Development of Hard X-ray Focusing Optics at Diamond Light Source

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Introduction
The short wavelength of X-rays makes them an excellent choice for probing materials on the nanometer scale and for crystallography of sub-micrometer crystallites. The objective of nanofocusing optics is to produce a small, focused beam size in order to obtain the highest X-ray flux on a small sample or as a fine spatial probe. Achieving nanometer-scale focused X-ray beam sizes puts great demands on the optical elements in an X-ray beamline—the optics must balance the requirements to de-magnify the electron beam X-ray source, to reduce the diffraction-limited focus size, and to minimize the contribution to the focus of aberrations in the optics while collecting the maximum X-ray flux into the focused beam. These requirements dictate that an extreme demagnifying geometry should be employed and that high-specification optical elements must be used. Nanofocusing optics has often been added as an upgrade to existing beamlines at Diamond, extending the range of science that can be carried out. Extreme nanofocusing also forms the basis of new beamlines at Diamond, such as the nanoprobe beamline (I14), which aims to provide sub-30-nm-dimension focused X-ray beams for mapping samples at high spatial resolution. The demand for nanometer-scale diffraction-limited X-ray beams is expected to grow at Diamond and requires corresponding advances in X-ray optics to exploit the present source and future lower emittance storage ring sources; for example, the proposed Diamond II upgrade, projected to give a factor 20 emittance reduction.

There is user demand for optics for routinely achieving these small spot sizes and also for the ability to change the focused beam size on a short timescale. Nanofocusing using refractive planar kinoform lenses and profiling of X-ray micro-focusing mirrors are two techniques being developed at Diamond Light Source for generating nanofocused beams and for the ability to change the focused beam size.

Development of Short Focal X-ray Lenses
Refractive lenses are conceptually simple optical systems for conditioning X-ray beams. Research and development in design and fabrication in the past 20 years has led to lenses being exploited successfully in many hard X-ray synchrotron beamlines. In the energy range of interest for the majority of beamlines at Diamond Light Source, low Z materials possess the best theoretical efficiency determined by the ratio of refractive power to X-ray absorption. Beryllium has successfully been used to make micro-focusing and long focal lenses, which are commercially available. The beryllium lenses are mature optics, with a performance very close to ideal. However, beryllium is a sintered material and its machining method is not likely to provide the very sharp radii required for nano-focusing. Single-digit nanometer focusing is theoretically possible with arrays of lenses with decreasing radii [1]; sharp focusing of order 50 nm using planar lenses is used on some beamlines [2]. The development of planar lenses for nano-beam experiments is less advanced than other devices, such as mirrors, zone plates, and multilayer Laue lenses. The main bottleneck in this development is that planar miniaturization techniques, which are satisfactory for industrial applications, do not achieve the desired aspect ratios to make high aperture lenses. A further limitation is that apertures are absorption limited.

Scattering from grain boundaries reduces flux and resolution in lenses made with polycrystalline materials. A technological breakthrough, like deep etching of diamond single crystals, would deliver a low absorption lens for X-ray nano-focusing. Such a lens would combine several advantages: in-line focusing; compact set-up; a theoretical superiority over mirror-based systems due to the large aperture without a shortened working distance.

Initially, we approached the problem of designing and fabricating X-ray lenses using single crystal germanium [3], silicon [4], and nano-and micro-crystalline diamond [5–7]. We demonstrated, for the first time, sharp focusing using diamond material. We achieved focusing down to about 200 nm FWHM with relatively large-aperture (50–100 μm), high-transmission refractive optics. Both silicon kinoform and diamond lenses fabricated at Diamond have the potential of achieving 50 nm focusing. A summary of the development of lenses in these two materials is presented in the following sections.

Lessons learned from fabrication challenges have led to designs of novel types of compound lenses whose efficiency is only limited by absorption and maximum aspect ratio [8]. These are the first aberration-free X-ray focusing lens designs available in the literature and are presented later in this article.

Silicon kinoform lenses
Single crystal silicon micro-machining is currently the method that delivers the most accurately fabricated lenses. We have achieved fabrication of low-absorption kinoform structures with aspect ratio as high as 50:1. The aspect ratio is defined as the ratio of lens height to lens minimum feature size normal to etch direction. The smallest feature sizes are 1–2 μm and the highest structures are close to 100 μm. We have experimentally tested these lenses and established that the effective aperture is above 50 μm with high transmission. The smallest focal...
spot measured was 225 nm FWHM (Figure 1) using a lens made in an older generation etcher, which showed significant side-wall structures known as scalloping. Later fabrication involved advanced e-beam lithography and the use of a fast cycle etcher, thus achieving curvature radii as small as several hundred nanometers. Such small radii provide single lens elements with focal lengths of order 50 mm in the energy range 8–20 keV, which are suitable for nano-focusing. According to an SEM study, the scalloping amplitude was below 20 nm and the lens walls reached a verticality of 89.9°. This figure cannot be easily improved, due to the nature of the etching processes normally used; i.e., Bosch cyclic etch or cryogenic etch. The conclusion of our development was that fabrication of a single element silicon lens with focal lengths down to 20 mm was at the limit of feasibility, and that arrays of lenses would have to be used to achieve such short focal lengths. This raised the problem of finding the ideal surfaces for the lens arrays, a theoretical problem which is easily solved.

Micro- and nano-crystalline diamond lenses

One of the most desirable materials for X-ray optics is single crystal diamond. Due to the well-known difficulties in sourcing and etching large single crystals of diamond [9], our development has focused on the deposition of micro- and nano-crystalline diamond (MCD and NCD, respectively) into silicon templates. The lenses were obtained by a final etch of the template away from the diamond. The aim was to achieve uniform deposition; i.e., complete filling of the silicon trenches. The highlight of this work is the achievement of perfect conformity of the material to the template. The SEM images in Figure 2 illustrate such accuracy. The superiority of diamond over silicon was easily inferred from the experimental data: both effective aperture and transmitted flux are twice as large with diamond [4, 7]. Scattering from the polycrystalline material has prevented a very sharp focus due to a large X-ray background at the focus, which also decreases efficiency. The X-ray flux transmitted by the 50 mm focal length lenses was of the order of...
30–50% in the central aperture of 50 μm. The focal spot measurements shown in Figure 2 (210 nm being the smallest spot size measured) indicate that the material drawbacks should not prevent obtaining sharper nano-focusing in the future. This could be achieved by a reduction of the amount of material in the lens using a kinoform design. This is a potentially important development for the NCD and MCD lenses, since one could obtain two important results at once: maximum lens transmission in the peak and minimum background. Finally, lens depth can be increased from the current 30–50 μm by longer deposition runs.

Prototype laser-milled single crystal diamond lenses, with sub-mm acceptance, are being fabricated by several groups [10, 11], but have yet not achieved micro-focusing. The availability of large diamond synthetic crystals, of high quality, is probably a few years in the future. This would drive the optimization of etch recipes capable of exploiting
physical and chemical processes for precise sculpting of diamond. In the short term, short-focal-length diamond lenses could be obtained by further optimization of the selective area deposition described in this article.

Aberration-free short focal X-ray lenses

A necessary condition for diffraction-limited focusing is the absence of optics aberrations. X-ray focusing lenses must be composed of a large number of refractive surfaces, due to the small refractive index decrement of materials. By applying Fermat’s principle, it is easily found that aberration-free refractive surfaces are of elliptical, hyperbolic, or Cartesian oval form only [12, 13].

Geometrical aberrations increase with lens apertures and surface curvature. Since a high aperture is the main requirement for an effective nano-focusing lens, we have developed a novel design in which an array of conical surfaces is capable of focusing the incoming beam to diffraction-limited size with zero aberrations [8].

The remaining challenge in nano-focusing lens design is maximizing the aperture while keeping the overall lens length well below the focal length. Possible compound lens designs are shown in Figure 3. These lenses can focus the synchrotron hard X-ray radiation to 20–50 nm FWHM, depending on energy (lens radii and absorption-limited apertures are energy-dependent). The proposed designs are easily applied to even shorter focal length lenses to approach the 10 nm focal spot; progress in the fabrication of such short focal lenses is currently hampered by the unavailability of a nano-lithography and nano-fabrication method for the material of choice, which is single crystal diamond.

Varying the Focused Beam Size

Micro-focusing optics is used with a range of techniques at Diamond Light Source, including diffraction, spectroscopy, and microprobe experiments. In the field of macromolecular crystallography, the biological samples can often only be obtained as small (sub-micrometer dimensions) crystals, which are frequently crystallographically imperfect and weakly diffracting. For high throughput applications, the beamline user requirement is for the ability to rapidly measure a series of crystals of differing sizes in an automated fashion. While demagnifying optics is able to achieve the sub-micrometer focal spot size required for the smaller crystals, for larger crystals the small focused beam size increases sample radiation damage, resulting in a degradation of the quality of the crystallographic data obtained. In such cases, it is highly beneficial to be able to use a beam size that is matched to the sample size.

For X-ray microprobe experiments, the smallest focal spot gives the best spatial resolution, allowing, for example, the distribution of particular chemical elements to be mapped at the highest resolution; however, the field of view becomes very small. A variable probe size would allow the spatial resolution and field of view to be varied, facilitating, for example, mapping of a large area before finer mapping of areas of interest. The ability to change the focal spot size at the sample would give increased flexibility in many experiments, allowing the beam to be matched to the sample size, allowing the beam to be used to study sample inhomogeneity, and reducing the sample radiation dose. Typical experiments require that the beam size be switched on a timescale of minutes; however, the demands of high throughput data collection for macromolecular crystallography make it desirable that the beam should be changeable from about 0.3 to 20 μm in a time scale of below one second. The focused beam size may be varied using multiple optical elements to generate an intermediate source, but for X-rays this results in a significantly greater complexity in optical alignment and results in a loss of X-ray flux at the sample. We have investigated the use of deformable mirrors and deliberate surface profiling of mirrors to give the capability of reproducibly changing the focused beam size without introducing a complicated optical arrangements and without sacrificing X-ray intensity.

Super-polished bimorph mirrors

A novel super-polished adaptive bimorph mirror (Figure 4) has been developed that provides variable focal distance and local figure control in the sub-mm range [14]. This has been achieved by bringing two state-of-the-art technologies together: super-polishing using Elastic Emission Machining (EEM) from JTEC (Osaka) [17] of an eight-channel,
piezo bimorph mirror from SESO (France). In-situ characterization with synchrotron radiation on B16, Diamond’s Test beamline [16], and optimization using at-wavelength metrology demonstrates the optics’ ability to provide distortion-free beams of variable size at variable focal distance. Figure 5 shows that an initially focused beam size of ~0.5 µm can be defocused in a controlled way to 25 µm with negligible structures in the beam profile, and even up to 50 µm with acceptable structures. The ripple on the flat top remains below 10%, even at a defocusing that increases the beam size 20-fold. This is made possible by the EEM mirror’s extremely small figure error (<1 nm rms), especially at mid-range spatial frequencies (10 µm to 1 mm in lateral size). This mirror compares extremely favorably to a typical X-ray focusing mirror, which produces unacceptable structures (50% or more ripple) in the beam intensity distribution if defocused to enlarge the focal spot by only two or three times. As far as we know, this is the first adjustable elliptical mirror with sub-nanometer figure errors. The results of this development have recently been applied to user experiments at the I24 MX micro-focus beamline, which has recently replaced the micro-focusing mirrors with super-polished ones.

Multi-lane mirrors

The EEM bimorph mirror allows fine control of the focused beam size and is easy to implement as an upgrade of existing optics systems. The time taken to change between different beam sizes depends on the relaxation time of the mirror piezo bending elements, which is at best of an order of a few minutes. As mentioned earlier, many applications require faster switching of the beam. For these experiments, we have developed a novel mirror design in which the basic elliptical mirror surface height profile is modified by a function calculated to broaden the focused beam profile by a known amount. A sequence of different beam sizes can then be obtained by fabricating the mirror with different surface modifications applied in parallel lanes running the length of the mirror. Switching to a different modification function is then simply a matter of translating the mirror laterally, moving the X-ray beam into a different lane, and this can be potentially achieved in less than one second. In order to reduce the amplitude of the mirror surface modifications required, the use of a periodic undulating surface profile was investigated [17]. By varying the wavelength of the surface modulation, it was shown, using physical optics simulations, that with short modulation wavelengths the modification acts like a diffraction grating, resulting in higher-order peaks separated from the main focus. With longer modulation wavelengths, physical and geometric optics simulation results converge and the geometric optics can be used to predict the focus size. A sine wave surface modulation function was modelled, but shows concentration of intensity at the edges of the focus. This indicates the appearance of caustic structures, familiar from visible light optics, caused by the wavefront folding back on itself in the image plane. The caustic peaks are the consequence of the second derivative of the sine wave passing continuously through zero for rays at the edges of the focal spot. A better surface modulation function consists of alter-
We designed a prototype mirror with three lanes which was then fabricated by JTEC (Osaka) [15]. The mirror had an active length of 90 mm, a focal length of 0.4 m, a glancing incidence angle of 3 mrad, and the reflecting surface was coated with rhodium [18].

On delivery, the mirror was measured by Diamond-NOM [19] and it was then tested on Diamond’s Test beamline [16]. The mirror surface profile measured by NOM was used to model the focused beam profile using wavefront propagation. Figure 6 shows the elliptical profile of lane 1 with the modifications that were applied in lanes 2 and 3, while the measured focused beam profiles are shown in Figure 7.

The profile for lane 1 shows good agreement with the modelled profile with an electron beam source size added as a Gaussian distribution, rms width 20 μm. The surface profile in lane 2 had a modulation of 20 nm peak to peak and, in lane 3, 100 nm peak to peak which led to focal...
Spot sizes of $\Delta = 0.5, 2.2,$ and $11.3 \, \mu m$. This prototype mirror demonstrated the potential use of this new type of optics but allowed only two modulation periods along the length of the mirror. A longer mirror could have a larger number of parabolic arcs and simulations, indicating that, by subtly varying the surface profile, the interference fringes between different waves can be made to partially cancel out, resulting in smoother focus profile.

**Conclusion**

There is a clear requirement on many beamlines at Diamond Light Source for high-intensity nano-focused X-ray beams for a range of experiments. With increasingly sophisticated experiments and potential improvements to the X-ray source characteristics, there is an imperative for parallel developments in nanofocusing X-ray optics.

Refractive lenses are operating well on Diamond beamlines, showing excellent beam stability and simple alignment. Improvements in the lens micro-fabrication process will include multi-step approaches to silicon etching in order to improve the aspect ratio and decrease the aberrations caused by non-vertical etch. These developments, together with expected increased brilliance of future X-ray sources, will allow the use of lenses in experiments that are currently flux limited.

It has also become apparent that the ability to reproducibly change the beam size at the sample enhances the range of measurements that can be made, whether for micrometer-scale crystallography or high-spatial-resolution X-ray probe experiments. The developments of new types of mirrors for changing the X-ray beam size at the sample are expected to have a significant impact on the quality of science produced on X-ray beamlines through the ability to change X-ray probe size and to rapidly change the beam size in order to reduce sample radiation damage.

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**Note**

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