DELIVERABLE REPORT

WP6-JRA1 Research on In-Operando and high throughput methods

D6.3

Soft X-ray Fresnel zone plates with a resolution of 10 nm or better for users

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Soft X-ray Fresnel zone plates with a resolution of 10 nm or better for users

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☒ P – Public
☐ PP - Restricted to other programme participants & EC: (Specify)
☐ RE - Restricted to a group (Specify)
☐ CO - Confidential, only for members of the consortium

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Contents

Executive Summary

Fabricating nanostructures with small lateral sizes and high aspect ratio is one of the key challenges, for instance, in developing highly resolving diffractive X-ray optics. While conventional lithography approaches break down below 20 nm feature size, growth processes can achieve extreme precision which reaches sometimes down to the atomic level. This approach has allowed for the production of diffractive X-ray lenses (Fresnel zone plates) with down to 12 nm line width in the past [1].

Targeting sub-10 nm resolution in X-ray microscopy, line widths have to be decreased to 8 nm or below. As direct fabrication of the required nanostructures is impossible with conventional lithography methods based on direct electron-beam writing, the line doubling method relaxes the demand of the lithography step to write every second line only. Utilizing atomic layer deposition (ALD) of iridium allows for doubling, or even multiplying the line density of the template structure.

In this deliverable, we report on the fabrication of Fresnel zone plates (FZPs) which feature 10 nm spatial resolution in soft X-ray microscopy.

We have successfully fabricated a set of zone plates with outermost zone widths of 8.8, 8.0, 7.2 and 6.4 nm at aspect ratios between 5 and 10 (see Deliverable 7.3 – Month 24). These FZPs have now been thoroughly tested and their microscopic resolution has been eveluated with dedicated test samples which exhibit 10, 9 and 8 nm metal lines and spaces.

The work described above will be presented on the X-ray Microscopy conference in Saskatoon, Canada, in August 2018. The fabrication process has been presented at the MNE2017 conference in Braga, Portugal, in September 2017, and published in the journal Microelectronic Engineering. The results from resolution tests will be submitted for publication in a high-impact journal. FZPs with 10 nm resolution are available for users at the Hermes beamline at Soleil and at the Pollux beamline at the Swiss Light Source.

1. Concept

X-ray microscopy and spectroscopy enable unique insights into condensed matter [2], magnetism [3], electronic materials [4], or biological specimen [5]. Self-evident in microscopy, high spatial resolution is essential for a multitude of methods based on X-rays. Despite of major achievements in recent years in shaping highly confined X-ray beams with spot sizes below 10 nm using mirrors [6], FZP lenses [7, 8] and multilayer Laue lenses [9, 10], this number remained a barrier for resolution values in X-ray microscopy.

Both scanning transmission X-ray microscopes (STXM), the PolLux STXM at the Swiss Light Source [11] and the Hermes STXM at Synchrotron [12] offer an energy range from 250 eV (λ = 5.0 nm) to 1.6 keV (λ = 0.8 nm). In an STXM, the incoming synchrotron radiation is focused with a diffractive X-ray lens, which is commonly a FZP [11-13]. The achievable minimum spot size of an X-ray beam focused with a FZP is close to the width of its outermost zone [1, 14]. The resolution of state-of-the-art STXMs is therefore constrained by the limitations in nanofabrication of these lenses. In order to achieve a diffraction-limited spot size of 10 nm in X-ray microscopy, we need to fabricate line widths of 8 nm or below. As outlined above, we achieved this with the line doubling approach as demonstrated in D 7.3. We now applied the FZPs fabricated with this method and tested their performance.

2. Fresnel Zone Plate Fabrication

By optimizing the nanofabrication process, we have successfully fabricated FZPs for soft X-rays with unprecedentedly small outermost zone widths well below 10 nm and high structural integrity. The preparation of these zone plates, which is described in detail in the report to deliverable D7.3, is illustrated in Fig. 1. In short, the method takes advantage of the high precision of ALD. In a first step, a sparse template pattern with only half line density of a FZP is exposed by EBL using the negative-tone resist hydrogen silsesquioxane (HSQ), developed and dried from supercritical CO_2. A typical template pattern with ~ 9 nm line width is show in Fig. 1e. Afterwards, the HSQ template is coated with iridium by ALD, see Fig. 1f. This step effectively doubles the lines of the template structure with a high refractive index material and thus yields a FZP with much smaller zone width than achievable with conventional EBL methods. In order to prevent absorption losses, the iridium caps and bottom are removed by milling the surface of the device with argon ions in normal incidence as shown in Fig 1g. As a final step, the template can be removed with hydrofluoric acid vapor, see Fig 1h. However, numerical simulations of our zone plates using rigorous coupled wave theory [15, 16] showed that the last step does not lead to a notable increase in efficiency and can be omitted. We thus used FZPs corresponding to Fig. 1c/g.

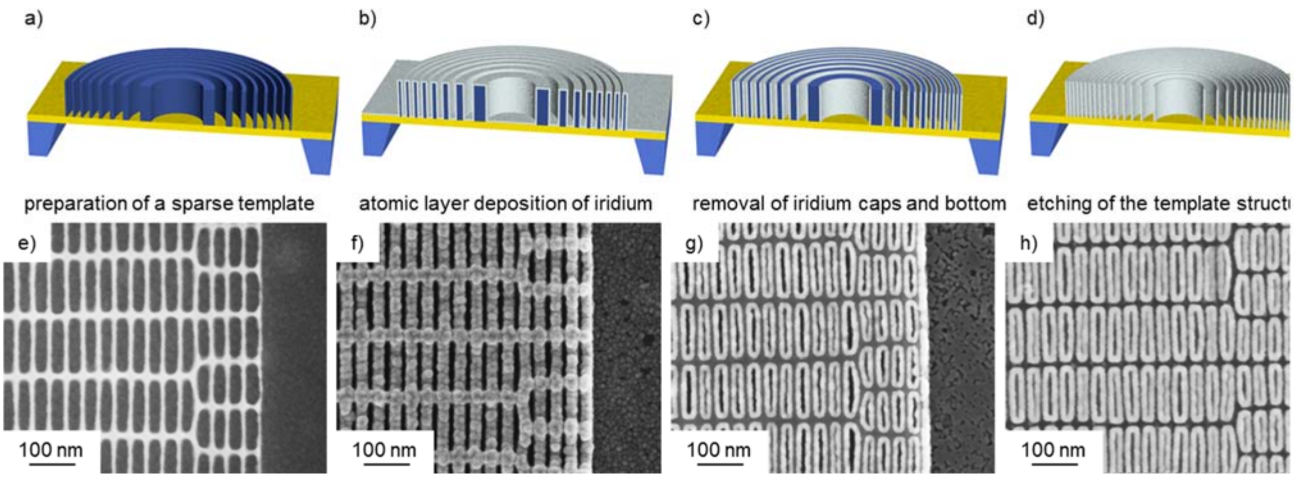

Figure 1: _Schematic illustration of zone plate fabrication using the line-doubling technique (a-d) and scanning electron micrographs of the resulting structures (e-h)_. a, e, A template pattern with half the line density of a Fresnel zone plate is lithographically prepared by exposure of a negative-tone resist to electrons, subsequent development and drying from supercritical CO2. b, f, The template pattern is then coated with 9 nm thick iridium in an atomic layer deposition step. c, g, The coated nanostructure is sputtered with argon ions to remove the iridium caps and the bottom layer to reduce absorption losses. d, h, The template structure is removed with hydrofluoric acid vapor.

2.1 Design Parameters of the Fresnel Zone Plates

The design parameters have been chosen to match exactly the conditions at the Pollux and Hermes beamlines, where the zone plates were tested. Especially the degree of coherence and the bandwidth at different energies had influence to the choice of design parameters.

2.1.1 Hermes

The following table gives an overview about the design parameters for sub-10 nm Fresnel zone plates for use at the Hermes beamline:

Table 1: the design parameters for sub-10 nm Fresnel zone plates for use at the Hermes beamline

outermost zone width [nm]	diameter [µm]	energy correction for [eV]	number of zones	focal length [mm] (at corr. E)	depth of focus [nm] (at corr. E)
8.8	240	750	6800	1.28	188
8.0	240	750	7500	1.16	155
7.2	240	750	8400	1.05	126
6.4	240	750	9400	0.93	99

2.1.2 Pollux

The following table gives an overview about the design parameters for sub-10 nm Fresnel zone plates for use at the Pollux beamline:

Table 2: the design parameters for sub-10 nm Fresnel zone plates for use at the Pollux beamline

outermost zone width [nm]	diameter [µm]	energy correction for [eV]	number of zones	focal length [mm] (at corr. E)	depth of focus [nm] (at corr. E)
8.8	100	850	2800	0.60	213
8.0	100	850	3100	0.55	176

2.2 Implementation in the Scanning Transmission X-ray Microscopes

To achieve the highest possible spatial resolution, the experimental geometry and mechanical stability of the STXM endstation has to be optimized, see Fig. 2. In particular, special attention has to be paid to the geometry of the order-sorting aperture and its holder due to the restricted focal length, and the mechanical stability of the sample stage. While the former challenge can be simply addressed by using a thin holder (150 µm) and pinhole (100 µm) to reduce the dimension along the beam propagation direction to approx. 250 µm, the latter one has to be tackled by carefully optimizing the interferometric control of the sample position. In this way, the maximum amplitude of sample displacement has been brought to ± 3 nm in both spatial directions at the Pollux microscope.

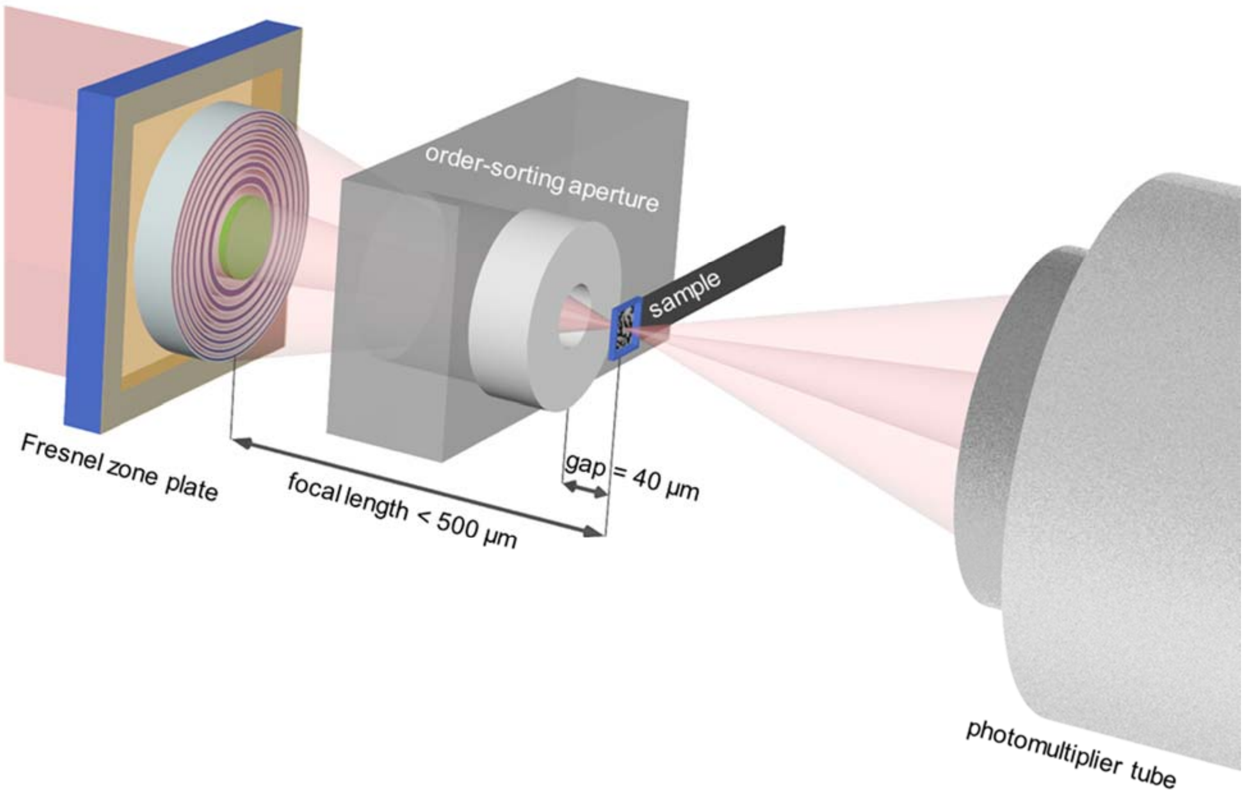

Figure 2: *Schematic illustration of the experimental implementation of a highly resolving Fresnel zone plate in the scanning transmission X-ray microscope at the PolLux beamline*. The incoming monochromatic X-ray beam (E_{ph} = 700 eV) is focused by means of a Fresnel zone plate with an outermost zone width of 8.8 nm with a focal length slightly below 500 µm. The gap between order-sorting aperture and sample is set to 40 µm. The transmitted intensity is recorded using a photomultiplier tube.

3. Performance of the 8.8 nm FZP

In order to test the spatial resolution of our high-resolution FZPs, we recorded test patterns with typical line widths of 10 nm and below. These patterns were prepared with a similar method as the FZPs, and thus consist of a periodic pattern of an iridium line, an HSQ line, a second iridium line, and a gap (compare Fig. 1g). The lateral dimension of each line or gap in different test samples is 10 nm, 9 nm, and 8 nm. We scanned the test objects point by point in steps of one nanometer. Closing the entrance and exit slits of the beamline ensures fully coherent illumination with an sufficiently small energy bandwidth. Fig. 3a shows the transmission on a 200 nm x 200 nm large area of the test pattern with 9 nm line width recorded with the PolLux microscope. The individual iridium lines are clearly visible, as well as the spaces in between. The microscopic resolution is so good that two collapsed lines (the darker, irregular structure visible on the left-hand side of the image) can be nicely distinguished from the regular pattern. To further enhance the accuracy of our setup, we saved the exact position of each recorded pixel from the interferometer feedback loop and interpolated the values at their programmed positions. The result is a slightly enhanced signal-to-noise ratio as shown in Fig. 3b. Determination of the spatial resolution was conducted using a one-dimensional Fourier shell correlation along the horizontal direction of the raw image shown in Fig. 3a [7, 17]. The result is shown in Fig. 3c, and yields a frequency cut-off at 0.13 nm^{-1} in Fourier space, or 7.9 nm in real space. The achieved spatial resolution sets a new benchmark in direct X-ray imaging without the need for image reconstruction, not only but particularly using X-rays with photon energies below 1 keV.

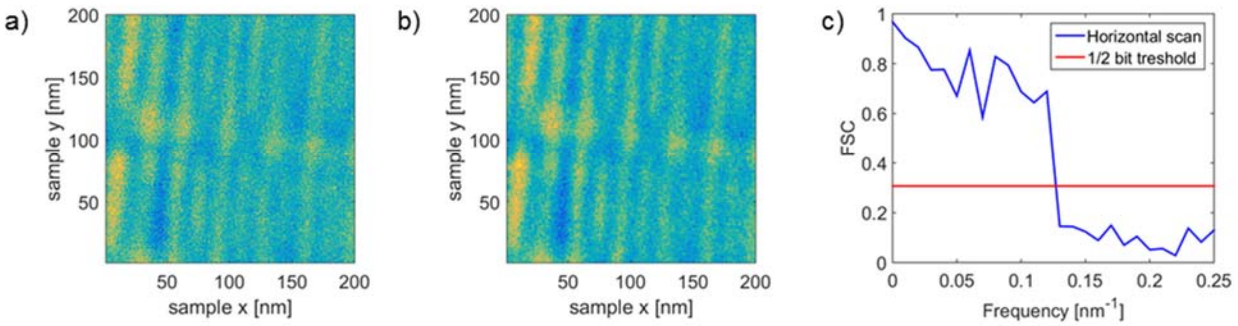

Figure 3: *Soft X-ray micrographs with sub-10 nm spatial resolution (a,b) and corresponding Fourier shell correlation*. a, Scanning transmission X-ray micrograph of an iridium test structure with 9 nm lines and spaces measured at the PolLux STXM (photon energy 700 eV, step size 1 nm). b, The microscopy image after position correction using the readout values from the interferometer at each scanned position. c, Fourier shell correlation along the x-axis of a). The analysis yields a cut-off at spatial frequencies of 0.13 nm^{-1}, corresponding to 7.9 nm.

The presented tests of an FZP with 8.8 nm outermost zone width on a test object with features of the same size encourages to take a step beyond to even smaller dimensions. As it turned out in various experiments, the requirements for FZPs with outermost zone widths of 8.0 or 7.2 nm are more and more demanding in terms of coherent illumination and energy bandwidth. Experiments using an 8 nm FZP at its diffraction-limited performance are ongoing and remain a technical challenge at current synchrotron sources. Using the FZP with 8.8 nm, we observe a breakdown of spatial resolutions at the test samples with 8 nm line width on the sample side. Fig. 4 shows a comparison of different test samples measured at the Hermes and PolLux STXMs. Whereas test samples with 10 and 9 nm line width can be nicely resolved with our high resolution zone plate (Fig. 4a,b), the ability to resolve individual lines breaks down at the determined cut-off value around 8 nm. The micrograph of such a structure is shown in Fig. 4c, where some areas appear not to be fully resolved whereas a part of the image still seems to contain information of individual lines (on the right hand side).

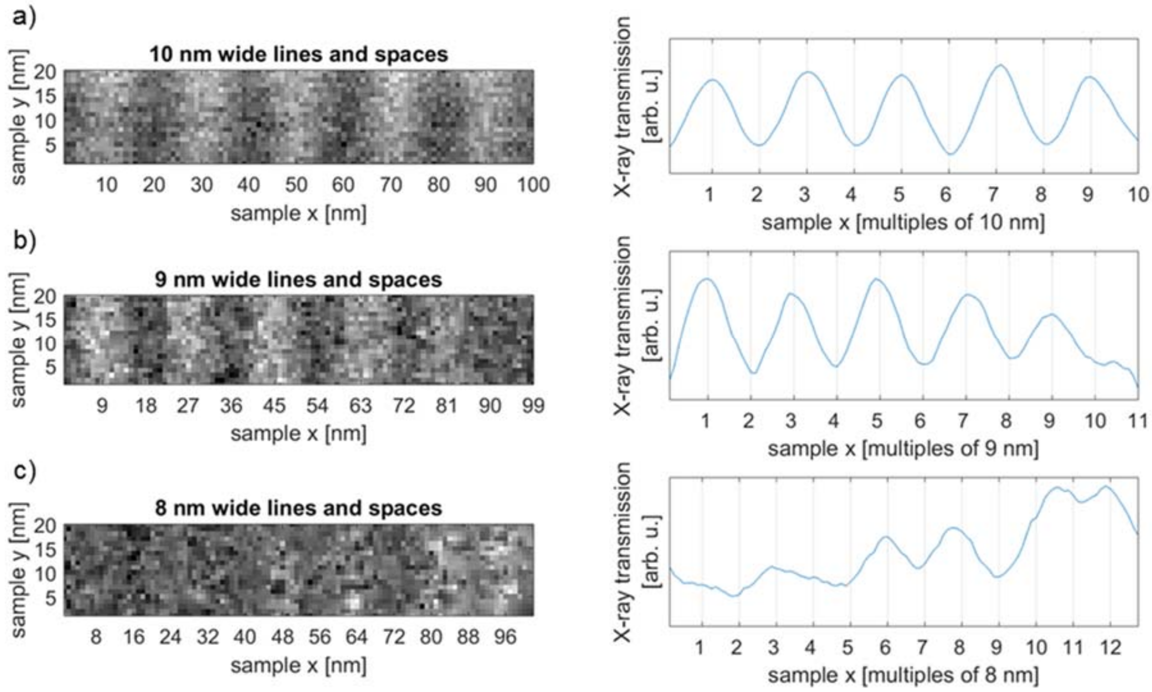

Figure 4: *Scanning transmission X-ray micrographs and corresponding profiles averaged in vertical direction of test objects with line width of 10, 9 and 8 nm*. a, 10 nm test structure imaged at the Hermes beamline using a photon energy of 850 eV. b, 9 nm test structure investigated at the PolLux beamline using a photon energy of 700 eV. c, 8 nm test structure imaged at the PolLux beamline using a photon energy of 700 eV.

4. Conclusions and perspectives

In order to increase microscopic resolution even further using existing FZPs with line width of 8.0 nm and 7.2 nm, we identify two main challenges: firstly, to control the distance between sample and zone plate with high accuracy, and secondly, to increase the quality of the incoming beam.

The first issue is currently addressed at the PolLux beamline by installation of an optical interferometer for the axis along the beam. Typically, requirements concerning positioning accuracy of sample with respect to the FZP lens are not crucial for common X-ray microscopy applications. However, the depth of focus and with it the required accuracy of the sample position along the optical axis for soft X-ray microscopy with sub-10 nm resolution is below 100 nm [18]. In order to avoid drifts out of focus, the z-position will be controlled with a feedback loop in the future.

The second challenge is, however, more demanding than to merely change the mechanical setup of the microscopes. During our experiments, we encountered that the coherent illumination of the FZPs is currently the limiting factor. In other words, the beamline slits had to be closed so much to ensure a fully coherent illumination that the signal-to-noise ratio is not sufficient for imaging anymore. Additionally, the required energy resolution of the incoming beam has to be on the order of several 1,000 due to the high number of zones. This implies further limits for the use of FZPs with practicably large diameters of 200 µm or above. In that sense, we expect new possibilities at the newly developed diffraction-limited X-ray sources [19] and free electron X-ray lasers. Their enhanced brilliance will provide a major advance on the capabilities to use highly resolving lenses for X-ray microscopy at a single-digit nanometer scale. This development is, however, clearly beyond the timescale for the current NFFA project and has to be addressed in the future.

The Hermes beamline at Soleil has received a set of 3 highly resolving FZPs with 8.8, 8.0 and 7.2 nm line width. These zone plates have comparably large diameters (240 µm) and offer therefore experimental conditions which are suitable for user operation. FZPs with 9 nm linewidth and capable of 8-9 nm spatial resolution are available at Hermes for future user experiments.

The Pollux beamline at the Swiss Light Source has received one FZP with 8.8 nm and 100 µm diameter. Another FZP with 8.0 nm is optimized for Pollux and available for implementation, but has not yet been tested in detail. We have to point out that these zone plates are extremely hard to use at the moment at Pollux, due to the very reduced flux of the beamline in completely coherent conditions.

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