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Polymer Compound Refractive Lenses for Hard X-ray Nanofocusing

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Abstract. Compound refractive lenses fabricated out of SU-8 negative photoresist have been used to generate a nanofocused, i.e. sub-µm sized X-ray focal spot at an X-ray nanodiffraction setup. X-ray microscopy and X-ray diffraction techniques have conceptually different demands on nanofocusing optical elements and so with the application of X-ray nanodiffraction in mind, this paper presents the results of an initial characterization of polymer lenses used as primary focusing device at an X-ray nanodiffraction synchrotron beamline. A beam size of ~ 600 nm was achieved at a photon energy of 13 keV following only a short initial alignment, with the focal spot representing a demagnified, direct image of the undulator source.

INTRODUCTION

Although already widely used in X-ray microscopy setups, polymer compound refractive lenses (CRLs) made out of SU-8 negative photoresist have not yet been applied to generate a nanofocused X-ray beam at an X-ray nanodiffraction setup. Nanofocusing, adiabatic, polymer compound refractive lenses combine the best of two worlds: the ease of alignment, common for most refractive X-ray lenses and a large effective aperture combined with a long working distance between exit aperture and focal plane even for hard X-rays, which is a feature usually attributed to reflective optics. This combination has long been desired in materials science related research and in particular in X-ray nanodiffraction. With this technique X-ray diffraction data is recorded using a nanofocused (i.e. sub-µm sized) hard X-ray beam. The recorded data then provide structural information with sub-µm spatial resolution from crystalline and semi-crystalline materials (e.g. metals, biomaterials, synthetic compounds). That way grain orientation, residual stress profiles, crystal structure or texture can be obtained with high spatial resolution in a non-destructive analysis. A key to the applicability of X-ray nanodiffraction for materials science related research – where extended in-situ sample environments are often required – is a sufficiently long focal distance. Provided a focusing element with a long focal distance and high focusing efficiency even at higher X-ray energies (>10 keV), high resolution nanodiffraction experiments can be performed even in extended sample environments (such as pressure cells or tensile rigs) and from strongly absorbing metallic samples.

LIGA FABRICATED POLYMER LENSES

For X-rays with photon energies in the range of many keV the refractive index of all materials is slightly below one. As a consequence, focusing refractive X-ray lenses have strongly curved biconcave parabolic surfaces [1]. The parabolic shape of the refracting surfaces avoids spherical aberration. In order to obtain a usable focal length f, despite the weak refracting power of an individual lens element, multiple lens elements are aligned along a common

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optical axis to form a compound refractive lens (CRL). The focal length f of a CRL is $f = R / (2N\delta)$ where δ is the refractive index decrement of the lens material and R the radius of the refracting surfaces in the apex of the parabola and N is the number of individual lenses. Suitable materials for refractive X-ray optics should have high refracting power in combination with low absorption. Thus, materials with a high ratio of δ/β are best suited, with the extinction coefficient β of a material with complex refractive index $n = 1 - \delta + i\beta$. Such materials are low-Z materials, for example Be, B, C (diamond), polymers or Al. To obtain the desired focal spot size in the range of below 200 nm, high quality structuring of the X-ray lenses is needed. At Karlsruhe Institute of Technology (Institute of Microstructure Technology), CRLs are made by precisely structuring the epoxy-resin based negative polymer resist SU-8 (subtype "mr-L-50" by mrt, Berlin, Germany) by deep X-ray lithography [2]. The process provides a patterning accuracy below 100 nm and allows for high aspect ratio structures with ratios up to above 100 for certain geometries. This fabrication method is an X-ray absorber mask based lithography process. The absorber mask is fabricated via 100 kV electron beam writing to generate the required geometric shape of the parabolic lenses, followed by electroplating gold as X-ray absorber on a thin titanium membrane. The SU-8-resist is exposed by shadow projecting the mask's structures into a 400 µm thick resist layer at the storage ring ANKA. The samples are developed to remove unexposed areas and dried after a post-bake step. The resulting substrate carries several parallel rows of line focusing (1D) CRLs. To get point focusing (2D) CRLs, the substrate is sawn to get single rows. Then one row is rotated by 90° around the optical axis and mounted with another row to form a 2D focusing CRL (see figure 1).

The SU-8 polymer resist, cross-linked during X-ray exposure, is not significantly modified by later X-ray exposures of the CRLs. These lenses have been proven to be long time radiation stable at multiple synchrotron radiation applications and in laboratory settings up to a deposited dose of 2 MJ/cm^3 [3]. Only some outgassing of small molecules takes place, modifying the focal length of a CRL in the range of one percent after a use of several months at synchrotron beam lines.

Figure 1 b) shows a set of planar parabolic refractive lenses with an effective aperture of 30 µm and minimal radii of curvature of 6 µm made out of SU-8 by deep X-ray lithography using synchrotron radiation generated at the ANKA storage ring in Karlsruhe, Germany. The finalized lenses can be operated in air at ambient conditions. A crossed geometry, intersected lens stack provides an efficient two-dimensionally focusing close to free of spherical and astigmatic aberration. At PETRA III synchrotron radiation source, the lenses have already been successfully used in full field microscopy setups (beamline P05, [4,5]).

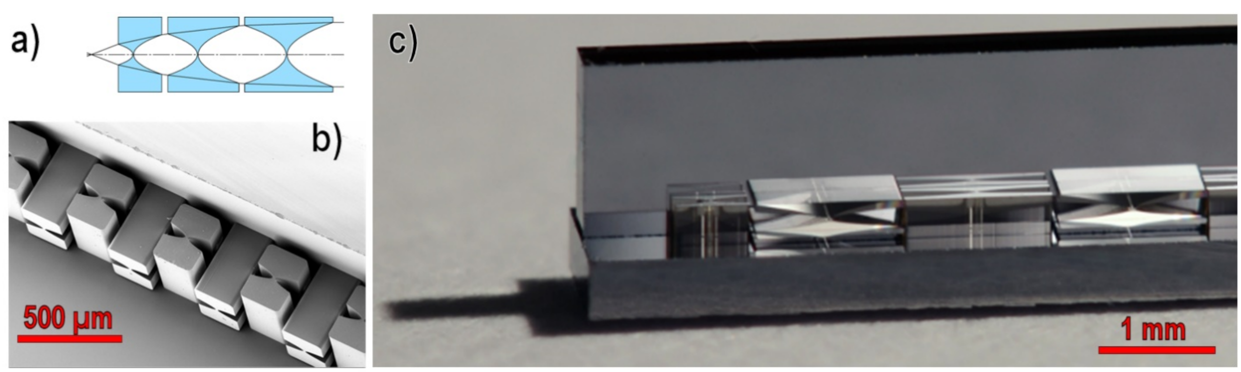


FIGURE 1. (a) Conceptual idea of an adiabatically focusing lens where the aperture and the apex radius of curvature continuously decrease along the beam path to account for the ever decreasing beam size. **(b)** SEM image and **(c)** optical image of a crossed geometry, 2D focusing lens. The two silicon wafers supporting the lens halves are glued together at a precise 90° angle.

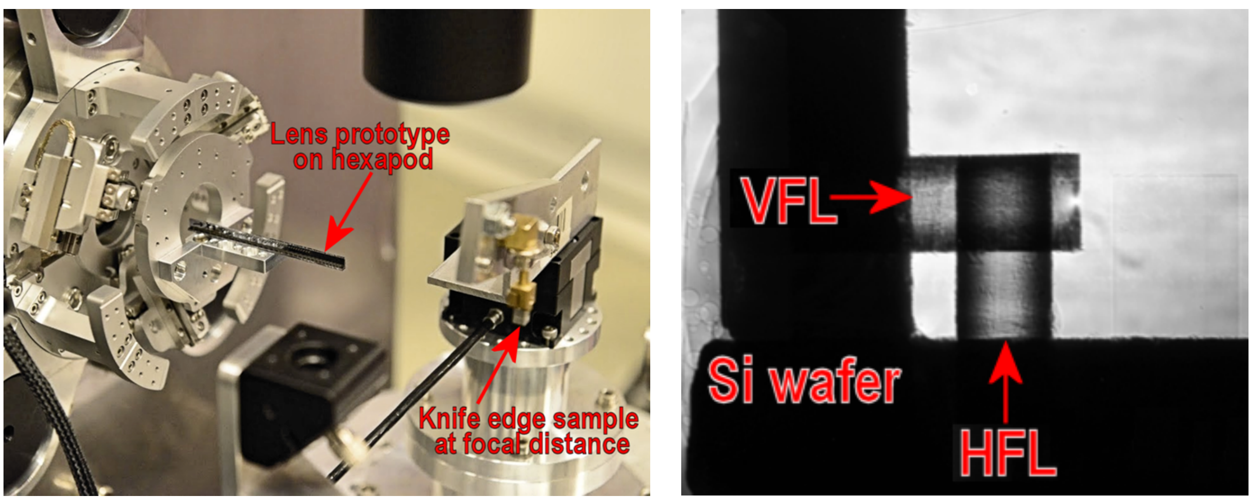
CHARACTERIZATION RESULTS

2D focusing lenses with adiabatic geometry have been installed at the Nanofocus Endstation of P03 beamline [6], at the PETRA III synchrotron source (DESY, Hamburg) for the purpose of initial characterization and for the determination whether this type of lenses is suitable as a nanofocusing element in an X-ray diffraction setup. The

optical element normally used at the Nanofocus Endstation is a pair of elliptical mirrors (KB mirror) and this optical system routinely generates a focal spot of 250 x 350 nm² at focal distance of 10 cm, throughout the entire energy range of the beamline (8-22 keV). The high focusing efficiency and the achromatic properties of a KB mirror system are strong benefits of this focusing concept and they are essential for the experiments routinely performed at the Nanofocus Endstation (specifically materials science related research). However, these benefits come at the price of tedious alignment and high sensitivity of KB mirrors to minute changes of incidence angles. Because of this, we are exploring different focusing schemes in order to identify viable alternative approaches for nanofocusing at P03.
Two adiabatic lens systems ("lens A" and "lens B") were used in our first tests and their geometric parameters are given in table 1. Both lenses had continuously decreasing apertures adapted to the decreasing beam diameter. In addition, lens B also had a continuously decreasing apex radius - in this way the exit numerical aperture of the lens stack is maximized and the focal spot diameter is minimized. At P03 beamline the lenses were used to directly demagnify the undulator source (horizontal size of 376 µm) at 13 keV.

	input aperture	input apex radius	exit aperture	exit apex radius	total length	working distance
lens A	170 µm	6 µm	111 µm	6 µm	74 mm	37.5 mm
lens B	110 µm	9.6 µm	90 µm	7.5 µm	17.5 mm	75 mm

TABLE 1. Geometric parameters of the two lenses used in the experiment. The working distance is the distance from the exit aperture to the focal spot (often erroneously referred to as focal length).



(a) (b)

FIGURE 2. (a) A 2D focusing lens was installed on a precise hexapod at the Nanofocus Endstation of P03 beamline (PETRA III, DESY, Hamburg). Beam direction is from left to right. The hexapod facilitated the linear and rotational alignment of the lens (74 mm long) while a video microscope (equipped with a fluorescent screen) was used downstream of the lenses focal plane to provide a real-time feedback for the alignment process. **(b)** The video microscope image of the readily aligned lens exhibits the horizontally focusing lens (HFL) and the vertically focusing lens (VFL) portions, next to their overlap where 2D focusing is achieved. The silicon wafer supporting the two lens halves is also clearly visible.

During the experiment, each lens was mounted on a miniaturized hexapod and the alignment of the lens into the X-ray beam was monitored using a video microscope that recorded the image from a fluorescent screen which was installed a few centimeters downstream of the lens focal plane (see figure 2 a+b). The aim of the initial alignment procedure was to have the optical axis of the lens coincide with the incident X-ray beam. When this goal was met, a scatter free pinhole ("Scatex" pinhole by INCOATEC, Geesthacht, Germany) was placed in front of the lens to act as an illumination aperture and a series of absorption scans ("knife-edge scans") was then performed to record the spatial change of the beam size along the beam direction. The full layout of the relevant beamline components is shown in figure 3. As shown in figure 4, the data recorded from lens A indicates a symmetrical beam waist with a distinct focal spot. The horizontal beam profile at the focal spot position can be fitted using a superposition of two Gaussians and notably no "tails" or fringes can be seen in the intensity profile. Although not shown, the vertical beam profile is nearly identical.

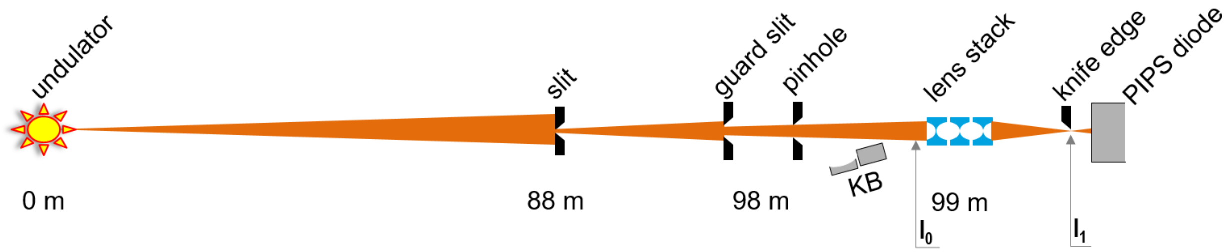


FIGURE 3. Conceptual sketch of the beamline components that are relevant for the data reported herein. Note that the monochromator (at 54 m) is not shown and that the KB mirror, normally used for nanofocusing at P03 Nanofocus Endstation, had been removed from the beam. A scatterless pinhole acts as the illumination aperture for the lens.

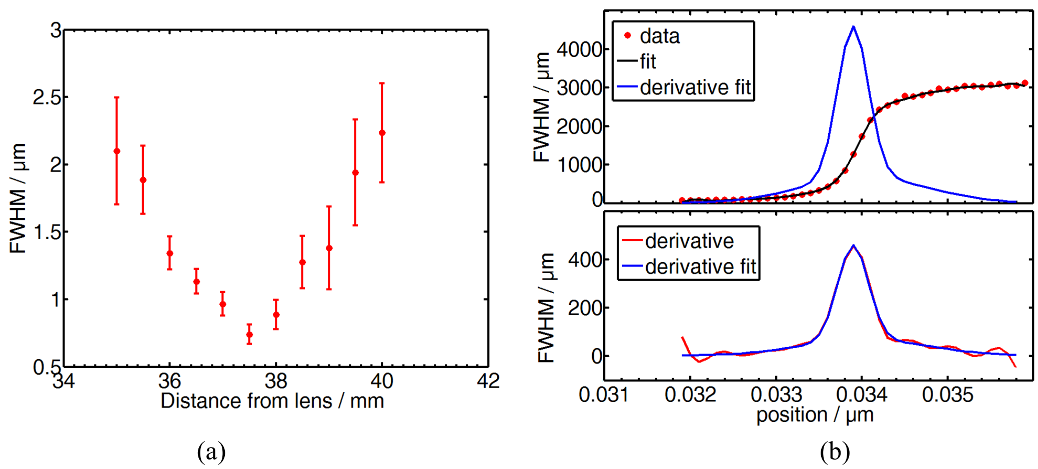
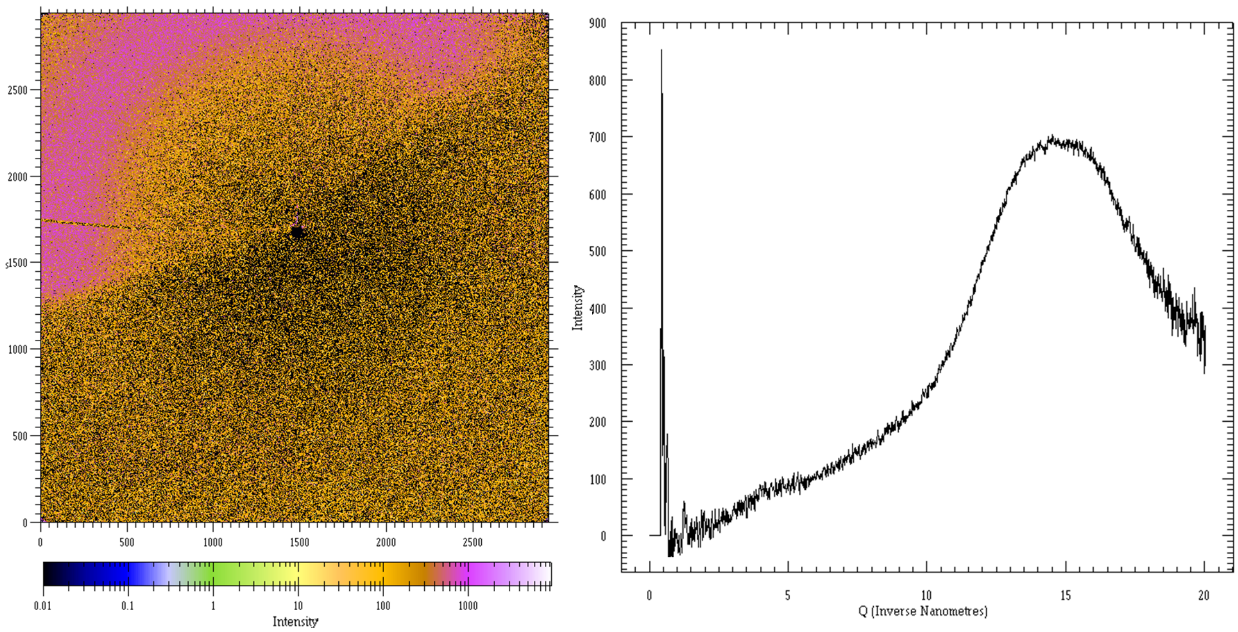


FIGURE 4. (a) The full width half maximum beam size (FWHM) was measured around the expected focal plane of lens A, using a gold knife edge probe and a symmetrical beam waist was found for both the vertical and horizontal beam profile (data shown is the horizontal profile). (b) The profile recorded at the minimum beam size position (37.5 mm from the lens exit) was fitted using a sum of two Gaussians to account for the focal spot as well as the wider and weaker background. At the minimum beam size position, the full-widths-at-half-maximum of the two Gaussian are 599 nm and 2.3 µm. Note that both panels share the same horizontal axis.

In order to further assess the applicability of these lenses for X-ray diffraction purposes, a 2D charge-coupled device (CCD) detector was used to record a near field image of parasitic scattering, caused by the lens itself. The data recorded from lens B is shown in figure 5 and it indicates a very clean low angle region and a distinct, broad diffraction ring at 15 nm^{-1} i.e. at high angles. It must be noted though, that this diffraction ring is only visible because the 2D lens was left un-covered on its open side. Consequently, the diffraction ring is not visible in the lower half of the CCD image simply because the silicon wafers absorb it. Consequently, a lens adequately covered on all sides would generate a CCD image without that high-angle parasitic scattering. In order to determine the focusing efficiency of each lens, the absolute flux was measured at both, the lens input (I_0) and in the focal spot of each lens (I_1, as indicated in figure 3). The ratio (I_1/I_0) of these two values was found to be 10% for lens A and 50% for lens B. The measurements of I_1 were performed using a calibrated PIPS diode (Passively Implanted Planar Silicon diode *Canberra* CAM-300AB) which was operated at a reverse bias of 10 V and placed 2 cm downstream of the focal spot. The photocurrent was amplified using a high gain current amplifier (*Femto* DLPCA-200) and converted into a photon flux according to the calculation provided in [8]. For the determination of I_0 the measurement was repeated with the lens removed from the beam and the guard slit aperture set to the size of the lens input aperture. Due to the large input size of the diode (300 mm²) the entire unfocused beam was recorded and the additional gas absorption was taken into account for a correct determination of the flux upstream of the lens position.



(a) (b)

FIGURE 5. (a) CCD detector image recorded from lens B at a distance of 37 cm. The detector image is background subtracted, i.e. it contains only scattering contributions from the lens itself. Note the very clean low angle region around the beam stop and the high angle parasitic scattering exiting the lens through its open side. (b) azimuthal integration of the detector image

CONCLUSION

2D focusing lenses with adiabatic geometry have been installed at the Nanofocus Endstation of P03 beamline, at the PETRA III synchrotron source, in order to determine their suitability as nanofocusing optics for X-ray diffraction experiments.

These lenses exhibit efficient focusing of X-rays at an energy of 13 keV with no detectable parasitic scattering at small angles, i.e. within the angular range that is relevant for SAXS (Small Angle X-ray Scattering) experiments. However, a broad diffraction ring was observed at larger angles. Its peak intensity, located at $q = 14.5\ nm^{-1}$, is constant along the detectable azimuthal range of about 150°. A potential source for this diffraction contribution is benzene [7], which is a major constituent of the randomly oriented SU-8 molecules. This parasitic scattering, however, can be removed very easily, by simply putting an adequate cover on the open side of the lens – doing so not only will remove the unwanted parasitic scattering. Equally important it will protect the fragile lens structure from mechanic damage and accumulation of dust particles in the lens - which otherwise is fit to be operated in air. A beam size of 599 nm in the horizontal direction was achieved, following only a short initial alignment procedure. This focal spot size is about twofold bigger than the ideal image size (283 nm) at the given image and source distances (74.5 mm and 99 m, respectively). The deviation reflects imperfections of the lens profile and is attributed to non-orthogonalities of the side walls of the lens elements (which is subject of improvement in future lenses).

Because of the long focal distances attainable with these lenses and the resulting low beam divergence, the angular resolution of an X-ray nanodiffraction experiment is not compromised, compared to KB-focusing. The inherent downside of any refractive X-ray optics is that the focal length is a function of X-ray energy (unlike with KB mirrors). Given the low cost of fabrication (ca. $3k for one 2D lens set) it is, however, viable to put a set of lenses on stock for the most commonly used X-ray energies. Such a set of lenses can be shipped with all lenses mounted on a single substrate, thus changing between different lenses can be done remote-controlled without having to access the lens plate.

Due to the relatively large apertures of these lenses and the fixed 90° configuration of the two lens halves it is relatively easy to align the lenses with the incident X-ray beam. Using a single 4D positioner (2 linear and 2 rotational freedoms of degrees; provided e.g. by a hexapod) and a video microscope the initial alignment procedure can be as short as one hour, making these lenses a viable alternative optical element at our experimental setup. Given the ease of installation these lenses are well suited to be used as a fallback option at P03 Nanofocus Endstation in case the KB-mirror based focusing cannot be used – for whatever reason.

ACKNOWLEDGEMENT

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