A combined Kirkpatrick-Baez mirror and multilayer lens for sub-10 nm x-ray focusing

A. Ruhlandt, T. Liese, V. Radisch, S. P. Krüger, M. Osterhoff, K. Giewekemeyer, H. U. Krebs and T. Salditt

1 Institut für Röntgenphysik, Universität Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany
2 Institut für Materialphysik, Universität Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany

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We have used a combined optical system of a high gain elliptic Kirkpatrick-Baez mirror system (KB) and a multilayer Laue lens (MLL) positioned in the focal plane of the KB for hard x-rays nano-focusing. The two-step focusing scheme is based on a high acceptance and high gain elliptical mirror with moderate focal length and a MLL with ultra-short focal length. Importantly, fabrication constraints, i.e. in mirror polishing and bending, as well as MLL deposition can be significantly relaxed, since (a) the mirror focus in the range of 200-500 nm is sufficient, and (b) the number of layers of the MLL can be correspondingly small. First demonstrations of this setup at the coherence beamline of the PETRA III storage ring yield a highly divergent far-field diffraction pattern, from which the autocorrelation function of the near-field intensity distribution was obtained. The results show that the approach is well suited to reach smallest spot sizes in the sub-10nm range at high flux.

Coherent x-ray imaging as a lensless technique holds the promise to overcome the resolution limits of x-ray objective lenses. In this context, ‘lensless’ means that no optical element is placed between sample and detector. This avoids limitations in the numerical aperture and allows highest possible dose efficiency. On the other hand, optical focusing of the illuminating beam is essential for obtaining high resolution images. Therefore, a high resolution experiment still calls for smallest possible beamsize or more precisely highest coherent flux enhancement. The condenser or illumination system (refractive, diffractive or reflective optics) depends on the specific experimental parameters and requirements, such as photon wavelength $\lambda$, bandwidth, focus size $\Delta$, efficiency etc. Importantly, as in other fields of optics, x-ray focusing can be optimized by use of combined systems. Here we present the combination of two very powerful optical components, preserving the respective advantages of each, to create a one-dimensional, high flux density, fully coherent x-ray line focus in the sub-10 nm range. In detail, we have used the high geometric acceptance and gain of a fixed elliptical shape Kirkpatrick-Baez mirror system (KB), with subsequent focusing by a nanoscale diffractive optical element, namely an off-axis multilayer Laue lens (MLL) of ultra-short focal length $f$ with a cross-section (aperture) adapted to the KB focal spot size.

MLLs have been developed as one- or two-dimensional zone plates fabricated by thin film deposition with interface positions according to $y_n = \sqrt{(n\lambda/2)^2 + n\lambda f}$ for each layer $n$. Compared to lithography, much smaller outermost zone widths down to $d_{r_n} \simeq 1$ nm can be achieved, allowing for large numerical aperture and correspondingly small spot size $\Delta \simeq 1.22d_{r_n}$. It is now well understood that this ideal value, calculated based on the projection approximation (thin diffraction structures compared to $f$), is constricted by diffraction within the zone plate (volume diffraction).
FIG. 1. (a) Schematic of the combined optics with KB pre-focusing followed by an off-axis MLL, in order to combine high gain and short focal width \( f \). (b) Optical path illustrating the short focal length \( f \approx 15 \, \mu m \) and the opening angle (14 mrad) of the first order diffracted beam. (c) Wave-optical simulation of the near-field behind the MLL (parameters see text), showing a 3.8 nm focal width under assumption of ideal ‘thin object’ conditions. (d) Simulation for the actual MLL fabricated and tested in this work.

However, numerical and theoretical studies\(^3,7,8\) indicate that values of \( \Delta \approx 1 \, nm \) can still be realized if the zones are tilted, with a tilt angle increasing with distance from the center. This is well below the limit of optics based on total external reflection (single coated mirrors, waveguides\(^9\)), and also below the smallest focus size reported so far (7 nm) achieved with an adaptive multilayer x-ray mirror system.\(^10\) Using a MLL which was tilted to an average Bragg angle, hard x-rays have been focused down to 16 nm,\(^4\) and more recently to 13.1 nm focal width (FWHM).\(^11\) Wedged designs with each zone satisfying its (local) Bragg condition have been addressed theoretically. A crossed arrangement of two 1D focusing MLLs with respective alignment tools was recently reported, leading to a 2D focal size of \( 25 \times 27 \, nm^2 \) at 12 keV and a focused flux of \( 2 \cdot 10^6 \) photons/sec.\(^12\) Two conclusions can be immediately drawn from the significant efforts and results of MLL optics, in particular with regards to further improvements down to smallest zone width/ highest resolution: (i) Adjusting a MLL to an average Bragg angle will work better for a small MLL segment with a smaller number \( N \) of illuminated zones. Along the same lines, fabrication of wedged zones will also be much easier, since all constraints in fabrication (stress, cumulative roughness) favor small MLL apertures. (ii) If small \( N \) MLLs are required without compromising high flux, an optimized focusing system has to be devised based on high gain pre-focusing. In other words, the MLL has to be illuminated by truncated plane waves in the focal plane of a second optical component, such as a high gain KB.

The schematic of the setup is shown in Fig. 1(a). Due to the small KB focal cross section (320 nm in the present experiment), only a segment of a MLL is illuminated, and must be fabricated. In general, the segment will not contain the central zone, and thus focus off-axis with respect to the optical axis of the pre-focusing system. In analogy to previous designs of condenser zone plates\(^13\) for x-ray microscopes we denote these structures by off-axis OA-MLL. The specific geometric values of the OA-MLL designed for the experiment along with the coordinate system (optical axis \( z \)) and the focal rays are indicated in (b). The center of the OA-MLL segment is aligned with the KB beam and set to \( y = 0 \). The fact that the first order focused beam of the OA-MLL diffracts off the KB optical axis, enables a partial or complete separation from the primary beam and higher order beams. The simulated intensity distribution behind the OA-MLL with parameters fixed to the experimental design is visualized in (c) for an ideal exit wave as calculated based on the ‘projection approximation’, and in (d) for the actual thickness of OA-MLL \( D = 3.26 \, \mu m \) based on a multi-slice algorithm. The simulations show that neither the (nano-size) illumination by the KB nor the
OA-MLL concept compromise sub-10 nm focusing. In fact, for the specific parameters (detailed below), the simulations yield a focus size of $\Delta = 3.8$ nm (intensity) calculated for the ideal exit wave shown (c), and $\Delta = 9.0$ nm (FWHM) for the exit wave resulting from the multi-slice treatment of volume diffraction. Note that the small focal length $f = 14.7 \mu m$ may withstand certain scanning applications needing large sample holders or environment. However, for projection propagation imaging (in-line holography), which is the main application driving the present work, the sample is positioned in a (downstream) defocus position, and a short $f$ is hence not overly restrictive. The design parameters and materials of the OA-MLL were selected in view of the experimental setup at beamline P10 at PETRA III (DESY, HASYLAB) and a photon energy of $E = 13.8$ keV, based on numerical simulations and the measured intensity distribution in the KB focal plane.\(^{14}\) The focused KB beam can be approximated by a Gaussian beam with a waist size of 320 nm $\times$ 200 nm (FWHM). Since the OA-MLL is placed in the focus of the KB, it is illuminated by a plane wavefront with a diameter corresponding to the Gaussian waist size. The height of the OA-MLL was 401 nm. For given $E$, tungsten and silicon are suitable materials because of the difference in their refractive indices. The ideal relative phase shift of $\Delta \varphi = \pi$ between the two multilayer materials requires an optical OA-MLL thickness of $D = 3.26 \mu m$. The fabrication process sets a constraint to the smallest zone width of $dr_N \geq 1.6$ nm. In a first simulation step, the Fraunhofer integral was solved numerically with the assumption of an infinitely thin structure, i.e. discarding volume effects. Two competing conditions emerge: On the one hand, the focal diameter decreases for shorter focal length, leading to foci down to $\simeq 3$ nm. On the other hand, the separability of the exit beams improves for larger focal length due to the limited $dr_N$. Under the given KB illumination condition, the simulation shows a good compromise at $f = 14.7 \mu m$. In a second step, the finite thickness of the OA-MLL was taken into account. We simulated the propagation through this structure by multi-slice Fresnel propagations. As shown in Fig.1(d), the finite depth has a crucial influence on the exit wavefield. It leads to a distortion of the exit wave, resulting in focus broadening, larger depth of focus (DOF), and higher background. However, for a first proof-of-principle of this combined optical scheme, fabrication was first carried out for the $\pi$-phase shifting value of $D = 3.26 \mu m$. In a next step, the thickness can be decreased to find an optimum value balancing the increase in phase shift with thickness against the detrimental effects of volume diffraction.

Based on the results of the simulation, we thus fabricated W/Si OA-MLLs using the combination of pulsed laser deposition (PLD) and focused ion beam (FIB) similar to the manufacturing process for soft x-ray transmission optics.\(^{15,16}\) The W/Si multilayer structure was laser deposited on Si$_3$N$_4$ according to the Fresnel zone plate law. It consists of the first 122 layers with heights between 36 nm and 1.6 nm leading to an overall height of 401.4 nm, starting on Si$_3$N$_4$ with the thinnest layer to avoid layer positioning errors and cumulative roughness.\(^{17}\) FIB processing was used to cover the multilayer by platinum, to cut a MLL structure to the desired thickness of 3.26 $\mu m$, to handle and manipulate the MLLs, including crossing two MLL lamellae for future 2D focusing, and to attach the MLL to the flattened tip of an Omniprobe needle for easier mounting, see Fig.2. For comparison, one half of the MLL was thinned down to 170 nm to also allow imaging measurements of a thin object.

The experiment was carried out at the nano-focus (KB) endstation of the coherence beamline P10 at the PETRA III storage ring of HASYLAB/DESY. The instrument and the KB focusing is described in.\(^{14,18}\) The MLL was mounted on a piezo $xyz$-stage positioned above a high precision rotation axis and additional $yz$ translations, and was aligned using an optical microscope (KB focus position), dark-field scans in the KB as well as in-line holography images in a waveguide beam which was pre-aligned to the optical axis. Figure 2(c) shows the hologram of the adjusted MLL illuminated by the diverging beam of a waveguide. The far-field pattern in Fig.3(a) was stitched together from three single images (horizontal shift) obtained with the detector PILATUS 300K (5.29 m behind the sample, 0.1 sec accumulation time), using a central beamstop (0.8 mm tantalum cylinder) to block the zero order. The diffraction pattern $I(q_x, q_y)$ extends over a very large $q$-range of $\geq 0.184$ $\text{Å}^{-1}$. In fact, the opening angle of the first order was not fully captured by a single detector frame, nor by a composite of three detector positions, since the flight tube obstructed larger diffraction angles. A horizontal diffraction cone with a half opening angle of 17.3 mrad was evidenced by an additional CCD detector (FDI, PhotonicScience), which was moved in front of the flight tube.
FIG. 2. (a) Electron microscopy image of the MLL placed on a tungsten tip. (b) Schematic of the MLL deposited on an approx. 1 μm thick Si₃N₄ membrane (bottom layer on the figure). Above the laser deposited W/Si multilayer (main part of the MLL), electron beam and ion beam Pt is deposited. (c) Hologram of the MLL, obtained by aligning the MLL in a waveguide beam. Further alignment of the MLL was performed by scanning the KB beam and recording the dark field contrast. (d) Electron microscopy image of a two-crossed MLL enabling the two-dimensional focusing of the x-ray beam.

In the vertical direction some intensity distribution is observed resulting from the tails of the KB beam. The horizontally very extended far-field diffraction pattern directly indicates very small real space intensity variations in the focal plane. Indeed, computing the 1D auto-correlation function $g(x) \propto FFT^{-1}[I(q_x)]$ from the farfield intensity distribution (Pilatus data shown in Fig.3(a)), a central full width of the autocorrelation function $FWHM_x = 6.8$ nm is obtained, see Fig.3(b)–3(d). Fig.3(d) shows the central peak of the one-dimensional autocorrelation function along the horizontal axis, which was obtained by integrating the pattern in Fig.3(c) along the vertical axis. This autocorrelation function, determined directly from the experimental far-field pattern, samples the central peak only with three pixels due to the detector geometry and the properties of the discrete Fourier transform. For higher sampling in the least-square fit, we therefore increased the sampling by zero padding of the intensity matrix, an operation effectively corresponding to an interpolation of the data. Fig.3(d) shows that the raw experimental autocorrelation data and the resampled curves are consistent, and can be fitted to a Gaussian with $FWHM_x = 6.8$ nm. Next, Fig.3(e) shows a comparison between the experimental result and the autocorrelation function expected from simulated field propagation. To this end, the far-field intensity corresponding to the parameters of Fig.3(d) was computed. After removal of the central pixels in the corresponding intensity matrix to mimic the beamstop, the autocorrelation was computed by an inverse Fourier transformation, showing an excellent agreement in the central peak. This central peak is maintained when the autocorrelation function is computed without ‘beamstop’, but resides on a broad peak, as shown in Fig.3(f). This broad peak is the result of
FIG. 3. (a) Measured far-field pattern of the MLL. The intensity is encoded logarithmically in the colormap ($I$ [cts]). Low $q$-values in the center are missing due to the beamstop. In horizontal direction, diffraction of the KB beam at the MLL leads to a wide spread of the far-field pattern. (b) Autocorrelation of the MLL exiting wave-field as determined from the far-field pattern (a) in logarithmic scale ($I$ [a.u.]). (c) Close-up of the autocorrelation in linear scale indicating a narrow intensity maximum in the center. (d) Horizontal profile of the autocorrelation function (dashed blue line), after vertical integration of the data shown in (c). The function is sharply peaked, therefore the central width is sampled only with three points. Padding of the diffraction data yields a consistent autocorrelation function of the same width (denoted as 'experimental resampled'), which can be fitted to a Gaussian (red line) of 6.8 nm FWHM. (e) Comparison of the experimental autocorrelation function (black line) with the wave optical simulation (red line). The central width is in excellent agreement. For comparison, a beamstop was assumed in the simulation to mimic the same filtering effect on the autocorrelation in the experimental analysis. (f) Acting as a high pass filter in the diffraction pattern, the beamstop suppresses the tails and broad features of the autocorrelation. Computed without beamstop, the sharp central peak (red arrows) resides on a broad maximum (blue arrows) corresponding roughly to the size of the beam in the focal plane.

The comparison between simulation and experiment can also be carried out in reciprocal space, by comparison of the measured $I(q_{x})$ with simulations, see Fig.4. The length of the simulations and the number of parameters impede least-square fitting, but semi-quantitative agreement can be readily reached, as shown in Fig.4(c), based on a reasonable parameter choice. The simulated far-field curve exhibits fine oscillations which are better resolved in the experimental data after closing the horizontal slits in front of the KB, from 0.38 μm to 0.08 μm, see Fig.4(a) and 4(b), respectively, leading both to a higher degree of coherence and a larger beam waist in the focal plane. The calculated far-field pattern shown in Fig.4(c) is based on the geometric design parameters of the MLL and beam propagation as in Fig. 1, but at the smaller KB slit setting, and with an additional tilt angle (misalignment) of 3.2 mrad taken into account. The difference between the experimental and simulated curve can possibly be attributed to further deviations from the idealized simulation, notably a positional shift of the MLL in the KB focus, or the lineshape of the focal intensity distribution which was assumed to be Gaussian.

The optical scheme proposed here provides a quasi-line (and quasi-point for the case of the crossed geometry) virtual source of sub-10 nm width and high intensity. In the present experiment, the integrated flux behind the MLL was measured with a calibrated pin diode to $7.9 \cdot 10^{10}$ photons/sec. Based on the multi-slice simulation of the deposited structure shown in Fig.1(d), the focus carries about 4.7% of this flux, corresponding to $3.7 \cdot 10^9$ photons/sec, which is significantly higher than the values obtained for sub-10 nm focusing based on single optical components. In the next step,
volume diffraction effects should be reduced by dicing the MLL to smaller width. Furthermore, an extension from 1D to 2D focusing can be envisioned by use of a crossed MLL. Finally, we stress that the performance of the compound optical system does not rely on the lens being positioned in the KB focus. For illustration, an example of a Fresnel zone plate positioned upstream from the focus is included as supplemental material (SOM), along with the simulation code for field propagation used in this work. We conclude that the compound KB-MLL optical system can fill a gap combining high coherent flux and smallest spot size, significantly extending the field of applications for full-field 3D phase contrast tomography at nanoscale resolution. Further improvements in view of higher MLL efficiency as well as focal size can easily be envisioned, challenging the current 7 nm record in x-ray focusing.

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20 See supplementary material at http://dx.doi.org/10.1063/1.3698119 for the simulation code, and a further example of beam propagation.