Fabrication of Fresnel zone plates for hard X-rays

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Abstract

A method to fabricate gold structures with high aspect ratio is presented. Fresnel zone plates with an outermost zone width of 100 nm and structures of 1 μm height are fabricated. Preliminary focusing results at an X-ray energy of 8 keV are presented and ways to improve the zone plate parameters are discussed.

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1. Introduction

The focusing of hard X-ray beams is an important prerequisite for many experimental techniques such as microfluorescence, microimaging, microspectroscopy, and microdiffraction. Currently, at many synchrotron sources, a significant effort is put into the reliable, efficient focusing of hard X-rays (photon energies above 8 keV) to submicron dimensions [1,2]. One of the most convenient devices for focusing X-rays is the Fresnel zone plate (FZP). FZPs have proven to give focal spots with excellent spatial resolution combined with low background. The diffraction limit for the resolution of a FZP is on the order of the smallest, outermost zone width, meaning that nanolithography processes with sufficient resolution have to be applied. In the soft X-ray range FZPs can reach a resolution below 20 nm [3]. However, the efficient focusing of hard X-rays is more difficult, as additional problems occur: to achieve acceptable diffraction efficiencies, the zone plate structures should be made from heavy materials. But even then the required height of the structures needs to be a micron or more. This means that the structures have to be 10 or even 20 times higher than their width, when both high resolution and efficiency is needed. This is the reason why the high resolution potential of FZPs could not be exploited in the hard X-ray regime so far.

In this paper we describe the fabrication process of gold FZPs, which is suitable for the fabrication of structures with an aspect ratio of more than 10. The feasibility of the process is demonstrated by the production of FZPs with an outermost zonewidth of 100 nm and a profile height of 1 μm. Preliminary results on the focusing of 8 keV X-rays at the MicroXAS beamline of the Swiss Light Source (SLS) with these FZPs are presented and future experiments are discussed.

2. Fabrication process

We fabricated FZPs using a three-level resist process and electroplating into a mold. The idea of the method was described before [4,5]. However, we made some modifications to fit it better to our experimental conditions. Main fabrication steps are shown in Fig. 1. A silicon chip with ~4 μm thick silicon membranes was coated by the following layers: Cr/Ge plating base (20/20 nm), polyimide (1 μm), Cr (20 nm), PMMA (50 nm). We patterned PMMA resist using the Leica LION LV-1 electron beam lithography tool at an electron energy of 2.5 keV. This system has a unique mode – continuous path control – which allows writing curved lines without stitching errors. In this...
mode the stage with the sample is moved under the electron beam, but inaccuracies, caused by mechanical movements of the stage are detected by a laser interferometer system and corrected by corresponding deflection of the electron beam. During the exposure of FZPs every zone is written by a single electron beam path, but the linewidth of the zones is defined by changing the defocus and the line dose [6]. After development of PMMA the underlying Cr layer was etched by reactive ion etching in Cl₂/CO₂ plasma. The Cr layer played a double role. It served as a conductive layer to prevent charging during electron beam exposure and is also used as an etching mask during the following etching of polyimide. The etching of polyimide was done in a Oxford Plasmalab 100 RIE system using pure oxygen as an etching gas, at room temperature, a pressure of 3 mTorr and a power of 200 W, which typically gives a DC Bias around 700 V. Then, the etched polyimide trenches were filled with gold by electroplating in a cyanide-based plating bath.

We found that after the plating, it appeared to be practically impossible to remove the Cr layer from the top of the polyimide by dry etching in Cl₂/CO₂ plasma. We assume that this was due to the oxidation of the Cr etching mask during the polyimide etching process. However, the Cr mask can be removed by wet-etching without damaging the Cr-layer in the Cr/Ge plating base and then the polyimide layer can be removed using an oxygen plasma. As an example a FZP with removed polyimide mold is shown in Fig. 2. We would like to stress here that the presence of a thin layer of Cr and polyimide mold between Au lines is not crucial for the performance of Fresnel zone plates in the hard X-ray range, since the refractive index of polyimide is much lower than that of gold. In our focusing experiments the zone plate with unremoved polyimide mold was used.

### 3. Focusing experiments at MicroXAS beamline

The microfocusing experiments were performed at the microXAS beamline of the Swiss Light Source synchrotron [7]. The radiation is obtained from an undulator source and monochromatized by a Si (111) double crystal monochromator. The very bright X-ray beam in the 5–20 keV energy range is used for the analysis of samples from environmental and materials science.

A schematic drawing of key components of the setup is shown in Fig. 3. Experiments were performed in air at atmospheric pressure using an X-ray energy of 8 keV. The X-ray beam passes through the 20 μm aperture and is focused by the FZP. In order to determine the focal spot
size gratings of different periods are scanned in the focal plane of the FZP and a modulation of the transmitted intensity is registered by the detector. In order to reduce the background caused by the divergent radiation of the higher diffraction orders a 5 μm order selecting aperture (OSA) is put 3 mm upstream of the FZP. The coherence length of the X-ray beam at this energy is ideally about 100 μm in vertical direction and 20 μm in horizontal direction. Therefore, in order to cut the incoherent part of the radiation we used a 20 μm coherence defining aperture in front of the FZP. We used a 30 μm FZP with outermost zonewidth of Δr_N = 100 nm in the experiment; therefore, only zones down to Δr_N = 150 nm were illuminated through the aperture as shown in Fig. 3. (Some Au spherical particles at the border and around zone plate has accidentally electroplated on top Cr layer and do not affect the performance of the zoneplate.) According to the Rayleigh criterion the focal spot size produced by a lens is δ = 0.61 × λ/2Δr_N. For a Fresnel zone plate NA ≈ λ/2Δr_N [8]. Therefore, in case of fully coherent illumination a focal spot of δ = 1.22 × Δr_N = 182 nm in the first diffraction order could be achieved in our case.

Fig. 4 illustrates examples of intensity modulations during grating scans in the focal plane of the FZP. Test gratings were 0.5 μm high gold structures plated into polyimide by the same method as for the fabrication of the FZPs. As seen from Fig. 4 the transmission modulation for a 5 μm period grating is about 18%, which is in good agreement with the transmission of 0.5 μm thick gold layer at 8 keV.

A comparison of measured and calculated visibility values of different test gratings are shown in Fig. 5. We define the visibility of a grating as v = (I_{max} − I_{min})/(I_{max} + I_{min}), where I_{max} and I_{min} are maximum and minimum transmissions through the grating, correspondingly. The solid line corresponds to the calculated visibility for the case of diffraction limited focusing of an X-ray beam. The plot was normalized by the value of the visibility of 5 μm period grating, assuming that the size of focused beam is much smaller than the half-pitch of the grating. The same value was also used as a reference in calculations of the visibility of gratings of higher spatial frequencies, assuming that the intensity distribution in the focal spot is described by a Bessel function and performing convolutions with the gratings of different periods. For small period gratings, the visibility goes down, because the “tails” of the Bessel distribution of the intensity in the focal spots are wider than the half pitch of the grating. However, still the grating lines are clearly
resolved. The experimental points are lower than the theoretical curve. This may be caused by slight defocusing of the beam, because the focal distance between the Fresnel zone plate and the test grating was not optimized. Unfortunately, for the resolution test we had only gratings with periods down to 500 nm available at the time of the experiment. Nevertheless, as seen from Fig. 5, even without optimizing the experimental conditions, the 250 nm half-period lines can clearly be resolved.

The theoretical diffraction efficiency of the gold FZP with zone height of 1 \( \mu m \) at 8 keV is 23%. The measured diffraction efficiency was 8.4%. The discrepancy is mainly caused by deviations in the duty cycle, as well as by absorption in the membrane, plating base and polyimide mold.

4. Conclusions

We developed a process for the fabrication of gold Fresnel zone plates with structures of 1 \( \mu m \) height and outermost zone widths down to 100 nm. The measured diffraction efficiency of the FZP is 8.4%. A focal spot below 250 nm was obtained. The results presented here are preliminary measurements of the first Fresnel zone plates fabricated by the described method. A number of improvements can be done to increase the aspect ratio of the structures of the FZPs. For example, the aspect ratio of mold structures can be significantly increased by introducing “bridges” [9] between lines, which increases their mechanical stability. In addition, the mechanical stability of the etched polymer mold during the electroplating step can be improved by electron beam hardening prior to the electroplating [10].

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