Upscaling of multi-beam x-ray ptychography for efficient x-ray microscopy with high resolution and large field of view

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ABSTRACT

Nondestructive imaging with both a large field of view and a high spatial resolution is crucial to understand complex materials and processes in science and technology. X-ray ptychography can provide highest spatial resolution but is limited in the field of view by the acquisition time and coherent flux at modern x-ray sources. By multi-beam ptychography, the sample can be imaged in parallel by several spatially separated and mutually incoherent beams. We have implemented this method using 3D nanoprinted x-ray optics to create tailor-made x-ray multi-beam arrays. The use of 3D printing allows us to create focusing optics with a minimum of nonfunctional support structures. In this way, large sample areas can be efficiently scanned in parallel with up to six illuminating beams.

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X-ray microscopy is an indispensable tool in many scientific fields due to its ability to image chemical and physical processes under realistic conditions. A key technique for high-resolution x-ray microscopy is scanning coherent diffractive imaging, also known as ptychography.¹ It allows us to solve various scientific questions concerning, e.g., biology,² catalysis,³ or nanotechnology.⁴ Many of these questions involve searching for a needle in a haystack, i.e., finding a small detail in a large object, for example, finding a local defect in a microchip. Searching for a small detail requires the imaging of a large field of view with high resolution. However, for scanning techniques such as ptychography, scanning large areas with high resolution requires long scan times. Demonstration experiments have been able to image on the order of 100 mega pixels per hour at a resolution of 40 nm.⁵ To push this to the level of imaging tera pixels at a resolution of 10 nm, as demanded, e.g., by the RAVEN program of IARPA,⁶ would increase the scan time by years.⁷ Here, we propose a multi-beam scanning approach that will significantly speed-up scan times. As conventional ptychography requires a coherent beam, the scanning time is ultimately limited by the available coherent photon flux. Multi-beam

ptychography can accept a larger incoherent part of the incoming beam, allowing us to scan large sample areas in parallel.

In multi-beam ptychography, multiple independent beams simultaneously scan different parts of the sample, allowing rapid scanning of large samples without sacrificing spatial resolution.⁸ This can be achieved because each beam only needs to be coherent with itself and can be incoherent to all other beams. Multiple groups have demonstrated multi-beam x-ray ptychography using pinholes⁹ or Fresnel zone plates¹⁰ to create the beams. However, these schemes leave large parts of the incoming beam unused, as both the pinholes and the zone plates require a significant dead area between the individual optics.

In this Letter, we report the use of additive 3D nanoprinting¹¹ to manufacture multi-beam x-ray optics. Taking advantage of this technology,^{12,13} we have fabricated lens towers that achieve sub-micron focusing of hard x-rays.¹⁴ Owing to the geometrical freedom of 3D printing, we can create multiple beams by printing multiple lens towers in proximity to each other. In the current work, we demonstrate a flexible multi-beam setup for x-rays that allows us to tailor the beam

configuration and beam separation and is ideally suited to scan large areas quickly.

We performed the experiment at the microprobe end-station of beamline P06¹⁵ of PETRA III at DESY in Hamburg at a photon energy of 7 keV. The experimental setup is illustrated in Fig. 1(a). We used an array of 3D-printed refractive lenses¹⁴ to create multiple parallel beams. Two lenses are sketched in Fig. 1(b), one intact and the other cut open to show the interior. A single lens has a square footprint of $45\ \mu\text{m} \times 45\ \mu\text{m}$ and a length of $600\ \mu\text{m}$ and contains four parabolic surfaces with a curvature of $2.5\ \mu\text{m}$ to focus x-rays. The refractive index decrement δ of the printed polymer at 7 keV is 5.54×10^{-6} . Each lens, thus, has a focal distance of 113 mm (at 7 keV) and a diffraction-limited spot size of 335 nm. The lenses are placed in a $47\ \mu\text{m}$ raster to avoid contact between neighboring lenses and to minimize manufacturing errors. This $2\ \mu\text{m}$ gap between the lenses leaves up to 90% ($=45^2\ \mu\text{m}^2/47^2\ \mu\text{m}^2$) of the incoming photon beam for measurements.

Using the slits in front of the lens array, we selectively illuminate lenses to create different multi-beam configurations. Background scattering from the optics is reduced by placing a tailor-made pinhole array between the lenses and the sample. This array of $30\ \mu\text{m}$ pinholes is made by laser ablation from a $100\ \mu\text{m}$ thick platinum foil. Some of the different possible beam arrangements are shown in Fig. 1(c). This

FIG. 1. (a) Experimental setup with the lens array (A) focusing multiple beams to a plane 20 mm upstream of the sample (C). Six of the beams are selected by the pinhole array (B) and scatter off the sample. The resulting diffraction patterns all overlap on the detector (D). (b) Not-to-scale sketches of the lens design, the top one is cut open to show the four parabolic surfaces. Each lens has a length L of $600\ \mu\text{m}$ and a footprint F of $45\ \mu\text{m} \times 45\ \mu\text{m}$. The openings are necessary to drain the unused precursor after the printing. (c) Five exemplary beam configurations; the pinhole array in combination with the upstream slits allows us to create one, two, three, four, or six beams.

particular arrangement of pinholes was chosen for this first experiment to test different beam separations of $47\ \mu\text{m}$, $94\ \mu\text{m}$, and combinations of both. A three by three beam array can be implemented for large-field-of-view imaging in future applications.

The sample is placed 20 mm behind the focal plane of the lenses. At the sample position, each beam has a diameter of $7.5\ \mu\text{m}$. As test samples we chose two NTT-AT (XRESO-50HC) Siemens star resolution charts that feature 50 nm structures. We use one test chart in the two-beam measurement and the other one for the six-beam case. After the beam is scattered off the sample, the diffraction patterns are detected using a single-photon counting, custom made in-vacuum Eiger 4M detector (pixel size $75\ \mu\text{m}$).¹⁶ The detector is mounted at the end of the evacuated flight tube in order to eliminate absorption and scattering in air.¹⁷ The distance z from the sample to the detector is 8050 mm.

In the first measurement, the most simple multi-beam setup is used comprising two beams. We select two vertically neighboring lenses [pinhole 1 and 2 in Fig. 1(c)], to create two beams with a separation of $47\ \mu\text{m}$. The test sample is scanned in a grid of $16 \times 31 = 496$ positions in $2\ \mu\text{m}$ steps with 1 s exposure per point. To avoid scan grid artifacts,¹⁸ each point is randomly offset horizontally and vertically up to $\pm 0.25\ \mu\text{m}$. The scan areas of the two beams overlap on a $30\ \mu\text{m} \times 13\ \mu\text{m}$ ($h \times v$) wide area.

We reconstruct the two-beam data set twice, first with single beam algorithms and then a second time with multi-beam algorithms. For both reconstructions, the Eiger images are cropped to 512×512 pixels, resulting in a reconstructed pixel size of $37.1\ \text{nm}$. For the single beam reconstruction, we use a combination of 500 iterations of the standard ePIE algorithm and 500 iterations of a parallel update, using the coupled object and probe equations described by Thibault *et al.*¹⁹ In contrast to ePIE, this update applies the changes from all diffraction patterns in parallel. The object and probe updates are, respectively,

$$O_{l+1}(\mathbf{r}) = O_l(\mathbf{r}) + \alpha \cdot \frac{\sum_j P_l^*(\mathbf{r} - \mathbf{r}_j) \phi_j(\mathbf{r})}{\sum_j |P_l(\mathbf{r} - \mathbf{r}_j)|^2 + \epsilon}, \quad (1)$$

$$P_{l+1}(\mathbf{r}) = P_l(\mathbf{r}) + \beta \cdot \frac{\sum_j O_l^*(\mathbf{r} + \mathbf{r}_j) \phi_j(\mathbf{r} + \mathbf{r}_j)}{\sum_j |O_l(\mathbf{r} + \mathbf{r}_j)|^2 + \epsilon}. \quad (2)$$

Here, ϵ is a small regularization constant ($\sim 10^{-3}$) and $\phi_j(\mathbf{r}) = \psi'_j(\mathbf{r}) - \psi_j(\mathbf{r})$ is the difference between the updated and the previous exit wavefront.

As the sample scanning stage did not reach the dialed positions with the necessary precision, a brute-force position refinement was used to improve the reconstruction.²⁰ The refined positions are used for all the following reconstructions of this scan.

In this single beam reconstruction, the reconstructed probe is a combination of the two beams. Similarly, the reconstructed object, shown in Fig. 2(b), is a combination of the two scanned regions. These combinations result in some features appearing twice in the reconstruction. Measuring the distance between the two copies of the same feature provides a more accurate estimate of the distance between the two beams. Here, the center of the Siemens star repeats after $45.5\ \mu\text{m}$. This slight deviation from the design value of $47\ \mu\text{m}$ can be caused by distortions in the wave front or manufacturing errors in the optics.

For the multi-beam reconstruction, we use a modified version of the incoherent multiplex algorithm from the study by Batey *et al.*²¹

FIG. 2. Reconstructions of the two-beam measurement. (a) and (c) use the multi-beam algorithm, and (b) uses the single beam ePIE algorithm. (a) shows the two probes in a complex colorscale. (b) shows the phase shift of the Siemens star reconstructed with ePIE. (c) shows the multi-beam reconstruction of the Siemens star, with the green rectangle indicating the area scanned by the first probe and the blue rectangle indicating the area scanned by the second probe. The zoom-in shows an enlarged view of the center of the Siemens star. All white scale bars represent $10\ \mu\text{m}$. The Siemens stars in (b) and (c) share the same colorscale.

This algorithm was designed to scan a sample at multiple wavelengths in a single measurement. Here, all probes have the same wavelength but scan different parts of the sample. To this end, each beam has a different optical axis and beam center on the detector. By initializing each beam in the sample plane with a suitable phase wedge factor, their footprint on the detector can be shifted individually. Adapting the multiplex algorithm for multi-beam ptychography is straightforward, and deriving a corresponding parallel multi-beam update is similarly possible.

With the modified multiplex algorithm, the two-beam data set is reconstructed for 500 iterations, followed by a further 500 iterations using the multi-beam parallel update.

The reconstructed probes are shown in Fig. 2(a). Although the nano-printing process is highly repeatable, the two probes are not

identical and show small differences. With the separation of the probes, the reconstruction is able to separate the two scan areas and reconstruct the full length of the Siemens star [see Fig. 2(c)]. In the central region, which was illuminated by both beams during the scan, very weak artifacts are visible. Outside this shared band, where only one of the two beams illuminated the sample, the artifacts are stronger, most prominently at the top and bottom of the reconstruction. These ghost artifacts arise because the two probes are partially coherent and interfere on the detector, creating interference stripes in the diffraction patterns. The stripes can be suppressed to make the beams effectively incoherent by choosing a beam separation that is an integer multiple of

$$D = \lambda z/p, \quad (3)$$

where λ is the wavelength of the x-rays, z is the distance between the sample and the detector plane, and p is the detector pixel size. Here, the characteristic beam separation D is $19\ \mu\text{m}$, which means that the nominal beam separation of $47\ \mu\text{m}$ corresponds to $2.5D$, halfway between the optimal values of $2D$ or $3D$. This suboptimal distance implies that the interference is only partially suppressed, here to about 15% of its maximum value. Correspondingly, the reconstruction algorithm cannot fully separate the two beams, giving rise to the aforementioned artifacts in the reconstructed sample. This problem is amplified by the similarity of the probes, which adds difficulty to the separation.

To estimate the resolution, we split the data set into half and reconstruct both halves separately. We align the two reconstructions²² and calculate the resolution with the Fourier ring correlation (FRC) and the half-bit criterion to $47\ \text{nm}$. The finest details of the Siemens star, the $50\ \text{nm}$ spikes in the center, are faithfully reconstructed, matching the FRC result.

Scanning this object with the same fluence using a single beam would have taken 1.8 times longer than this two-beam scan. The improvement is smaller than the number of beams due to the shared band that was scanned by both beams. Using three lenses to create three beams, an even larger area could be scanned in the same time. The gain in this case compared to a single beam would be 2.6.

Even greater gains are possible with more parallel beams. To this end, the slits in front of the optics were opened to fully illuminate the lens array, creating six parallel beams behind the pinhole array as shown in Fig. 1(c) on the far right. The nominal separation between the outermost beams is $94\ \mu\text{m}$, twice the distance between two individual lenses. In a second ptychographic experiment, a damaged Siemens star was scanned across the beams in a raster of 120×120 steps of $1\ \mu\text{m}$, so that together, the multi-beams scan an area of $(120\ \mu\text{m} + 94\ \mu\text{m})^2$. Each scan point was randomly offset in the horizontal and vertical direction up to $\pm 0.25\ \mu\text{m}$, and the detector was exposed each time for 0.3 s. As the scan range is larger than the maximum beam separation, the individual scan areas of each beam overlap in the center of the Siemens star.

For the reconstruction, we combine 1000 iterations of the multi-beam adapted sPIE algorithm^{23,24} with a 2×2 upsampling and then the corresponding parallel update for a further 1000 iterations. The use of sPIE allows us to reconstruct a larger field of view for each probe [compare Figs. 2(a) and 3(a6)]. As reconstructing six beams is more challenging than one or two beams, the weak aberrations of the lenses have more influence on the reconstruction quality and make enlarging the field of view necessary. We also use a brute-force position refinement. After the two-beam scan and similar measurements, all showed

FIG. 3. Reconstruction with 6 beams. (a1)–(a6) The six reconstructed probes, with the arrangement according to the corresponding pinholes. The white scale bars represent $5\ \mu\text{m}$. Only (a6) shows the full probe, (a1)–(a5) show only the central part. Their field of view is indicated by the blue dashed square in (a6). (b) The reconstructed phase shift of the Siemens star. The area indicated by the blue rectangle is shown enlarged in Fig. 4. The black scale bar represents $50\ \mu\text{m}$.

deviations to smaller beam separations, centered around $46\ \mu\text{m}$, and we reconstruct the probes with this separation (accordingly $92\ \mu\text{m}$ for the isolated sixth beam). Consequently, the area that was scanned by the beams shrinks slightly to $212\ \mu\text{m} \times 212\ \mu\text{m}$. The six reconstructed beams are shown in Figs. 3(a1)–3(a6) and are arranged in the same order as they illuminate the sample [see Fig. 1(c)]. The reconstructed object is shown in Fig. 3(b) with the Siemens star in the center, structures of lines and spaces above and below it, and squares of different sizes to the left and right. In a previous experiment, the tantalum

FIG. 4. Comparison of the damaged $500\ \text{nm}$ lines and spaces on the Siemens star sample: (a) the six-beam reconstruction and (b) SEM image of the same area.

structures on the sample were in parts destroyed, creating the irregular features. The most damaged area is on the top right, indicated by the blue square in Fig. 3(b). This detail is shown enlarged in Fig. 4, illustrating the reconstruction quality.

Scanning this area with the same fluence using just a single beam would have taken more than thrice as long. Ideally, the six beams could achieve a speedup as high as six. Here, the overlap between the individual scan areas reduces the speedup. Scans with a smaller overlap are difficult to reconstruct because the high similarity between the six beams makes it difficult for the reconstruction to correctly separate the individual contributions of each beam. With little or no overlap, the reconstruction algorithm has no direct way to discern, which beam creates a certain diffraction feature. To achieve the full speedup in future experiments, the beams should be individualized.

In conclusion, we have demonstrated how 3D nanoprinting offers a more adaptable way to construct x-ray multi-beam optics. By using this technology, we could create a dedicated multi-beam setup with a flexible beam arrangement. As refractive x-ray lenses can be placed closer to each other than previous multi-beam optics, they are better suited to utilize more of the incoming photon flux. Using six beams with a maximum beam separation of $92\ \mu\text{m}$, we could image an area of $212\ \mu\text{m} \times 212\ \mu\text{m}$, more than three times larger than the mechanical scan area of $120\ \mu\text{m} \times 120\ \mu\text{m}$. It is still open how high the number of simultaneous beams can be pushed, leaving the potential for even higher scan speedups. This would allow us to image samples of millimeter size for needle-in-the-hay-stack problems in a useful time span. Potential limiting criteria can be the size of the flat beam, the lifetime of the lenses,²⁵ or the count rate of the detector.

Currently, the high similarity between the different beams is limiting the speedup and has proven to be a challenge for the reconstruction. Due to the highly reproducible printing process, the lens towers and their respective probes are essentially clones, and the only differences are introduced due to small manufacturing errors. Disentangling the diffraction from many nearly identical beams in the reconstruction requires high iteration numbers. To allow larger beam numbers in the future, this can be prevented by making the probes more unique, ideally even mutually orthogonal, for example, by adding a particular orbital angular momentum to each beam.²⁶

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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