Refractive Phase Plates for Aberration Correction and Wavefront Engineering

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The short wavelength of X-rays allows in principle the creation of focal spot sizes down to a few nanometers and below. At the same time, this short wavelength and the resulting interaction with matter puts stringent requirements on X-ray optics manufacturing and metrology. With the transition from third-generation synchrotron sources to diffraction-limited storage rings of the fourth generation, more beamlines will operate at higher spatial coherence. Thus, more instruments will work with smaller focal spot sizes that are increasingly dominated by diffraction effects instead of a demagnification of the X-ray source. Consequently, the requirements of X-ray optics will increase to ensure best beam characteristics via diffraction-limited optics. Simultaneously, X-ray optics manufacturing strives to achieve higher numerical apertures to provide ever decreasing beam sizes. On the forefront of this development are highly specialized nanofocusing beamlines with X-ray optics that push focusing toward 10 nm [1–4] and have the ambitious goal to reach 1 nm spot sizes [5]. The fabrication of X-ray optics requires the most advanced technologies, such as lithographic nano-fabrication for diffractive [6] and refractive optics [7], surface figuring with atomic precision for total reflection and multilayer mirrors [8], and thin-film technologies for multilayer optics [9]. All of these technologies have been developed over decades and further advances are expected in the future. Minuscule fluctuations or process anisotropies can cause shape deviations of the X-ray optic with a significant impact on focusing performance.

Refractive phase plates in combination with a focusing optic are one solution to overcome these technological limitations. While the weak interaction of hard X-rays with matter and the resulting refractive index decrement $\delta$ on the order of $10^{-6}$ pose a challenge for the fabrication of X-ray lenses [10], they allow for very precise control of the induced phase shift via thickness variations in a corrective optical element based on refraction. Even with a conservative shape accuracy of 1 μm for such an optical element, a correction of the wavefield error to $<0.05\lambda$ is possible. This exceeds both the Rayleigh criterion for the peak-to-valley wavefront error $<0.25\lambda$ and the Maréchal criterion for the RMS wavefront error $<0.07\lambda$ for a diffraction-limited optic. A key aspect for this correction scheme is an exact wavefield characterization upon which the design of the corrective element is based.
**Technical Report**

**At-wavelength aberration characterization**

For a successful correction of aberrations, it is crucial to measure the wavefront error with confidence and achieve both a high spatial resolution and phase sensitivity. Many techniques like speckle tracking [11–14] and various grating interferometry techniques [15–18] exist. They have low requirements on transverse coherence and are thus also suited for lab sources. In addition, some only require a single detector image, making them suited for fast online feedback and X-ray free electron laser (XFEL) application. However, these techniques often suffer from at least one of the following issues: a limitation in spatial resolution, problems with strongly divergent beams, or boundaries in the measurable magnitude of the phase gradient.

Here, we are focusing on another well-established method for wavefront characterization based on scanning coherent X-ray diffraction imaging, namely ptychography [19]. Due to its requirement of longitudinal and transverse coherence, the method is mainly applicable for synchrotron radiation sources of the third and fourth generation, but also successful XFEL nanobeam characterization has been reported [20]. Since the method reconstructs both the complex-valued transmission function of the sample and the complex-valued wavefield at the same time within a self-consistent and overdetermined iterative algorithm [21–23], the method is very robust and allows the reconstruction of confined X-ray beams, ranging from moderately focused beams to strongly divergent beams of any X-ray optics [24–30]. Thus, the method has become a reference standard for wavefront characterization in recent years.

To characterize the X-ray beam, a sample is positioned close to the focal plane and scanned transversely through the beam. Ideally, this sample is well-known, has features in any spatial direction to uniformly probe the wavefield, and scatters strongly to maximize the spatial resolution in real space, which directly depends on the magnitude of the scattering angle. A Siemens star pattern is ideally suited and often used. The sample is scanned in a raster fashion across the X-ray beam, ensuring a sufficient overlap between neighboring scan points [31]. Typically, a couple of hundred patterns are collected on a photon-counting pixel detector placed in the far-field. This analysis step is denoted by (1) in Figure 1. Together with the known scan positions and sample-to-detector distance, the diffraction patterns are fed into the iterative algorithm, which is available in several software packages today [32–36]. Depending on position accuracy and data quality, the algorithm typically converges within a couple of hundred iterations, taking only a few minutes on modern GPU systems. At the end of this iterative process, denoted as step (2) in Figure 1, the complex-valued wavefield at the sample plane is retrieved. In step (3), the wavefield is propagated with the Fresnel-Kirchhoff diffraction formula to a freely chosen plane in which the refractive phase plate shall be placed. A spherical term is fitted and subtracted from the phase of the propagated field in order to retrieve the phase error $\varphi_z$ of the wavefront. The thickness profile $z_{pp}$ of the phase plate in step (4) is designed to compensate the phase error $\varphi_z = k_0 z_{pp}(k) z_{pp}$, where $k$ is the wavenumber and $\delta_{pp}(k)$ is the refractive index decrement of the phase plate material. When the manufactured phase plate is placed at the chosen plane in step (5), the refraction in the phase plate material compensates the aberration induced by upstream optics.

**Fabrication techniques for refractive phase plates**

As refractive phase plates compensate aberration by directly reproducing them into a thickness profile, the requirements on manufacturing are strongly linked to the type of aberration that shall be corrected. Often, complex two-dimensional and three-dimensional structures need to be fabricated and precise shape control in the micrometer range can be a challenge. The first demonstration of refractive aberration correction in the X-ray regime was thus demonstrated by a two-dimensional polymer structure made with the LIGA process [37] to correct a one-dimensionally focusing X-ray mirror [38]. This lithographic process allows an accurate manufacturing of large-scale planar structures, especially suited to correct large-aperture mirror optics. The transition from two-dimensional phase plates to three-dimensional structures in

![Figure 1: Principle of wavefront characterization via ptychography and subsequent aberration correction with a refractive phase plate. The X-ray beam interacts with a focusing optic, where aberrations are introduced to the wavefront by shape inaccuracies of the optic. The aberrated beam is focused onto a strongly scattering characterization sample (e.g., a Siemens star), which is placed in the vicinity of the focal plane. (1) Several diffraction patterns from overlapping sample positions are recorded in the far field. (2) The ptychographic algorithm iteratively reconstructs the complex-valued wavefield at the sample position from the collection of diffraction patterns. (3) The wavefield can be propagated to a plane further upstream using the Fresnel-Kirchhoff diffraction formula. A phase error is retrieved by fitting and subtracting a spherical wave from the aberrated wavefront. (4) The wavefront error is converted into a thickness profile that cancels the error by refraction in the phase plate material. An optical element with this thickness profile is manufactured. (5) The refractive phase plate is mounted in the plane freely chosen in step (3). Aberrations are compensated for by the tailor-made optical element.](image-url)
order to correct rotational-symmetric refractive X-ray lenses (CRLs) focusing in two dimensions was made a few months later with a fused silica phase plate [39] made by laser ablation [40, 41]. Such a structure is depicted in Figure 2a. A challenge for X-ray optical elements that are structured via laser ablation is the high surface roughness visible in Figure 2a. However, as only one corrective element is typically needed, the induced small-angle scattering is often negligible. The ablation process can also be applied to diamond, a highly promising material for refractive X-ray optics due to its high density in combination with low atomic number, high thermal conductivity, and radiation hardness. Both X-ray lenses [42–44] and corrective phase plates [45] were manufactured. Recently, mechanical polishing of diamond lenses in order to significantly reduce the surface roughness was reported [46]. So far, the technique can only polish surfaces with a rotationally symmetric geometry and the polishing of more complex surfaces of phase plates strongly depends on the mechanical manufacturing capabilities for the polishing bit. Additive polymer printing via two-photon absorption in a photo resist [47] has enabled the manufacturing of smooth three-dimensional polymer structures with high spatial resolution. This technique is not only suitable to print refractive lenses [48, 49]. It is also well-suited for corrective phase plates with high aspect ratios for harder X-rays in the 20 keV to 40 keV range [45] or complex microstructures; e.g., for the aberration correction of diffractive multilayer Laue lenses (MLLs) [50]. Such a phase plate to correct a crossed pair of MLLs is shown in Figure 2b, where the complex inner part is the corrective shape and the outer rectangular structures are alignment markers and stabilizing elements.

Another possibility is a direct on-chip aberration correction of nano-focusing X-ray lenses (NFLs), which are typically made by lithographic processes in silicon [51, 52], silicon carbide [53], or diamond [54, 55]. Due to etching anisotropies, the parabolic lens shape is often not perfectly reproduced. As many tens to even hundreds of single lenses are stacked on a single lens row on these wafers, even small etching errors of only a few nanometers add up while the beam propagates through the thick lens stack. These errors can be compensated subsequently with an on-chip phase plate, as shown in Figure 2c made by FIB milling. Two of these wafers are typically combined in a crossed geometry to focus in two dimensions. Consequently, each of the two wafers needs to be compensated individually by such a structure [56]. Today, FIB processing also allows the fabrication of three-dimensional structures; e.g., for rotational-symmetric diamond micro lenses [57].

Aberration correction

Refractive phase plates are capable of correcting aberrations in all types of X-ray optics. So far, the correction of reflective mirrors [38, 58], refractive lenses [39, 45, 56, 59, 60], and diffractive multilayer optics [50] has been demonstrated. In contrast to adaptive correction optics relying on shape-morphing mirrors, the structure of a refractive phase plate is fixed upon manufacturing and the concept is based on a reproducible and well-characterized aberration pattern of the optic. In return, the instrumentation for phase plates is economical and space-saving. Figure 3 shows implementations of phase plates at the PETRAIII beamline P06 [61]. The phase plate is typically in the direct vicinity of the

![Figure 2: Refractive phase plates can be used with different focusing optics and fabricated by various manufacturing techniques. (a) A rotational-symmetric phase plate made by laser ablation into fused silica for the compensation of spherical aberrations in beryllium CRLs. (b) Complex three-dimensional polymer phase plate created by additive printing. The structure is created via two-photon polymerization in a photo resist and compensates the aberration in a crossed pair of MLLs. (c) The image shows the end of a row of one-dimensionally focusing parabolic NFLs etched into a silicon wafer. At the exit on the right side, a phase plate profile was created by FIB milling to compensate etching errors within the lens stack.](image-url)
focusing optic and requires a minimum of two transverse translation stages for lateral alignment. For phase plates that are not rotationally invariant, more degrees of freedom are required, as shown in Figure 3b, where the rotation along the optical axis is visible at the top of the image. Figure 3a shows a stack of beryllium CRLs that is corrected with a phase plate positioned downstream from the lens stack. In this configuration, the calculation of the phase plate can be achieved as described before and shown in Figure 1. However, as refractive lenses are chromatic and change their focal length $f \propto E^2$ with X-ray energy $E$, the longitudinal position of the phase plate is important and refractive lenses can be corrected over a larger energy range if the phase plate is shifted along the optical axis accordingly [59]. Positioning the phase plate upstream of the focusing lens can avoid a repositioning along the optical axis when the energy is varied. As both the refractive index decrement $\delta$ of the lens and the phase plate scale with $E^2$, good correction results over wide energy ranges can be obtained without reposi-
tioning. If the focusing element is optically thick, this is no longer the case. The aberration signature will slightly change with varying energy, as the beam path within the optical element is altered significantly. In general, the same degree of aberration correction with a shifting phase plate downstream of the lens [59] can be obtained as by a fixed phase plate upstream of the thick focusing lens. But the latter simplifies instrumen-
tation and avoids user interaction and realignment. However, a correct lateral alignment of the phase plate is crucial to match the features of the phase plate with the aberration. The required positioning accuracy strongly depends on the spatial variations of the aberration.

Changing aberrations are a challenge for refractive phase plates, as their shape is typically fixed during manufacturing and cannot be adapted later on. For aberrations that can be separated into one-
dimensional sinusoidal components, a team from Diamond Light Source (DLS) demonstrated an adaptive correction via two refractive elements with sinusoidal shape that can be shifted relative to each other [58]. This approach allows the control of both amplitude and phase for a single frequency. By switching to another structure with different period, the frequency can also be adapted. Combining multiple of these elements along the optical axis allows for the modeling of complex phase errors by the means of a Fourier series, which is a summation of sine and cosine functions. The method is especially suited for one-
dimensionally focusing optics in a single or crossed configuration like Kirkpatrick-Baez mirrors, NFLs, and MLLs.

An example of the aberration correction effect from refractive phase plates is shown in Figure 4. Here, a stack of 50 beryllium CRLs at a photon energy of 8.2 keV was corrected at beamline I13-1 of DLS [45]. Due to the mechanical manufacturing process by a rotationally symmetrical stamp via a coining process, each lens surface exhibits a small shape error in the range of 0.5 μm [39, 59]. This leads to significant spherical aberration for larger lens stacks in nanofocusing applica-
tions, shown by the horizontal beam caustic in Figure 4a. In the focal plane, shown in Figure 4b and highlighted by the dashed line in Figure
4a, the central speckle with a size of 69 nm FWHM is surrounded by ring-shaped side lobes. This leads to an increase of the point-spread function, which is critical for scanning microscopy applications and X-ray heating experiments at XFEL sources. The RMS wavefront error was measured to be 0.75 λ. With the help of a laser-ablated diamond phase plate, the RMS wavefront error improved to 0.25 λ, resulting in the horizontal beam caustic shown in Figure 4c. While a laser-ablated diamond phase plate shows a higher surface roughness than printed polymer phase plates, its material properties and radiation hardness are ideally suited for intense XFEL applications. The focal plane is presented in Figure 4d with a clean focal speckle of 76 nm, containing a high fraction of the total intensity. This is reflected by the Strehl ratio, a measure for the peak intensity in focus compared to an ideal optic. It improved from 0.1 to 0.7, slightly below the threshold at 0.8 for diffraction-limited focusing.

Wavefront engineering and beam shaping

The manufacturing techniques for refractive phase plates described earlier also enable other applications in the domain of X-ray wavefront control, where a refractive optical element is used to manipulate physical or geometrical properties of the wavefield. One widely used technique for visible light is the creation of light carrying orbital angular momentum (OAM) with a diverse field of applications [62]. First experiments in the X-ray regime were reported by researchers from Australia with a stepped vortex plate [63]. Later on, diffractive X-ray optics for OAM beams were also reported [64, 65]. Figure 5a shows the measured height map of a refractive vortex phase plate structured by laser ablation into fused silica with a smooth phase ramp and a sharp discontinuity [66]. The phase plate is positioned close to the focusing optic, either upstream or downstream, similarly to the corrective phase plate shown in Figure 3. When the beam propagates toward the focal plane, a donut-shaped intensity profile is created. Its diameter depends on the topological charge, which is equal to the phase shift induced by the height ramp in multiples of 2π. The resulting complex wavefield and intensity distribution in the focal plane is presented in Figure 5b and Figure 5c, respectively. The example shows that laser ablation in radiation hard materials can enable wavefield manipulation not only at synchrotron radiation facilities, but also at intense XFEL sources. Another alternative to create ring-shaped intensity profiles without OAM are axicon lenses [67], where the whole focusing lens is replaced by a series of parabolic axicons in analogy to the CRL concept or specifically shaped X-ray mirrors [68].

Depending on the individual experimental requirements, tailored made phase plates, as shown in Figure 5d, can also be manufactured. The structure shows four cones with a flat central part to create alternating phase ramps at the outer diameter of the X-ray beam intended for interference measurements. In the focal plane, this phase plate creates a crosshair-shaped beam, which has been characterized by ptychography. Its complex representation is shown in Figure 5e and the intensity distribution in Figure 5f.

Concluding remarks

Aberration correction by refractive phase plates has become a well-recognized technique to improve the performance of existing X-ray optical systems toward diffraction-limited operation. It has been employed at many facilities, including DLS, PETRAIII, ESRF, EuXFEL, LCLS, and PAL-FEL, where reflective, refractive, and diffractive focusing optics were successfully corrected for aberration. The field of wavefront engineering in the hard X-ray domain is still emerging, but current fabrication techniques for refractive phase plates offer a lot of potential. Similar considerations were made for beam shaping with diffractive optics [69]. One application that utilizes beam-shaping phase plates is multibeam ptychography, where different phase plates for each beam are used to create a set of diverse probes that are distinguishable by the iterative algorithm [70]. The creation of special intensity profiles in the focal plane—e.g., a homogeneous intensity distribution—could be another use case of interest.

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