Diffractive lenses for photon energies ranging from the extreme ultraviolet to hard x-rays

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ABSTRACT

Diffractive optics for the x-ray range have to meet the various requirements of experimental set-ups at synchrotron or other light sources. In the case of focusing elements it is essential that the devices are matched to parameters such as the photon energy and spatial coherence of the source, as well as the required spatial resolution, working distance, and diffraction efficiency. In some cases, a suppression of disturbing diffraction orders requires additional features such as an opaque central stop integrated into the lens.

The Laboratory for Micro- and Nanotechnology provides the essential technologies necessary for the design and fabrication of diffractive x-ray lenses for a wide range of photon energies and applications. Over the past years, a large variety of optics tailored to the specific needs of x-ray optical experiments have been fabricated and tested. These include transmission Fresnel phase zone plate for microscopy, microprobe, or beam monitoring applications, as well as condensers to increase the flux in waveguiding experiments in the hard x-ray range. An overview of the nanofabrication technologies and a selection of experiments demonstrating the devices performance are presented.

Keywords: Zone plate, electron beam lithography, microscopy, x-ray fluorescence, EUV lithography

1 INTRODUCTION

Fresnel zone plates have been employed for several decades to focus x-rays. While the initial developments were driven by the development of soft x-ray microscopes,¹ optimised diffractive lenses are at present also required for more and more other applications using various regions of the x-ray spectrum. These applications include microanalysis, microspectroscopy, and microdiffraction. The diffraction-limited resolving power of a zone plate depends on its outermost zone width, dr_n . The resolution δ_R according to the Rayleigh criterion is given by $\delta_R = 1.22 dr_n$. The Rayleigh model describes the imaging of two infinitely small incoherently illuminated point sources. The resolution limit is reached when the intensity dip $I_{\min}$ between the two superimposed Airy disks in the image amounts to 73.5% of the maximum intensity $I_{\max}$.² In this case, the contrast $(I_{\max} - I_{\min}) / (I_{\max} + I_{\min})$ is 15.3%. Although this Rayleigh contrast is somewhat arbitrary, as many microscopes can resolve smaller contrasts, it is useful to have a generally accepted criterion for the benchmarking of imaging systems. The testing of zone plates according to this model would require an object consisting of pairs of bright spots on a dark background with diameters which are small compared to the expected resolution. As such objects are difficult to obtain, resolution tests are often performed by imaging grating structures. The half pitch observable with a Rayleigh contrast of 15.3% is $\delta_{\text{grid}} = 0.7 dr_n$.³ This means, that the diffraction limit of zone plates allows imaging of periodic structures with widths just below one outermost zone width. The best resolution values reported so far have been achieved using zone plates with outermost zone widths below 30 nm^{4,5} optimised for operation in the “water window” between 284 eV and 543 eV photon energy.

In many applications a zone plate is used to generate a small, intense focal spot by forming a demagnified image of the x-ray source. To avoid a deterioration of the zone plate resolving power, the geometrical source image size S' should be well below the zone plate diffraction limit. For large demagnification factors, S' is given by $S' \sim S \cdot f/g$, where S is the effective source size, f the zone plate focal length, and g the effective source-to-zone plate distance. With $f \lambda = dr_n d$ and the size of the

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diffraction limited spot $\delta \sim dr_n$, the condition $d < g \lambda/S$ for diffraction limited imaging is obtained. The maximum value of d describes the size of the coherently illuminated area (i.e. the transverse coherence length) in the zone plate plane. In general, the source sizes of synchrotron sources are quite different in horizontal and in vertical direction. In consequence, the obtained spot size may be diffraction limited only in one direction when the zone plate diameter has a value between the horizontal and vertical transverse coherence lengths. Depending on the specific application, it may be useful to apply zone plates with diameters significantly larger than the coherently illuminated area. Although in this case, the obtained spot size will be larger than the outermost zone width, more flux is collected compared to a smaller zone plate with the same focal length. The diffraction efficiency, i.e. the fraction of incident radiation that is diffracted into the order used for imaging, depends on the shape of the zone structures and the optical constants of the zone plate material. The material should have a strong phase shift, to avoid high aspect ratios of the outermost structures, coupled with low absorption. For a given photon energy and zone plate material, the maximum obtainable efficiency and the necessary zone height to obtain this value can be calculated using scalar diffraction theory developed by Kirz⁶ and the complex refractive index $n=1-\delta-i\beta$ tabulated for most elements by Henke et al..⁷ For diffractive structures with a square wave profile, the first order diffraction efficiency E_1 at a given x-ray wavelength λ is given by:

$$E_1 = \frac{1}{\pi^2} (1 + e^{-2k\phi} - 2e^{-k\phi} \cos \phi)$$

where $k = \beta/\delta$, and $\phi = 2\pi\delta/\lambda$ denotes the phase shift caused by diffracting structures with a height of t . The maximum diffraction efficiency is high for a favorable ratio of absorption to phase shift, i.e. for small values of k . In the case of the ideal phase zone plate ($k=0$), a maximum first order diffraction efficiency of $4/\pi^2 = 40.5\%$ is obtained, while for high absorption ($k \rightarrow \infty$), $1/\pi^2 = 10.1\%$ can be achieved. The upper graph in Figure 1 depicts the maximum obtainable efficiency of a diffractive optical with binary structures as a function of photon energy from 30 eV up to 30 keV for a selection of elements. The four thick curves belong to the materials used in the experiments described in this article, while the thinner curves belong to other materials commonly used in zone plate fabrication. It can be seen that the maximum diffraction efficiency is generally high for photon energies just below an absorption edge, whereas it shows a huge drop for energies just above an edge. A

Figure 1: Maximum obtainable diffraction efficiency of a diffractive optical element with binary structures for a selection of materials as a function of photon energy (top graph). The vertical lines indicate energies addressed in the different sections of this article. The lower graph represents the necessary height of the diffractive structures to obtain 80% of the maximum efficiency values.

very important issue for the fabrication process is the structure height required to give a good efficiency for a given material and photon energy, as it is proportional to the aspect ratio of the zone structures. Because the maximum of the diffraction efficiency as a function of the structure height, $E_I(t)$, is not very sharp, it is advantageous to accept a small loss in obtainable efficiency as this can significantly reduce the necessary structure height. Therefore, we plotted the structure heights required to obtain 80% of the maximum possible diffraction efficiency in the lower graph of Figure 1. Except for energies close to an edge, the phase shift is proportional to the energy resulting in regions with parallel lines in the logarithmic plot.

These curves can be used to find a good zone plate material for a specific set-up. The techniques to pattern the selected material can be divided into two groups. One is based on a selective deposition of material by electrodeposition. Excellent zone plates have been fabricated by plating of Ni ^{5,8-10} or Au ^{8,11-12} into polymer moulds. The other technique uses dry etching for a selective removal of the zone plate material. Semiconductors like Si ¹³ or Ge ^{4,5,14,15}, as well as some metals like Ta ^{15,16} or W ¹⁷ are also used because they can be structured with high aspect ratios by reactive ion etching.

The zone plate patterns are mostly defined by electron beam lithography. State-of-the-art exposure systems are capable of producing spot sizes of a few nanometers and provide pattern placement accuracies of a few tens of nanometers. Our zone plates were exposed with a LION e-beam lithography system from Leica Microsystems Jena using an electron energy of 2.5 keV. The LION has some special features which make it particularly suited for the generation of optical patterns. Its continuous path control mode allows the exposure of curved or straight lines by moving the sample stage continuously underneath the electron beam. This makes it possible to generate patterns over large areas without any field stitching. Curved lines are not approximated by polygons, instead they are described by Bézier-polynomials, which results in smooth curves and a very compact data format. Each zone is exposed by a single sweep of the electron beam. We have developed a method to predict the line width as a function of line dose and defocus, which allows us to write diffractive optical elements with good line width control for structure sizes ranging from many microns down to below 50 nm.¹⁸

2 ZONE PLATES FOR THE TESTING OF EUV-LITHOGRAPHY OPTICS

The most favored technology for the future fabrication of electronic circuits with critical dimensions in the 50 nm range is extreme ultraviolet lithography (EUVL) using radiation with a photon energy around 90 eV (13 nm wavelength). Photo stepper systems for this range are presently under development, and many severe technological challenges have to be tackled in context with the development of suitable sources, photo masks, illumination optics and projection optics. The latter will probably consist of a set of aspherical Mo/Si multi-layer mirrors. To meet the system performance specifications required by the semiconductor industry, the error budgets of these mirrors with respect to surface fidelity is very stringent. Carl Zeiss, one of the main suppliers of optical components for lithography tools is presently developing an interferometer technique for the testing of EUV optics using synchrotron radiation, which requires a lens to generate a spherical wave front. For this purpose we developed zone plates for the EUV range.

At 90 eV photon energy, chromium shows a comparatively low maximum efficiency but, compared to other routinely used materials, it only requires a very low zone height as can be seen from Figure 1. This enables us to use a simple, robust technique for the fabrication of the 1.6 mm diameter Cr zone plates. Silicon nitride membranes with a thickness of 100 nm were coated with 60 nm thick Cr by thermal evaporation. The zone plate pattern was exposed into a 80 nm thick PMMA resist layer using the continuous path control mode described above. We applied reactive ion etching in a Cl_2/CO_2 plasma for the pattern transfer into the Cr layer. In Figure 2 a lens with 1.6 mm diameter, 2000 zone pairs, and 100 nm outermost zone width is shown. Each zone was divided into 3 arcs in such a way, that a set of 3 narrow radial Cr spokes remained unetched to provide a conducting connection between all zones. This is essential for an inspection of the structures after reactive ion

Figure 2: A zone plate for EUVL optics testing consisting of 60 nm thick Cr structures on a 100 nm thick Si_3N_4 membrane. a) visible light microscope image showing an overview of the 1.6 mm lens with a 200 μm central stop. b) SEM image of the outermost 100 nm wide zones. Central region before (c) and (d) after a self-aligned deposition of a central stop consisting of 1.2 μm thick photo resist.

etching to avoid severe electrostatic charging. The inner region was left unpatterned to provide an opaque central stop of 200 μm in diameter. The transmission for 90 eV radiation of this central region is in the order of 5%. To enhance the absorption, we reinforced the central stop using a self aligning procedure: the zone plate is coated by 1.2 μm thick positive tone photo resist and exposed from the back side using a mask aligner. As the visible light transmission of the patterned part of the zone plate is significantly higher than that of the unpatterned Cr layer, the resist is removed only in the patterned regions. In Figure 2 the central region of a zone plate (c) without and (d) with the patterned resist is shown. The remaining transmission of the reinforced stop is in the order of 10^{-5} .

To determine the efficiency values obtainable with the described technique, a set of linear gratings fabricated using the same method was investigated at the Center for X-ray Lithography (CXrL) in Madison, Wisconsin, US. For gratings with 100 nm lines and spaces, absolute first order diffraction efficiencies up to 4.5% were measured. Taking into account the comparatively low transmission of the support membrane of about 40%, this corresponds to 11.3% efficiency of the Cr structures which is close to the theoretical value of 13.5 % for 60 nm thick Cr. Future improvements of the total efficiency will be attempted by using Si support membranes which have significantly less absorption.

3 SOFT X-RAY ZONE PLATES FOR RADIOBIOLOGICAL APPLICATIONS

In order to improve the understanding how radiation interacts with living cells and tissues, an x-ray microprobe using the characteristic carbon K-radiation from a graphite x-ray tube (278 eV) has been developed recently by the Gray Cancer Institute, Northwood, UK. With this system it is possible to deliver precise doses of radiation to selected individual cells *in vitro*, or to pre-selected targets within cells. A more detailed description of this system and experimental results are given at this conference¹⁹.

We developed Fresnel zone plates optimized for operation at 278 eV. Two types of lenses with focal lengths of 9 mm have been fabricated: a) with 400 μm diameter and 100 nm outermost zones and b) with 800 μm diameter and 50 nm outermost zones. The lenses are mounted at about 170 mm from the source, which has a size of about 5 μm . Since the source is only demagnified by a factor of 18, the size of the focal spot is of the order of 500 nm, and thus not limited by the outermost zone width. The larger diameter has merely the advantage of collecting a larger solid angle, and delivering a higher flux in the spot.

The zone plates were made from 250 nm thick silicon nitride membranes with a size of 2 mm $\times$ 2 mm coated with 15 nm of Cr and a 50 nm top layer of PMMA resist. In contrast to the EUV zone plates described in the previous section, the Cr layer serves different purposes. Firstly, electrostatic charging during the pattern definition by electron beam lithography is prevented. Secondly, it serves as a hard mask during the pattern transfer process into the Si₃N₄ membrane (see Figure 3 left). The optimum height of the zone structures is about 200 nm, so that the 250 nm thick membranes have to be etched almost completely through. In consequence, the membranes tend to break due stress induced by the heat load during the reactive ion etch process. We were able to prevent this by interrupting the process every 15 seconds to allow for cooling of the substrates. The efficiency of both types of lenses was investigated using the x-ray microprobe. Efficiencies of up to 14% and 11% for the small and the large diameter devices respectively were measured. The lower value for the large zone plate can be explained by the fact, that the smaller zone structures down to 50 nm in size were transferred with less pattern fidelity. Nevertheless, the four times larger solid angle collected by the 800 μm diameter lenses leads to an increase in flux by a factor of 3.

Figure 3: Fabrication process by electron beam lithography and reactive ion etching of Si₃N₄ (left). Visible light microscope image of a 400 μm diameter lens with 100 nm outermost zone width (center). SEM image of the 100 nm wide outermost zones (right).

4 A ZONE PLATE BASED BEAM MONITOR FOR THE SLS

Source imaging can provide valuable information about the size, shape, position, and stability of 3rd generation synchrotron radiation sources, and allows for an optimization of the brilliance by minimizing the coupling parameter²⁰⁻²². At Swiss Light Source, a bending magnet beamline is reserved for diagnostics purposes. At this location electron beam size, emittance and coupling measurements are planned. In addition, bunch purity and bunchlength measurements must be performed at the same beamline using visible synchrotron radiation. For beam size measurements we are expecting horizontal and vertical 1-sigma beam size of about 50 μm for 1% emittance coupling. However, the storage ring design allows much smaller coupling values for different lattice modes. Emittance coupling of 0.1% would already lead to a vertical 1-sigma beam size of 13 μm . Thus a resolution in the range of some microns is required which is a value, that cannot be obtained using pin-hole cameras²⁰.

Apart from the required resolution, some practical constraints have to be taken into account. Firstly, it is desirable to have no reflective components in the imaging system that could introduce aberrations and thus affect the source size measurements. Secondly, it is advantageous to have no components inside the beamline's front end. This allows for easier access and alignment, and removal of the set-up from the beam without complications so that the beamline can be used for other experiments. In our case, this means that the first component of the monitor has a distance from the source of at least $g=11$ m. Generating a magnified image of the source would thus require a very long, potentially unstable set-up. To keep the set-up short, a demagnifying geometry was chosen.

The separation of two distant point sources that can be distinguished according to the Rayleigh-criterion is limited by the diffraction at the optics aperture²³. This means that the minimum distance between two source points that can be resolved according to the Rayleigh criterion is $G = 1.22 \cdot \lambda/\alpha$, where $\alpha = 2\pi/g$ the solid angle collected by an optics with the radius r situated at a distance g from the source. Thus, unlike the situation in microscopy applications, the resolution of a demagnifying set-up does not depend on the outermost zone width. The angular divergence of our beam line is in the order of 0.5 mrad which limits the useful optics diameter to 5 mm at 10 m distance from the source. This results in a diffraction-limited resolution of 1.7 μm at 1.8 keV photon energy (0.7 nm wavelength).

Figure 4: Scanning type set-up for beam monitoring planned at the Swiss Light Source

There are two principal ways of imaging a synchrotron source. Either the source image is collected by a spatially resolving detector like a phosphorous screen or a CCD camera. In this case, the granularity of the detector requires a 1:1 or even a magnifying set-up. To avoid these difficulties we chose the principle based on scanning a pinhole through the image plane of a zone plate as shown in Figure 4. The source is imaged by zone plate and the (demagnified) image is recorded by scanning a pin-hole in the image plane while detecting the transmitted flux. An order selecting aperture (OSA) and a central stop remove all radiation from other diffraction orders. Since the zone plate is highly chromatic, the focused source image for the wavelength corresponding to the distance between zone plate and pinhole is superimposed by the defocused images of other wavelengths. To suppress these defocused contributions, a multilayer mirror or a crystal can be used as a band-pass filter. This filter can be very small in size, and it does not have to meet any further demands, e.g. with respect to its flatness. As a detector a simple photo-diode can be applied. The set-up is easy to align since all optical components except for the detector are on the optical axis.

Table 1: Parameters of the planned SLS beam monitor

wavelength	0.7 nm	outermost zone width	0.35 μm
photon energy	1.8 keV	Central stop diameter	2 mm
distance source – zone plate	10 m	number of unobstructed zone pairs	1070
zone plate diameter	4 mm	required bandwidth $\lambda/\delta\lambda$	500
diffraction limit of resolution	2.1 μm	pinhole diameter	0.4 μm
focal length	2 m	field of view ($3\times$ expected FWHM)	300 μm
distance zone plate - source image	2.5 m	range of pinhole scanner	75 μm
demagnification factor	4	Number of pixels	256 x 256

The most important decision that has to be taken is which wavelength to use. Although the diffraction limit of the obtainable resolution becomes more favorable for shorter wavelengths, radiation in the 0.8 nm wavelength range (1.8 keV photon energy) will be used for a number of practical reasons: firstly, this energy range is very well suited for the fabrication of zone plate optics with the required dimensions and high diffraction efficiency. Particularly silicon, a material easy to pattern by reactive ion etching, has an excellent ratio of phase shift and absorption. This means that phase zone plates with efficiencies up to 35% can be fabricated (see Figure 1). Secondly, the proposed method requires a pinhole for image formation with a sub-micron diameter. For hard x-rays such pinholes are extremely difficult to obtain with high optical contrast. For 0.7 nm radiation, the transmission of a 1 μm thick Ta layer is already below $1\cdot 10^{-3}$, which allows for the application of thin film pinholes fabricated by Focused Ion Beam technology²⁴ (see Figure 5). Compared to lower energies, 1.8 keV radiation has the advantage of a higher transmission through filters and support membranes (93% transmission for 1 μm thick Si). Taking into account the above considerations, the layout of the beam monitor was planned as given in Table 1.

Figure 5: SEM images of the imaging components of the planned beam monitor for the Swiss Light Source. Left image: central structures of a 4 mm diameter zone plate etched into silicon. Central image: 350 nm wide outermost zone structures. Right image: half-micron diameter pinhole milled through a 5 μm thick Ta foil (right) that will be used to scan the source image.

The zone plates for the described set-up are presently made at the LMN from 250 micron thick Si substrates. We use photo lithography and deep reactive ion etching to form approximately 5 μm thick membranes. No etch stop layer is used and the membrane thickness is merely controlled by the etching time. The zone plate pattern is then exposed onto the membrane and etched 2-3 μm deep into the silicon by reactive ion etching using a 25 nm thick Cr hardmask.

5 ZONE PLATES OPTIMIZED FOR SULPHUR X-RAY FLUORESCENCE MICROSCOPY

For many samples the fluorescent x-ray emission resulting from absorption of the incident beam offers useful chemically sensitive information about the system under study. In particular, the vastly reduced Brems-strahlung background confers a trace element sensitivity which can be significantly better than that possible using electron excitation. Use of this fluorescence emission allows the study of bulk, non-conducting samples and is therefore also of interest for systems which cannot be prepared in suitably thin sections for standard absorption contrast microscopy. On the ESRF x-ray microscopy beamline ID21, a scanning microscope (SXM) is in operation²⁵ which is capable of detecting fluorescence signals at medium photon energies from 2-8 keV giving access to K-absorption edges of P, S, K and Ca. Sulphur (K-lines X-ray fluorescence at 2307–2464 eV) is of great importance in applications such as biology, medicine, and geochemistry.¹ The main problem in using fluorescence signals from sulphur is the low fluorescence yield for K-line emission of only 0.078²⁶. Therefore, this technique depends upon the quality of the microfocusing optics which must deliver high fluxes into the smallest possible probe size. We chose germanium as zone plate material which, at 2.4 keV, allows up to 23% of the incoming radiation to be dif-

Figure 6: (a) SEM image of a *Ge* zone plate with 100 nm outermost zone width optimized for S fluorescence x-ray microscopy. (b) SEM image of a ZnS test object with structure widths ranging from 50 nm to 1 μm . (c) SEM image of ZnS structures ranging from 75 nm to 175 nm. (d, e) S-fluorescence image of the test object, structures down to below 100 nm line width can be resolved.

fracted into the focal spot. However this would require a structure height of 1350 nm, leading to a very challenging aspect ratio of 13.5 for 100 nm wide outermost zones. Therefore the zone height was chosen to be 900 nm, which limits the obtainable efficiency to 18.3% (see Figure 1) but greatly facilitates the fabrication process. The flux that can be achieved in the focal spot of the lens also depends on the light collecting area. For diffraction limited probe sizes the maximum useful diameter of the zone plate, d , depends on the degree of spatial coherence provided by the source. In the SXM at the beamline ID21 of ESRF, the zone plate diameter can be as big as 1000 μm and still provide diffraction limited resolution in the vertical direction, while in the horizontal direction, only 180 μm of the zone plate is coherently illuminated. In order to have reasonable count rates, the zone plate diameter was chosen to be 300 μm with a focal length of 60 mm at 2.4 keV photon energy. In consequence, diffraction limited resolution can only be expected in the vertical direction.

The zone plates are supported by 250 nm thick silicon nitride membranes which have a transmission of 90% at 2.4 keV photon energy. The membranes are coated by thermal evaporation with a 10 nm thick adhesive layer of chromium, 900 nm of germanium as phase shifting material, and a 30 nm thick top layer of chromium. A 60 nm thick layer of polymethylmethacrylate (PMMA) is spincoated as an electron beam resist. The resist pattern is first transferred into the 30 nm thick Cr layer by reactive ion etching in a Cl_2/CO_2 plasma. The thin Cr layer serves as a hard mask for etching of the Ge layer using a CBrF_3 plasma. Figure 6a shows scanning electron micrographs of the resulting Ge structures. The diffraction efficiency of the zone plates was measured to be up to 9.6% at 2.4 keV photon energy.

To test the properties of the ID21 SXM using sulphur K line X-ray fluorescence emission as the imaging signal, zinc sulphide (ZnS) Siemens stars were generated by electron beam lithography and lift off techniques. The structures shown here have a thickness of 26 nm, their structure widths vary from 1 μm down to 50 nm (Figure 6b and 6c). The sample was scanned with radiation just above the sulphur K-absorption edge at 2472 eV. To limit the background signal from the uncoated regions, a Ge substrate was chosen. Scanning X-ray fluorescence images of the test structures are shown in Figure 6d and 6e. The smallest structures that could be resolved have a half pitch of just below 100 nm. However, close to the resolution limit the contrast is deteriorated by significant shot noise. Considering a 100 nm-diameter probe, this signal originates from less than $3 \cdot 10^{-16}$ g (300 age) of sulphur and demonstrates the enormous sensitivity of the method. More detail on these experiments can be found elsewhere²⁷.

6 SILICON BRAGG-FRESNEL LENSES

Bragg-Fresnel lenses (BFL) provide an interesting method for focusing of hard x-rays ($E > 6$ keV). The principle of operation combines the Bragg-reflection of a crystal with the focusing properties of a Fresnel zone plate pattern etched into the crystal²⁸. Round lenses in near-normal incidence for two dimensional focusing have been applied e.g. in scanning systems for microanalysis²⁹ and for source imaging in order to monitor the electron beam of synchrotron sources^{30,31}. The disadvantage of this configuration is the restriction to a very narrow wavelength range given by the crystal. A more flexible set-up that can be used over a wide wavelength range is based on linear BFLs³² to generate a line focus. Although silicon BFLs are the most common, other crystals like Ge, GaAs, InP, and mica have been reported. Furthermore, Bragg-Fresnel Multilayer Lenses (BFMLs) based on patterned multilayer mirrors have been used successfully for x-ray focusing and imaging.

The phase shift between x-rays passing through the trenches of a BFL and those passing through the ridges should be equal to π for maximum efficiency. Using silicon as the lens material, it can be calculated that the optimum zone height is $1.24 \mu\text{m}$ ³³. This value does not depend on the photon energy, because the wavelength dependency of the optical constants is compensated by the change of the Bragg angle which in turn increases the optical path through the zone structures. Therefore, the same device can be used for a wide range of photon energies.

We fabricated various linear BFLs on Si $\langle 111 \rangle$ substrates with 100 nm outermost zone width which is significantly smaller than for any other Si BFL reported so far. The structures were etched into the substrate to a depth of more than $1 \mu\text{m}$ by reactive ion etching. Figure 7 shows a cross-section through a silicon BFL. The lenses have dimensions ranging from $125 \mu\text{m}$ to $1000 \mu\text{m}$ in width and 5 mm to 10 mm in length. The diffraction efficiency was measured at the dipole beam line BM 5 of ESRF. We obtained efficiency values of up to 26% at 13.25 keV photon energy¹³. The combination of wide aperture angle, high efficiency and large size of our devices results in a significant increase in achievable x-ray flux.

Figure 7: Cross section through a Bragg-Fresnel lens with 100 nm outermost zones etched into a $\langle 111 \rangle$ oriented silicon substrate.

7 WET ETCHED LINEAR TRANSMISSION LENSES AS CONDENSERS FOR WAVEGUIDING EXPERIMENTS

The main drawback of BFLs is due to the fact that the optical axis of the setup has to make a large additional angle which - in contrast to mirror optics - changes with the photon energy. Moreover the lens's Bragg condition makes it only suitable for a narrow bandwidth and difficult to align when used in combination with a crystal monochromator. These problems become even more severe when a two-dimensional focusing by a combination of two linear BFLs with orthogonal orientation is envisioned. Therefore, transmission lenses avoiding these difficulties would be most useful in such applications. A possibility for the fabrication is presented in this section.

A novel method for the investigation of the fundamental mechanisms of lubrication and friction phenomena at the nanometer scale has been developed in the past years^{34,35}. Small angle scattering of hard x-rays is used to reveal ordering phenomena in liquids confined in a small gap between two opposing plates. It has been conjectured that the ordering effects within the fluid induced by the confinement strongly modify the fluid's lubricating and rheological properties³⁶. When the gap between the plates is in the order of a few hundred nm, the confining surfaces can be used as a waveguide for x-rays with the liquid acting as the waveguide core. Depending on the gap, w , the wavelength and the angle of incidence, θ_{in} , discrete waveguiding modes can be excited which determine the intensity distribution in the far field behind the waveguide exit (see figure 8). Ordering of the spatial arrangement of the fluid's constituents (molecules or suspended colloidal particles) within the waveguide gap can be detected as mode coupling, i.e. as scattering of x-rays into modes that would not have been excited in a disordered system. The distribution of intensity over the modes exiting the waveguide is detected in the far field and from the intensity distribution we reconstruct the density profile of the fluid across the gap. Using this method, a layer-wise ordering in solutions of colloidal nanoparticles confined in gaps down to a few hundred nanometer has been observed recently³⁵.

Figure 8: Schematic of the waveguiding geometry. For illustration, the waveguide is filled with two layers of order fluid and a TE_2 mode is drawn at the entrance (left). Focusing the beam onto the entrance of the waveguide using a linear Fresnel zone plate can significantly increase the signal detected in the far field behind the waveguide set-up (right). Angles and distances are not to scale.

The present research activities focus on photon correlation spectroscopy (XPCS) measurements in the time domain, enabling us to determine, for example the anisotropic diffusion coefficients of colloidal particles in the directions along and perpendicular to the confining directions. Such investigations are extremely “photon hungry”. Even at state-of-the-art insertion-device synchrotron beamlines, the obtainable temporal resolution is not sufficient to clearly identify the relevant dynamical effects with sufficient signal-to-noise ratio. We therefore developed condenser lenses to enhance the intensity coupled into the waveguide gap. The linear lenses were fabricated from $\langle 110 \rangle$ oriented silicon substrates by electron beam lithography and selective wet etching. SEM images of a wet etched linear silicon Fresnel lens with approximately $13\text{ }\mu\text{m}$ high structures are given in Figure 9. The outermost zone width is width 300 nm , corresponding to an aspect ratio of about 40. The used fabrication technology is described in more detail elsewhere³⁷.

Figure 9: SEM images of a linear silicon lens fabricated by electron beam lithography and wet etching. The structures are approximately $13\text{ }\mu\text{m}$ high; the outermost zone width is 300 nm , resulting in an aspect ratio of about 40.

For 13.3 keV photon energy as used in our waveguiding experiments, it can be calculated that binary Si structures should be $17\text{ }\mu\text{m}$ high to give the highest diffraction efficiency. We used various lenses and optical arrangements to increase the flux coupled into the waveguide set-up. The highest gain we achieved was a factor of 50. This value was obtained using a $200\text{ }\mu\text{m}$ aperture lens with 750 mm focal length and 350 nm outermost zones installed at the undulator beam line ID 22 of the ESRF. Developments for the fabrication of wet etched linear silicon lenses with outermost zone widths down to 100 nm optimized for coherence matched micro-focusing are ongoing³⁸.

8 BLAZED DIFFRACTIVE X-RAY OPTICS WITH STAIR-CASE PROFILES

Even in the case of zero absorption, diffractive optical elements consisting of binary structures are limited to a maximum diffraction efficiency of 40.5%. Higher values can only be obtained by using asymmetric, i.e. blazed structure profiles. The structures should ideally provide a phase shift of the light passing through which gradually increases from zero to 2π to give

Figure 10: SEM image of a blazed Si zone plate with 480 nm outermost period, 1.5 μm structure height and 100 μm diameter (left). Cross-section of 550 nm pitch structures with a blazed profile (right).

100% diffraction efficiency. Such “saw tooth” profiles are very difficult to make, especially when high aspect ratios are required. A reasonable approximation of a continuous profile can be obtained by stair-cases of discrete height levels. The achievable efficiency is limited to 68%, 81%, and 95% for 3, 4, and 8 levels respectively, when absorption is neglected. Until recently transmission diffractive optics for the x-ray regime only consisted of binary structures. The reason for this lies in the fact that wavelengths in the nanometer range or below require very small structures to maintain acceptable diffraction angles. In addition comparatively high structures are required to provide sufficient phase shift or absorption for reasonable diffraction efficiencies. The first blazed zone plates with efficiencies clearly beyond those obtainable with binary structures have been fabricated by Di Fabrizio et al. for 7 keV x-rays using aligned x-ray lithography steps and electroplated nickel structures¹⁰. These zone plates have widths of the top (4th) height level down to 500 nm, i.e. an outermost pitch of 2 μm. We pursue a technique based on selective reactive ion etching of silicon to obtain discrete height levels described elsewhere in more detail³⁹. An example of the obtained structures is shown in Figure 10. We achieved structures with height levels widths down to 120 nm, i.e. 480 nm pitch, and structure heights of 1.5 μm. These devices are well suited photon energies around 1 keV. With four height levels, efficiencies almost twice as high as for a binary structure can be obtained. The required structure height increases drastically when approaching the absorption edge at 1.8 keV. Below 1 keV, less than 2 μm height is sufficient. When applied to linear diffractive structures, the presented fabrication method should be applicable for hard x-rays, when the devices are tilted with respect to the incident x-ray beam. A dedicated paper dealing with the possibilities of this technique is presented in these proceedings³⁸. The developed selective etching process of stair-case profiles can also be applied to other materials in Addition to *Si*. Especially for the hard x-ray range, blazed optical elements etched into heavier elements such as *Ta* would provide excellent efficiency at moderate aspect ratios.

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REFERENCES

1. B. Niemann, D. Rudolph, G. Schmahl, *Optics Communications* **12**, 160 (1974)
2. D. Attwood, *Soft X-Rays and Extreme Ultraviolet Radiation*, Cambridge University Press, Cambridge (1999) p. 357
3. M. Born and E. Wolf, *Principles of Optics*, 4th edition, Pergamon Press (1970) p. 415
4. G. Schneider, T. Schliebe, H. Aschoff, *J. Vac. Sci. Technol.* **B 13** 2809 (1995)
5. S.J. Spector, C.J. Jacobsen, D.M. Tennant, *J. Vac. Sci. Technol.* **B 15** 2872 (1997)
6. J. Kirz, *J. Opt. Soc. Am.* **1**, 301 (1974)
7. B.L. Henke, E.M. Gullikson, J.C. Davis: *Atomic Data and Nuclear Data Tables* **54**, 181 (1993)
8. E. Anderson and D. Kern, in: *X-Ray Microscopy III*, Eds.: A.G. Michette, G.R. Morrison, C.J. Buckley, 75 Springer Verlag, Berlin (1990) p. 75
9. D. Weiss, M. Peuker, G. Schneider, *Appl. Phys. Lett.* **72**, 1805 (1998)
10. E. Di Fabrizio F. Romanato, M. Gentili, S. Cabrini, B. Kaulich, J. Susini, R. Barrett, *Nature*, **401**, 895 (1999)

11. M. Panitz, G. Schneider, M. Peuker, D. Hambach, B. Kaulich, S. Oesterreich, J. Susini, G. Schmahl, in: *X-Ray Microscopy: Proceedings of the Sixth International Conference*, Eds.: W. Meyer-Ilse, T. Warwick, D. Atwood (AIP, Melville N.Y., 2000), p. 676
12. W. Yun, B. Lai, Z. Cai, J. Maser, D. Legnini, E. Gluskin, Z. Chen, A.A. Krasnoperova, Y. Vladimirovsky, F. Cerrina, E. Di Fabrizio, M. Gentili, *Rev. Sci. Instrum.* **70**, 2238 (1999)
13. C. David, A. Souvorov: *Rev. Sci. Instr.* **70**, 4168 (1999)
14. C. David, J. Thieme, P. Guttmann, G. Schneider, D. Rudolph, G. Schmahl, *Optik* **91** 95 (1992)
15. C. David, B. Kaulich, R. Medenwaldt, M. Hettwer, N. Fay, M. Diehl, J. Thieme, G. Schmahl, *J. Vac. Sci. Technol.* **B13** 2762 (1995)
16. A. Ozawa, T. Tamamura, T. Ishii, H. Yoshihara, T. Kagoshima, *Microelectron. Eng.* **35**, 525 (1997)
17. P. Charalambous, in: *X-ray Microscopy: 6th International Conference*, Eds.: W. Meyer-Ilse, T. Warwick, D. Attwood, AIP 458 (2000) p. 625
18. C. David and D. Hambach, *Microelectron. Eng.* **46** 219 (1999)
19. M. Folkard, G. Schettino, B. Vojnovic, A.G. Michette, C. David, S.J. Pfauntsch, K.M. Prise, B.D. Michael, this conference
20. P. Elleaume, C. Fortgang, C. Penel, E. Tarazona, J. Synchrotron Rad. **2**, 209 (1995)
21. W.B. Peatman, K. Holldack, J. Synchrotron Rad. **5**, 639 (1998)
22. Z. Cai, B. Lai, W. Yun, E. Gluskin, D. Legnini, P. Illinski, G. Srajer, *Rev. Sci. Instr.* **67**, 1 (1996)
23. M. Born, E. Wolf, "*Principles of Optics*", Pergamon Press (1980) p. 459
24. C. David, C. Musil, A. Souvorov, B. Kaulich, in: *X-ray microscopy, 6th International Conference*, Eds.: W. Meyer-Ilse, T. Warwick, and D. Attwood, AIP (2000) p. 704
25. R. Barrett, B. Kaulich, M. Salomé, J. Susini, in: *X-ray Microscopy: 6th International Conference*, Eds.: W. Meyer-Ilse, T. Warwick, D. Attwood, AIP (2000) p. 458
26. M.O. Krause, *J. Phys. Chem. Ref. Data.* **8**, 307 (1979)
27. C. David, B. Kaulich, R. Barrett, M. Salomé, J. Susini, *Appl. Phys. Lett.* **77**, 3851 (2000)
28. V.V. Aristov, Y.A. Basov, A. Snigirev: *Rev. Sci. Instr.* **60**, 1517(1989)
29. S.M. Kuznetsov, I. Snigireva, A. Snigirev, P. Engstrom, C. Riekel, *Appl. Phys. Lett.* **65**, 827 (1994)
30. E. Tarazona, P. Elleaume, J. Chavanne, Ya. Hartman, A. Snigirev, I. Snigireva, *Rev. Sci. Instrum.* **65**, 1959 (1994)
31. K. Holldack, A. Erko, T. Noll, W.B. Peatman, *Nucl. Instrum. Methods Phys. Res.* **A365**, 40 (1995)
32. V.V. Aristov, Y.A. Basov, A. Snigirev, V.A. Yunkin, T. Ishikawa, S. Kikuta, *Nuc. Instrum. Methods Phys. Res.* **A308**, 413 (1991)
33. S.V. Babin, A.I. Erko, *Nuc. Instrum. Methods Phys. Res.* **A282**, 529 (1989)
34. M.J. Zwanenburg, J.F. Peters, J.H.H. Bongaerts, S.A. de Vries, D.L. Abernathy, J.F. van der Veen, *Phys. Rev. Lett.* **82**, 1696 (1999)
35. M.J. Zwanenburg, J.H.H. Bongaerts, J.F. Peters, D.O. Riese, J.F. van der Veen, *Phys. Rev. Lett.* **85**, 5154 (2000)
36. B. Bushan, J.N. Israelachvili, U. Landman, *Nature* **374**, 607 (1995)
37. C. David, E. Ziegler, B. Nöhammer, *J. of Synchrotron Rad.* **8**, 1054 (2001)
38. C. David, *Microelectronic Engineering* **53** (2000) 677 - 680
39. C. David, B. Nöhammer, E. Ziegler, O. Hignette, this conference