Ptychographic characterization of the wavefield in the focus of reflective hard X-ray optics

Cameron M. Kewish a, Pierre Thibault a, Martin Dierolf b,1, Oliver Bunk a, Andreas Menzel a, Joan Vila-Comamala a, Konstantins Jefimovs c, Franz Pfeiffer b,1

a Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland
b Technische Universität München, D-85748 Garching, Germany
c EMPA, CH-8600 Dübendorf, Switzerland

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Abstract
A technique for quantitatively characterizing the complex-valued focal wavefield of arbitrary optics is described and applied to reconstructing the coherent focused beam produced by a reflective/diffractive hard X-ray mirror. This phase-retrieval method, based on ptychography, represents an important advance in X-ray optics characterization because the information obtained and potential resolution far exceeds that accessible to methods of directly probing the focus. Ptychography will therefore be well-suited for characterizing and aligning future nanofocusing X-ray optics.

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1. Introduction

Nanofocused X-ray beams promise to provide high spatial resolution for X-ray spectroscopy, scanning microscopy and diffraction. X-ray optics, such as zone plates [1], refractive lenses [2], total reflection mirrors [3], and multilayer optics [4,5], have been used to focus hard X-ray beams to <100 nm, and in some cases below 40 nm. These focal spot sizes represent significant achievements in the spatial resolution of hard X-ray microscopy, but the technological developments to achieve the ultimate wavelength-limited resolution with X-rays are still far out of reach.

When trying to increase the resolution it is crucial to characterize the aberrations in the focus, to give insight into the relationship between fabrication tolerances and optical performance. Precise measurement of the intensity profile in the vicinity of the focus, or ‘probe’, becomes more difficult as the beam size decreases. Conventional ‘knife-edge’ scans are expected to fail at higher photon energies and numerical apertures, because the depth of focus can be shorter than the X-ray attenuation length in the knife-edge material. Alternative procedures have been employed to measure probe intensity profiles of the order of 25–100 nm-wide, e.g., using fluorescence from nanofabricated structures [4–6], or modulation transfer function measurements from periodic samples [7]. However, these methods are limited to providing linear profiles of the beam intensity, and are therefore insensitive to wavefront aberrations and are restricted to working directly in the focal plane. A procedure similar to autocollimation was used to infer wavefront aberrations introduced by a hard X-ray mirror in one-dimension (1-D), measuring beam displacements during slit-scanning [4]. This can be extended to two dimensions (2-D) by scanning a pinhole rather than a slit, but the resolution is limited by the pinhole size and diffraction between the aperture and the optics.

Phase retrieval algorithms are often used to reconstruct complex-valued wavefields from far-field coherent diffraction patterns. So-called ‘lensless’ imaging replaces the X-ray image-forming system with computations and produces images with resolution dictated by the range of diffraction angles measured. These techniques are usually based on iterative algorithms [8,9], although analytical methods are being developed (see e.g., [10]). Once the phase problem is solved, the far field exit wave can be Fourier transformed to yield the projection of the complex refractive index distribution of a sample. Phase retrieval calculations incorporating optical metrology data were used to refine the phases of focal plane intensity measurements at high resolution in 1-D [11,12], but the nanostructure used to probe the focus restricted the technique to obtaining linear wavefront profiles.
An iterative method based on curved wavefronts was proposed for reconstructing the 2-D probe wavefield from the far-field intensity distribution [13,14]. It relies on an accurate knowledge of the wavefront curvature and the dimensions of the focusing optic, or the pupil function, to provide the real-space support constraint for the reconstruction algorithm. However, for reflective optics, which necessarily operate at grazing incidence angles, or volume diffractive optics, the exit pupil plane is not as easily defined.

Recently, an iterative phase-retrieval method related to ptychography was reported, which makes use of multiple exposures to solve the phase problem [15,16]. Combining redundant information from overlapping illuminations on an extended sample additionally allows one to recover the 2-D probe wavefield in an essentially arbitrary plane perpendicular to the focused beam between the optic and the detector [17–19]. By numerically propagating the reconstructed wavefield along the optical axis, it is possible to diagnose aberrations in the optical system resulting from astigmatism, or misalignment. The information obtained in this process can allow optimization of a beamline optics, alignment of a focusing system for microscopy or scanning probe applications. Propagating the wavefield back to the focusing optic can further allow an assessment of the fabrication errors that are present, by analysis of the wavefront aberrations produced by the optics. We have applied ptychography to assess the focusing performance of a curved multilayer X-ray focusing optic, as a demonstration of principle, to assess whether this method will be useful to characterize future X-ray optics providing nanometer-sized hard X-ray probes.

2. Experimental procedure

The experiments were carried out at the cSAXS beamline at the Swiss Light Source, an undulator beamline that is optimized for coherent X-ray scattering techniques and small angle-scattering in the range of wavelengths between 0.65 and 3.0 Å. A schematic of the experiment geometry is shown in Fig. 1.

A double-crystal Si(111) monochromator selected 1.54 Å wavelength radiation from the undulator spectrum, which was incident at a grazing angle of 0.23° on a Rh-coated mirror to reject >97.5% of the higher-harmonics, prior to the beam entering the experiment hutch. The radiation source is approximately 20 μm (vertical) by 200 μm (horizontal) in size at the undulator, which at the chosen wavelength of 1.54 Å provides coherent radiation over an area of ~250(ν) × 25(ν) μm² transverse to the beam in the experiment hutch 34.5 m downstream [20].

The X-ray focusing optic used for this experiment is a commercial 2-D focusing device [21], whose paraboloidal reflective surface is coated with a laterally graded multilayer optimized for X-rays of this wavelength. This mirror was designed to collimate the divergent beam from a laboratory X-ray source into a parallel beam. Instead, the mirror was used in a reversed geometry, to focus the undulator beam to a size of the order of a micron at a distance approximately 120 mm from the center of the mirror.

A coherent portion of the beam was selected by a 20 μm-diameter pinhole located approximately 12 mm upstream of the entrance window of the mirror chamber. The apertured beam was incident upon the mirror at a grazing angle of θ = 1.3°, corresponding to the Bragg angle of the multilayer coating at this wavelength. In the focal plane, a nanofabricated sample was scanned with a high-precision piezoelectric positioning stage (nominal 0.3 nm resolution with <2 nm repeatability). The sample stage was mounted on an optical bench that was rotated to an angle of 2θ with respect to the incident beam.

The radiation transmitted and diffracted by the sample traveled through a He-filled ‘flight tube’, to minimize absorption and scattering along the 7.26 m path to a PILATUS 2M detector [22]. This temperature-stabilized detector samples the far-field intensity with 1475 × 1679 pixels of size 172 × 172 μm², having 20-bit dynamic range, no readout noise or dark current, and a point-spread function of one pixel [23]. Despite the large dynamic range of the detector, and in order to avoid the use of a beam-stop, we had to attenuate the incident beam with a 200 μm-thick Si single-crystal such that no pixel was subjected to more than a few million counts per second per pixel, corresponding to the dead-time of the detector [24].

For hard X-rays traversing thin samples, the exit wave is to a good approximation equal to the product of the probe wavefield, P(r) and the complex transmission function of the sample, O(r), e.g.,

$$\psi_j(r) = P(r-r_j)O(r),$$  (1)

where $$\psi_j(r)$$ represents the exit wave resulting from the j th sample position $$r_j$$. The intensity in the far-field, $$I_j$$, can be written as

$$I_j(q) = |\psi_j(q)|^2,$$  (2)

where the tilde denotes the two-dimensional Fourier transform of the exit wave, and the diffraction vector q is the reciprocal-space coordinate dual to the sample plane coordinate r. Eqs. (1) and (2) comprise the constraints used to guide the iterative search to recover the phases that are lost when measuring the far-field diffraction pattern. When multiple intensity measurements from partially overlapping probe positions are given as input to the ptychography algorithm, a unique solution can be obtained. Such a dataset also contains sufficient information to refine the probe function from an initial assumption. This procedure, described in detail by Thibault et al. [18], consists of numerically solving the simultaneous equations

$$\hat{O}(r) = \sum_j \hat{P}(r-r_j)\psi_j(r),$$  (3)

$$\hat{P}(r) = \sum_j \hat{O}(r+r_j)\psi_j(r+r_j),$$  (4)

where the circumflex represents the discrete, reconstructed images of the exit wave and the probe, respectively, and the...
superscript asterisk denotes complex conjugation. The ptychography algorithm used here assumes that the illumination function is coherent, without allowing for the possibility of partial coherence resulting from the finite source size.

A ptychography scan consisted of collecting a series of diffraction patterns from a sample illuminated at various overlapping positions. The locations of the scan points were chosen to avoid the ‘raster pathology’ which can arise when ptychography data are collected from sample positions which are periodic, as on a rectangular grid [18]. Specifically, the scan points were located on concentric circles, whose radii varied from 0.5 to 5 μm in steps of 0.5 μm. The scan started in the center, and each circle contained five positions more than the previous, requiring a total of 330 positions to scan a ~10 μm-diameter area and ensure at least 50% overlap of neighboring points. Two diffraction patterns were collected at each of the scan positions, integrating for 0.1 and 30 s, respectively. The values in the center of the shorter exposure were then used to correct overexposed pixels in the center of the longer exposure.

3. Results

For probe retrieval it is important to employ a strongly scattering sample, which includes fine details that will diffract to sufficiently high angles to achieve the desired resolution. As a sample we used a nanofabricated Snellen chart, consisting of Au structures produced by electron beam lithography and electroplating [1] into a polyimide mold resist layer on a silicon nitride supporting membrane. The sample plane was positioned near the focal plane of the mirror, determined by scanning the opaque edge of the sample-holder through the beam. Fig. 2(a) shows the reconstructed complex exit wave in the sample plane. The phase difference of approximately π/3 rad introduced by the Au structures compared to the surrounding polyimide layer indicates a Au thickness of around 0.4 μm ± 25%, where larger structures toward the top of the chart are on the upper end of this range. Although the Au thickness could not be directly measured, this range of thicknesses is reasonable because the polyimide layer was 1 μm thick, and the Au coating process was halted well below this thickness to avoid overplating. As a result, some of the structures were unfiled or partially-filed, and the average thickness was expected to be well below a micron. This is also evidenced by a scanning electron microscope (SEM) image shown in Fig. 2(b), where the light colored structures are Au and the darker background is the polyimide layer on the membrane.

The central 512 × 512 pixels were extracted from the measured full-frame diffraction patterns for reconstruction. For the experiment geometry outlined above, this results in reconstructed pixels of side-length 12.72 nm. The complex probe wavefield is reconstructed with pixels of the same size, shown in Fig. 2(c), where the image width corresponds to 6.51 μm. This resolution allows us to see the detailed structure in the beam, resulting primarily from aberrations and roughness present in the surface of the focusing mirror.

Intensity profiles were calculated by integrating all pixels in the detector at each scan point, as a thick (opaque) knife-edge was translated by ± 2.5 μm in 20 nm increments across the beam, in both the X and Y directions. Taking the derivative of these profiles, we find the full-width at half-maximum (FWHM) to be \( w_x = 1.32 \mu m \) and \( w_y = 1.26 \mu m \) in the X and Y directions, respectively. To compare the reconstructed probe with the knife-edge scans of the focus, we find the FWHM of the probe intensity integrated in the perpendicular direction, i.e.

\[
i(x) = \int |\tilde{p}(x,y)|^2 \, dy.
\] (5)

which reveals that the reconstructed beam is slightly smaller than the directly measured beam, as shown in Fig. 2(d). The size of the reconstructed focus in the X and Y directions is \( w_x = 0.77 \mu m \) and \( w_y = 0.78 \mu m \), respectively. The difference is due to the convolution of the beam size with the knife-edge shape function.

Measurements were also taken with the sample out of focus, to demonstrate that this beam characterization method is robust with respect to positioning of the sample and different wavefront curvatures. Since we reconstruct a complex-valued wavefield, we can numerically propagate it along the optical axis. Fig. 3(a) shows a sagittal view of the probe wavefield shown in Fig. 2(c), where the horizontal axis is the direction of propagation. Comparing this directly with a coronal view of the wavefield (not shown) reveals that there is no astigmatism in the focus, as the horizontal and vertical foci coincide. The beam waist can clearly be seen and the location of the original scanning plane, shown with a dashed line in the center, indicates that the sample was well inside the focus. A different sample, a 175 μm-thick silicon chip with integrated circuits, was scanned at distances of \( \Delta z = +8.64 \mu m \) (positive indicating downstream) and \( -16.28 \mu m \) from the previous scanning plane.

![Fig. 2.](image-url) (a) Reconstructed complex-valued image of a nanofabricated test object, showing Au structures deposited on a silicon nitride membrane. (b) Scanning electron micrograph of the sample. (c) Reconstructed illumination function obtained simultaneously to the exit wave. (d) Comparison of line-profiles (solid lines: X-axis shown in blue, Y in red) through the normalized intensity of the reconstructed probe, and the intensity calculated from knife-edge scans (circles). The color wheel in (c) shows the mapping for complex values in (a) and (c) displayed with phase mapped to hue and amplitude to value. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
Fig. 3 (b) shows the image of the complex exit wave reconstructed for the chip sample in the downstream position, indicated by the dashed line in Fig. 3 (a). This reconstruction used 128 x 128 pixels from each of the measured diffraction patterns, resulting in a pixel size of 50.86 nm. The top interconnect layer and via structures, of 0.25 mm dimensions, are the dominant features in the sample reconstruction. The reconstructed probe compares qualitatively very well with the probe numerically propagated by \( D \) from the focal plane [Figs. 3 (c) and (d)], and a line profile through the two wavefield intensities shows the quantitative agreement wherein the RMS difference is of the order of 2% [Fig. 3(e)]. The RMS difference between the phases of these wavefields, in regions where there is non-negligible amplitude, is approximately 0.5 rad corresponding to a wavefront reconstruction accuracy of about \( \lambda / 12.7 \). Similar agreement was obtained for the upstream position.

4. Discussion

The potential of ptychography for beam characterization in X-ray optics is demonstrated by the agreement between the wavefields reconstructed in different planes, and for different samples. The full complex wavefields before and after the sample can be recovered at high resolution without a priori knowledge of the focal length, or the sample position relative to the focus along the optical axis. Only the wavelength, sample-to-detector distance and the sample positions perpendicular to the optical axis need to be known accurately. The resolution enhancement of ptychography can be seen clearly in the details of the reconstructed wavefields. The fine details in the reconstructed sample images agree well with SEM images: despite the focal spot size of the mirror being rather large, we can resolve the ‘holes’ in the numerals 6 and 8 that can be seen in Fig. 2(b) are 0.70 nm in diameter. The spatial resolution is limited in principle only by the highest detection angle which receives statistically significant coherent diffraction intensity. In practice, however, the resolution of the probe (and object) reconstruction is ultimately limited by the accuracy to which one knows the sample positions, or can refine them from the data.

Comparing the intensity of the reconstructed probe wavefield with the knife-edge scans shows that the directly measured values overestimate the beam size. This results primarily from the convolution of the knife-edge shape with the beam profile. In the present case, the effect is quite noticeable, because the knife-edge intersecting the beam was not optimized for the measurement. In fact, an edge of the 3 mm-thick sample holder was used, and therefore the measurement represents a beam size measurement at up to 3 mm downstream from the sample position. Simply propagating the focused probe by this distance increases its FWHM by approximately 40%, which improves the agreement between the two measurements, but indicates that there is a further contribution, e.g., from the roughness of the edge. This highlights a difficulty of physically probing a focused beam, even using specialized knife-edges and scanning techniques, since the beam size and depth-of-focus will decrease appreciably with diffraction-limited hard X-ray nanofocusing optics.
5. Summary and outlook

The objective of this experiment was to characterize the focus of an X-ray mirror using ptychography. The reconstruction algorithm allows the unique recovery of the complex wavefield in the plane of a specimen which is scanned through a coherent beam. The reconstructed wavefield can then be numerically propagated along the focused beam path, making available detailed information about the optical system, including astigmatism and other aberrations which are not obtainable from knife-edge scans. We reconstructed exit waves from samples located at several positions relative to the focus of an X-ray mirror, along with a self-consistent set of illumination functions. The beam size obtained is smaller than that measured with knife-edge scans of the intensity profile, a result which can be attributed to instrumental broadening. Ptychography does not require accurate knowledge of the dimensions of the focusing mirror or the form of the pupil function, instead relying on very accurate sample positioning and far-field intensity measurements collected with a high dynamic range, low noise detector. We conclude that this technique represents an important advance in so-called ‘at-wavelength’ X-ray optics characterization which does away with the need to directly probe the focus. Ptychography will therefore be useful in the testing and alignment of beamlines employing nanofocusing optics, and assessing the effect of optics on the X-ray wavefronts by propagating the beam along the focused beam path, making available detailed information about both metrology and fabrication.

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