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High-resolution differential interference contrast X-ray zone plates: Design and fabrication ☆

Magnus Lindblom [a,*], Tomi Tuohimaa [a], Anders Holmberg [a], Thomas Wilhein [b], Hans M. Hertz [a], Ulrich Vogt [a]

[a] *Biomedical and X-ray Physics, KTH/Albanova, SE-106 91 Stockholm, Sweden*
[b] *University of Applied Sciences Koblenz, Rhein Ahr Campus Remagen, Suedallee 2, D-53424 Remagen, Germany*

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Abstract

Differential interference contrast is a potentially powerful technique for contrast enhancement in soft X-ray microscopy. We describe the design and fabrication of single-element diffractive optical elements suitable as objectives for high-resolution differential interference contrast microscopy in the water-window spectral range. A one-dimensional pattern calculation followed by an extension to two dimensions results in a pattern resolution of 1 nm, which is well below fabrication accuracy. The same fabrication process as for normal zone plates is applicable, but special care must be taken when converting the calculated pattern to a code for e-beam lithography.

Keywords: Diffractive optics; Differential interference contrast; X-ray microscopy

1. Introduction

In the X-ray region nearly all materials exhibit stronger phase-shifting properties than absorption. Therefore phase-sensitive imaging can enhance contrast compared to absorption-based imaging in X-ray microscopy. We recently demonstrated the first compact water-window interferometer based on a single diffractive optical element (DOE) [1]. In the present paper we describe how single-element DOEs can be designed and fabricated with high resolution for use as objectives for differential interference contrast (DIC) soft X-ray microscopy, both in synchrotron-based instruments and in compact microscopes [2,3].

Different phase-sensitive imaging techniques have been applied in X-ray microscopy, e.g. Zernike phase contrast [4,5] and the use of segmented detectors for scanning X-ray microscopy [6]. Zernike phase contrast suffers from imaging artifacts and severe alignment requirements and the use of segmented detectors is not applicable for full-field imaging. Another technique, which is compatible with both scanning and full-field microscopy, is differential interference contrast. Since the gradient of the phase is imaged, DIC is most beneficial when imaging small-sized and weakly absorbing samples. Furthermore, there is no need for additional components aside from the objective. Thus, absorption losses are minimized, which is especially important for operation with compact sources that have limited X-ray flux. DIC microscopy was demonstrated at 4 keV by Wilhein et. al. [7] using a twin-zone plate (TZP) as the microscope objective. Although relatively easy to use, a TZP is hard to fabricate. Di Fabrizio et al. [8] simplified matters by designing DIC objectives consisting of a single-diffractive optical element, whose functionality was also verified at 4 keV. In this work we extend this concept to allow high-resolution DOEs to be fabricated. A simplified pattern calculation enables the generation of patterns not only with higher resolution, but which can also be accurately exposed with e-beam lithography.

In the following sections, we describe the design and fabrication for single-element DOEs, suitable for DIC, with special emphasis on pattern design and the adaptation of the pattern to e-beam lithography.

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* Corresponding author.
E-mail address: magnus.lindblom@biox.kth.se (M. Lindblom).

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2. Design and fabrication

2.1. Fabricated optics

The fabricated DOEs are similar to ordinary nickel zone plates [9] but were designed to focus an incident plane wave into two focal spots instead of just one. An example is shown in Fig. 1. Two types of DOEs have been fabricated that give foci separated either along the optical axis (axial shear) or perpendicular to it (lateral shear). Both types were fabricated with an outermost zone width of 50 nm. A zone plate for DIC microscopy should generate two overlapping images of the object separated by a distance less than the resolution limit. In the pattern calculation, described in Section 2.3, the focal spot separation was therefore chosen to be 75% of the outermost zone width for the lateral shear zone plates. For the axial shear zone plates it was set to 75% of the depth of focus. DOEs with outermost zone width of 100 nm and a lateral spot separation of 600 nm have also been fabricated for use in interferometric experiments with soft X-rays [1]. The typical nickel height was about 200 nm in the center and 160 nm in the outermost zones. The diameter was 75 μm for all DOEs. These optics have been tested using a compact liquid-nitrogen laser-plasma source [1] and at the TWINMIC microscope at the ELETTRA synchrotron [10,11]. The results indicate that DOEs generate two focal spots as described in this paragraph.

2.2. Fabrication process

The fabrication process is briefly described here. For a more detailed discussion, see [9]. The structures, which consist of electroplated nickel, were fabricated on 50 nm thin Si_3N_4-membranes using a tri-level resist scheme. The patterning was performed with e-beam lithography at 25 keV (Raith 150). The steps of the process are illustrated in Fig. 2. A Si_3N_4-membrane prepared for patterning is covered by a stack of materials. Closest to the membrane a plating base, consisting of a 5 nm layer of chromium and 10 nm of germanium, is vapor-deposited. On top of this a 150–200 nm thick layer of plating mold (ARC XL-20, Brewer Science) and a 30 nm layer of e-beam resist (ZEP-7000, Nippon Zeon) are spin-coated. These two layers are separated by 5 nm of vapor-deposited titanium, which serves as etch-mask later in the process. After the e-beam exposure, which is discussed in more detail in Section 2.4, the pattern is transferred into the titanium etch-mask by BCl_3 reactive ion etching (RIE). The final pattern transfer into the plating-mold material is done by O_2 RIE. Nickel is then electroplated into the mold in a nickel–sulfamate solution (Lectro-Nic 10-03, Enthone Inc.). To remove the titanium etch-mask and the plating mold the two RIE steps are repeated. In the end, only the nickel zones of the DOE and the plating base remain on the Si_3N_4-membrane.

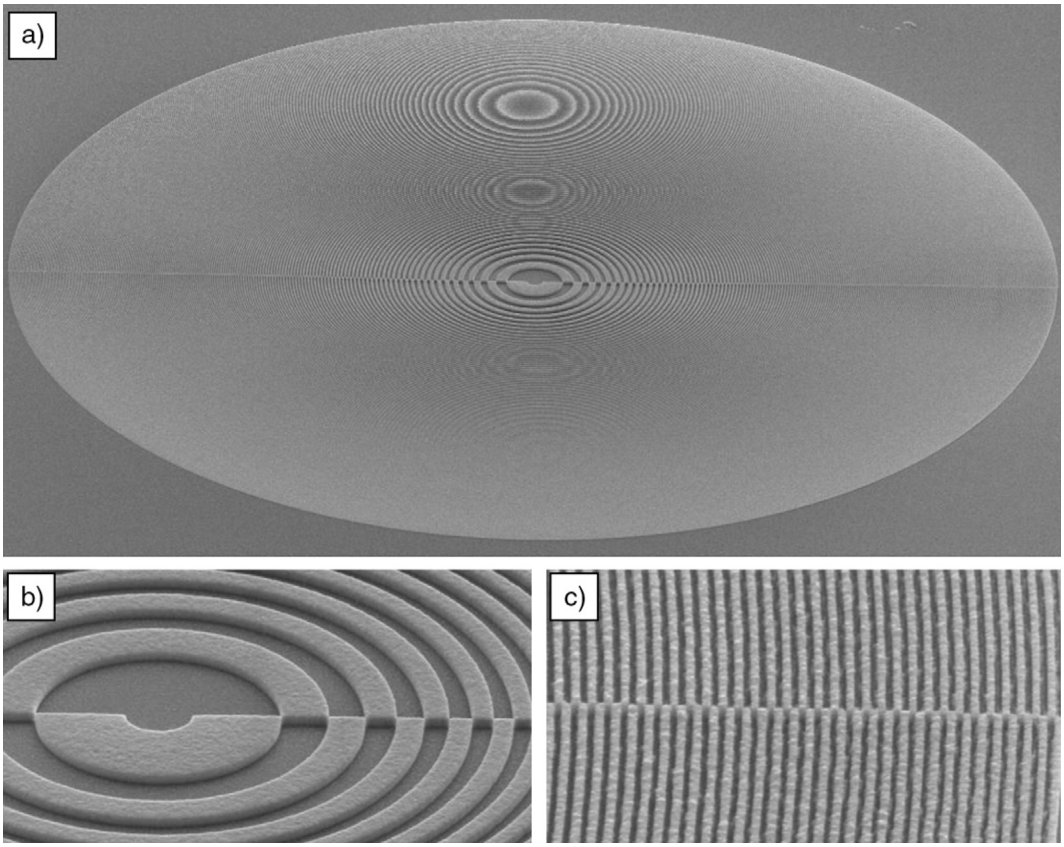

Fig. 1. (a) An electron microscope image of a nickel DOE, with a diameter of 75 μm, that generates two laterally separated focal spots. The circular patterns away from the center of the DOE are Moiré patterns and not actual features. (b) Detail from the central part. (c) Detail from the edge with 50 nm lines and spaces. In all three images the DOE was tilted by 60°.

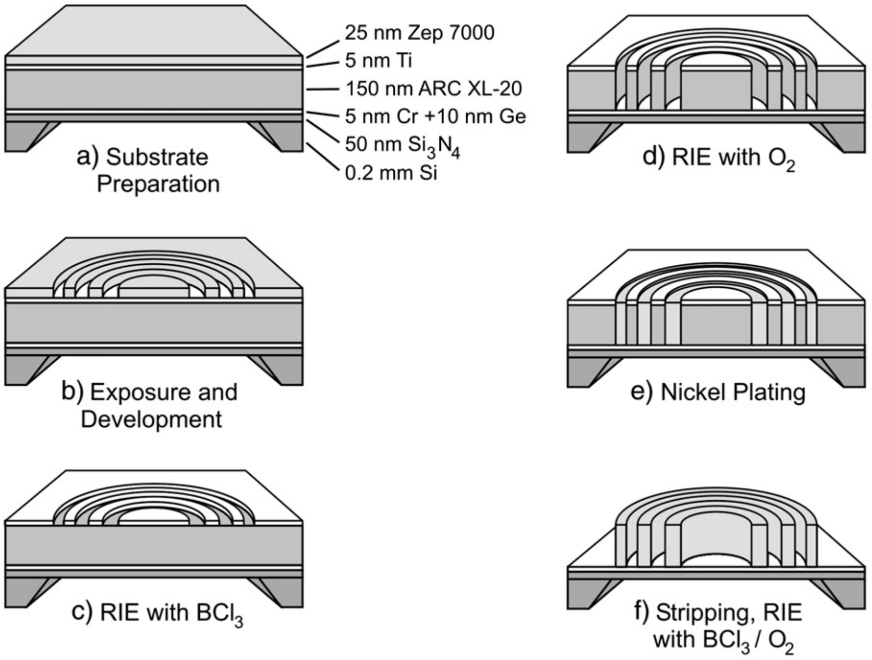

Fig. 2. The steps of the zone plate fabrication process.

2.3. Pattern calculation

A DOE can be thought of as a physical realization of a transmission function that transforms the incoming radiation from a source into a desired outgoing wave. If the DOE is assumed to be thin then the outgoing wave, U_{out}, is related to the incoming wave, U_{in}, through the relation $U_{\mathrm{out}}(x,y)=t(x,y)\cdot U_{\mathrm{in}}(x,y)$, where x and y are coordinates in the plane of the DOE and $t(x,y)$ is the transmission function in this plane. If furthermore, the absorption is neglected the transmission function can be written as $t(x,y)=\exp(i\Phi(x,y))$, where $\Phi(x,y)$ is the phase function which in turn is the difference in phase between the outgoing and the incoming wave: $\Phi(x,y)=\arg(U_{\mathrm{ou}}t(x,y))-\arg(U_{\mathrm{in}}(x,y))$ (modulo 2π). With these approximations it is the phase function that needs to be physically constructed in order to realize the desired transmission function. For that purpose the phase function needs to be binarized so that an exposable pattern for e-beam lithography is obtained.

This approach, which has been applied earlier for design of hard-X-ray DOEs by Fabrizio et al. [8], offers high flexibility since the incoming and outgoing waves can be chosen freely. However, it is difficult to apply this method to generate high-resolution patterns since it requires operations on very large data sets. We carry out the calculation in only one dimension. A one-dimensional pattern is then achieved which is extended to two dimensions by symmetry considerations and simple approximations. In this way a high pattern resolution can be achieved and the pattern will automatically lend itself to e-beam lithography. The flexibility is of course compromised but as long as the desired outgoing wave is not very irregular there is no need for two-dimensional calculations.

The DIC-zone plates described in Section 2.1 were designed to focus an incident plane wave into two foci. The first step in the pattern calculation is to determine the outgoing wave at the plane of the DOE. This was done by replacing the foci with two spherically emitting monochromatic point sources. At this stage the focal spot separation and the bias (the phase relation between the two foci) were defined. As mentioned in Section 2.1, two configurations of the focal spots were realized, lateral and axial shear, which correspond to focal spot separation perpendicular to and parallel to the optical axis respectively. The waves from the point sources were then propagated to the plane of the DOE and superpositioned to give the outgoing wave in one dimension. With the illumination assumed to be a plane wave the one-dimensional phase function, Φ_{1D}, was then calculated by the method described above and binarized using the condition: $\Phi_{\mathrm{1D}}=0$ if $0<\Phi_{\mathrm{1D}}<\pi$ and $\Phi_{\mathrm{1D}}=1$ otherwise. Then, by letting $\Phi_{\mathrm{1D}}=1$ represent a zone with material and $\Phi_{\mathrm{1D}}=0$ an empty zone, the phase function was translated to a one-dimensional binary DOE pattern.

The transfer of the one-dimensional pattern into a two dimensions was realized by symmetry considerations. For axial shear patterns the symmetry is fully rotational, but for lateral shear the conversion is not as straightforward and a couple of approximations are needed. The symmetry is broken since the outgoing wave at the DOE is a superposition of the spherical waves originating from two laterally separated point sources. The first of the approximations was that the zone boundaries were assumed to be circular. The second approximation concerned the geometry of the pattern inversions, see Fig. 1. The actual location of the inversion is where the spherical waves from the point sources are π out of phase but in our approximation they were assumed to occur along straight lines across the DOE. With these two simplifications the conversion to two dimensions is straightforward and the error will be negligible as long as the spot separation is small in comparison to the focal length. In our case the error was below fabrication accuracy even for the DOEs with a focal spot separation of 600 nm and a focal length of 3 mm. Note that even if the spot separation would be sufficiently large to induce an important error there would still be no need to do a two-dimensional calculation of the phase function. Instead, one would simply calculate the shape of the zone boundaries and determine the location for the pattern inversion for each zone. An interesting observation is that the pattern inversions, which occur with a periodicity determined by the spot separation, correspond to a phase shift of π. In view of that a lateral shear DOE can be considered to be a zone plate with a superimposed phase grating. This observation introduces another method to generate patterns for high-resolution lateral shear DOEs by combining a grating and a zone plate pattern [12,13].

As an evaluation, numerical simulations were performed by propagating a plane monochromatic wave through the two-dimensional binary patterns. Fig. 3 displays results from such simulations as well as crude examples of calculated DOE patterns. These simulations were done only with low-resolution patterns containing few zones but the results show that the DOE patterns generate the two expected focal spots. The patterns later used for fabrication had a diameter of 75 μm and were calculated with a 1 nm resolution. Due to their size the simulations had to be restricted to one dimension for these patterns.

2.4. E-beam lithography

Equally important as the pattern calculation is the transfer to an exposable file for electron beam lithography. The Raith 150 e-beam lithography system works with the GDSII stream format and it is the composition of the GDSII file that determines in what way the exposure is performed. Due to the high degree of symmetry in the calculated patterns it is relatively easy to convert them into an exposable file. Nevertheless, the method of conversion is still important for the exposure results.

We have applied two different exposure schemes which both work but which still give very different results. The first approach made use of a feature of the Raith system that lets the user approximate arbitrary lines with polygonal paths by specifying their nodes and widths. Each zone segment (the zones are not full circles) was defined as a polygonal line of that type. The order of exposure in this case is such that the beam stops at each node and scans the full width of the polygon line before moving onto the next node. Although simple, this method is associated with a long exposure time and a DOE with a diameter of 75 μm required 50–60 min of writing time. The thermal drift during such a long exposure causes pattern distortion, which results in coma aberration in the optical element. In addition, the freedom of patterning is limited and in this case the sharp edges at the end of each zone segment could not be generated. Another drawback was the occurrence of systematic pattern defects. The reasons are presently unclear to us and may be an issue for the individual system. The better approach is to manually define each zone segment as a collection of polygonal lines with zero width. The system then writes each zone segment by tracing out the lines in their full lengths one by one. In this way, the exposure time was reduced to about 5 min for the DOEs discussed here, which is a ten fold improvement. Furthermore, all details of the pattern can be faithfully reproduced and the exposures were free from defects.

3. Conclusion

We have presented a pattern generation method that allows fabrication of high-resolution single-element DOEs. Experiments performed at KTH Stockholm, using a liquid-nitrogen laser-plasma source, and at the TWINMIC microscope at the Elettra synchrotron, verify their functionality [1,11]. The results imply that they are suitable for soft X-ray DIC microscopy. The next step

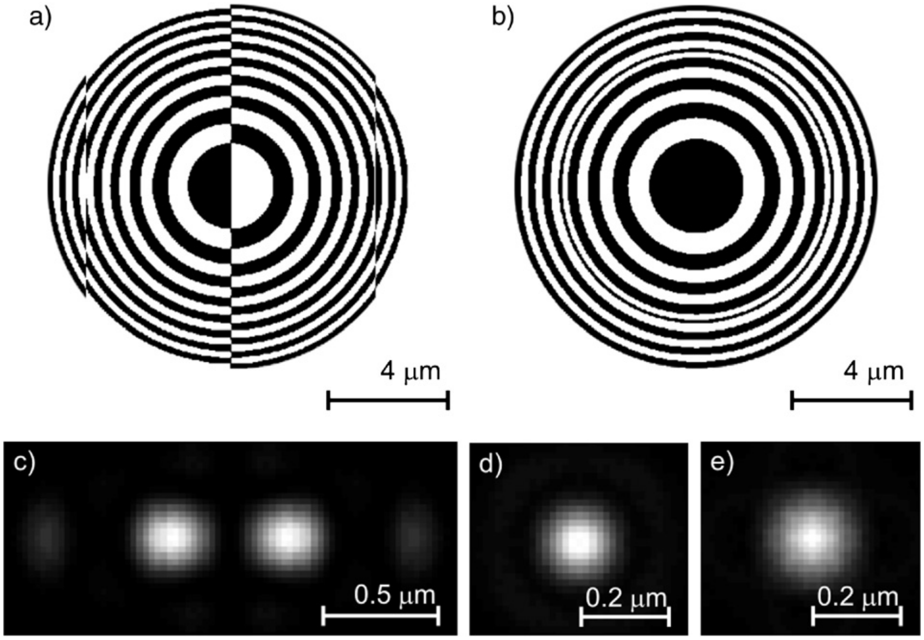

Fig. 3. Examples of (a) lateral- and (b) axial shear DOE patterns. The focal spot separation was calculated to be 0.5 μm (a) and 63 μm, about four times the depth of focus (b). The propagation distance was 1 mm in both cases. Parts (c), (d) and (e) show the intensity distributions calculated by wave-field propagation of a monochromatic wave with $\lambda = 2.48$ nm: (c) in the focal plane of the lateral shear DOE, (d) and (e) at the two focal positions from the axial shear DOE.

is to apply these optics in full-field water-window microscopy. This will be realized at the TWINMIC microscope and in the compact soft X-ray microscopes at KTH Stockholm. We also intend to fabricate new DOEs with higher resolution. An improvement to 25 nm outermost zone width is realistic on short term.

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