High Spatial Resolution STXM at 6.2 keV Photon Energy

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Abstract. We report on a zone-doubling technique that bypasses the electron-beam lithography limitations for the production of X-ray diffractive optics and enables the fabrication of Fresnel zone plates with smaller outermost zone widths than other well-established approaches. We have applied this method to manufacture hard X-ray Fresnel zone plates with outermost zone widths of 25 and 20 nm. These lenses have been tested in scanning transmission X-ray microscopy (STXM) at energies up to 6.2 keV, producing images of test structures that demonstrate a spatial resolution of 25 nm. High spatial resolution STXM images of several biological specimens have been acquired in transmission, dark-field and differential phase contrast modes.

Keywords: X-ray microscopy; X-ray diffractive optics; Electron-beam lithography; Atomic layer deposition

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INTRODUCTION

X-ray microscopy has been demonstrated to be a powerful imaging technique for the inspection of both inorganic [1, 2, 3] and biological [4, 5, 6] specimens, as it provides high spatial resolution in combination with high penetration power and chemical sensitivity. Indeed, the demands on high spatial resolution have been a driving force for the development of X-ray optics. Mirrors [7, 8] and refractive lenses [9] are commonly used to concentrate hard X-rays, and spot sizes down to 50 nm have been reached. Diffractive optics, mainly Fresnel zone plates (FZPs), have also been used for hard X-ray imaging with sub-50 nm spatial resolution [10, 11, 12].

FZP-based X-ray microscopes have achieved sub-20 nm resolution in the soft X-ray regime [13, 14], but the fabrication of FZPs for hard X-rays is more challenging as it requires high aspect ratio structures to ensure an efficient performance of the device. FZPs are commonly prepared by using electron-beam lithography (EBL) to write structures with nanometer precision. After the exposure the lens pattern has to be transferred to generate the functional high aspect ratio structures. While features in the sub-20 nm range can be prepared by state-of-the-art EBL systems, the pattern transfer to generate high aspect ratio sub-50 nm structures still remains as a complex task.

Recently, we demonstrated the feasibility of bypassing the EBL limitations for production of soft X-ray diffractive optics by using a zone-doubling technique [15]. While in the previous experiments we demonstrated sub-12 nm spatial resolution at 1.0 keV photon energy [14],

we now show that this method is also applicable to the fabrication of FZPs for the multi-keV photon energy range. We have prepared FZPs with effective outermost zone widths of 25 and 20 nm and aspect ratios of about 15. These lenses have been used as focusing elements for scanning transmission X-ray microscopy (STXM) at the coherent Small Angle X-ray Scattering (cSAXS) beam-line of the Swiss Light Source (Paul Scherrer Institut, Villigen, Switzerland).

ZONE-DOUBLED FRESNEL ZONE PLATES

The preparation of the zone-doubled FZPs relies on the conformal deposition of a high refractive index material (iridium) by atomic layer deposition (ALD) onto the sidewalls of a prepatterned template made from a low refractive index material (silicon). This modified sort of FZPs, their geometry and their diffraction efficiency have been extensively addressed in [15]. The template made of silicon consists of a FZP pattern in which the line density is decreased by a factor of two at the outer regions of the structure. The template is later coated by an iridium layer that is uniform over the whole structure and matches the outermost zone width (figure 1).

Diffraction Efficiency Calculations

Due to the particular geometry of the zone-doubled FZPs, the well-established formula for the diffraction

FIGURE 1. Drawing of zone-doubled FZP made of silicon and iridium. The silicon template is coated by a uniform layer of iridium of thickness w , which matches the outermost zone width of the FZP.

efficiency of an ordinary FZP have to be recalculated. Following the expressions derived in [15], figure 2 plots the expected diffraction efficiency of zone-doubled FZPs made of iridium and silicon. An effective outermost zone width of 25 nm is considered for a photon energy of 6.2 keV.

In comparison to an ordinary FZP, in the zone-doubled lens the diffraction efficiency depends not only on the material height of the structures, h , but also on the sidewall material thickness, w . For the parameters considered in figure 2, the highest theoretical diffraction efficiency would be achieved with material thickness of 1050 nm and sidewall material thickness of 32 nm. The latter value is slightly shifted above the effective outermost zone width of 25 nm because a higher value maximizes the constructive interference over a larger area of the zone-doubled FZP pattern.

Fabrication Method

The fabrication method has been described in detail in [14]. The manufacturing of the zone-doubled FZPs started with the exposure of the template pattern by electron-beam lithography. Subsequently, the pattern was transferred to a chromium layer by a chlorine-based plasma, that was used as mask during the pattern transfer into silicon by reactive ion etching [16]. The template is then coated by a conformal uniform layer of iridium by ALD [17]. This technique can grow iridium thin films with almost atomic accuracy through self-limiting, sequential surface reactions. This method is well-suited to the preparation of FZPs for the multi-keV regime because simplifies the fabrication of very high aspect ratio structures that are required at these photon energies. In addition, it ensures reproducibility and an efficient fabrication yield since it only requires a single lithography step.

To date, we have been able to prepare zone-doubled FZPs with outermost zone widths of 25 and 20 nm and a structure height of 350 nm. Thus, aspect ratios of >15 have been reached and a maximum theoretical diffraction

FIGURE 2. Calculated diffraction efficiency for a zone-doubled FZP with an effective outermost zone width of 25 nm as function of the material height, h , and the sidewall material thickness, w . Silicon is considered as low refractive index template material while iridium is taken as high refractive index coating material. The red dash line indicates a material height of 350 nm corresponding to the zone-doubled FZPs that have been manufactured.

efficiency of about 5% can be expected, as emphasized in figure 2.

SCANNING TRANSMISSION X-RAY MICROSCOPY

A zone-doubled FZP was used to create a nanofocused beam for STXM imaging at an undulator beamline. The STXM setup consisted of the zone plate, with a central stop and order sorting apertures positioned to select the first-order focus, and a sample mounted in the focal plane. The experiment was mounted on a 15 mm-thick stainless steel baseplate to prevent relative displacements of the optics in the case of vibrations. The detector could be positioned either 2.75 or 7.0 m downstream of the focal plane, behind a He-filled 'flight' tube to avoid excessive absorption or scattering by air. A PILATUS 2M detector, operated in single photon counting mode [18, 21], was used to record the divergent radiation cone emanating from the sample. The variation in position and intensity of the beam footprint during a scan of the sample was processed to give absorption, differential phase contrast and dark field images of the sample [19].

During the experiments, several zone-doubled FZPs with diameters of 100 and 50 μm and outermost zone widths of 25 and 20 nm were mounted. The energy

FIGURE 3. STXM micrographs of test structures with 25 nm lines and spaces acquired using a zone-doubled FZP with a diameter of 50 μm and an outermost zone width of 25 nm. A step size of 6 nm and a dwell time of 100 ms were used. The structures are completely resolved in transmission, dark field and differential phase contrast modes (DPCx and DPCy). The data for these images were acquired at 5.0 keV photon energy.

of the incoming X-rays was set at 5.0 and 6.2 keV for the images presented. Due to the small outermost zone widths, typical focal lengths were ranging from 12 down to 6 mm. Central stops made of 15 μm -thick gold layers and diameters of 20 and 30 μm were located on independent stages in front of the FZP. Pinholes of 10 and 20 μm in diameter were used as order selecting aperture depending on the diameter of FZP in use.

Figure 3 shows the STXM micrographs of a test structure with 25 nm lines/spaces that was fabricated by the same method to produce the zone-doubled FZPs. This image was recorded by a zone-doubled FZP with a di-

FIGURE 4. STXM images of cytoskeleton filaments of pancreatic cancer cell. The data were acquired with step size of 20 nm and dwell time of 50 ms at 6.2 keV photon energy.

FIGURE 5. STXM images of blood cells infected by malaria. (a) Overview scan over a large area of $300 \times 90 \mu\text{m}^2$. (b) High resolution image a blood cell that ruptured due to the infection. The data were acquired with step size of 20 nm and dwell time of 50 ms at 6.2 keV photon energy.

ameter of $50 \mu\text{m}$ and an outermost zone width of 25 nm. Transmission, dark field and differential phase contrast images have been obtained from the acquired data. The image clearly shows that 25 nm lines and spaces are resolved. The data were recorded using a step size of 6 nm and dwell time of 100 ms at photon energy of 5.0 keV.

In addition, a few biological specimens were inspected. During these scans a zone-doubled FZP with a diameter of $100 \mu\text{m}$ and an outermost zone width of 20 nm was used. Figure 4 shows a human cell (pancreatic cancer cell, cell line Panc1), which was prepared to observe the cytoskeleton filaments by scanning electron microscopy [20]. The STXM data were recorded at 6.2 keV photon energy using a step size of 20 nm and a dwell time of 50 ms. The integrated phase was calculated from the differential phase contrast images following the expressions derived in [19].

Figure 5 shows a STXM image of human red blood cells infected by malaria. Figure 5(a) shows a large scan over an area of $300 \times 90 \mu\text{m}^2$ that was used to locate the position of the blood cells. Figure 5(b) shows high resolution images of a blood cell that ruptured because of the infection. The data were acquired with a step size of 20 nm and a dwell time of 50 ms. Finally, figure 6 shows STXM images of $1 \mu\text{m}$ -thick mouse liver cell slice. The transmission image contains some artifacts

FIGURE 6. STXM image of a mouse liver cell slice. Both transmission and integrated phase signals are shown. The data were acquired with a step size of 20 nm and a dwell time of 50 ms.

due to beam intensity fluctuations during the acquisition of the data. However, these variations are not observed on the integrated phase image because the differential phase signals are insensitive to intensity fluctuations of the incoming X-ray beam.

The diffraction efficiency of a $100 \mu\text{m}$ zone-doubled FZP with an outermost width of 20 nm was measured

to be 1.5%. This represents only 40% of the expected value and it is lower most likely due to the defects in the fabricated zone-doubled structures.

CONCLUSIONS

We have demonstrated the possibility of applying the zone-doubling technique for the fabrication of FZP for the multi-keV photon energy range. The manufactured zone-doubled FZPs had outermost zone widths of 20 and 25 nm and a material height of 350 nm. They have been used to obtain high resolution STXM images of several biological specimens. We believe that the results shown here can be improved further in terms of spatial resolution and diffraction efficiency.

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