High-efficiency Bragg–Fresnel lenses with 100 nm outermost zone width

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In this article, we describe the manufacture and characterization of linear silicon Bragg–Fresnel lenses (BFLs) with outermost zone widths of 100 nm. This is, to our knowledge, a factor of 3 smaller than any other device of this kind reported so far. Furthermore, we give the first detailed description of an accurate and robust method to measure the diffraction efficiency of BFLs. The first-order diffraction efficiency of our lenses at 13.25 keV photon energy was $26\% \pm 0.5\%$. The combination of wide aperture angle, high efficiency, and large size of our devices results in a significant gain in achievable x-ray flux. © 1999 American Institute of Physics.

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I. INTRODUCTION

Bragg–Fresnel lenses (BFLs) provide a versatile 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 crystal.¹ BFLs have been used successfully in various setups. Round lenses in near-normal incidence for two-dimensional focusing² [Fig. 1(a)] have been applied, e.g., in scanning systems for microanalysis³ and for source imaging in order to monitor the electron beam of synchrotron sources.^{4,5} The disadvantage of this configuration is the restriction to a very narrow-wavelength range given by the crystal. A more flexible setup that can be used over a wide-wavelength range is based on linear BFLs (Ref. 6) to generate a line focus [Fig. 1(b)]. Additional focusing in the second direction can be achieved by bending of the crystal as shown in Fig. 1(c).⁷

Although silicon BFLs are the most common, other crystals like Ge,⁸ GaAs, InP,⁹ and mica¹⁰ have been reported. Furthermore, it should be mentioned that a variety of Bragg–Fresnel multilayer lenses (BFMLs) based on patterned multilayer mirrors have been used successfully for x-ray focusing and imaging.^{5,11,12} In this article, however, we restrict ourselves to BFLs based on crystals.

The light-collecting power of a lens is basically given by three factors: First, the lens should have a large aperture in order to collect a large fraction of the incoming beam. Second, the transmitted radiation should be delivered into a small spot. Third, the losses of the lens should be as low as possible. This means that the bandwidth of the radiation transmitted by the lens should be as large as possible, and within this band, a maximum diffraction efficiency is desirable. The diffraction limit of a BFL's spatial resolution is given by $d = 1.22dr_n$, where dr_n is the outermost zone width. However, in many cases where the BFL is used to focus x rays into a spot, the source image, given by the

source size and the (de)magnification factor of the setup determines the achievable spot size. Therefore, it is advantageous to use BFLs with a short focal length, which (for a given aperture and wavelength) can only be achieved by decreasing dr_n .

The spectral bandwidth of a BFL is determined by the width of the Bragg reflex, i.e., typically in the range of $\delta\lambda/\lambda = 10^{-4} - 10^{-5}$. For diffraction-limited focusing of a BFL, the required bandwidth is reciprocal of the zone number. Therefore, a bandwidth on the order of $10^{-2} - 10^{-3}$ is sufficient. For many applications of x-ray focusing, where a small spectral bandwidth is not necessary, it would be desirable to increase the width of the crystal's Bragg reflex in order to enhance the obtainable flux. Several experiments have been carried out which increase the reflected bandwidth by distorting the crystal, e.g., by ion implantation¹³ or ultrasonic excitation.¹⁴ Alternatively, the application of strained, epitaxially grown, alloy layers, e.g., in the Si/SiGe material system, seems to be possible.

The diffraction efficiency of a BFL depends on the shape and height of the zone plate structures. For zone structures with a rectangular profile, the first-order diffraction efficiency reaches its maximum for a duty cycle of 0.5 (line-to-space ratio of 1:1). The phase shift ϕ between x rays of wavelength λ passing through the trenches of a BFL and those passing through the ridges should be equal to π for a maximum diffraction efficiency of 40.4%. It is given by

$$\phi = \frac{4\pi}{\lambda} \frac{\delta h}{\sin \theta},$$

where h is the height of the zone structures, $1 - \delta$ the real part of the complex refractive index $n = 1 - \delta - i\beta$ of the zone plate material, and θ is the Bragg angle. Using silicon as the lens material, it can be calculated¹⁵ that the optimum zone height is $1.24 \mu\text{m}$. This value does not depend on the photon energy,¹⁶ as the wavelength dependency of δ is compensated by the change of θ . The influence of the zone profile on the diffraction properties of a BFL is described in detail by Hart-

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FIG. 1. Three basic setups for x-ray focusing using BFLs: two-dimensional focusing in near-normal incidence (a), one-dimensional focusing using a linear BFL (b), and two-dimensional focusing using a linear BFL on a bent crystal (c).

man, Yunkin, and Snigirev.¹⁷ However, experimental data on the efficiency of these lenses is very sparse. To our knowledge, the only report on the efficiency of a linear silicon BFL with 500 nm outermost zone width is given by Aristov *et al.*¹⁸ Here, an efficiency of “just below 30%” was derived by microdensitometry on a photographic plate, a method that can be expected to be fairly inaccurate.

II. ZONE PLATE MANUFACTURE

The main difficulties in the fabrication process of a BFL are twofold. First, the pattern generation has to provide high-precision accuracy for a large number of lines with continuously varying linewidth. Second, the structures have to be transferred into the phase-shifting material. For small outermost zone widths, this results in challenging aspect ratios of the structures.

A. Pattern definition by electron-beam lithography

The size of linear BFLs is generally larger than the deflection field of the standard electron-beam system. As a consequence, the BFL pattern has to be stitched together by a

number of subfields with sufficient accuracy. The LION-LV1 e-beam lithography system of the Paul Scherrer Institute (PSI) (Leica Microsystems Lithography Jena) used in our work has some special features which make it particularly suited for the generation of diffractive optics. Its continuous path control mode (CPC) can produce an 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.

For every line element, the exposure dose can be set individually by adjusting the stage speed, and the beam can be defocused in a defined way. It is, therefore, possible to generate lines with well-defined width by a single sweep in the CPC mode, which can considerably reduce the total exposure time. We have developed a method to predict the linewidth as a function of line dose and defocus, which allows us to write diffractive optical elements with accurate linewidth control for structure sizes ranging from several microns down to below 100 nm.¹⁹ This method was applied to the writing of linear BFL patterns with lengths up to 10 mm, widths up to 1 mm, and zone numbers of up to 2500 zone pairs. These dimensions are significantly larger than those of any other BFL reported so far. The exposures were performed at 2.5 keV electron energy on 50-nm-thick polymethacrylate (PMMA) resist. An overview of the generated zone plate patterns is given in Table I.

B. Pattern transfer by reactive ion etching

In order to give a high diffraction efficiency, the BFL pattern in PMMA has to be etched into the Si <111> substrate to a depth of more than 1 μm , which results in an aspect ratio of the outermost 100-nm-wide structures of more than 10. For this purpose, we developed a highly anisotropic and highly selective reactive ion etching (RIE) process. The resist pattern is first transferred into a 20-nm-thick Cr layer by reactive ion etching in a Cl_2/CO_2 plasma. The thin Cr layer then serves as a mask for a second etching step in a gas mixture of 5 sccm (standard cubic centimeters) O_2 , 4 sccm SF_6 , and 35 sccm CBrF_3 at a pressure of 20 mTorr and 200 V of self-bias voltage.

TABLE I. Parameters of the BFLs patterns fabricated by e-beam lithography.

Zone plate parameters	BFL1	BFL2	BFL3	BFL4
Length	3 mm	3 mm	3 mm	10 mm
Width	0.125 mm	0.250 mm	0.5 mm	1 mm
Number of zone pairs	312	625	1250	2500
Outermost zone width	100 nm	100 nm	100 nm	100 nm
Focal length at 13.25 keV photon energy (0.0936 nm wavelength)	0.16 m	0.33 m	0.66 m	1.32 m
Numerical aperture at 13.25 keV photon energy	7.6×10^{-4}	7.6×10^{-4}	7.6×10^{-4}	7.6×10^{-4}

FIG. 2. Scanning electron micrographs of the cross section through the silicon structures of the BFL used in the efficiency measurements. The outermost zone width is 100 nm (200 nm period). The depth of the zone structures is $1.2\ \mu\text{m}$ for the central zones; it gradually decreases to $1.05\ \mu\text{m}$ for the outermost zones.

Figure 2 shows scanning electron micrographs of a cross section through the resulting structures. Compared to other etching processes used for BFL fabrication,¹⁷ only a very weak dependency of the trench depth on the structure width was observed. The depth ranges from $1.20\ \mu\text{m}$ for the central zones to $1.05\ \mu\text{m}$ for the 100-nm-wide outermost zones. In the model of a rectangular zone profile and a duty cycle of 0.5, this corresponds to a diffraction efficiency of 40.4% for the central and 37.8% for the outermost structures. The shape of the smaller structures deviates significantly from a rectangular shape, although this should not reduce the obtainable efficiency significantly.¹⁷ In between the wider zones, the formation of “grass,” i.e., extreme roughness due to micro-masking, can be observed. This may lead to losses in efficiency and increased diffuse scattering, which we were not able to quantify in our experiments.

III. EXPERIMENTAL CHARACTERIZATION

A. Experimental setup

The BFLs were tested at the Optics Beam Line BM5, which is installed at a bending magnet of the European Synchrotron Radiation Facility (ESRF) facility. The measurements were performed without any monochromator, employing only a Pt-coated Si mirror at 0.3° incident angle to suppress higher the harmonics of the BFL crystal. This angle corresponds to a cutoff energy of 15 keV. The BFL crystal was mounted to deflect the beam in the horizontal direction. The glancing angle was set to 8.6° , resulting in a $\langle 111 \rangle$ Bragg reflection at 13.25 keV photon energy. It can be calculated,²⁰ that the reflectivity of the illumination mirror is at least a factor of 300 less for the $\langle 333 \rangle$ reflection (39.75 keV), so all contributions of higher harmonics can be neglected.

The measurements described below were carried out using the lens shown in Fig. 2. The geometrical parameters of the lens (type BFL3) used here are given in Table I. The resolution and efficiency of the BFL were measured by ver-

tically scanning a slit with adjustable width through the reflected beam at a distance equal to the focal length for 13.25 keV radiation. The advantage of using a (nearly) white beam is the fact that the exact Bragg condition is always fulfilled. Thus, artifacts caused by intensity fluctuations induced through the coupling of the slit position with the Bragg angle are greatly reduced. The slit length is not important in our setup and we chose a length of approximately $10\ \mu\text{m}$, which greatly facilitates the alignment of the slit axis with respect to the line focus, and which still provides sufficient flux to allow for high signal to noise. For alignment purposes, a high-resolution charge-coupled device camera was used while the measurements were carried out using a silicon photodiode. The diode currents in our efficiency measurements ranged from 1 nA to $20\ \mu\text{A}$, which are well beyond the diode’s dark current of 11 pA. Furthermore, the values lie well within the linear range of the detector. The incident x-ray intensity varied slightly along the scan range of the measurement slit. This was compensated by moving the lens out of the beam and recording a scan signal across an unpatterned part of the Si crystal. The maximum variation of the illumination was 5%. In order to minimize the effect of this variation on the obtained efficiency values, all data used in the efficiency calculations were submitted to a flat-field correction.

B. Obtainable resolution

The width of the line focus results from the convolution of the geometrical source image with the BFL’s point spread function (PSF) and with the slit function. The geometrical image of the source can be calculated from the source size and the demagnification factor. On the BM5 bending magnet, a vertical source size of $100\ \mu\text{m}$ at full width at half maximum (FWHM) is expected for 13.25 keV radiation. From the focal length $f=0.66\ \text{m}$ and the source distance $g=40\ \text{m}$, we calculate a demagnification factor of $d=60$,

FIG. 3. Resolution measurement obtained by scanning a $0.5\text{-}\mu\text{m}$ -wide slit perpendicular to the zone plate's line focus. The obtained peak width of $2.4\text{ }\mu\text{m}$ represents the size of the (demagnified) image of the synchrotron source.

which yields a geometrical source image on the order of $2\text{ }\mu\text{m}$.

The width of the PSF should be on the order of the outermost zone width, which is about 20 times smaller. Therefore, the BFL's resolving power cannot be determined in our setup. Figure 3 shows the diode signal of a scan across the line focus with a slit width of approximately $0.5\text{ }\mu\text{m}$. The obtained width of the line focus, $2.4\text{ }\mu\text{m}$ at FWHM, is dominated by the geometrical source size. The logarithmic intensity scale reveals a large number of symmetrical satellite peaks, which can be explained by the sidelobes of the PSF of a diffractive lens with a finite number of diffracting structures.²¹

C. Efficiency measurements

For a reliable measurement of the efficiency, it is important to separate the contributions of the various diffraction orders. Figure 4 illustrates the intensity distribution in the focal plane of the first-order radiation for the converging (positive) and diverging (negative) diffraction orders up to $m = \pm 3$. Higher orders generally exist, but their intensity in the first-order focal plane can be neglected for two reasons.

FIG. 4. Schematic view showing the lateral intensity distribution for diffraction orders of a diffractive lens up to $m = \pm 3$ in the first-order focal plane. The illuminated areas have widths that are integers of the lens diameter $2r$.

FIG. 5. Schematic view of the expected detector signal when scanning a slit across the line focus of a linear diffractive lens.

First, for a duty cycle of 0.5 even orders do not exist, while for the uneven orders the efficiency decays with m^{-2} . Second, the radiation for higher orders is spread out over an area proportional to $m-1$ for converging and proportional to $m+1$ for diverging orders.

If we take into account only $m = \pm 1$ and if we assume a constant efficiency over the lens aperture, then we expect a signal shape like the one shown in Fig. 5 when scanning a slit across the line focus. The central peak (dashed line) contains the flux diffracted into the $+1$ st order, its width represents the slit size. The flux diffracted into the -1 st order is distributed over a defocused line twice as wide as the lens (dotted line). In the range from $-r$ to $+r$ excluding the central peak, the measured signal represents the sum of the -1 st order and the zero-order flux (i.e., the undiffracted radiation).

In areas more than $2r$ away from the focus, we find the flux reflected from unpatterned areas of the crystal, which has to be taken as a reference signal for the incoming flux F_0 . It is the product of the slit length l , the slit width w , and the incoming flux density D_0 within the rocking curve of the Bragg crystal:

$$F_0 = l w D_0. \quad (3a)$$

The flux F_1 diffracted into the first-order focus stems from a small area across the BFL whose width is defined by the length l of the measurement slit. Moreover, it is equal to the area under the peak curve $S(z)$ (dashed curve in Fig. 5) divided by the slit width, provided that the slit width is large compared to the source image.²²

$$F_1 = 2r l D_0 E_1 = \frac{1}{w} \int_{\text{peak}} S dz. \quad (3b)$$

From Eqs. (3a) and (3b) we derive the first-order diffraction efficiency E_1 :

$$E_1 = \frac{1}{2r F_0} \int_{\text{peak}} S dz. \quad (3c)$$

The advantage of this method is that it is not necessary to know the slit length or slit width used in the measurement. Consequently, the diffraction efficiency can be calculated

FIG. 6. Measured data of scans across the line focus of a 500- μm -wide Bragg-Fresnel lens for five different slit widths. The data have undergone a flat-field correction and they were normalized to the ring current.

from a single scan. Furthermore, no flux from diffraction orders other than the +1st can contribute to the peak signal, as these are taken into account when subtracting out the background in the proximity of the peak.

Using this method, the data plotted in Fig. 6 were evaluated. The curves were obtained using five different slit widths, and the values for the widths given in the plot were obtained from the FWHM values of the peaks. The principal shape of the curves matches the one expected, except that the region within 250 μm from the central peak is not as flat as indicated in Fig. 5. This is because the zero-order efficiency of the central regions of the BFL is higher than that for the outer regions with smaller structures. This, in turn, indicates a drop in first-order efficiency for decreasing zone width, which can also be observed in the sloped signal between $-2r$ and $-r$ stemming from the -1st diffraction order.

Table II gives an overview of the results. It can be seen that the measured values of F_0 are proportional to the slit width w as expected, and that the resulting first-order efficiencies do not depend on the slit width. We obtained a first-order diffraction efficiency of $26\% \pm 0.5\%$. The error is estimated from the inaccuracy of F_0 which may contain small contributions of the higher diffraction orders that are difficult to quantify. However, such additional contributions would increase the measured values for F_0 above the true reference signal. According to Eq. (3b), this would result in efficiency values below the true performance of our BFLs. Additionally, the -1st -order efficiency was evaluated from the curves by the integration of the broad -1st -order signal (corresponding to the dotted line in Fig. 5), which had to be interpolated in the interval between $-r$ and r . The obtained

values are significantly higher than 26%. The difference can be explained by contributions of higher diffraction orders passing through the measurement slit. This interpretation is confirmed by the trend of a linear increase of the -1st -order efficiency values with the slit width.

The described method to measure the diffraction efficiency appears to be very robust, since it does not require detailed knowledge of parameters like slit dimensions or an exact match of a monochromator to a Bragg angle. The resulting efficiency values are close to the ones reported previously, although the widths of the diffracting structures are significantly smaller. For a given focal length and wavelength, the aperture of our optics is a factor of 3 higher than that of a BFL with a 300 nm outermost zone width. This means that a corresponding gain in obtainable flux can be expected. Furthermore, our capability of producing BFLs with larger total area may result in additional gain, depending on the geometry of a specific setup.

The diffraction limit of the BFL's resolution should also improve proportionally to the smallest zone width. To demonstrate this, we need to perform resolution tests with BFLs having short focal length and with a source of reduced source size (i.e., closed primary slits), which are no longer limited by the geometrical source image. Furthermore, experiments to increase the accepted bandwidth by using epitaxially grown SiGe alloys with a gradient in Ge concentration are planned in the near future.

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TABLE II. Data used to determine the BFL diffraction efficiency according to Eqs. (3b) and (3c).

Slit width w (μm)	F_0 (a.u.)	F_0/w (a.u. μm^{-1})	$\int_{\text{peak}} S dz$ (a.u. μm)	+1st order efficiency (%)	-1st order efficiency (%)
15.6	1605	102.9	122.1	26.2	29.3
24.7	2567	103.9	208.5	25.9	31.2
33.9	3481	102.7	287.4	26.1	31.8
59.1	6106	103.3	508.8	25.8	32.1
78.9	8077	102.4	693.1	26.0	33.0

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