Wavelength tunable diffractive transmission lens for hard x rays

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We report on the fabrication and testing of linear transmission Fresnel zone plates for hard x rays. The diffractive elements are generated by electron beam lithography and chemical wet etching of ⟨110⟩ oriented silicon substrates. By tilting the cylindrical lenses with respect to the x-ray beam, the effective path through the phase shifting zones can be varied. This makes it possible to optimize the diffraction efficiency over a wide range of photon energies, and to obtain effective aspect ratios not accessible with untilted optics. The diffraction efficiency of such a lens was measured as a function of the tilt angles for various energies between 8 and 29 keV. Values close to the theoretical limit were obtained for all energies. Because of the coherence preserving properties of diffractive optics, the method opens up opportunities for experiments using coherent hard x rays. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1379364]

The focusing of hard x rays ($h\nu > 8$ keV) to submicrometer dimensions is an important prerequisite for many of techniques such as microanalysis, microimaging, microspectroscopy, and microdiffraction. Reflective, grazing incidence optics are most commonly used for this purpose, but submicron spot sizes can only be obtained currently with great difficulty due to aberrations stemming from imperfections of the mirror surfaces.[1] Furthermore, high quality ellipsoidal focusing mirror systems are difficult to align and expensive. Due to their complexity, they are usually an integral component of a synchrotron beamline, which limits their flexibility.

The first devices to achieve submicron hard x-ray focusing in a routine fashion were Bragg–Fresnel lenses (BFLs).[2] These devices usually consist of a Bragg crystal patterned with a diffractive Fresnel lens. Although BFLs with zone widths down to 100 nm and good diffraction efficiency have been made,[3] there are some severe difficulties related with their practical application. First, 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 lense's Bragg condition makes it only suitable for a narrow bandwidth and difficult to align when used in combination with a crystal monochromator. Therefore, BFLs are only used in very few experimental setups. Focusing transmission lenses avoiding these difficulties would be most useful in a wide range of applications.

Some years ago it was recognized that compound refractive lenses (CRLs) might provide an alternative way to focus hard x rays without an additional angle in the optical axis. CRLs were first proposed and patented by Tomie[4] and shortly later experimentally verified by Snigirev *et al.*[5] Two alternative arrangements are presently in use. The first type consists of a linear array of holes along the optical axis drilled into a block of low absorbing material, preferentially Be. The cylindrical shape of the refracting surfaces leads to inevitable spherical aberrations, which limit the obtainable spot size to some microns. The second consists of low absorbing disks stacked along the optical axis with the refractive shape embossed into the surfaces of each disk. This method gives the opportunity to avoid spherical aberrations by using parabolic surfaces, and submicron focusing has been demonstrated using Al as lens material. However, for acceptable focal lengths the high absorption of Al limits the transmission of these devices to about one percent.[6] The fabrication of aspherical refractive lenses from Be with adequate quality has not been possible up to now due to its hardness and brittleness.

Diffractive transmission lenses (Fresnel zone plates) have been used successfully for x-ray focusing and imaging. Spot sizes in the range of tens of nanometers for soft x rays[7–11] and of about 100 nm for the multikilo-electron-volt region[12] and for hard x rays[13] have been achieved. The diffractive patterns are mostly generated by electron beam lithography. State-of-the-art lithography tools are capable of positioning the zone structures with accuracies in the order of nanometers. As the required accuracy to avoid aberrations is in the order of one outermost zone width, the resolving power of a zone plate is diffraction limited. Thus, the obtainable spot size is approximately equal to the width of the outermost zones when the lens is irradiated with spatially coherent light. This means, that in contrast to refractive or reflective hard x-ray optics, the coherence of the incoming light is preserved. This feature becomes increasingly important in experiments taking advantage of the high coherence provided by modern synchrotron sources.[14]

The efficiency of a zone plate, i.e., the fraction of the incoming radiation diffracted into the focal spot, depends on the phase shift and attenuation caused by the diffractive structures. 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 the scalar diffraction theory developed by Kirz[15] and the complex refractive index $n = 1 - \delta - i\beta$ tabulated for most elements by Henke *et al.*[16] For diffractive structures with a

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TABLE I. Maximum first order efficiencies E_{max} and optimum structures heights t_{opt} of a binary diffractive optical element for a selection of elements commonly used in zone plate fabrication at different photon energies.

	10 keV		15 keV		20 keV	
Element	E_{max} (%)	t_{opt} (μm)	E_{max} (%)	t_{opt} (μm)	E_{max} (%)	t_{opt} (μm)
C	40.4	13.6	40.5	27.6	40.5	34.0
Si	38.7	12.6	39.7	19.3	40.1	26.0
Ni	30.1	3.4	34.9	5.1	37.0	6.9
Ge	38.0	6.8	32.7	9.3	35.5	12.5
Ta	26.4	2.4	29.5	3.2	33.0	4.3
Au	32.7	2.0	27.3	2.9	31.4	3.7

square wave profile, the first order diffraction efficiency E_1 at a given x-ray wavelength λ is given by

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

where $k = \beta/\delta$, and $\phi = 2\pi\delta t/\lambda$ denotes the phase shift caused by the diffracting structures. 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 $4/\pi^2 = 40.5\%$ can be obtained for $\phi = \pi$.

Especially for energies just above the K edges of medium weight elements like Ni or Ge and the L edges of heavy materials like Ta, or Au, a significant fraction of radiation is lost by absorption as can be seen from Table I. The obtainable efficiency of light elements like C or Si in this energy range is close to the theoretical limit of an ideal phase zone plate due to the absence of absorption edges. Unfortunately, the required zone plate thickness to obtain a phase shift of π is more than 10 μm, which results in enormous aspect ratios for submicron structure widths. The fabrication of nanostructures using standard technologies like reactive ion etching is limited to aspect ratios in the order of 10 due to a limited control of the sidewall angles.

We used a simple technique to fabricate diffractive lenses with significantly higher aspect ratios. The lenses were made from ⟨110⟩ oriented Si substrates. First, membranes of 15–20 μm thickness were formed by photolithography and reactive ion etching from the backside of the substrate. The linear zone plate patterns were then defined by electron beam lithography and transferred into 30-nm-thick chromium structures by a lift-off process. On such substrates, lines oriented in the ⟨112⟩ direction have sidewalls with ⟨111⟩ orientation. Using an ethylendiamine pyrocathecol solution as orientation-selective wet etch resulted in structures with vertical sidewalls.[17] Figure 1 shows a linear zone plate with 324 nm outermost zone width (half-pitch) and 13 μm structure height. A more detailed description of the lens fabrication process is given elsewhere.[18]

This method is obviously limited to producing linear diffractive structures and thus to one-dimensional focusing [see Fig. 2(a)]. To achieve two-dimensional focusing, we suggest aligning two linear lenses with different focal lengths and orthogonal orientation along the optical axis as indicated in Fig. 2(b). In addition, diffractive optical elements with linear structures leave an additional degree of freedom. By a tilt of φ around an axis perpendicular to the diffractive structures and the optical axis, the effective path through the structures can be increased by a factor of $1/\cos\varphi$ [Figs. 2(c)–2(d)].

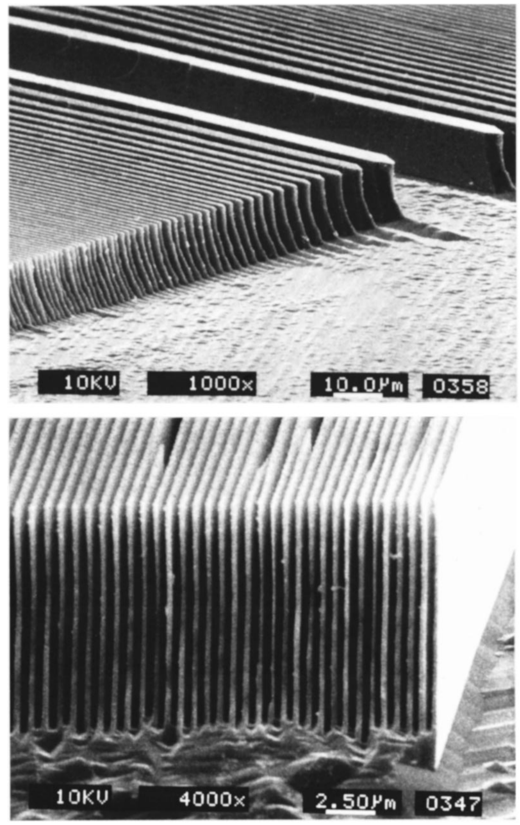

FIG. 1. Scanning electron microscopy images of linear Fresnel zone plates fabricated by electron beam lithography and wet chemical etching of ⟨110⟩ oriented silicon substrates. The outermost zone width is 324 nm, the zone height is 13 μm, corresponding to an aspect ratio of 40.

The diffraction efficiency of a wet etched Fresnel zone plate fabricated by this technique with 5.5 μm high Si struc-

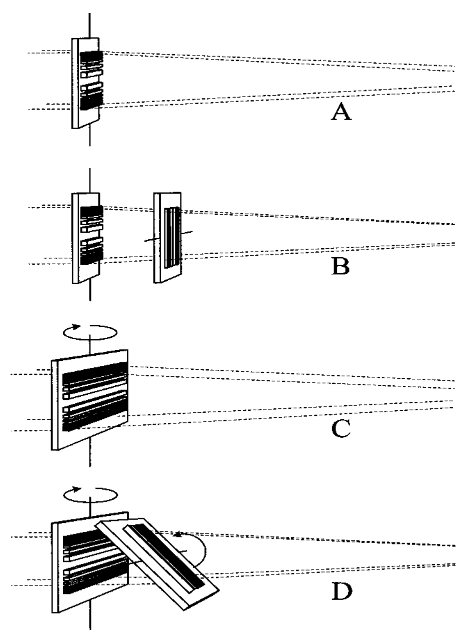

FIG. 2. A linear zone plate can be used to produce a line focus (a), while a combination of two such devices will result in two-dimensional focusing (b). By tilting of the lenses, the effective path through the phase shifting structures can be varied (c, d) in order to match the phase shift over a wide energy range and to increase the obtainable aspect ratio.

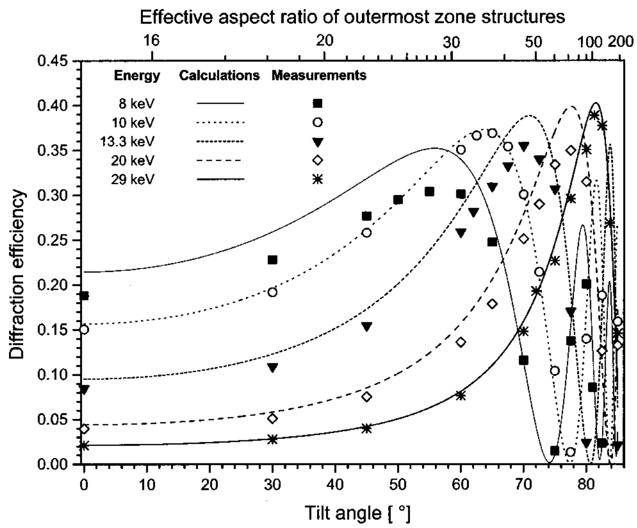

FIG. 3. First order efficiency of a linear Fresnel zone plate with 5.5 μm high Si structures for photon energies between 8 and 29 keV as a function of the zone plate tilt angle. The lens aperture was 200 μm. The upper horizontal axis indicates the effective aspect ratio of the 350 nm wide outermost zones.

tures, an aperture of 200 μm, a length of 2.5 mm, an outermost zone width of 350 nm, and 5 μm support membrane thickness was measured at the BM5 bending magnet beamline of the European Synchrotron Radiation Facility (ESRF). The diffraction efficiency for the untilted lens is expected to be optimum at 4.9 keV photon energy. Due to air absorption, we were not able to measure the diffraction efficiency for energies below 8 keV. In addition we were restricted by the geometry of the Si⟨111⟩ double crystal monochromator to values below 29 keV. The lens was mounted with the diffracting structures oriented horizontally, thus, focusing in the vertical direction. A slit with an opening of approximately 20 μm height and 200 μm width was scanned through the focal plane of the lens while recording the transmitted signal using a photodiode. The first order diffraction efficiency can be obtained by evaluating the integral over the focal peak. This method was previously developed to characterize Bragg–Fresnel lenses.[3] It has proven to be very accurate, as it does not require exact knowledge of the slit dimensions.

The calculated efficiency data and the measured values for 8, 10, 13.3, 20, and 29 keV photon energy are plotted in Fig. 3. The curves reach their maxima at 55.8°, 63.6°, 70.9°, 77.5°, and 81.5° tilt angle, respectively, which corresponds to a phase shift of π. Oscillations at higher tilt angles with minima for values equivalent to even integers and maxima for odd integers of π phase shift can be observed. We were able to measure the diffraction efficiencies up to tilt angles of 85°, corresponding to an aspect ratio of 150. The measured data for 10 and 29 keV photon energy very closely follow the theoretical curves, whereas for the other energies, the measured data are somewhat lower than expected. This is probably due to a slight angular misalignment around the axis perpendicular to the optical axis and the tilt axis, which would lead to a tilt of the structures sidewalls with respect to the beam.

The wet etched Si lens used in a tilted arrangement can thus be used over a wide energy range without any complicated realignment. Although the aspect ratio of the untilted device is already fairly high (15.7), the wet etching technique is capable of exceeding this value at least by a factor of 2 as shown in Fig. 1. It is, therefore, possible to fabricate lenses with higher structures for even harder photon energies or to reduce the outermost zone width and, thus, the achievable spot size down to the 100 nm range. The technique to increase the aspect ratio by tilting could also be applied to zone plates with blazed zone profiles.[19,20] This would be of special interest for two-dimensional focusing using two linear lenses. A combination of two binary lenses limits the total efficiency to a maximum value of $(40.5\%)^2 = 16.4\%$, while a combination of ideal blazed zone plates could give up to 100% efficiency. Furthermore, it should be noted, that the described techniques are also suited to produce other diffractive optical elements than merely lenses. Such devices could include wavefront-shaping elements or linear gratings to be used as beam splitters for interferometric[21,22] or holographic applications.

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