Ultimate limitations in the performance of kinoform lenses for hard x-ray focusing

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Diffractive x-ray lenses suffer from limited focusing efficiency, which can be improved by replacing the binary nanostructures of the lenses with kinoforms. Here we present the first example of kinoform lenses for x rays and compare their efficiency with those of binary Fresnel zone plates (FZPs), realized through gray-scale-focused Xe ion beam lithography. Unexpectedly, experimental results indicate lower focusing efficiency with those of binary Fresnel zone plates (FZPs), for example of kinoform lenses for x rays and compare their features of the lenses with kinoforms. Here we present the first approach may suggest that it is not the path to increasing focusing efficiency. © 2019 Optical Society of America under the terms of the OSA Open Access Publishing Agreement

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Cylindrical refractive [1] and kinoform [2–4] lenses made of light elements were shown to successfully focus multi-keV x-ray beams. These optical elements produce a line focus, and 2D focusing requires an alignment of two lenses in a crossed configuration. Binary Fresnel zone plates (FZPs) are popular diffractive x-ray optical elements because they are compact and easy to align and use. Their major drawback, however, is a limited focusing efficiency (40.5% at most for an ideal FZP made of nonabsorbing material with phase-inverting zones). Instead, the use of parabolically shaped zones [Fig. 1(a)] ensures that rays from within each full-period zone are phase-shifted to constructively interfere at the focal spot, hence maximizing the efficiency of the first diffraction order. Such a kinoform lens can theoretically have 100% focusing efficiency; however, fabrication of these optical elements with a complex surface relief using conventional lithography methods is extremely challenging. To overcome these difficulties, the parabolic or sawtooth-like zones are sometimes approximated by multistep (staircase) zone shapes to achieve the blazing effect.

The multistep FZPs can be produced (i) by multiple aligned lithography steps [5–9], or (ii) by stacking individual binary zone plates either in close proximity to each other or at a designed separation [10–14]. However, it may be challenging to perfectly align the two binary zone plates, whether temporarily or permanently, and misalignments, zones’ imperfections or distortions result in decreased focusing efficiency [10]. Furthermore, despite a possible gain in focusing efficiency, the requirement of high-precision alignment of separate optical components makes the stacked zone plates more complex to use and less attractive compared to monolithic FZPs, characterized by their simplicity of handling. Complex surface profiles of kinoforms can be realized by various three-dimensional (3D) micromachining techniques, such as high-resolution gray-scale electron beam lithography [15–19] and a variety of other methods [20].

Focused ion beam (FIB) milling is a direct-write method that was demonstrated to be capable of 3D surface micromachining to realize complex 3D shapes (such as kinoforms [21–24]) in a variety of substrates. In this method, a finely focused beam of ions (typically 30 keV Ga+) is scanned across a surface. The impinging ions sputter away the substrate atoms. By varying the time that the ion beam dwells at every point (and hence the milled depth), a complex surface relief can be chiseled out in the substrate surface. The disadvantage of the FIB milling is its relatively slow speed due to the pixel-by-pixel pattern transfer. Recently introduced plasma-FIB systems based on high-brightness inductively coupled plasma ion sources allow speeding up the milling process significantly by replacing Ga with heavier Xe ions. The increased sputter yields of Xe (e.g., >35% higher for Au) and more than an order of magnitude higher ion currents as compared to Ga-based FIBs can decrease the milling times by more than an order of magnitude, albeit at the expense of reducing the resolution. Here, we fabricated both binary FZPs and kinoform lenses in 800-nm-thick Au and 1.1-μm-thick W substrates with focused Xe beams and compared their focusing efficiencies in the 5–7 keV x-ray energy range. Au is a common material used in diffractive optics elements due to its strong interaction with x rays and ease of fabrication. W is an attractive material for high-power applications.
due to its extremely high melting point. While W is a challenging material for microfabrication, gray-scale FIB milling of W is straightforward.

The lenses were fabricated on 1-μm-thick Si₃N₄ membranes coated with 10 nm Ti serving as an adhesion promoter. The membranes were sputter-coated either with 800 nm Au or 1.1 μm W. The milling was performed using focused 30 keV Xe ions at 30–1000 pA currents from a ThermoFisher Helios P-FIB UX-G4 system. The beamspot size varies between 25–150 nm for these beam currents, with the resolution currently limited to >100–200 nm, while the achievable aspect ratios are <3–5. Due to the sharp features in the kinoform pattern, the deviations from the target profile are greater for the kinoform lenses, as observed in Figs. 1(e) and 1(h). The desired surface profiles (kinoforms or binary zones) were digitized into 8-bit maps with pixel coordinates corresponding to the position within the lens, and the pixel value proportional to the depth of the milled profile. P-FIB milling using Xe was previously shown to generate smooth and high-quality surface profiles in hard substrates [25,26]. Here, we followed similar procedures to generate the lenses in Au and W with a 50-μm diameter and with an outermost zone of 220 nm and corresponding full-period zone of 440 nm for binary and kinoform lenses, respectively [Figs. 1(b) and 1(g)]. The first-order diffraction efficiency (DE) measurements were performed at the x-ray fluorescence microscopy (XFM [27]) beamline of the Australian Synchrotron. The intensity shaped by the lenses was monitored by scanning a 10-μm pinhole in the focal plane using a p-i-n diode [Fig. 2(a)]. The resulting intensity map is a convolution of the probe and the intensity profile with various diffraction orders clearly visible. The shape of the intensity map is discussed in detail in Ref. [28].

![Fig. 2. Kinoform and binary FZP lenses DE measurements.](image)

DE was determined from the ratio of the focused intensity and the total intensity illuminating the lenses while taking into account the absorption in the supporting membrane estimated from mapping the incident beam. Figures 2(b) and 2(c) show the measured DE for Au and W lenses compared with theoretical values. The measured efficiencies are comparable within each energy range and are relative through the energy range, with the differences likely due to varying systematic offsets. Hence, the measurements broadly match the theoretical values.

The theoretical efficiency value can be estimated using procedures described in Ref. [7]. Here, the zone plate annular radii, \( r_n \approx \sqrt{n2\lambda f} \), where \( n \in N \) are the full-period zone indices, \( \lambda \) is the wavelength, and \( f \) is the focal length, have a period of \( 2\lambda f \) in \( r^2 \) coordinates. The transmittance of the zone plate in a thin-lens approximation is therefore,

\[
t(r) = \sum_n C_n e^{-2\pi i n / \lambda f}; \quad C_n = \int_0^{2\lambda f} t(r) e^{2\pi i n r^2 / \lambda f} dr^2.
\]  

(1)

For a binary zone plate with transparent and opaque zones and with a thickness \( h \) made of a material with a refractive index \( 1 - \delta + i\beta \) (\( \delta \) and \( \beta \) are dispersive and absorptive properties of the material) the \( n \)th order efficiency is, therefore,

\[
|C_n|^2 = \left| \frac{1}{2\lambda f} \int_0^{2\lambda f} \exp\left(2\pi i n^2 / 2\lambda f\right) dr^2 \right|^2 + \left| \frac{1}{2\lambda f} \int_{2\lambda f}^{2\lambda f} \exp\left(2\pi i n^2 / 2\lambda f - i2\pi \delta h / \lambda - 2\pi \beta h \right) dr^2 \right|^2.
\]  

(2)

![Fig. 1. Au kinoform and binary FZP lenses. (a) Schematic view of kinoform and binary FZP lenses zone cross section profiles; (b)–(d) and (e)–(g) kinoform lenses and binary FZP milled in Au, respectively; (d) and (g) outermost zones were coated with Pt and cross sections milled to reveal the profiles of the zones. See also Supplement 1, Fig. S1.](image)
Similarly, for a lens with kinoform full-period zones, the efficiency is

\[ |C_{al}|^2 = \left| \frac{1}{2\lambda f} \int_0^{2\pi f} \exp \left( \frac{2\pi in r^2}{\lambda f} - i \frac{2\pi n f r^2}{\lambda f} \right) dr^2 \right|^2. \]  

(3)

Using tabulated x-ray optical constants \[29\], the above integrals were evaluated numerically. The results are presented as solid lines in Figs. 2(b) and 2(c). Efficiencies were estimated from simulated wavefront propagation \[Figs.2(b) and 2(c)\]. The experimentally measured and theoretical values are generally in good agreement. We attribute the discrepancies mainly to the imperfections in the lens zones. The differential milling \[31\] of sputtered polycrystalline Au and W results in rough surfaces. The surface roughness and zone height variations lead to random phase variations and spurious scattering that decrease the apparent efficiency. Both measured and theoretical efficiencies of kinoform lenses in this study are considerably lower compared to the corresponding binary FZPs. This is explained by the nonoptimal full-period zone height of the kinoform lenses. These lenses are most efficient if the phase shift within each full-period zone unwraps radially throughout the zone from 0 to \(2\pi\) rad, ensuring that the rays from different parts of the lens constructively interfere in the focus. However, at the 6 keV x-ray energy of the maximum efficiency of the binary FZPs in this study, the heights of the lenses, 800-nm Au and 1.1 \(\mu\)m of W, generate only 0.66\(\pi\) and 0.9\(\pi\) rad phase shifts, respectively. As shown next, the <\(\pi\) rad phase shift is not optimal for the performance of the kinoform lenses.

Figure 3 compares theoretical efficiencies of an Au kinoform and a corresponding binary FZP lens at 6 keV photon energy both for ideal, purely phase lenses and real lenses with absorption. In the absence of absorption, the binary FZP reaches its fundamental efficiency limit of 40.5\% when its zones are 1.2-\(\mu\)m thick, imparting \(\pi\) rad phase shift, thereby inverting the phase of the incident beam. However, when the absorption of Au is taken into account, the maximum achievable efficiency is reduced to 27\%. The efficiency maximum occurs at the zone height of 1.1 \(\mu\)m, such that the 0.9\(\pi\) rad phase shift and the absorption are at their optimal combination. Similarly, an absorption-free kinoform lens reaches 100\% focusing efficiency when the kinoform heights are 2.4 \(\mu\)m, corresponding to the maximum phase shift of 2\(\pi\) rad.

However, since the kinoform lens is twice as thick as the corresponding binary FZP, the absorption is nearly doubled, such that the maximum achievable efficiency is significantly reduced from 100\% to 41.5\%. This is less than double the efficiency of the binary FZP. The optimal Au kinoform lens thickness at 6 keV from Fig. 3 is 2.1 \(\mu\)m, corresponding to 1.7\(\pi\) rad phase shift.

Through the comparison of the calculated efficiencies for binary FZP and kinoform lenses made of gold and nickel \[typical lens materials for hard and soft x-ray energies \(\text{Fig. 4}\), it is shown that kinoform lenses outperform binary FZPs primarily at higher x-ray energies where the material absorption is lower, below the absorption edges. The kinoform lens only outperforms the binary FZP when the lens thickness surpasses the thickness at which the full-period zones produce >\(\pi\) rad phase shifts \(\text{Fig. 3}\). At lower and more achievable thicknesses, particularly where the phase shift is <\(\pi\) rad, the efficiency may be lower than that of a binary FZP, as a large fraction of the incoming intensity is transmitted \(\text{the zeroth order}\). The efficiency gain is, however, reduced at lower energies, where the absorption is higher.

The binary FZP efficiency peaks at 33.8\% with 2.47-\(\mu\)m-thick Au structures at 11.6 keV, whereas the Au kinoform lens maximum efficiency is twice as high, at 69.6\%. This efficiency gain, however, comes with a staggering requirement for the optimal lens thickness >4.8 \(\mu\)m, which currently presents a considerable fabrication challenge. Comparatively lighter materials, e.g., Cu or Ni, appear to be more suitable for efficient multi-keV x-ray focusing \(\text{see Supplement 1, Figs. S4–S6}\), although even greater material thickness requirements make the realization of such optical elements extremely difficult. The increased thickness of the diffractive lens further implies that the thin-lens approximation used to design the zone plates becomes less valid as volume diffraction effects become more prominent \[32\].

The successful fabrication of a multimicrometer thick kinoform lens, therefore, does not guarantee its optimal focusing performance. The cost of manufacturing 3D high-aspect ratio structures, for extremely high-resolution x-ray nanofocusing, implies that the factor of 2 theoretical gain in the efficiency of kinoform lenses compared to conventional binary FZPs may not provide sufficient return on investment. Hence, it is not beneficial to realize the complex kinoform surface when a simpler-to-fabricate FZP with binary zones achieves better performance.
To conclude, we fabricated, characterized, and compared first-order focusing efficiencies of kinoform lenses and binary FZPs realized in 800-nm Au and 1.1-μm W layers by means of direct-write gray-scale focused Xe ion beam lithography. Both theoretical and experimental results indicate reduced focusing efficiencies of kinoform lenses compared to binary FZPs in the 5–7 keV x-ray energy range, due to the nonoptimal phase-shifting capability (<π rad) of the kinoform full-period zones. Theoretical investigations that incorporate absorption revealed that blazing of the binary FZPs zones results in only a limited boost in the DE in higher (multiple keV) x-ray energies and for impractical multimicro-meter material thicknesses of the zones, making binary FZPs a preferred option both in terms of ease of their fabrication and focusing efficiency.

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See Supplement 1 for supporting content.

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