolution [19,26,32]. Consequently, the elongation factor of existing machines is 30–60 times, which may result in unexpected artifacts.

To investigate elongation artifacts of extended structures in a technologically relevant context, a toy structure is built as shown in Fig. 6a, which mimics a high-Z catalyst particle on a low-Z hollow support. Real-world examples measured by tilt-series tomography can be found in [2,33], which clearly show when particles lie inside or outside the support. In contrast, from the simulated depth-sectioning reconstruction (Fig. 6b), it is very difficult to tell where the catalyst particle was relative to the support. Indeed it appears that instead of sitting on the support, the catalyst particle is inside the cavity. This is despite the fact that the support is thicker than the DoF of the microscope and still the particles appear to lie inside the cavity. This illustrates the major consequence of the elongation factor – that larger objects are distorted over larger distances than small objects. This type of artifact is likely to be very common when depth sectioning small particles in extended amorphous media by aberration-corrected STEM.
In a depth-sectioning experiment, users would likely terminate a through-focal series when the main features start to become blurred. In this case, elongation effects of big features are not fully noticeable because only a limited range is imaged. However, any features larger than two times the probe size \((> 2 \times \alpha_{\text{max}}/\lambda)\) are elongated in a similar fashion even though it may not be readily apparent, especially when the objects are embedded in amorphous matrix. This is very dangerous if one interprets the reconstructions without realizing that all the features are elongated.

The elongation effect and the missing-cone problem considerably limit the application of STEM depth sectioning for extended objects. However, for isolated point-like/1D/2D objects – such as individual atoms, on-axis atomic columns and atomic planes, the situation is less bleak. One might naively expect that a point object remains point-like even after the elongation factor is applied. A more rigorous argument as to why the missing-cone problem can be neglected for resolvable 1D and 2D objects can be obtained in reciprocal space. These 2D objects are infinitely long in reciprocal space. These 2D objects – such as individual atoms, on-axis atomic columns and atomic planes, the object must be smaller than the smallest probe dimension. This would be the case for a dopant atom in an amorphous matrix. While in principle, depth sectioning could be applied to locating dopant atoms inside a crystalline matrix, channeling effects confound the interpretability of the local-ization of dopant atoms in the z dimension \([17,18,34–36]\).

It is important to note that the previous discussion assumes very little or no prior knowledge of specimens. However, in some cases, prior knowledge is available. For example, a catalyst particle analysis specimen can be prepared by spraying catalyst particles over a carbon grid and, in this instance, it is known before beginning characterization that the particle is sitting on the grid \([37]\). In STEM, one can focus on a particle first and then on the substrate to reveal the height of the particle in the beam direction; this method has depth precision of several nanometers, limited by counting, scan and read-out noise, chromatic blur and the DoF. Quantitative calculations and simple formula for these limits can be found in \([18,19]\).

**Discussion**

ADF-STEM depth sectioning, at the current stage, is subject to severe elongation effects and artifacts from the missing cone of information. It is very difficult to obtain a directly interpretable 3D reconstruction with this technique without prior knowledge of the specimen. However, in addition to ADF-STEM, current microscopes can be operated in BF-STEM/TEM, hollow-cone illuminated ADF-STEM and SCEM modes. We examine the performance of these modes for focal-series tomography in the following sections.

**BF-STEM/TEM**

If a weak-phase object and single scattering is assumed, a BF-STEM/TEM image can be approximated by

\[
I \approx 1 + 2\sigma_i v(x, y, z) \otimes \text{PSF}_{BF}(x, y, z)
\]

where \(\sigma_i\) is the impact factor, \(v(x, y, z)\) is the atomic potential and \(h(x, y, z)\) is the 3D PSF for BF imaging in weak-phase approximation \([38]\). The CTF can then be expressed as

\[
\text{CTF}_{BF}(k_x) = \int \text{PSF}_{BF}(x, y, z) \exp(i2\pi k_x x) \, dx
\]

\[
= \int \sin \left[ \chi(k_z, z = df - d_f) \right] \exp(i2\pi z k_z) \, dz
\]

\[
= \int \sin \left[ \chi_0(k_z) + \pi \lambda^2 k^2 z \right] \exp(i2\pi z k_z) \, dz
\]

\[
= \frac{1}{\pi} \left\{ \exp \left[ -i \left( \chi_0(k_z) + \pi \lambda^2 k^2 z d_f \right) \right] \delta \left( k_z - \frac{1}{2} \lambda k^2 \right) \right\} - \exp \left[ i \left( \chi_0(k_z) + \pi \lambda^2 k^2 z d_f \right) \right] \delta \left( k_z + \frac{1}{2} \lambda k^2 \right)
\]

(11)

Thus, it is found that the BF-TEM only transfers information on the two parabolic surfaces \((k_z = \frac{1}{2} \lambda k^2)\) and \((k_z = -\frac{1}{2} \lambda k^2)\) (Fig. 8a). With partial coherence included, the two surfaces will be smeared out \([38, 39]\), allowing for more information to be transferred – not enough, however, for a
reliable 3D reconstruction, or to be competitive with the other methods. Nevertheless, in the extreme case that the TEM illumination angle \( \alpha_{\text{max}} \) is comparable or even bigger than the aperture angle \( \alpha_{\text{max}} \) of the post-specimen objective lens, where the coherent length of the illumination is smaller than the resolving power of the objective lens, the image formation becomes incoherent [15, 38] and the 3D CTF is the same as that of ADF-STEM [39]. This is an upper bound on the CTF, and for partially coherent illumination, the 3-D BF CTF will always lie within these limits.

**Hollow-cone illuminated STEM**

Hollow-cone illumination in STEM can reduce channeling artifacts [36] when the probe-forming aperture is increased above 50 mrad. This result is physically intuitive because a high-angle incident electron would tend to leave the column rather than staying on the column due to the higher transverse momentum. By only allowing the weakly confined electrons to probe the sample, channeling is significantly reduced. Following the derivation in appendix C of [19], the CTF of hollow-cone illuminated ADF-STEM can be expressed as

\[
\text{CTF}(k_x, k_y) = \frac{8\pi^2}{k_{\text{max}}^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{k_{\text{min}}}^{k_{\text{max}}} d\kappa_z d\kappa_x d\kappa_y \\
\times \int_{\kappa_{\text{min}}}^{\kappa_{\text{max}}} d\kappa_z \delta(2\pi k_x \kappa_z - J_0(2\pi k_x r) J_0(2\pi k_y r) \\
\times \exp\left[i \pi \lambda (k_x^2 - k_y^2)\right] \exp\left[i (\chi_r(k_x) - \chi_r(k_y))\right] \\
\times \exp\left(2\pi i k_z z\right) k_x k_y k_z \\
\int_{-\infty}^{\infty} dz \exp\left(i 2\pi k_z z\right) \exp\left[i \pi \lambda (k_x^2 - k_y^2) z\right]
\]

where \( k_{\text{min}} = \alpha_{\text{min}} / \lambda \) and \( k_{\text{max}} = \alpha_{\text{max}} / \lambda \). The integral with respect to \( z \) is

\[
\int_{-\infty}^{\infty} dz \exp\left(i 2\pi k_z z\right) \exp\left[i \pi \lambda (k_x^2 - k_y^2) z\right] \\
= \frac{k_x + \frac{\lambda}{2} (k_x^2 - k_y^2)}{k_x^2 - k_y^2}.
\]

So, the CTF is only non-zero for \( k_z = \frac{\lambda}{2} (k_x^2 - k_y^2) \). Because \( k_{\text{min}} < |k_z| < k_{\text{max}} \) and \( k_{\text{min}} < |k_z| < k_{\text{max}}, \) the range of \( k_z \) for non-zero CTF is \( -\frac{\lambda}{2} (k_{\text{max}}^2 - k_{\text{min}}^2) < k_z < \frac{\lambda}{2} (k_{\text{max}}^2 - k_{\text{min}}^2) \). The boundary for hollow-cone illuminated STEM is the same as that of the uniformly illuminated STEM but with the top and bottom cut-off (Fig. 7b). Therefore, the depth resolution is a factor of \( 1 - \alpha_{\text{min}}^2 / \alpha_{\text{max}}^2 \) worse than ADF-STEM and it is subject to the missing-cone problem as well.

**SCEM**

From light optics, it is known that for coherent imaging, the PSF of BF-SCEM is the product of the BF PSFs of the pre- and post-specimen lenses [40–46]:

\[
\text{PSF}_{\text{BF,confocal}} = \text{PSF}_1(x, y, z) \text{PSF}_2(-x, -y, -z) \\
= \int_{k_x}^{k_x_{\text{max}}} \exp\left[i \chi_r(k_x, r) + i \pi \lambda k_x^2 z\right] J_0(2\pi k_x r) d\kappa_x r \\
\times \int_{k_y}^{k_y_{\text{max}}} \exp\left[i \chi_r(k_y, r) + i \pi \lambda k_y^2 z\right] J_0(2\pi k_y r) d\kappa_y r.
\]

If the pre- and post-specimen lenses use the same illumination aperture, i.e. \( k_{\text{max}} = k_{\text{max}} \), then the derivation of the boundary of the CTF follows that for ADF-STEM as laid out in appendix C of [19]. Even though SCEM uses one more lens, it has exactly the same information limit as ADF-STEM in reciprocal space. From a reciprocal space information-transfer point of view, BF-SCEM and ADF-STEM have the same vertical information limit \( (1/k_{\text{c,max}} = 2\lambda/\alpha_{\text{max}}^2) \). However, from a real space point of view, the ADF-STEM PSF is much more localized than the SCEM weak-phase PSF. The most important conclusion is that the CTF of BF confocal has the same parabolic shape and is subject to the same missing-cone problem (Fig. 8c). This was observed experimentally when Nellist et al. recorded the elongation effect of a 3.1-nm gold particle in a recent experiment using a double-corrected SCEM with a stage-scanning system [47]. In another experiment, Takeguchi et al. showed atomic resolution images (gold [111]) acquired by an aberration uncorrected stage-scanning SCEM (200 keV, \( C_1 = 1.0 \) mm, \( \alpha_{\text{max}} = 10 \) mrad) [48]. This work reported a series of three-dimensionally scanned BF-SCEM images of platinum nanoparticles on a carbon film. The nanoparticles remain visible when the stage is displaced by \( \pm 260 \) nm from the in-focus position even though the displacement is 5-fold bigger than the DoF (DoF \( \approx 1/k_{\text{max}} = 2\lambda/\alpha_{\text{max}}^2 = 50 \) nm). This is again a consequence and demonstration of the elongation effect in BF-SCEM.

In the case of incoherent imaging [20, 40–45], the SCEM PSF can be expressed as a product of the incoherent PSFs of the pre- and post-specimen lenses:

\[
\text{PSF}_{\text{SCEM,confocal}} = |\text{PSF}_1(x, y, z)|^2 |\text{PSF}_2(-x, -y, -z)|^2.
\]
Fig. 9. Comparison of different imaging modes in depth-sectioning reconstruction (200 keV, \( \alpha_{\text{max}} = 40 \text{ mrad} \), \( C_1 = -0.0235 \text{ mm} \), \( C_3 = 14.70 \text{ mm} \), \( C_5 = 0 \text{ mm} \), \( d_{\text{fo}} = -70.5 \text{ Å} \)): (a) the cross sectioning of the original object. The object is infinitely long in the direction perpendicular to the plane of the paper (a–f) 3D reconstruction by (b) BF-STEM, (c) ADF-STEM, (d) hollow-cone illuminated ADF-STEM, (e) coherent BF-SCEM and (f) incoherent SCEM.

difficult to interpret. In contrast, incoherent SCEM is the only mode of those investigated in which the missing cone is filled. As shown in Fig. 9, by filling the missing cone in reciprocal space, the incoherent SCEM reconstruction in real space closely reproduces the features of the original object. However, the reliance on core shell inelastic scattering may considerably reduce the dose efficiency, signal and hence resolution for SCEM.

Conclusion

We observe a strong elongation effect and missing-cone problem with ADF-STEM depth sectioning, both in real and reciprocal space. The effects dominate in theory, simulation and experiments. The elongation factor is approximately \( \sqrt{\frac{1}{\alpha_{\text{max}}}} \). Thus, for existing aberration-corrected STEMs operated at optimal imaging conditions, the elongation factor is larger than 30, which can yield unexpected artifacts, such as particles outside a hollow shell appearing to lie inside the shell. Thus, one needs to be very cautious in interpreting 3D depth-sectioning reconstructions if no prior information is available.

We have also examined other S/TEM-based imaging modes in this work. The analytic boundaries of the 3D CTFs of BF-STEM/TEM, hollow-cone illuminated ADF-STEM, coherent BF-SCEM and incoherent SCEM are derived. It is found that BF-STEM/TEM only transfers information on two parabolic surfaces, and both hollow-cone illuminated STEM and coherent BF-SCEM have the same missing-cone problem as ADF-STEM. In contrast, incoherent SCEM is the only mode of those investigated in which the missing cone is filled. However, the reliance on an inelastic scattering mechanism in SCEM greatly reduces the dose efficiency of the method.

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