Supporting Online Material for

High-Resolution Scanning X-ray Diffraction Microscopy

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High-Resolution Scanning X-Ray Diffraction Microscopy: Supporting Online Material

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Materials and Methods

Apparatus and specimen

The experiment was conducted at the coherent small-angle x-ray scattering (cSAXS) beamline at the Swiss Light Source (SLS). X-rays were produced by an undulator source and filtered with a Si(111) double-crystal monochromator. The hard x-ray (6.8 keV) beam was focused using a 50 µm diameter zone plate (SI) with outermost zone width of ∆r = 100 nm. A 20 µm diameter pinhole was placed upstream from the zone plate to ensure good coherence of the beam. This reduces the numerical aperture and thus the diffraction-limited focusing power by a factor of 2.5, resulting in a spot size of 300 nm. The zeroth and higher orders of the zone plate were filtered out by displacing the pinhole out of the zone plate optical axis, and using an order-sorting aperture close to the specimen plane. This off-axis geometry produces a small deviation from the main beam’s propagation axis, resulting in a slight shift of the diffraction
pattern in the detector plane. The tilt of the probe propagation can be completely removed in
the reconstruction by proper recentering of the diffraction patterns.

The specimen was a 30 µm diameter Fresnel zone plate with 70 nm outer zone width (S1). Fabrication of the zones involved etching trenches in a 1 µm thick polyimide mold, and filling them with gold by electroplating. The plating was continued so that additionally a continuous gold coating formed on top of the structure. The reconstructions agree very well with the 1 µm depth of the trenches, and give a thickness of about 0.9 µm for the overcoat.

The specimen was mounted on a piezo-electric stage with a 2 nm nominal displacement precision. The flux incident on the sample was approximately 1.2 × 10^7 photons/s. The diffraction patterns were measured using the Pilatus pixel array detector (S2), a single-photon counting detector having no readout noise and no dark-current. The detector has 172 × 172 µm^2 pixels. It was placed 2.2 m away from the specimen. Although the Pilatus detector covered a solid angle of more than 100 × 100 mrad^2, essentially all the signal was concentrated within 10 × 10 mrad^2. Scattering at larger angles can be detected with longer exposure times or higher incident flux. Despite possible experimental difficulties such as the increase of incoherent scattering or the larger dynamic range, data collection at larger scattering angles translates directly into a higher resolution in the reconstruction.

Reconstruction Algorithm

Let Ψ be an element of a high-dimensional space S built from the direct product of spaces of each individual views: Ψ = (ψ_1, ψ_2, . . . , ψ_N). S is a Euclidean space since, in practice, all views are sampled on a discrete grid. The problem to solve is finding the unique (up to trivial rescaling and translation) Ψ that satisfies simultaneously two constraints. The Fourier constraint is given by

\[ I_j(q) = |\tilde{\psi}_j(q)|^2, \]  

(S1)
where $\tilde{\psi}(q)$ is the 2-dimensional Fourier transform of $\psi(r)$,

$$\tilde{\psi}(q) = \frac{1}{2\pi} \int \psi(r)e^{-ir \cdot q} \, d^2r. \quad (S2)$$

The overlap constraint, given by Eq. 1 in the main text, requires that there exist two functions $P$ and $O$ such that, for all $j$,

$$\psi_j(r) = P(r - r_j)O(r). \quad (S3)$$

The conditions under which this factorization is valid are discussed below.

The relative simplicity of the constraint sets defined by the above equations makes possible the definition of projections onto them. Given any $\Psi \in S$, the projection onto a constraint space $\Pi(\Psi)$ is the element that satisfies the constraint while minimizing the distance $\|\Psi - \Pi(\Psi)\|^2$.

Since Eq. S1 is decoupled in $j$, the Fourier projection is a parallel version of the Fourier projection commonly used in various phase problems:

$$\Pi_F(\psi_j) : \psi_j \rightarrow \psi_j^F = p_F(\psi_j). \quad (S4)$$

Here, $p_F$ is the single-dataset Fourier modulus projection which involves replacing the computed Fourier amplitudes with the measured ones while keeping the phases:

$$p_F(\psi) : \tilde{\psi}(q) \rightarrow \tilde{\psi}^F(q) = \sqrt{I(q)\frac{\tilde{\psi}(q)}{|\tilde{\psi}(q)|}}. \quad (S5)$$

The overlap projection is given by

$$\Pi_O(\psi) : \psi_j \rightarrow \psi_j^O(r) = \hat{P}(r - r_j)\hat{O}(r), \quad (S6)$$

where $\hat{P}$ and $\hat{O}$ are the solution of the coupled system

$$\hat{O}(r) = \sum_j \hat{P}^*(r - r_j)\psi_j(r) \quad \sum_j |\hat{P}(r - r_j)|^2, \quad (S7)$$

$$\hat{P}(r) = \sum_j \hat{O}^*(r + r_j)\psi_j(r + r_j) \quad \sum_j |\hat{O}(r + r_j)|^2. \quad (S8)$$
\( \Pi_\Omega \) is also decoupled in the individual views unless there is non-zero overlap between adjacent probe positions. The coupling between adjacent views is the essential ingredient that gives the method its efficiency.

In general cases, the difference map algorithm requires four distinct arrays of the size of \( \Psi \). Thanks to the structure of the projections, only one such array is needed for the current implementation. With a standard choice of parameters [\textit{i.e.} \( \beta = 1 \) in Elser’s formulation (S3)], the iteration loop takes a simple form. Given an initial random state \( \Psi \), the two following steps need to be computed in turn.

1. If the number of iterations is smaller than a predetermined threshold, typically between 20 and 50, compute only \( \hat{O}(r) \) with equation S7, using an initial guess for the probe function. Updating the probe immediately was observed to be numerically unstable and to lead to very long convergence times. This is not a limitation as the initial probe need not be accurate.

If the number of iterations has exceeded the threshold, update both the probe and the object functions by a few iterations of equations S7 and S8, starting with the probe from the previous step. At each step, apply a threshold on \( \hat{O} \) to maintain all its amplitudes smaller than 1. This operation limits the relative amplitude ambiguity \( PO = (\alpha P)(\alpha^{-1}O) \), where \( \alpha \) can be any complex number.

2. Update each view according to

\[
\psi_j^{(n+1)}(r) = \psi_j^{(n)}(r) + p_F \left( 2\hat{P}(r - r_j)\hat{O}(r) - \psi_j^{(n)}(r) \right) - \hat{P}(r - r_j)\hat{O}(r), \tag{S9}
\]

This formulation is dictated by the difference map; it can be seen that \( \psi_j^{(n+1)}(r) = \psi_j^{(n)}(r) \) only if \( \hat{P}(r - r_j)\hat{O}(r) \) satisfies the Fourier constraint — the goal to attain.
Convergence is monitored with the progress of the error

\[ \epsilon_{n+1} = \| \psi^{(n+1)} - \psi^{(n)} \|^2 \]  

(S10)

Both projections given by equations S4 and S6 are easily implemented on a computer. Depending on the accuracy of the initial probe, and on other factors such as the scattering strength of the specimen, as little as 10 iterations can be sufficient to obtain reasonable convergence. We have observed that longer transients, when they occur, are caused by the slow dynamics of phase vortices, which are either expelled to the edges of the image or annihilated in a vortex/anti-vortex pair without manual intervention. In terms of memory, the method is more demanding than the ptychographic iterative engine (S4). It can however be easily cast for parallel computation. The second step is completely decoupled in the individual views, and the first step contains only a few standard parallel operations (“broadcast” and “gather”). Future implementation of the algorithm will use this feature, which is expected to reduce the computation time substantially.

For the reconstructions presented in this report, 61 × 61 diffraction patterns were used, each being sampled on a 128 × 128 array. Accordingly, the “state vector” \( \Psi \) was a 3721 × 128 × 128 complex-valued array. The initial probe was the calculated wavefield at the focal plane of an ideal lens with a 20 \( \mu \)m circular pupil. The final object image shown on figure 3 of the main text is the average of 30 reconstruction estimates taken from a single run at intervals of 5 iterations. Averaging is a reliable way to ensure uniqueness of a reconstruction, though generally at the cost of a slight decrease in resolution. The computing time for a complete 300 iteration reconstruction was about 3 hours.
Validity of the wave factorization assumption

A fundamental assumption of ptychography is the possibility of factorizing the exit wave into two independent functions, the probe $P$ and the object $O$:

$$\psi(x, y) = P(x, y)O(x, y). \quad (S11)$$

The conditions for this assumption to be valid have been discussed by Rodenburg \(S5\) in terms of Ewald sphere constructions. Here we present an alternative derivation carried in real space.

Let $\Psi_0(x, y, z)$ be the wavefield produced by the apparatus. In absence of the specimen, the field propagates freely and thus satisfies the homogeneous time-independent wave equation

$$\nabla^2 \Psi_0 + k^2 \Psi_0 = 0. \quad (S12)$$

When the specimen is present, the wavefield is given by the Ansatz

$$\Psi(x, y, z) = T(x, y, z)\Psi_0(x, y, z), \quad (S13)$$

where it is required that $\Psi$ satisfies the inhomogeneous wave equation

$$\nabla^2 \Psi + k^2 n^2 \Psi = 0. \quad (S14)$$

$n(x, y, z)$ is the space-dependent complex index of refraction of the specimen. The specimen has a finite extent $L$ in the direction of propagation, so that $n$ is equal to unity except for $0 < z < L$. We want to find under which conditions a function $T$ independent of $\Psi_0$ can satisfy Eq. $S14$ within the specimen depth. With $P(x, y) = \Psi_0(x, y, L)$ and $O(x, y) = T(x, y, L)$, these conditions are obviously those that also guarantee the validity of Eq. $S11$.

Substituting Eq. $S13$ into Eq. $S14$, and using $\Psi_0 = \psi_0 \exp(ikz)$ yields

$$\left(\nabla^2 T + 2ik\partial_z T + k^2(n^2 - 1)T\right)\psi_0 + 2\nabla \psi_0 \cdot \nabla T = 0. \quad (S15)$$
The transfer function $T$ can be independent of $\psi_0$ if the rightmost term is small enough to be neglected, in which case $T(x, y, z) \exp(ikz)$ is seen to satisfy the wave equation,

$$\nabla^2 [T \exp(ikz)] + k^2 n^2 T \exp(ikz) = 0. \quad (S16)$$

As $T = 1$ for $z < 0$, this equation has a unique solution, equal to the wave resulting from a plane wave illumination of amplitude one. The last term between the parentheses in Eq. S15, of order $k^2$, is always dominant. A sufficient condition for the approximation to be valid is

$$k |\psi_0 \partial_z T| \gg |\nabla \psi_0 \cdot \nabla T|. \quad (S17)$$

A rough but general estimate of this inequality is obtained with scaling arguments. From the scalar product, only the transverse terms are important since $|\partial_z \psi_0| \ll k |\psi_0|$. The transverse gradient of $T$ scales at most with the inverse resolution $R^{-1}$ of the measurement: $|\nabla T| \sim R^{-1} |T|$. In the $z$ direction, $|\partial_z T|$ is at least as large as $L^{-1} |T|$. Thus, one gets

$$R/L \gg \lambda/a, \quad (S18)$$

where $a$ is the extent of $\psi_0$ in the specimen plane. In the case of a focused probe, Eq. S18 can be written $R/L \gg N$, with $N$ the numerical aperture of the focussing device. Below are observations on this result and its consequences.

1. In optimal cases, $R$ is the resolution of the imaging system — only function of the angle subtended by the detector. However, weakly scattering specimens can produce a much narrower angular range of usable intensity. In such cases, $R$ should be defined in terms of this “intrinsic resolution”.

2. $Z = Ra/\lambda$ gives roughly the maximum extent over which the wavefield factorization approximation is valid. As a consequence, the imaged region of the specimen needs to lie entirely within this depth. This limits not only the thickness of a specimen, but also
the tolerance of the method to a tilt of the specimen plane. $Z$ also represents the “depth of field” of the method: the focus plane of the reconstructed image lies within this range.

3. Because the method relies on the overlap of adjacent probe translations, $a$ is generally much larger than $R$. The finite depth of focus of the probe, $a^2/\lambda$ is therefore always larger than $Z$, and the probe can always be assumed to be constant within the specimen.

4. When the condition Eq. S18 is satisfied, $T$ is given by the wave equation, without limitation on the scattering regime. The specimen need not be a weak phase object, and the Ewald sphere curvature need not be negligible. Even strong dynamical scattering in the specimen does not compromise the reconstruction. This result also has important consequences for the interpretation of scanning transmission x-ray microscopy images. The validity of the probe factorization assumption confirms that the interpretation given to the first moments analysis is sound [see Fig. 2 and note (21)].

For the experiment presented in this report, the maximum thickness given by Eq. S18 is around 50 $\mu$m. With a field of view of only a few microns, the factorization approximation holds even if the specimen is tilted by a few degrees.

**Reconstruction of a different region of the zone plate specimen**

A reconstruction of the zone plate’s center and its vicinity is displayed in Figure S1. The retrieval of a complex-valued image readily allows the simulation of any traditional imaging method — a principle coined “omni-microscopy” by some authors (S6). One such possible post-reconstruction treatments is illustrated in Fig. S1C: the derivative of the phase along $x$ simulates the image that would be obtained in a differential phase contrast imaging experiment. The gain in resolution provided by the SXDM reconstruction is apparent when the image is compared to Fig. S1D, which was obtained from the STXM analysis of the data.
Figure S1: Reconstruction of the region surrounding the center of the zone plate specimen. (A), (B) Amplitude and phase of the reconstructed optical transfer function, from 50 × 90 diffraction patterns. (C) Gradient of the phase along x for the region outlined in B. (D) The same region from the differential phase contrast image shown in Fig. 2 in the main text. (E) SEM image of the same region. The defects in the zone plate are in part visible as portions of the polyimide structure emerging from the gold layer.

References


