Image reconstruction of nanostructured nonperiodic objects only from oversampled hard x-ray diffraction intensities

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X-ray-diffraction microscopy images nanocrystals and nonperiodic objects by directly reconstructing from oversampled diffraction intensities. Successful image reconstruction of nonperiodic objects has so far required additional experiments to supplement the missing data in the diffraction intensities. Reconstruction only from diffraction data is desirable. We show that image reconstruction of nonperiodic objects can be done without any supplemental experiments by applying a modified hybrid input-output algorithm to experimental hard x-ray-diffraction data of a nanostructured pattern.

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X-ray-diffraction microscopy is a newly developed imaging technique to determine the structures of nonperiodic objects and nanocrystals by using coherent x rays and the oversampling method.1–9 When a coherent beam of x rays illuminates a finite object, the Fraunhofer diffraction intensities of the object can be sampled at a spacing finer than the Nyquist interval. When the sampling interval is fine enough so that the oversampling ratio, a parameter to characterize the degree of oversampling, is 2 or more, a nontrivial unique phase set is in principle embedded inside the diffraction intensities,10 and can be directly retrieved by using global minimization methods such as an iterative method.11 Fienup’s hybrid input-output (HIO) algorithm shows good convergence among algorithms of the iterative methods.12 Due to a high penetrating power of x rays, x-ray-diffraction microscopy can be applied to imaging relatively thick objects. Furthermore the method does not require x-ray optics with a high numerical aperture and can achieve higher spatial resolution than conventional x-ray imaging methods. These features make it potentially an important imaging technique with broad application in nanomaterials science3,4,6–9 and biology.5,13

In x-ray-diffraction microscopy, missing near-forward diffraction data due to a beamstop have been a major difficulty that has been preventing us from reconstructing nonperiodic objects only from the diffraction intensities. The missing data problem is essentially unavoidable, since the unscattered transmitted x rays contribute in addition to the diffracted x rays, and hence the diffraction intensities at the exact forward pixels are not directly observable. The beamstop has to be sufficiently large not to overload the detector with a limited dynamic range. Furthermore, the parasitic scattering, e.g., from a pinhole, also makes it difficult to measure near-forward data with a high signal-to-noise ratio.

The near-forward data correspond to low spatial frequencies of the electron density of the sample, and determine an approximate shape of the reconstructed image. In previous works Fourier transform of lower resolution images by, e.g., soft x-ray microscopes14 was used to estimate the near-forward diffraction intensities.3–5 It should however be noted that the estimated intensities were somewhat different from the correct ones, because soft x-ray microscopes only provide absorption contrast images. In another work, a scanning electron microscope image was used to define a detailed shape of the support.9 Here a support is the region where the sample electrons are expected to exist, and is needed to be defined for image reconstruction. When a sample consists of a small number of isolated pieces, the autocorrelation (Patterson) function can be used to define the support,9 but the technique is difficult to be applied to general objects with complex structures. Also near-forward data are necessary to calculate the autocorrelation function, and hence the missing data problem can not be avoided. If a sample has spatial periodicity such as a nanocrystal, the problem can be circumvented by recording diffraction intensities near a Bragg peak.6,7 In this experimental setup, measurement is relatively easier due to less contribution of the parasitic scattering and no contribution of the transmitted x rays.

The loss of near-forward data is an extremely challenging problem in practical applications of x-ray-diffraction microscopy. The required supplemental experiments to recover near-forward data take extra time for measurement and hence cause extra sample damage. The sample damage problem is especially crucial in biological applications.5 With x-ray free electron lasers, serious sample damage may not allow us to perform supplemental experiments.8 In time-resolved experiments to follow structure changes, it will be difficult to carry out supplemental experiments with the same sample condition. In this paper we show that image reconstruction of nonperiodic objects can be carried out without requiring any supplemental experiments by applying the modified HIO (MHIO) algorithm to simulated data and experimental hard x-ray-diffraction data of a nanostructured pattern.

In the iterative methods, the phases are recovered by iteratively applying Fourier transform and inverse Fourier transform. At each iteration, constraints are applied in both reciprocal space and real space. In reciprocal space, the absolute values of the estimated structure factor $|F(K)|$ are set to the experimental values $|F(K)|$ while keeping the estimated phases. The constrained structure factor $G_\alpha(K)$ is thus given by
The constrained electron density $n$ is typically between 0.5 and 1.

In Fienup's HIO algorithm, the electron density is negative. There are different algorithms for the real-space constraints. In Fienup's HIO algorithm, the constrained electron density $\rho_{n+1}(r)$ is obtained by

$$\rho_{n+1}(r) = \begin{cases} \rho_n(r) & (r \in S) \land (\rho_n(r) > 0) \\ \rho_n(r) - \beta \rho_n(r) & \text{otherwise}, \end{cases}$$

where $S$ represents the support region and $\beta$ is a constant typically between 0.5 and 1.

The iterative method starts with a guessed electron density $\rho_0(r)$ as the initial input. In the studies, we used for $\rho_0(r)$ a random electron density inside the support and no electron density outside the support. Here a problem arises in the normalization of $\rho_0(r)$. The total number of electrons in a sample is unknown when the diffraction intensity at the central pixel is unknown. This is because the structure factor at the central pixel determines the total number of electrons. We propose to normalize the Fourier transform $G_i(K)$ of $\rho_0(r)$. The normalization is made so as to equalize the maximum of $G_i(K)$ in $K \in D$ to the experimental value,

$$G_0(K) = \begin{cases} t G_i(K), & K \in D \\ m_i(K) G_i(K), & K \in D, \end{cases}$$

where

$$t = \max(|F(K)|)_{K \in D} \left/ \max(|G_i(K)|)_{K \in D} \right.,$$

and $m_i(K)$ is given by Eq. (2) with $n = i$. The inverse Fourier transform of $G_0(K)$ finally gives the normalized initial electron density $\rho_0(r)$.

In order to clarify the problem of the HIO algorithm with missing near-forward data, we first carried out reconstructions with simulated data. We took an image shown in Fig. 1(a) as a two-dimensional real sample with 91×45 pixels. Figure 1(b) shows simulated diffraction intensities with 129×129 pixels. The diffraction data at the central 37×37 pixels were removed to simulate the data loss due to a beamstop. In Fig. 1(b) the diffraction intensities were oversampled, which corresponds to surrounding the electron density area with a no-density area. The support size was set to the same as the image size (a tight support). The oversampling ratio, defined by $\sigma = \text{total area}/\text{support area}$, was 4.0. $\beta$ in Eq. (3) was set to be, throughout the analyses, 0.6 in the top and left regions outside the support and 0.8 otherwise. We used Eq. (4) for the normalization of the initial electron density. The image reconstruction using the HIO algorithm was not successful as shown in Fig. 1(c). Figure 2 (a) shows the reconstruction error as a function of the iteration number, which is defined by

$$E_n = \left( \sum_{r \in S} |\rho_n^i(r)|^2 \right) \left/ \sum_{r \in S} |\rho_n^i(r)|^2 \right. \right)^{1/2}.$$

Although the reconstruction error in the HIO algorithm gradually decreased with the iteration number, it does not guarantee that the reconstruction proceeded toward the correct solution. As shown in Fig. 2(b), the estimated total number of electrons increased with the iteration number, which reduced the reconstruction error. But the quality of the recon-

![Image](318x523 to 558x733)

![Image](318x130 to 558x228)
the experimental one, and conversely when the estimated diffraction pattern is broader than the experimental one, and the estimated total number of electrons was 3.3 times higher than the true value. The estimated total number of electrons changed rapidly to 1.5 times higher than the true value after the first 50 iterations of the HIO algorithm. Reasonable reconstruction using the HIO algorithm was obtained after $2 \times 10^4$ iterations when the missing near-forward data size was up to $9 \times 9$ pixels, which was 17 times smaller than that of the MHIO algorithm.

We then applied the HIO and MHIO algorithms to experimental hard x-ray-diffraction data. The experiment was performed at BL29XUL (Ref. 17) of SPring-8 using the same experimental setup as the one used in Refs. 4, 5, and 18. A two-dimensional nanostructured gold pattern with the size of $2.5 \times 2.0 \mu m^2$ was used as a sample [see Fig. 2(a) of Ref. 18 for the scanning electron microscopy image]. The sample was made on a $Si_3N_4$ membrane by electron-beam lithography. Hard x-rays with a wavelength of 2.13 Å were used to achieve high spatial resolution. In x-ray-diffraction microscopy experiments, coherent x-rays are required. The condition for the transverse coherence is $(\Delta \theta)_{\text{rms}} \approx \lambda / (4 \pi Oa)$, and for the longitudinal coherence is $(\Delta \lambda / \lambda)_{\text{FWHM}} \approx \sqrt{2 \ln 2} d / (\pi Oa)$. Here $\Delta \theta$ is the angle that the source subtends at the sample, $\lambda$ the wavelength, $O$ the oversampling degree, $a$ the sample size, and $d$ the desired single pixel size of the reconstructed image. In our case the conditions were $(\Delta \theta)_{\text{rms}} \approx 2.4 \mu rad$ and $(\Delta \lambda / \lambda)_{\text{FWHM}} \approx 3.7 \times 10^{-4}$. The required transverse coherence was achieved by a small vertical source size ($\sim 10 \mu m$) (Ref. 19) and by setting the horizontal front-end slit size to be $150 \mu m$ (rms: $43 \mu m$), where the x-ray source and the front-end slit were located at 54-m and 25-m upstream of the sample, respectively. The required longitudinal coherence was achieved by using a Si (111) double-crystal monochromator with $(\Delta \lambda / \lambda)_{\text{FWHM}} \approx 1.3 \times 10^{-4}$. A $20 \mu m$ diameter pinhole and a corner were placed at 38.1-mm and 12.7-mm upstream of the sample, respectively, to reduce the background scattering from the $Si_3N_4$ mem-
brane and to reduce the parasitic scattering from the pinhole. The diffraction intensities were measured by a direct illumination liquid-nitrogen-cooled charge-coupled device (CCD) detector with $1152 \times 1242$ pixels placed at 743-mm downstream of the sample. The single pixel size of the CCD detector was $22.5 \times 22.5 \mu m^2$. Due to a beamstop in front of the CCD detector, data were missing at some pixels. Part of the missing data was recovered by using centrosymmetry of the diffraction pattern (Friedel’s law), but the data at the central $61 \times 61$ pixels were still lost after the symmetry operation.

Figure 3(a) shows the experimental diffraction pattern ($1001 \times 1001$ pixels) with missing data at the central $61 \times 61$ pixels. The data are those used in Ref. 18. The size of the diffraction pattern and other experimental parameters determined the single pixel size of the reconstructed image to be $7 \times 7$ nm$^2$. The support size was set to be $2.6 \times 2.0 \mu m^2$ (363,290 pixels). The oversampling ratio was calculated to be $\sigma = 9.5$. Figure 3(b) shows the reconstructed image after $5 \times 10^4$ iterations using the HIO algorithm with the initial normalization of Eq. (4), which was not recognizable. This is because the estimated total number of electrons, as observed in the simulations, increased with the iteration number, which indicates that the reconstructed image using the HIO algorithm did not improve with the iteration number. Figure 3(c) shows the recognizable reconstructed image of the sample pattern by using the MHIO algorithm. The estimated total number of electrons converged to a certain value in the MHIO algorithm. To refine reconstruction we used a lower quality reconstructed image of Fig. 3(c) to set an improved support. Figure 3(d) shows a reconstructed image with an improved support in the MHIO algorithm after $5 \times 10^4$ iterations. The improved support was defined as the region where the value of the Gaussian filtered image of Fig. 3(c) with $\sigma = 20$ pixels was more than 23% of the maximum of the Gaussian filtered image. Further refinement can be done in a similar manner.

In summary, we have successfully reconstructed the image of a two-dimensional nanostructured pattern only from the oversampled hard x-ray-diffraction intensities with missing near-forward data by using the MHIO algorithm. The algorithm does not require additional experiments to supplement the missing intensities for x-ray-diffraction microscopy, and consequently reduces radiation damage to samples. We anticipate that the algorithm will greatly broaden the applications of x-ray-diffraction microscopy, especially in biological applications, in time-resolved measurement, and in measurement with x-ray free electron lasers, where it is difficult to supply the missing near-forward data with supplemental experiments.

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