optica

Hard X-ray multi-projection imaging for single-shot approaches: supplementary material

P. Villanueva-Perez[1,2,*], B. Pedrini[1], R. Mokso[1,3], P. Vagovic[2], V. A. Guzenko[1], S. J. Leake[4], P. R. Willmott[1], P. Oberta[5], C. David[1], H. N. Chapman[2,6,7], and M. Stampanoni[1,8]

[1] Photon Science Division, Paul Scherrer Institut, 5232 Villigen, Switzerland
[2] Center for Free-electron Laser Science (CFEL), DESY, Notkestrasse 85, 22607 Hamburg, Germany
[3] Max IV Laboratory, Lund University, SE-221 00 Lund, Sweden
[4] ESRF, The European Synchrotron, 71 Avenue des Martyrs, 38000 Grenoble, France
[5] Institute of Physics, Academy of Sciences of the Czech Republic v.v.i., 818221 Praha, Czech Republic
[6] Department of Physics, University of Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany
[7] Centre for Ultrafast Imaging, Luruper Chaussee 149, 22761 Hamburg, Germany
[8] Institute for Biomedical Engineering, UZH/ETH Zürich, 8093 Zürich, Switzerland
[*] Corresponding author: pablo.villanueva-perez@cfel.de

Published 29 November 2018

This document provides supplementary information to "Hard X-ray multi-projection imaging for single-shot approaches," https://doi.org/10.1364/optica.5.001521. We further elaborate on the beam splitter concept introduced in the main text. We also discuss in detail the data analysis and the near-field and far-field experiments.

1. CRYSTAL BEAM SPLITTERS

We propose the use of face-centered-cubic crystals as beam splitters due to their high degree of symmetry, i.e. they can simultaneously generate multiple deflected beams. Specifically, we focus on diamond and silicon crystals. Diamond is a good candidate for X-ray free-electron laser optics due to its low X-ray absorption, high damage threshold, and good heat conductivity. On the other hand, silicon crystals can be produced with high purity, low strain, and no defects. Their low cost means that they can be considered consumable during high-flux experiments at a free-electron lasers, whereas diamond crystals are expected to survive those experiments [1]. Both crystals are commercially available with subnanometer roughness and submicron scale thickness control being optimal for X-ray multi-projection imaging. Table S1 reports the deflection angles for a few allowed silicon and diamond reflection families whose corresponding wavelengths are in the hard X-ray regime and with practical deflection angles. The energy to set a family of planes in Bragg condition is given by

$$E = \frac{hc}{2d\sin\theta}, \tag{S1}$$

where E is the energy, h is the Planck constant, and c is the speed of light in vacuum. The alignment of silicon and diamond crystals to generate the multiple beams will be performed below the damage threshold [1, 2]. The damage threshold for biological or other radiosensitive samples will also be taken into account [3]. This alignment procedure will be crucial for the experiments performed in diffraction-before-destruction conditions [4] at X-ray free-electron lasers.

2. SIMULTANEOUS ILLUMINATION CONSTRAINTS

In order to image the sample simultaneously by all the generated beams, two conditions have to be satisfied. First, we impose a geometric condition, which requires that all the beams fully illuminate the sample. Therefore, the sample has to be positioned downstream the crystal at a position $L \leq L_g$, where the maximum geometrically allowed distance L_g for a thin crystal is constrained by the beam diameter S, the maximum transverse dimension of the sample t, and the deflection angle 2θ:

$$Lg \leq \frac{1}{sin(2\theta)}\left(\frac{S\cos(2\theta) - t}{2}\right). \tag{S2}$$

Refl.	Symmetry direction	Reflection multiplicity	E (keV)	Deflection angle (2θ)
Si(131)	(001)	8	12.56	35.1°
Si(331)	(001)	8	13.70	48.2°
C($\bar{3}$11)	(011)	6	13.52	50.5°
C(1$\bar{1}\bar{1}$)	(111)	3	9.03	38.9°
C($\bar{1}$1$\bar{3}$)	(111)	6	11.04	63.0°

Table S1. Bragg-reflection families of diamond and silicon cubic crystal structures suitable for multi-beam generation in the photon energy range (E) between 2 and 14 keV, with a deflection angle between 20° to 65°.

Second, the direct and the deflected beams have different optical paths, thus they do not illuminate exactly at the same time the sample. Thus given a maximum tolerable time delay (Δt) which ensures the immutability of the sample, the maximum tolerable distance between the crystal and the sample is given by

$$L_t = \frac{c\Delta t}{\frac{1}{cos(2\theta)} - 1}\,, \tag{S3}$$

where c is the speed of light. At XFELs, the maximum tolerable time delay between deflected and the direct beam is constrained by time interval between the arrival of the imaging pulse and the observation of radiation damage. For example, for a biological sample like a Lysozyme crystal imaged by 3×10^{12} photons at 12 keV focused on a 100×100 nm^2 the aforementioned interval is below 10 fs [4] and the maximum distance is of the order of 10 μm. Finally, the sample to crystal distance L is chosen to be smaller than the minimum of the geometrical L_g and temporal L_t constraints. To ensure the overlap of the different beams at the sample position during actual experiments, the beam diameter on the crystal will be adjusted accordingly. For micrometer and submicrometer samples in order to facilitate the overlap a beam-recombiner setup will be designed as described in Ref. [5].

3. PROPAGATION-BASED PHASE CONTRAST

Phase-contrast imaging techniques exploit the phase change of the exit wavefront after transmitting through a sample rather than the change of transmission due to absorption. Such techniques are specially useful to distinguish between two materials with similar transmission or transparent materials to the probing radiation. The high penetration power of hard X-rays, specially for low-Z materials, makes them a perfect radiation type to exploit phase-contrast techniques. In the context of this work, we exploit propagation-based phase contrast, i.e. we use the free-space propagation to observe the phase change of the exit wavefront as intensity variations in the recorded images [6]. The phase information is retrieved from the intensity variations using phase-retrieval algorithms. The main solutions to this inversion problem in the near-field regime linearize either the transmissivity of the sample as exploited by the contrast-transfer function approach (CTF) [7] or the propagator as transport-of-intensity equations (TIE) [8] do. As the presented images are acquired at propagation distances of the order of the depth of focus of the used microscope, we can exploit TIE algorithms, such as that presented in Ref. [9].

4. TOMCAT SETUP AND DATA COLLECTION

The phase-contrast near-field experiments were performed at the TOMCAT beamline of the Swiss Light Source. The X-rays provided by a bending magnet source were monochromatized by a multilayer monochromator to 12.6 keV with approximately a 2 % bandwidth. The natural divergence of the bending magnet X-ray beam at TOMCAT is about 2 mrad in the horizontal and 0.6 mrad in the vertical direction, which is larger than the Darwin width of the used crystal. A 100 μm thick Si(001) crystal was illuminated by a 10×6 mm^2 beam after conforming it with three sets of slits. The crystal was mounted on a triple-axis goniometer to generate simultaneously several reflections. Behind the crystal a moth was positioned to be illuminated by the different generated beams. At 10 cm from the moth the detector was positioned to record the different phase-contrast images. For each of the images the detector was translated in a plane perpendicular to the incoming beam. The detector was an X-ray 1:1 (Optique Peter) microscope with a high efficiency scintillator, which converts X-rays to optical photons. The camera used was a pco.edge 4.2 CMOS detector with a pixel size of 6.5 μm and 2048×2048 pixels. Each of the moth projections was acquired with 4×10^{11} ph/mm^2 on the crystal beam splitter [10]. Around 63 % of the fluence on the crystal beam splitter contributed to form the direct-beam image, while the contribution to the silicon (111) and (131) images was of 4 % and 2 %, respectively.

5. COHERENT DIFFRACTION IMAGING

Coherent diffraction imaging (CDI) [11] is a lensless technique. The sample is illuminated by plane waves, and the diffraction patterns are recorded in the far-field regime. CDI provides a reconstruction of the complex X-ray transmissivity of the sample by means of phase-retrieval procedures based on iterative transform algorithms [12, 13]. The achievable resolution is given by the largest diffraction angle at which the intensity of the diffraction pattern exhibits sufficient signal-to-noise in order to reliably reconstruct the phase [6], so that the ultimate resolution limit is set by the wavelength of the incident radiation. The reconstructions presented in Fig. 3(c), (d), and (e) are obtained after averaging 20 reconstructions, where each of them was obtained after 2800 iterations of shrink-wrap algorithm [14] and 1200 iteration of hybrid input-output [13] with $\beta = 0.9$ combined with an error reduction algorithm [12].

6. ID01 SETUP AND DATA COLLECTION

The CDI experiments were performed at the ID01 beamline at ESRF. The beam was monochromatized by a double crystal monochromator at 12.56 keV with a $\sim 4 \times 10^{-4}$ bandwidth as required to efficiently generate eight beams by a silicon crystal (Table S1). The coherent portion of the beam was focused by compound refractive lenses to a focal spot of 1 μm^2 on the sample. The sample was composed of a 100 μm thick Si(001) crystal

and 500 nm gold nanostructures attached to it. The sample was mounted on a hexapod stage capable to precisely orient the sample to generate the eight deflected beams. The intensity of the deflected beams was 10 % less intense than the transmitted beam. Once the crystal was aligned, a SmarAct 3D piezo system was used to position precisely the gold nanostructures to be illuminated by the deflected beams. As the beam was tightly focused around the sample and the sample was not exactly positioned behind the crystal, we translated around 50 μm the sample from the position in the direct beam to the position in each of the diffracted beams. The diffractometer was positioned on each of the three accessible beams to record the diffraction patterns. The detector used was a Maxipix with 55 μm pixel size and 512 $\times$ 512 pixels. The Maxipix was positioned 2.37 m from the sample on the diffractometer arm and a vacuum pipe with a beamstop was installed between the sample and the detector to reduce the air scattering. The direct-beam image was recorded with a fluence of 1.1×10^{10} ph/μm^2 illuminating the crystal beam splitter, but only 38 % of that fluence illuminated the sample. The deflected-beam images were recorded for both reflections with a fluence of 3.0×10^{11} ph/μm^2 with an efficiency of 4 %.

7. 3D RECONSTRUCTION

The 3D reconstruction was retrieved by using the filtered back-projection algorithm. As the experimental data recorded at ID01 was limited to only three projections, we have applied symmetry constraints [15]. First, we applied a four-fold symmetry constraint around the beam direction, i.e. perpendicular to the view in Fig. 3(c). Second, we applied a mirror symmetry constraint around a mirror plane defined perpendicular to the projection in Fig. 3(c). Once the 3D model was reconstructed, a histogram constraint vetoing the outlaiers was applied. The 3D data visualization has been obtained using ParaView software [16].

8. SAMPLE PREPARATION FOR CDI

Gold nanoparticles for CDI experiments were manufactured on 250 nm thick Si_3N_4 membranes by means of two steps: electron-beam lithography and electroplating [17, 18]. First, a metal stack of Cr/Au/Cr (5nm/10nm/5nm) was evaporated on a silicon nitride membrane. Subsequently, a negative tone e-beam resist HSQ (FOX16, Dow Corning, 1:1 dilution with MIBK) was spin-coated at 3000 rpm, resulting in a film thickness of about 250 nm. In the first e-beam lithography step, an array of micro-rings were exposed using Vistec EBPG5000Plus direct writing e-beam lithography system, operated at 100 kV accelerating voltage. After development of the exposed HSQ in NaOH buffered solution (MICROPOSIT 351, Rohm and Haas) and rinsing in deionized water, silicon dioxide discs with inner/outer diameter of 250 nm/4 μm, respectively, and the distance between the neighboring particles of 100 μm were defined at specified locations. For the following e-beam exposure, approximately 900 nm thick layer of positive tone resist (PMMA 950k, 8% in anisole, MicroChem Corp.) was spin-coated on the membrane at 4000 rpm and baked out at 175 °C on a hotplate. By performing the second overlay exposure and developing the samples in IPA:DI H_2O (7:3) solution, structures with various manifold rotational symmetry, fitting into a circle of 500 nm diameter, were created exactly above the SiO_2 discs. This way, molds for electroplating gold nanoparticles with well-resolved 3D shape control were defined. After a short Cl_2-based plasma etching step (required to remove the upper Cr layer), the mold was filled with gold during the electroplating step to a height of 500 nm. After removing the PMMA layer and subsequently HSQ discs in acetone and BOE (buffered oxide etch), respectively, individual nanoparticles with complex 3D shape along the rotational symmetry axis anchored on the Si_3N_4 membrane were fabricated, an example is shown in Fig. 3(b).

REFERENCES

1. F. Uhlén, D. Nilsson, A. Holmberg, H. M. Hertz, C. G. Schroer, F. Seiboth, J. Patommel, V. Meier, R. Hoppe, A. Schropp, H. J. Lee, B. Nagler, E. Galtier, J. Krzywinski, H. Sinn, and U. Vogt, "Damage investigation on tungsten and diamond diffractive optics at a hard x-ray free-electron laser," Opt. Express **21**, 8051–8061 (2013).
2. T. Koyama, H. Yumoto, Y. Senba, K. Tono, T. Sato, T. Togashi, Y. Inubushi, T. Katayama, J. Kim, S. Matsuyama, H. Mimura, M. Yabashi, K. Yamauchi, H. Ohashi, and T. Ishikawa, "Investigation of ablation thresholds of optical materials using 1-μm-focusing beam at hard x-ray free electron laser," Opt. Express **21**, 15382–15388 (2013).
3. M. R. Howells, T. Beetz, H. N. Chapman, C. Cui, J. M. Holton, C. J. Jacobsen, J. Kirz, E. Lima, S. Marchesini, H. Miao, D. Sayre, D. A. Shapiro, J. C. H. Spence, and D. Starodub, "An assessment of the resolution limitation due to radiation-damage in x-ray diffraction microscopy." J. electron spectroscopy related phenomena **170**, 4–12 (2009).
4. R. Neutze, R. Wouts, D. van der Spoel, E. Weckert, and J. Hajdu, "Potential for biomolecular imaging with femtosecond X-ray pulses." Nature **406**, 752–757 (2000).
5. K. E. Schmidt, J. C. H. Spence, U. Weierstall, R. Kirian, X. Wang, D. Starodub, H. N. Chapman, M. R. Howells, and R. B. Doak, "Tomographic Femtosecond X-Ray Diffractive Imaging," Phys. Rev. Lett. **101**, 115507 (2008).
6. K. A. Nugent, "Coherent methods in the X-ray sciences," Adv. Phys. **59**, 1–99 (2010).
7. J.-P. Guigay, "Fourier-transform analysis of Fresnel diffraction patterns and in-line holograms," Optik. **49**, 121–125 (1977).
8. M. R. Teague, "Deterministic phase retrieval: a Green's function solution," J. Opt. Soc. Am. **73**, 1434 (1983).
9. D. Paganin, S. C. Mayo, T. E. Gureyev, P. R. Miller, and S. W. Wilkins, "Simultaneous phase and amplitude extraction from a single defocused image of a homogeneous object." J. Microsc. **206**, 33–40 (2002).
10. G. Lovric, R. Mokso, C. M. Schlepütz, and M. Stampanoni, "A multi-purpose imaging endstation for high-resolution micrometer-scaled sub-second tomography," Phys. Medica **32**, 1771–1778 (2016).
11. J. Miao, P. Charalambous, J. Kirz, and D. Sayre, "Extending the methodology of X-ray crystallography to allow imaging of micrometre-sized non-crystalline specimens," Nature **400**, 342–344 (1999).
12. R. W. Gerchberg and W. O. Saxton, "A practical algorithm for the determination of phase from image and diffraction plane pictures," Optik **35**, 237–246 (1972).
13. J. R. Fienup, "Iterative Method Applied To Image Reconstruction And To Computer-Generated Holograms," Opt. Eng. **19**, 193297 (1980).
14. S. Marchesini, H. He, H. N. Chapman, S. P. Hau-Riege, A. Noy, M. R. Howells, U. Weierstall, and J. C. H. Spence, "X-ray image reconstruction from a diffraction pattern alone," Phys. Rev. B **68**, 140101 (2003).
15. R. Xu, H. Jiang, C. Song, J. A. Rodriguez, Z. Huang, C.-C.

Chen, D. Nam, J. Park, M. Gallagher-Jones, S. Kim, S. Kim, A. Suzuki, Y. Takayama, T. Oroguchi, Y. Takahashi, J. Fan, Y. Zou, T. Hatsui, Y. Inubushi, T. Kameshima, K. Yonekura, K. Tono, T. Togashi, T. Sato, M. Yamamoto, M. Nakasako, M. Yabashi, T. Ishikawa, and J. Miao, "Single-shot three-dimensional structure determination of nanocrystals with femtosecond x-ray free-electron laser pulses," Nat. Commun. **5**, 4061 (2014).

16. J. Ahrens, B. Geveci, and C. Law, "ParaView : An End-User Tool for Large Data Visualization," Energy. **836**, 717–732 (2005).
17. S. Gorelick, J. Vila-Comamala, V. Guzenko, R. Mokso, M. Stampanoni, and C. David, "Direct e-beam writing of high aspect ratio nanostructures in PMMA: A tool for diffractive X-ray optics fabrication," Microelectron. Eng. **87**, 1052–1056 (2010).
18. V. A. Guzenko, B. Pedrini, A. Menzel, and C. David, "Fabrication of nanoparticles with 3D shape control for X-ray scattering experiments," Microelectron. Eng. **121**, 127–130 (2014).