Multislice does it all—calculating the performance of nanofocusing X-ray optics

KENAN LI,¹ MICHAEL WOJCIK,² AND CHRIS JACOBSEN²,3,4,*

¹Applied Physics, 2145 Sheridan Road, Northwestern University, Evanston, IL 60208, USA
²Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439, USA
³Department of Physics and Astronomy, 2145 Sheridan Road, Northwestern University, Evanston, IL 60208, USA
⁴Chemistry of Life Processes Institute, 2170 Campus Drive, Northwestern University, Evanston, IL 60208, USA
* cjacobsen@anl.gov

Abstract: We describe an approach to calculating the optical performance of a wide range of nanofocusing X-ray optics using multislice scalar wave propagation with a complex X-ray refractive index. This approach produces results indistinguishable from methods such as coupled wave theory, and it allows one to reproduce other X-ray optical phenomena such as grazing incidence reflectivity where the direction of energy flow is changed significantly. Just as finite element analysis methods allow engineers to compute the thermal and mechanical responses of arbitrary structures too complex to model by analytical approaches, multislice propagation can be used to understand the properties of the real-world optics of finite extent and with local imperfections, allowing one to better understand the limits to nanoscale X-ray imaging.

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References and links
1. Introduction

Significant advances have been achieved in the development of nanofocusing x-ray optics, leading to demonstrations of x-ray beams focused to dimensions approaching 10 nm or better [1–3] and focal spots two or three times larger in practical experiments. While this does not approach the resolution available in electron microscopes, x-rays are able to penetrate materials hundreds of micrometers thick, and due to the lack of bremsstrahlung background they offer a thousandfold improvement in trace element detection in thick specimens [4]. As a result, these advances in x-ray nanofocusing are providing new, complementary scientific insights in biomedical and materials research [5, 6]. These advances have been coincident with a worldwide investment in new free-electron laser and diffraction-limited storage ring x-ray sources, so that the brightness
of x-ray sources has been increasing faster than Moore’s “law” in microelectronics.

As impressive as these results are, in all cases the achieved optical performance is inferior to what would be predicted from simple theory. In addition, simple theory does not suffice for describing more advanced x-ray optics. For Fresnel zone plates, scalar diffraction theory applied to the total complex transmittance of coarse zone structures works well for thin optics [7], but already there are nanofabrication advances [8–10] that produce zone structures approaching small multiples of the x-ray wavelength \( \lambda \) in width, and with a thickness such that a simple projection of material thickness is no longer valid for predicting the wavefield exiting the optic and converging to the focus. Realizing this, approaches such as coupled wave theory (CWT) have been shown to be necessary for accurate predictions of the properties of thicker zone plates [11] and in particular for zone structures that are at an angle relative to the illumination direction [12, 13]. The more recent development of multilayer Laue lenses [14], where thin film deposition techniques are used to produce volume grating structures tilted to meet the Bragg condition, provides another example where CWT-based approaches have been required. Together these analytical models are providing a good deal of insight into design approaches for nanofocusing x-ray optics.

However, analytical models fall short of dealing with the complexity of real optics. Methods such as CWT assume a simple, geometrically-definable grating of infinite width and constant periodicity. While this can serve as an approximation of a local region of a diffractive optic, in Fresnel zone plates and in multilayer Laue lenses the width \( dr_n \) of the \( n \)th zone changes as \( dr_n = \sqrt{A f / (2 \sqrt{n})} \), so as the integer zone index \( n \) (and the radius \( r_n^2 \approx n \lambda f \)) changes so does the grating period (though approximations can be used to accommodate zone period variations [15]). In addition, because these diffractive zones are often so small that they are at the limit where nanolithographic processes begin to fail, or x-ray mirrors suffer from surface roughness and figure errors which degrade their performance, analytical approaches applied to structures described with perfect geometry are incapable of representing real, as-fabricated nanofocusing optics. To take an analogy, basic physics tells us how to analytically calculate heat transfer or bending radii in simple objects such as rectangles and cylinders, but it is inadequate for calculating the properties of more complex objects with irregular machined shapes; in such cases, one must turn to numerical approaches such as finite element analysis to obtain a realistic picture.

2. Multislice wave propagation

Multislice methods have been used in several calculations involving weak, forward direction optical interactions in electron [16, 17] and x-ray [18–21] optics. It has some similarities to partial wave solutions that have been applied to Fresnel zone plate simulations [22, 23] and to image formation from low-contrast specimens [24]. We show below that the multislice approach can be applied to an even broader range of phenomena including grazing incidence x-ray reflectivity [25] and x-ray standing waves, as well as high aspect ratio, tilted, and imperfect (as-fabricated) diffractive optics.

The multislice approach is illustrated in Fig. 1. A 3D object is discretized into a set of voxels at positions \( (x, y, z) \), each of which is filled with a medium with refractive index

\[
n(x, y, z) = 1 - \delta(x, y, z) - i \beta(x, y, z),
\]

where \( \delta(x, y, z) \) represents the phase shifting part of the x-ray refractive index and \( \beta(x, y, z) \) represents the absorptive part [27]. When an incident plane wave \( \psi_j(x, y) \) encounters a slab with a thickness \( \Delta z \), the wavefield is modified by the projected net optical response of the slab according to

\[
\psi'_j(x, y) = \psi_j(x, y) \exp \left( \frac{2 \pi \Delta z}{\lambda} \left( \delta(x, y, z_j) + i \beta(x, y, z_j) \right) \right)
\]
Fig. 1. Schematic representation of the method of multislice propagation, used to simulate a wavefield propagating though a non-homogeneous refractive medium [26]. Along the beam direction, the object is represented by a series of slices separated by distances $\Delta z$. At the entrance of a slice, the incident wavefield $\psi_j$ is first modulated by the refractive effects of the slab of material, leading to a modified wavefield $\psi'_j$ at the same plane. This wavefield is then propagated to the next slab entrance, yielding the next slice’s wavefield of $\psi_{j+1}$. Eventually one arrives at the exit wave leaving the medium, which can then be transferred to a far-field diffraction pattern using free-space propagation.

where $z_j$ represents the position of the $j^{th}$ slab in an extended object. This modulated wavefield is then brought to the entrance of the next, $(j + 1)^{th}$ slab using free space propagation [28] as given by

$$
\psi_{j+1}(x, y) = \mathcal{F}^{-1}\left\{ \mathcal{F}\{\psi_j(x, y)\} \cdot \exp\left[-i \frac{2\pi \Delta z}{\lambda} \sqrt{1 - \lambda^2 (u_x^2 + u_y^2)} \right] \right\}
$$

where $\mathcal{F}\{\cdot\}$ represents a Fourier transform and $\mathcal{F}^{-1}\{\cdot\}$ its inverse, $\lambda$ is the x-ray wavelength, and $u_{x,y}$ are spatial frequencies. In the case of discrete sampling over $N_t$ pixels of transverse sizes $(\Delta x, \Delta y)$, the spatial frequencies go up to a maximum Nyquist sampling limit of $u_{x,\text{max}} = 1/\Delta x$ and $u_{y,\text{max}} = 1/\Delta y$, and the pixel sizes $(\Delta x, \Delta y)$ remain constant in the process of multislice propagation. When subsequently propagating a wavefield in vacuum over a distance longer than $N_t (\Delta x \Delta y)^2 / \lambda$, sampling considerations [28, 29] dictate the use of an alternative propagation approach of

$$
\begin{align*}
\psi_{j+1}(u_x \lambda z, u_y \lambda z) & = & \mathcal{F}\left\{ \psi_j(x, y) \cdot \exp\left(-i\frac{x^2 + y^2}{\lambda z} \right) \right\} \\
& & \cdot \frac{i}{\lambda z} \exp\left[-i\pi \frac{\lambda^2 (u_x^2 + u_y^2)}{\lambda z} \right].
\end{align*}
$$

Multislice propagation calculations require that the object interacting with the x-ray beam be described by a three-dimensional refractive index distribution $n(x, y, z) = 1 - \delta(x, y, z)$ –
$i\beta(x, y, z)$. For a numerical calculation, this means that the object must be mapped onto a discrete grid of voxels; this is discussed further in Sec. 6. The main limitation is that the multislice approach cannot represent backwards propagation, though Bragg diffraction at $2\theta$ angles greater than $90^\circ$ can be handled by tilting both the beam axis and the optical structure relative to the propagation axis.

In order to apply the multislice method to a finely-gridded cylindrically-symmetric optic such as a zone plate, it is far more efficient to use a one-dimensional Hankel transform [28–31] in the radial direction rather than a two-dimensional Fourier transform in Cartesian coordinates. This approach is applied to the zone plate simulations in Sec. 5 below. The calculation grid size involved $4 \times 10^5$ pixels of width $\langle \Delta r \rangle = 1$ nm, and $N = 64$ slice steps along the beam propagation direction.

Alternative approaches to multislice propagation include the finite-difference time-domain (FDTD) method [32–34] which is well suited to dealing with structures smaller than or somewhere on the order of the electromagnetic wavelength. For x-ray calculations on nanoscale amorphous materials described by a refractive index distribution (thus neglecting Bragg diffraction from crystalline planes), this level of detail is unnecessary. In such cases, the multislice approach is less computationally demanding than FDTD because it is a position-evolved method based on slice-by-slice wavefield propagation with gridding at the length scale of the finest features in the optical structure, whereas FDTD is time-evolved and requires that the entire computational domain be gridded on a transverse and longitudinal length scale that is small compared to the x-ray wavelength.

3. X-ray reflectivity

Having discussed the multislice method, we show here the surprising result that it can be applied to complicated phenomena with stronger net effects on an x-ray wavefield. One of the strongest modifications one can apply to a wavefront is its reflection, where the Poynting vector describing electromagnetic energy flow is completely redirected. Reflectivity is normally calculated using boundary conditions for both the electromagnetic and magnetic fields, but magnetic effects are usually weak in x-ray optics and we show here that the multislice method can be used without explicitly ensuring that electric field boundary conditions are met.

Because the x-ray refractive index [35] of $n = 1 - \delta - i\beta$ differs only slightly from unity with $\delta \approx 10^{-5}$, the normal incidence reflectivity $\delta^2/4$ from a single refractive interface is vanishingly small [25] except for the case of normal incidence synthetic multilayer structures [36]. In grazing incidence x-ray reflectivity, the $n < 1$ nature of the x-ray refractive index means that what would be total internal reflection of visible light at the interface from a medium to vacuum becomes total external reflection of x-rays at the interface from vacuum to a medium. The critical angle $\theta_c$ for grazing incidence x-ray reflectivity is given by [25] the grazing angle $\theta_c = \sqrt{2\delta}$. Detailed calculations [35] of the x-ray reflectivity versus grazing incidence angle $\theta$ show that reflectivities of 80% or more can be achieved at grazing incidence angles below $\theta_c$ in excellent agreement with experimental observations. We show in Fig. 2(b) a comparison between the well-known theoretical result and the result obtained by multislice propagation; it shows that the multislice approach accurately represents the outcome of x-ray grazing incidence reflectivity, even though it starts from a different basis than the electric and magnetic field boundary conditions of classical Fresnel reflectivity theory. The multislice approach allows one to see multiple x-ray optical effects in one calculation. In Fig. 2(c,d), we show a 10 keV plane wave incident onto a slit aperture, followed by a grazing incidence reflective surface of gold. This figure shows four different manifestations of x-ray optical interactions simultaneously: 1) diffraction from the slit, with diffraction fringes increasing in strength and widening the beam as it propagates; 2) the angular dependence of x-ray reflectivity, with poor reflectance at 11 mrad grazing angle of incidence since it is above the critical angle of $\sqrt{2\delta} = 7.73$ mrad; 3) standing wave phenomena above the
Fig. 2. Calculating x-ray reflectivity using multislice propagation. The method of the calculation is shown in (a), where complications of diffraction from beam-defining apertures (as shown in c and d) are removed. A 10 keV x-ray plane wave is incident at the same grazing angle $\theta$ on gold reflective surfaces angled from both the bottom and the top. Within regions $I_1$ and $I_3$, a fraction $r$ of the beam is reflected off of mirror surfaces, while the beam in region $I_3$ is unaffected. The output region $I_4$ contains the interference between $rI_1$, $I_2$, and $rI_3$ which leads to local maxima and minima due to beam interference, but the total energy in $I_4$ is unaffected by these local redistributions. As a result, we have $r(I_1 + I_3) + I_2 = I_4$, or $r = (I_4 - I_2)/(I_1 + I_3)$ for the reflectivity of the optical surfaces. The resulting reflectivity curve $r(\theta)$ obtained by multislice propagation is shown in (b), plotted alongside the classical formula obtained by Henke [35] for unpolarized radiation (polarization effects are unnoticeably small in this case). As can be seen, the two curves are in excellent agreement, with only small differences of about 2–3% reflectivity at grazing incidence angles a bit below the critical angle of $\sqrt{2}\delta = 7.3$ mrad for Au at 10 keV. This shows that multislice propagation can reproduce even very strong x-ray beam effects due to refractive index variations. In (c) and (d), a 10 keV x-ray plane wave is incident on a 2 $\mu$m wide slit, followed by a gold reflective surface set at either 4 mrad (c) or 11 mrad (d) grazing angle of incidence. Dashed lines indicate the mirror surfaces. Multislice shows multiple optical effects in one calculation: diffraction from the slit, the angular dependence of x-ray reflectivity, standing wave phenomena above the surface of the mirror (magnified in the inset sub-images), and x-ray beam penetration over a limited distance into the mirror surface.
Table 1. Critical angles in mrad for grazing incidence x-ray reflection as calculated first using the theoretical result of $\sqrt{2}\delta$, and then as calculated at the maximum slope in the multislice reflectivity curve generated as shown in Fig. 2.

<table>
<thead>
<tr>
<th>Material</th>
<th>X-ray energy</th>
<th>6 keV</th>
<th>10 keV</th>
<th>30 keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Au</td>
<td>Multislice</td>
<td>12.97</td>
<td>7.76</td>
<td>2.70</td>
</tr>
<tr>
<td></td>
<td>$\sqrt{2}\delta$</td>
<td>13.06</td>
<td>7.73</td>
<td>2.67</td>
</tr>
<tr>
<td>Ir</td>
<td>Multislice</td>
<td>14.00</td>
<td>8.30</td>
<td>2.91</td>
</tr>
<tr>
<td></td>
<td>$\sqrt{2}\delta$</td>
<td>14.07</td>
<td>8.27</td>
<td>2.87</td>
</tr>
</tbody>
</table>

The calculation of reflectivity in Fig. 2(a) was carried out in a way that avoided the diffractive spreading of energy that follows a wave’s interaction with an aperture, which is why mirror surfaces were used at the top and the bottom of the transverse/longitudinal or $x-z$ field of view of the calculation. The calculation of reflection in Fig. 2(c) involves a 1D array with 1 nm transverse pixel size over a width of $10^4$ pixels, and $10^5$ steps in the propagation direction with a distance increment of 10 nm per step. Since Eq. (3) shows that two Fourier transforms are required for each propagation step, the calculation time is set in part by the need for $2 \times 10^5$ fast Fourier transform or FFT operations on a $10^4$ pixel 1D array, which takes less than 1 min on a standard laptop computer.

The reflectance versus grazing angle calculation of Fig. 2(b) showed a slight discrepancy between the multislice approach and the classical result [35], and this slight discrepancy is also illustrated in Table 1. We believe that this discrepancy comes primarily from the discretization of the multislice method, even when partial voxel filling is used as discussed in Sec. 6. In order to understand discretization effects further, we have carried out simulations of both a discretized but otherwise smooth mirror, and mirrors with deliberately-induced profile “roughness.” For a mirror with Gaussian-distributed random surface height errors characterized by a root mean square or RMS roughness of $\sigma$, the mirror reflectivity will be reduced by a Debye-Waller type factor $\eta_{\sigma}$ of

$$\eta_{\sigma} = \exp \left[ -\left( \frac{2\pi 2\sigma \sin \theta}{\lambda} \right)^2 \right] = \exp \left[ -(4\pi\sigma \sin \theta/\lambda)^2 \right]$$

where $\theta$ is the angle of grazing incidence. In Fig. 3, we show a simulation of reflectivity from several rough mirror surfaces, compared with the theoretical reflectivity [35] multiplied by $\eta_{\sigma}$. In the simulations, the rough mirror surface is represented by an array of Gaussian-distributed random numbers after a series of smoothing processes, which is locally smooth (within average range of 2-4 $\mu$m) and globally rough, as shown in Fig. 3(a). The simulation is carried out in the way similar to Fig. 2(a), and involves a 1D array with $\Delta x = 0.1$ nm transverse pixel size over a width of 30,000 pixels, and a distance increment of $\Delta z = 10$ nm in the propagation direction. As can be seen in Fig. 3(b), the simulation matches theoretical results very well for reflectivity of rough mirror surface. Fig. 3(c) shows the angular distribution of the reflected beam, indicating that rough mirror surfaces blur the angular distribution so as to cause some portion of the beam to be reflected into other directions. Such simulations tell us how roughness will affect the reflectivity thus the performance of x-ray mirror. This again demonstrates that the multislice approach is able to simulate optics with specific flaws in a way that analytical approaches cannot.
Fig. 3. Simulation of the effects of surface roughness on x-ray reflectivity. Because the multislice method can deal with any arbitrary optical object, in (a) we show three simulated surface profiles with Gaussian-distributed departures from a flat with standard deviations of $\sigma = 0$, 0.5, 1.0, and 2.0 nm RMS. They come from an array of Gaussian-distributed random numbers followed by doing rolling average over 5 points and rescaling to the desired standard deviations. Then two more times of rolling average over 250–1000 points are carried out for better local smoothness and then the arrays are rescaled again. These four different surface profiles were then used in a multislice calculation ($\Delta x = 0.1$ nm and $\Delta z = 10$ nm) for grazing incidence reflection of 10 keV x-rays from a gold surface using the approach of Fig. 2(a), yielding the multislice reflectivity curves shown here in (b); shown on the same plot is the theoretical reflectivity [35] multiplied by the roughness correction factors of Eq. (5) for each value of $\sigma$. The exit wave was then propagated to the far-field, yielding an angular distribution of light set first by the finite mirror width giving a $\sin(\theta)/\theta$ distribution for $\sigma = 0$ nm, and then further redistribution of light into larger angles as the roughness is increased to $\sigma = 0.5$, 1.0, and 2.0 nm.
4. Comparison with coupled wave theory

We next compare the multislice approach to coupled wave theory (CWT) for high aspect ratio zone plate structures [11]. We carried out our own CWT calculation using the approach described by Schneider et al. [13] to recreate their Fig. 8.10 result for non-1:1 line:space ratio zones slanted so as to meet the Bragg diffraction condition; our CWT results shown in Fig. 4(a) appear identical to those earlier published results. This figure also includes the multislice calculation results obtained using a grid with a transverse pixel size of $\Delta x = 3$ nm, and a slice step length of $\Delta z = 5$ nm in the propagation direction. The multislice results shown in Fig. 4(a) are essentially identical to the CWT results, again showing the validity of the multislice approach.

As noted above, CWT assumes a volume grating structure of infinite width and constant grating characteristics. However, real optics always have finite size, and Fresnel zone plates and multilayer Laue lenses have grating periods that vary across the optic; multislice propagation is able to handle these real-life circumstances. As one example, in Fig. 4(c) we show a simulation of a 2 $\mu$m wide grating with 29 nm:29 nm line:space ratio and a thickness of gold of $t = 6 \mu$m, as illuminated by 10 keV x-rays. The finite grating has 34.5 periods, and was simulated using $N = 64$ slices with $\Delta z = 31.25$ nm. As can be seen in Fig. 4(c), the multislice method and CWT calculations for the exit wave match very well except at the edges, which CWT cannot depict.

In Fig. 4(d), the exit waves are carried forward to the far-field diffraction pattern; the first order diffraction efficiency is almost identical, with a slight decrease in transmittance due to the edge effects shown in Fig. 4(c).

A crucial consideration in multislice propagation is the number $N$ of slices the sample should be divided into along the propagation axis (the beam axis and the propagation axis do not have to coincide, as illustrated in Fig. 2(c,d)). Since the transverse distance from an edge to the first Fresnel fringe in propagation scales as $\sqrt{\lambda z}$ where $z$ is the propagation distance, we wish to have the transverse pixel size $\Delta x$ be a small fraction $\epsilon_1$ of this distance, or $\Delta x = \epsilon_1 \sqrt{\lambda z}$. Nyquist sampling would suggest $\epsilon_1 \leq 0.5$ and the Rayleigh quarter wave criterion would suggest $\epsilon_1 \leq 0.25$. Since the depth of focus of an optic goes like $\lambda/\theta^2$, the transverse sampling condition implies that the longitudinal sampling be some small fraction $\epsilon_2$ of $z = \Delta x^2/(\epsilon_1^2 \lambda)$, or

$$\Delta z = \frac{\epsilon_2 \Delta x^2}{\epsilon_1^2 \lambda}. \quad (6)$$

We do not yet have a more precise analytical model for finding critical values of $\epsilon_1$ or $\epsilon_2$; instead, our practice is to increase the number of depth slices $\Delta z$ (thus effectively choosing smaller values of the ratio of $\epsilon_1/\epsilon_2^2$ in Eq. (6) and see how the calculation results approach an asymptote. If we assume $\epsilon_1 = \epsilon_2 = 0.1$ as reasonable sampling values, for a pixel size of $\Delta x = 1$ nm at 10 keV we obtain a slice thickness of $\Delta z = 81$ nm. Thus for a grating with $t = 2 \mu$m thickness we would expect that about 25 slices would be required to obtain accurate results, and for $t = 6 \mu$m about 75 slices would be required. In order to test this scaling, we show in Fig. 4(b) the results of a multislice calculation for a 29 nm:29 nm line:space grating with no Bragg angle tilting while varying the number $N$ of slices. This figure also shows the result of a CWT calculation. As can be seen, the multislice result shows excellent agreement with the CWT result as long as $N = 64$ slices are used, consistent with this simple estimate; however, it is best to check for convergence as $N$ is increased. We show in Fig. 5 the RMS deviation of the multislice approach from the CWT exit wave over one grating period. This also serves as an example of the convergence test to determine the slice step.

5. Imperfect optics

Having verified the validity of multislice propagation even in cases with energy redirection (x-ray reflectivity) and dynamical diffraction effects (thick, Bragg-tilted volume zone plates), we now...
Fig. 4. Comparison of multislice and CWT calculations of diffraction from gratings and zone plates. The CWT result was calculated using the approach described by Schneider et al. [13] for conditions in which simple scalar diffraction theory gives inaccurate results. In (a) we show that multislice can reproduce the results of an example CWT calculation [13, Fig. 8.10] showing non-scalar soft x-ray diffraction efficiency from a constant period grating slanted at the Bragg angle that would be used in a zone plate with an imaging magnification of $M = 1000 \times$. In (b) we show the diffraction efficiency for a $t = 6 \, \mu m$ thick Au grating oriented along the beam direction (that is, without the grating tilted to meet the Bragg condition) as a function of grating bar width, and with an increasing number $N$ of depth slices used for the multislice calculation. As can be seen, increasing the number of slices to $N = 64$ (with a slice thickness of $\Delta z = 94 \, nm$, as expected from Eq. (6)) allows one to nicely reproduce the CWT results. In (c) we show the comparison for CWT for the 10 keV x-ray exit wave from an infinite grating of $t = 6 \, \mu m$ Au with 29 nm:29 nm line:space ratio, against the end effects of a finite width grating (a situation that CWT cannot address). In (d) we show the effects of propagating this exit wave to the far field where one can measure both total transmittance and first-order diffraction efficiency for the infinite grating using CWT and the finite grating using increasing number of slices $N$ in the multislice approach. Again, multislice with $N = 64$ nicely reproduces the CWT result (and differs from the simple scalar diffraction result), with a slight loss in transmittance due to the edge effects shown in (c).
Fig. 5. Comparison of the multislice approach with increasing number of slices $N$ against CWT for a case where scalar diffraction theory gives inaccurate results. This calculation is done for a $t = 6 \, \mu m$ thick Au grating oriented along the beam direction (that is, without the grating tilted to meet the Bragg condition) with 10 keV x-ray illumination; at this thickness, scalar diffraction theory gives incorrect results for small grating periods. In (a), we show the calculated exit wave (the wavefield leaving the downstream side of the grating) as a function of increasing the number $N$ of depth slices (or, equivalently, decreasing the multislice slab thickness $\Delta z$ of Eq. (6)) along with the CWT result. In (b) we show the RMS error in intensity and phase of the exit wave compared with the CWT result for several different grating thicknesses $t$. The phase error shows the most systematic trend of error versus thickness, with $N = 64$ slices already achieving a RMS phase error of less than 50 mrad in all cases. The diffraction efficiency calculation as a function of zone width $d_{rn}$ for 1:1 line:space ratio, along with the scalar diffraction efficiency result [7], was shown in Fig. 4(b), which also showed that $N = 64$ slices gave accurate results for these parameters.

consider another case for which analytical results are difficult to apply: imperfect zone structures designed to be representative of actual flaws in as-fabricated Fresnel zone plates. These flaws can result from effects such as increased electron sidescattering with depth in photoresists, or imperfect directionality in reactive ion etching, or capillary forces during removal of liquid baths used in photoresist development or electroplating, or numerous other factors which come into play at the limits of fine linewidth, high aspect ratio nanofabrication. Analytical methods such as CWT are not easily applied to even simple mathematical approximations of imperfect structures, whereas with the multislice approach all one needs to do is to map any flawed diffractive structure onto a gridded array of the refractive index of $n(x, z)$ or $n(x, y, z)$ as appropriate (Eq. (1)). Modeling of zone placement errors within the limits of scalar diffraction theory has been carried out by numerous investigators [37,38], while a parabolic wave equation approach has been used to model the difference in x-ray focusing produced by sharply-defined versus “fuzzy” or soft-edged zones [23]. In our case, to illustrate one example of the ability to deal with arbitrary zone plate manufacturing flaws, we show in Fig. 6(a) some examples of partially collapsed zone structures that are representative of what we have seen in one example nanofabrication process involving electroplating [39]. The “collapsed plating” case represents what can happen after electroplating when the mold is removed; capillary forces can cause adjacent metal zones to collapse towards each other. The “collapsed mold” case represents what can happen when the photoresist is developed and the plating mold itself collapses between adjacent structures, prior to electroplating. These effects happen more frequently with higher aspect zone structures; thus the finest, outermost zones are affected most strongly. In Fig. 6(b) we show the result of a multislice calculation of the optical properties of a zone plate with an increasing fraction of zones affected
by these two example manufacturing flaws. Because the spatially-averaged projected thickness of absorbing material is unaffected by zone collapse, the total transmittance of the zone plate is unaffected; however, the diffraction efficiency is reduced and the width of the focal spot is increased.

Another example of an optic to which CWT cannot be applied is a zone-doubled zone plate which involves varying zone width with more than one material varying in depth. Zone doubling involves electron beam fabrication of low-electron-density structures, followed by atomic layer deposition (ALD) of a high-electron-density layer to produce diffractive structures on either side of the fabricated structures [9]. This can provide a factor of two improvement in the spatial resolution relative to what could otherwise be achieved by electron beam fabrication of the finest, outermost zones. However, zone doubling has a limitation in that the line:space ratio for inner zones in a Fresnel zone plate becomes far from ideal (the ALD “zones” are of fixed width throughout the zone plate structure). Because Fresnel zone plates have varying zone width, and because zone-doubled zone plates have both the low-electron-density structures as well as the ALD layer, partial voxel filling is required to accurately represent either type of optic.

We have calculated the exit waves for two simulated zone plates: a conventional Fresnel zone plate fabricated in t = 2 µm of gold, and a zone-doubled zone plate with a t = 2 µm thick nanofabricated structure of silicon followed by atomic layer deposition of 20 nm of iridium. Multislice propagation was carried out for 10 keV x-rays using the Hankel transform over N = 64 propagation slices within the zone plate structures. In Fig. 7(a), we show the intensity and phase profiles of exiting wave from the innermost zones, where the line:space ratio for zone-doubled zone plates is far from the usual 1:1 ratio. Differences in wave profiles can be seen near the interface between silicon and iridium, and between iridium and vacuum. These exit waves were then propagated to the zone plate focal plane, again using the Hankel transform approach, with results shown in Fig. 7(b). As can be seen, the conventional Fresnel zone plate would have higher focusing and diffraction efficiency than the zone-doubled zone plate, while the zone-doubled zone plate has a somewhat smaller focus spot due to the stronger weighting given to the finest, outermost zones due to their high diffracting efficiency by having nearly a 1:1 line:space ratio. In practice, the zone-doubling method allows one to produce higher spatial resolution zone plates due to the relaxation of the grating density that needs to be fabricated using electron beam lithography [9].

6. Optical structures at multislice grid boundaries

Consider the case of boundaries between two media with refractive indices n_a and n_b where the boundary occurs not at the edge of a voxel, but within; this is shown in Fig. 8(a). In this case our approach is to calculate the wavefield modulation by the mixed voxels (the volume fraction c occupied by material a, and the rest by material b), whether or not the interface is perpendicular (i), or parallel (ii), or in some other direction relative to the propagation direction which can be always categorized in (i) or (ii) as long as the voxels are small enough. In case (i), x-rays pass through material a first and then material b; therefore the wavefield is modulated within a voxel first by cΔz thickness of material a and then by (1 − c)Δz thickness of material b. In other words, the exit wavefield from a voxel ψ’ can be calculated from the incident wavefield ψ by Eq. (7) which is

$$\psi' = \psi \cdot \exp \left[ i \frac{2\pi c\Delta z}{\lambda} (\delta_a + i\beta_a) \right] \cdot \exp \left[ i \frac{2\pi (1-c)\Delta z}{\lambda} (\delta_b + i\beta_b) \right]$$  \hspace{0.5cm} (7)

In case (ii), part of the x-ray wavefield passes through material a, and part through material b simultaneously; therefore the wavefield is split and modulated by a thickness Δz of materials a and b respectively and then superposed within a voxel, so that ψ’ can be calculated by Eq. (8)
Fig. 6. Multislice propagation allows one to simulate various flaws in the nanofabrication process, and see what their effects are on Fresnel zone plate diffraction efficiency. In (a), we show “normal” zones with the desired profile, surrounded on either side by two different examples of zone collapse: “collapsed plating” where the as-fabricated zones collapse on each other after electroplating, or “collapsed mold” where the plating mold collapses prior to electroplating of zone structures. In (b), we show the results of an increasing fraction of zones suffering either type of collapse, leading to a reduction of diffraction efficiency and an increase in the focal spot size when the zone plate is illuminated by a plane wave. The focus size is determined by the position of the first minimum from the axis. This calculation was carried out for Au regular zone plate of 2 µm thick, 40 µm diameter and 20 nm outermost zone width, illuminated by 10 keV x-rays.
Fig. 7. Multislice propagation simulations from a regular Fresnel zone plate, and from a zone-doubled zone plate [9]. This is representative of a calculation for which CWT cannot be used, since the zone width (and, in the case of zone-doubled zone plates) the projected material composition within a zone changes from zone to zone. In this example two zone plates with $t = 2 \mu m$ thickness and $dn = 20$ nm outermost zone width for use with 10 keV x-rays were compared: a standard Fresnel zone plate fabricated in Au, and a zone-doubled zone plate fabricated in Si with 20 nm of Ir deposited using Atomic Layer Deposition (ALD). In (a) we show the exit waves from these two zone plates calculated using multislice propagation with $N = 64$ slices, revealing very different characteristics. In (b) we show the radial intensity profile, as well as the radially-integrated energy distribution, indicating that zone-doubling produces a somewhat smaller focus spot due to lower diffraction efficiency from the coarser, inner-most zones with a line:space ratio that is far from the desired 1:1 value. In practice, the zone-doubling method allows one to produce higher spatial resolution zone plates because the electron beam fabricated structures can have lower grating density with fewer limitations due to electron beam sidescattering in photoresists and structure collapse during the nanofabrication process. The same radial intensity profile is shown as a 2D image in (c).
which is

$$
\psi' = \psi \cdot \left\{ c \cdot \exp \left[ \frac{2\pi \Delta \varepsilon}{\lambda} (\delta_a + i\beta_a) \right] + (1 - c) \cdot \exp \left[ i\frac{2\pi \Delta \varepsilon}{\lambda} (\delta_b + i\beta_b) \right] \right\},
$$

(8)

Fig. 8. Partial filing of voxels through which the interface between two media passes. The fraction $c$ of the voxel filled with material $a$ is calculated, whether or not the interface is parallel (i), or perpendicular (ii), or in some other direction relative to the voxel faces. This approach was used in generating Fig. 2. As a test, a 1 $\mu$m-thick gold grating structure illuminated by 10 keV x-rays was shifted by 0.0, 0.5, and 1.0 pixels relative to the voxel boundaries with $\Delta x = 2$ nm pixel size and $\Delta z = 2$ nm step size in the propagation direction. The grating exit waves (b) and far-field diffraction intensities (c) show the same characteristics no matter where the actual refractive index boundaries lie, as expected.

With this approach, one can model refractive index boundaries at arbitrary positions or even angles relative to the propagation calculation direction. Arbitrary positions were used for the continuous variation of zone displacement or zone width in Figs. 6 and 7, and arbitrary angles were used in the calculations shown in Figs. 2, 3, and 4. We use the partial voxel filling to the calculation of reflectivity and simulation of x-ray reflection on mirrors, which the mirror
boundary changes extremely slowly in transverse relative to a pixel. In our simulation of x-ray reflection on 4 mrad mirror with 1 nm pixel size, the boundary passes to the next transversal pixel after propagating for at least 25 slices. Also partial voxel filling applies to regular zone plates for that the zone period changes with radius, and to zone plates with defects such as zone collapses where small position variation needs to be represented over the multislice propagation. In order to more explicitly test the validity of this approach, we show in Fig. 8(b) a test of the exit wave and in Fig. 8(c) far-field diffraction pattern resulting from a grating with boundaries at, and in-between, pixel edges. As shown in Fig. 8(b), a grating structure is shifted by 0.0, 0.5 and 1.0 pixel relative the voxel boundaries. The intensity and phase distributions of the exit wave from the grating are the same in all cases, but there shows sub-pixel shifts for the grating shifted by 0.5 pixel. Fig. 8(c) shows that the far-field diffraction intensities are overall identical for the partial filling case of a shift of 0.5 pixels. Partial voxel filling is desired for arbitrary optical structures to represent sub-pixel position variation of materials in both transverse and longitudinal directions.

The requirement that objects be mapped on to a three-dimensional grided array \( n(x, y, z) \) can be reduced in cases where there are no material variations in one direction; indeed, most calculations shown here are on grids of \( n(x, z) \).

7. Conclusion

We have shown here that the multislice propagation method can be used for calculations in x-ray optics in situations far wider than previously realized, including cases where the Poynting vector is changed considerably (grazing incidence reflectivity) and where methods like CWT have been thought to be the only valid approach (such as in very high aspect ratio gratings and Fresnel zone plates). The power of the multislice approach is that it can be applied to any structure where one can define a three-dimensional refractive index distribution \( n(x, y, z) \), including x-ray nanofocusing optics with real flaws that cannot be described by simple mathematical functions that apply over an infinite extent. This enables the realistic modeling of as-fabricated x-ray nanofocusing optics, so that by understanding the effects of various flaws one can place effort on the most important ones and thus enable advances in the studies of materials properties at the nanoscale.

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