Supplemental material for: Structured illumination for tomographic X-ray diffraction imaging

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1 Theoretical model

To simulate the system, we construct a forward model relating the object [defined in terms of the position-dependent coherent scatter form factor $f(r_o, q)$] and measurement $y(E, t, r_d)$ spaces. Here r_d are the positions of the detector pixels relative to the source and r_o is the object position, q is the momentum transfer, and t is the time. Because of the large design space associated with SICSI, we do not derive here a general model; instead, we describe the experimental configuration discussed in the manuscript. Namely, we model a modulated fan beam composed of a series of periodically-spaced pencil beams oriented along a line. An x-ray source is located at the origin, and a single, energy-sensitive detector pixel is located at $r_d = (x_d, z_d)$.

The differential cross section describing the scatter of an x-ray with energy E into a unit solid angle is given by

$$\frac{d\sigma_{coh}(E)}{d\Omega} = \frac{r_e^2}{2}\left[1 + \cos(\theta)^2\right] f(q). \tag{1}$$

Here r_e is the classical electron radius and $f(q)$ is the square of the coherent scatter form factor, which modifies the Thompson cross section of a free electron by taking into account the effect of nearby scatters. The intensity of the scattered x-rays at a given detector pixel is therefore given as

$$y(E, t, \mathbf{r}_d) = \int dx\, dy\, dz\, dq\, n\, T(E, \mathbf{r_o}, \mathbf{r_d}) \frac{d\sigma_{coh}(E)}{d\Omega}\, \Delta\Omega\Phi(E, x, y, z)\delta\left[E - \frac{hcq}{\sin(\theta/2)}\right]. \tag{2}$$

Here $\theta = \cos^{-1}(\mathbf{r}_d \cdot \mathbf{r}_o/|\mathbf{r}_d||\mathbf{r}_o|)$ is the scatter angle, $\Phi(E, x, y, z) = \phi(E)c(xd/z, yd/z)$ is the structured incident illumination, n is the density of scatterers, and $T(E, \mathbf{r_o}, \mathbf{r_d})$ is the transmission along a path connecting the source, $\mathbf{r}_o$ and $\mathbf{r}_d$. For ease of analytic evaluation, we choose the coded aperture pattern to be a train of delta functions $c(x, y) = \sum_j \delta(x - x_j)\delta(y)$. We note that one can easily model the effects of using finite-sized apertures by integrating over several such codes with different relative shifts. In addition, we make the assumption that the transmission is roughly uniform across all paths and equal to T_o. While this approximation holds for uniform and optically thin objects, one can correct for spatially-dependent attenuation by simultaneously recording a tomographic transmission image.

Looking at the impulse response when $f(r_o, q) = f_o \delta[\mathbf{r} - \mathbf{r}_o(t)]\delta(q - q_o)$ yields a measurement

$$y(E, t, \mathbf{r}_d) = \sum_j nT_o f_o \delta\left(\frac{x_j z_o}{d} - [x_o - v_x t]\right) \delta\left(E - \frac{hcq_o}{\sin[\theta_j/2]}\right) L(\theta_j)\phi(E), \quad (3)$$

where $\theta_j = \theta[(x_j z_o/d, 0, z_o), \mathbf{r}_d]$ and $L(\theta_j) = \left(1 + \cos(\theta_j)^2\right) \Delta\Omega(\theta_j)$. One can see from this expression that z_o, x_o, and q_o are encoded in the temporal modulation frequency, absolute temporal location of the signal, and slope of the resulting curve in E-t space, respectively.

We descritize the forward model by considering rectangular bins in the detector integration time and pixel size, and assume a Gaussian energy response of the detector (which represents an approximation to the true response of an energy-sensitive detector). This yields

$$\begin{aligned} y_m &= \int dE\, dt\, dr_d\, y(E, t, r_d) rect\left(\frac{t - t_m}{\Delta t}\right) rect\left(\frac{x_d - x_m}{\Delta x_d}\right) exp\left[-\left(\frac{E - E_m}{\Delta E}\right)^2\right] \\ &= \sum_j \int dr_d\, nT_o f_o\, rect\left(\frac{[x_j z - x_o d]/vd - t_m}{\Delta t}\right) rect\left(\frac{x_d - x_m}{\Delta x_d}\right) \times \\ &\qquad exp\left[-\left(\frac{hcq_o/\sin(\theta_j/2) - E_m}{\Delta E}\right)^2\right] \phi\left[\frac{hcq_o}{\sin(\theta_j/2)}\right] L(\theta_j), \end{aligned} \quad (4)$$

where Δx_d, Δt, ΔE are the effective detector pixel width, integration time, and energy resolution, respectively, and x_m, t_m and E_m are the centers of the respective bins. By evaluating y_m for a range of different input point objects, one obtains the forward matrix $\boldsymbol{H}$. By considering vectorized versions of the measurement and object $\boldsymbol{y}$ and $\boldsymbol{f}$, respectively, one can solve the linear problem $\boldsymbol{y} = \boldsymbol{H}\boldsymbol{f}$ for $\boldsymbol{f}$ using the algorithm discussed in Sec. 3.

We typically choose to sample the object space with a voxel size of 10 mm in z, 0.5 mm in x, and 0.05 nm^{-1} in q. In the measurement space, we typically consider energies between 20 and 90 keV (with energy bins of 1 keV) and record approximately 60 time steps (enabling the object to pass through approximately 8-10 primary beams). For these parameters, the matrix $\boldsymbol{H} \in \mathbb{R}^{N \times M}$ is rectangular with the number of estimated object voxels exceeding that of measurements. Here $N = N_t N_E$ and $M = N_z N_x N_q$ are the number of measurements and object points, where t, E, z, x, and q correspond to the time, energy, range, cross-range and momentum transfer dimensions. For the estimates shown in the paper, we use a compression ratio of $N/M = 5 - 10$.

2 Resolution

To estimate analytically the resolution of the system in the absence of noise, we take an approach similar to that described in [1]. We start by looking at Eqs. 3 and 4 for the case of a flat input spectrum. One can see that the signal for a point object consists of curve

$$E(t) = \frac{hcq_o}{\sin[\theta_j(t)/2]} \tag{5}$$

in E-t space. This curve asymptotes to $E \to \infty$ for $\theta \to 0$, which occurs when

$$t = \frac{z/z_d x_d - x_0}{\mathrm{v}}. \tag{6}$$

Due to the presence of the mask, the curve is modulated with a frequency of

$$f_{mod} = \frac{z_m \mathrm{v} k_m}{z}, \tag{7}$$

where k_m is the spatial frequency of the mask pattern.

Working backwards, one can estimate Δz (the uncertainty in z) using Eq. 7 and the time-bandwidth relationship of Fourier transforms. We find that Δz is reduced for codes with smaller feature sizes and large source-to-detector distances. Given an estimate of z, one can then use Eq. 6 along with the finite extent of the detector pixel and integration time to estimate Δx. Using the definition of θ_{jk} in terms of z and x and the detector energy resolution ΔE, one can use Eq. 5 to estimate the uncertainty in q. For the parameters used in our experiment, we find that Δz ~10 mm, Δx ~1 mm, and Δq ~0.01 $1/\AA$. This does not represents a fundamental limit, though; by using smaller mask features and a larger z_d, one can in theory achieve a resolution of Δz ~1-2 mm, Δx ~0.5 mm, and Δq ~0.005 $1/\AA$ for a range of locations and materials.

3 Inversion

We recover the molecular information of the objects illuminated by the x-rays by modeling the multiplexed measurements $\boldsymbol{y}$ using a Poisson noise model and solving a maximum a posteriori (MAP) optimization problem using a nonlinear inversion algorithm. Specifically, we let $\boldsymbol{y} \sim \text{Poisson}(\boldsymbol{Hf} + \boldsymbol{\mu}_b)$ where $\boldsymbol{f} \in \mathbb{R}^{M\times 1}$ is the underlying volumetric image that we wish to reconstruct and $\boldsymbol{\mu}_b$ is any unmodeled background scatter from the experimental system. To recover $\boldsymbol{f}$, we assume that the underlying image in (x, z, q) space is piecewise smooth spatially (along the (x, z) coordinates) and piecewise polynomial spectrally (along the q coordinate). This assumption enables us to regularize potential estimates and, along with a maximum likelihood criterion, ensures that the resulting estimate is as representative of the measured data as possible and also piecewise smooth spatially and spectrally.

We implement the reconstruction algorithm using a generalized expectation-maximization framework where the generalization comes from the fact that the unmodeled background $\boldsymbol{\mu}_b$ is unknown in practice and needs to be estimated from a realization of the background $\boldsymbol{b} \sim \text{Poisson}(\boldsymbol{\mu}_b)$. Mathematically, our MAP optimization problem can be described as follows:

$$\widehat{\boldsymbol{f}} = \underset{\widetilde{\boldsymbol{f}} \in \Gamma_M}{\arg\min} \left(-\log p \left(\boldsymbol{y} \middle| \boldsymbol{H}\widetilde{\boldsymbol{f}} + \widehat{\boldsymbol{\mu}}_b \right) + \tau \text{pen} \left(\widetilde{\boldsymbol{f}} \right) \right) \tag{8}$$

where $\widehat{\boldsymbol{\mu}}_b$ is an estimate of $\boldsymbol{\mu}_b$ obtained using the multiscale Poisson denoising algorithm in [2], Γ_M is a collection of estimates obtained using a recursive, dyadic, partition-based framework also discussed in [2], $\text{pen}\left(\widetilde{\boldsymbol{f}}\right)$ is a penalization term that is proportional to the complexity of $\widetilde{\boldsymbol{f}}$, and τ is an user-defined parameter that balances the log-likelihood term and the penalization term. We solve the above optimization problem using a straightforward extension of the GEM algorithm discussed in [2] to accommodate the background. The key advantage of this method is its ability to adapt to varying smoothnesses along the spatial and spectral coordinates. Since the spectrum corresponding to each material has a piecewise polynomial structure while the objects spatially display piecewise smooth or blocky structures, the algorithm refrains from using a sparsity basis (*e.g.*, a 3d wavelet basis) that assumes uniform smoothness along all three coordinates.

References

[1] Joel A. Greenberg, Kalyani Krishnamurthy, and David Brady. Snapshot molecular imaging using coded energy-sensitive detection. *Opt. Express*, 21(21):25480–25491, Oct 2013.

[2] Kalyani Krishnamurthy, Maxim Raginsky, and Rebecca Willett. Multiscale photon-limited spectral image reconstruction. *SIAM Journal on Imaging Sciences*, 3(3):619–645, 2010.