Phase retrieval from exactly oversampled diffraction intensity through deconvolution

Changyong Song,[1] Damien Ramunno-Johnson,[1] Yoshinori Nishino,[2] Yoshiki Kohmura,[2] Tetsuya Ishikawa,[2] Chien-Chun Chen,[3] Ting-Kuo Lee,[3] and Jianwei Miao[1,*]

[1]*Department of Physics and Astronomy and the California NanoSystems Institute, University of California, Los Angeles, California 90095, USA*

[2]*SPring-8/RIKEN, 1-1-1, Kouto, Mikazuki, Sayo-gun, Hyogo 679-5198, Japan*

[3]*Institute of Physics, Academia Sinica, Nankang, Taipei, 11529, Taiwan*

(Received 1 August 2006; revised manuscript received 5 October 2006; published 4 January 2007)

We have shown that, when the linear oversampling ratio ⩾2, exactly oversampled diffraction patterns can be directly obtained from measured data through deconvolution. By using computer simulations and experimental data, we have demonstrated that exact oversampling of diffraction patterns distinctively improves the quality of phase retrieval. Furthermore, phase retrieval based on the exact sampling scheme is independent of the oversampling ratio, which can significantly reduce the radiation dosage to the samples. We believe that the present work will contribute to high-quality image reconstruction of materials science samples and biological structures using x-ray diffraction microscopy.

DOI: 10.1103/PhysRevB.75.012102 PACS number(s): 61.10.Nz, 42.30.Rx, 68.37.Yz, 61.82.Rx

Shannon introduced a sampling theorem in 1949, which states that when a function is band limited within, $[0,a]$, its Fourier transform sampled at the Nyquist interval, $1/a$, fully and exactly determines the function itself.[1] Three years later, Sayre pointed out that the Bragg peaks diffracted from a crystal are exactly sampled at the Nyquist interval.[2] Bragg peaks alone (i.e., without phases), however, cannot be used to uniquely determine the crystal structures. Based on the argument that the autocorrelation function of any object is twice the size of the object itself, Bates extended Sayre's work and concluded that if and only if a diffraction pattern is sampled at a frequency twice finer than the Nyquist interval in each dimension (i.e., oversampled), phases can be uniquely retrieved from the intensity.[3] In 1998, Miao *et al.* showed that Bates' conclusion is overly restrictive and proposed a different explanation.[4] They showed that when an isolated object is coherently illuminated, the diffraction intensity points are correlated. When the correlated and independent intensity points are more than the number of unknown variables of the sample structure, phases are in principle encoded in the intensity.[4] Based on the oversampling phasing method, a novel type of microscopy (i.e., x-ray diffraction microscopy) has been developed for high-resolution two-dimensional (2D) and three-dimensional (3D) imaging of nanocrystals and noncrystalline specimens by using x rays[5–18] or electrons.[19,20]

It is, however, impossible to exactly sample diffraction intensity in experiments. This is because, in measuring the intensity, detectors integrate the scattered particles within specific solid angles. Intensity integration is different from exact sampling and hence reduces the contrast of the interference intensity.[21] The diffraction intensity with reduced contrast introduces artifacts in phase retrieval and in some cases the errors can be as large as 20%. This may seriously limit the applications of x-ray diffraction microscopy in the fields where small density variations are critical to the understanding of the structures and functions. In this paper, we showed that when the linear oversampling ratio ⩾2, exactly oversampled diffraction intensity can be obtained from the measured data through deconvolution. Both our computer simulation and the experimental results have shown that the quality of phase retrieval is distinctively improved by exact sampling of diffraction intensity.

When the far-field waves scattered from a finite object fulfill the Born approximation, the exactly sampled diffraction intensity, $I_S(k)$, is the Fourier transform of the autocorrelation function of the object

$$I_S(k) = \frac{I_e}{M}\sum_{x=0}^{N-1}[\rho(x)\otimes\rho^*(-x)]e^{-2\pi ikx/M},$$

$$k = 0,1,\ldots,M-1, \tag{1}$$

where x and k are the coordinates in real and reciprocal space, I_e the scatted intensity by an electron, $\rho(x)$ the electron density of the object, $\rho(x)\otimes\rho^*(-x)$ the autocorrelation function, and $\sigma_x = M/N$ the linear oversampling ratio in the x axis.[4] In practice, a detector used for measuring a diffraction pattern integrates the intensity within specific solid angles, which is determined by

$$\begin{aligned}I_M(k) &= \int_{k-\Delta/2}^{k+\Delta/2} I_S(k)dk \\ &= \frac{I_e}{M}\sum_{x=0}^{N-1}[\rho(x)\otimes\rho^*(-x)]\frac{\sin(\pi x\Delta/M)}{\pi x/M}e^{-2\pi ikx/M},\end{aligned} \tag{2}$$

where $I_M(k)$ is the measured diffraction pattern and Δ is the detector fill factor which is between 0 and 1. If $\Delta=0$, $I_S(k)$ is equal to $I_M(k)$. However, it is experimentally desirable to collect all the coherently scattered particles (i.e., $\Delta=1$) to enhance the signal-to-noise ratio. When $\Delta\neq 0$, one cannot in general obtain $I_S(k)$ from $I_M(k)$ by using Eqs. (1) and (2). This is because the autocorrelation function of any object is twice the size of the object itself. When the linear oversampling ratio <2, the autocorrelation function is wrapped up with itself. Only when the linear oversampling ratio ⩾2 (i.e., $M \geqslant 2N$),[22] we can obtain $I_S(k)$ by

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$$I_S(k) = FT\left\{\frac{FT^{-1}[I_M(k)]}{\mathrm{sinc}(x/M)}\right\}, \quad \mathrm{sinc}(x/M) = \frac{\sin(\pi x/M)}{\pi x/M}, \tag{3}$$

where FT and FT^{-1} represent the Fourier transform and the inverse Fourier transform and $\Delta=1$. While we have derived the equation for obtaining exactly oversampled diffraction patterns in one dimension, the extension to two or three dimensions is straightforward.

To verify Eq. (3), we have carried out a series of numerical simulations. Diffraction patterns based on intensity integration and exact sampling were generated to study the quality of phase retrieval. A 2D detector was simulated for intensity integration with $\Delta=1$. The phases were then retrieved from the simulated diffraction intensity by using an iterative process called the hybrid input and output algorithm (HIO).[23] Figures 1(a) and 1(b) show the reconstructed images from integrated diffraction patterns with a linear oversampling ratio of 2 and 10, respectively. The blurred structures in Fig. 1(a) are due to intensity integration. Although the intensity-integrated pattern with a larger linear oversampling ratio somewhat improves the quality of phase retrieval shown in Fig. 1(b), it is experimentally undesirable as higher incident coherent flux is needed. For example, to achieve the same signal-to-noise ratio per pixel, the diffraction pattern for Fig. 1(b) requires $\sim$25 times higher incident coherent flux than that for Fig. 1(a). By using Eq. (3), we performed exact sampling of the intensity-integrated diffraction patterns. Figures 1(c) and 1(d) show the reconstructed images from exactly oversampled diffraction patterns with the linear oversampling ratio=2 and 10, respectively. The quality of both images is perfect. We have also systematically studied the quality of phase retrieval from intensity-integrated diffraction patterns with respect to the linear oversampling ratio. For each diffraction pattern, we have carried out five trials with different initial random phase sets. The discrepancy between the original and reconstructed images was calculated by $R_d=\Sigma|\rho_{org}-\rho_{rec}|/\Sigma|\rho_{org}+\rho_{rec}|$, where ρ_{org} and ρ_{rec} are the original and reconstructed images. The averaged discrepancy, R_d, is plotted in Fig. 1(e) as a function of the linear oversampling ratio. The error bar was calculated from the standard deviation. The quality of phase retrieval improves with the increase of the linear oversampling ratio, which is consistent with Eq. (3). For a larger linear oversampling ratio, the term of x/M in Eq. (3) becomes smaller and the sinc function is close to 1.

We further verified Eq. (3) by using experimental data. An oversampled diffraction pattern (792×792 pixels) was experimentally obtained from a GaN nanocrystal particle with a linear oversampling ratio of $\sigma_x=\sigma_y=18$.[13] To reduce the oversampling ratio for the diffraction pattern, we numerically integrated the intensity by binning 9×9 pixels into 1 pixel. The new diffraction pattern is an 88×88 pixel array with $\sigma_x=\sigma_y=2$. Phase retrieval was carried out by using a guided HIO algorithm (GHIO).[24] GHIO started with 16 image reconstructions on each diffraction pattern with 16 random phase sets as the initial input. Each image reconstruction was iterated back and forth between real and reciprocal space. In

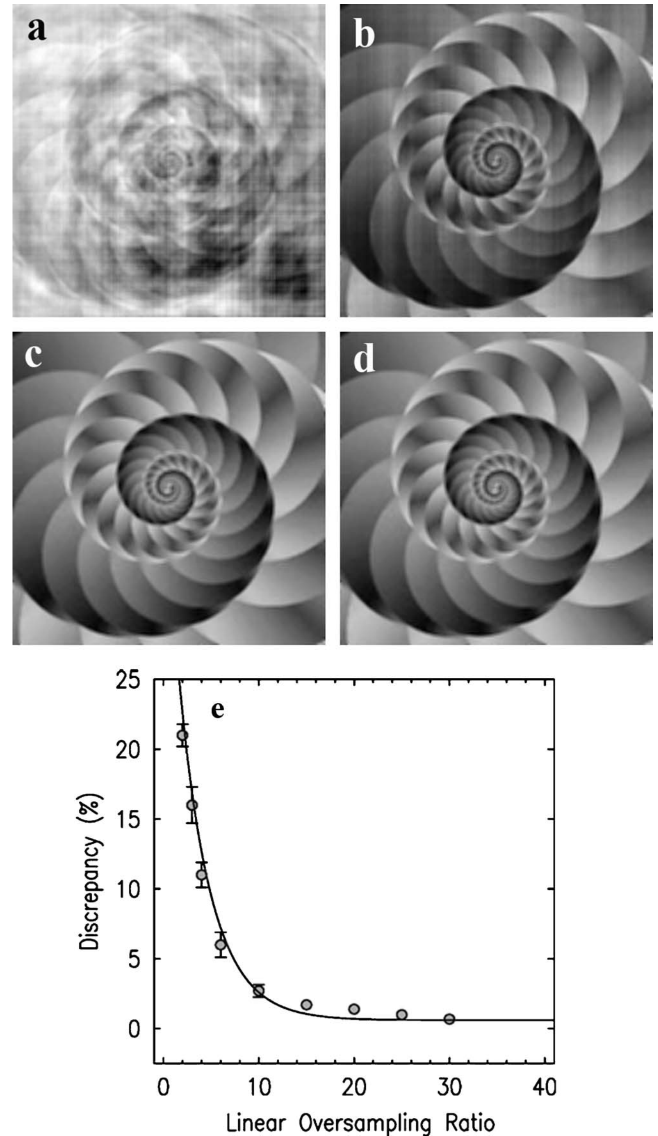

FIG. 1. Images reconstructed from intensity-integrated diffraction patterns with (a) $\sigma_x=\sigma_y=2$ and (b) $\sigma_x=\sigma_y=10$, and from exactly oversampled diffraction patterns with (c) $\sigma_x=\sigma_y=2$ and (d) $\sigma_x=\sigma_y=10$. (e) The discrepancy error (R_d) of phase retrieval as a function of the linear oversampling ratio.

real space, both the electron density outside a support and the negative density inside the support were slowly pushed close to zero, where the support represents a boundary somewhat larger than the sample envelop. In reciprocal space, the measured magnitude of the Fourier transform (i.e., the square root of the diffraction pattern) was enforced in each iteration. After 2000 iterations, 16 images were obtained, which represents the 0th generation of the reconstruction. An error was calculated for each image based on the difference between the calculated magnitude of Fourier transform and the measure one. A best image was chosen corresponding to the smallest error. By multiplying the best image with each of the 16 aligned images and taking the square root of the product, a new set of images were obtained which were used for the next generation. This step was to reduce the electron density fluctuation inside the images. We repeated the proce-

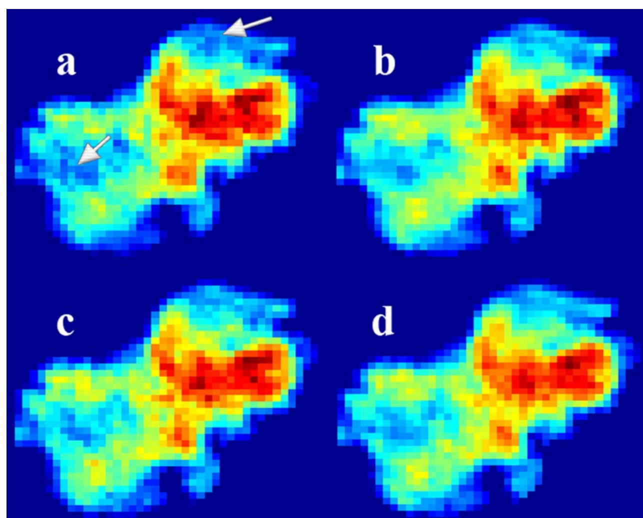

FIG. 2. (Color online) Images reconstructed from measured diffraction patterns with (a) $\sigma_x=\sigma_y=2$ and (b) $\sigma_x=\sigma_y=18$, and from exactly oversampled diffraction patterns with (c) $\sigma_x=\sigma_y=2$ and (e) $\sigma_x=\sigma_y=18$. The red, yellow and blue colors represent the high, the medium and the low electron density.

dure for each generation and, after 9 generations, the 16 reconstructed images were consistent. Figures 2(a) and 2(b) show the best images after 9 generations, reconstructed from the diffraction patterns with a linear oversampling ratio of 2 and 18, respectively.

We performed exact sampling of the diffraction patterns based on Eq. (3). The central missing data were filled in by applying the inverse FFT to Fig. 2(b), and were removed after exact sampling to assure the same reconstruction conditions. Image reconstruction was carried out by using GHIO. Figs. 2(c) and 2(d) show the reconstructed images from the exactly oversampled diffraction patterns with a linear oversampling ratio of 2 and 18, respectively. In comparison of Figs. 2(a) and 2(b), Figs. 2(c) and 2(d) are more consistent to each other. Furthermore, due to the intensity-integration and a lower oversampling ratio, Fig. 2(a) shows noticeable structural degradation as indicated by the arrows. The quantitative discrepancy between Figs. 2(a)–2(d) was calculated by $R_{ij}=\Sigma|\rho_i-\rho_j|/\Sigma|\rho_i+\rho_j|$, where ρ_i or ρ_j represents one of the four images. Table I shows the values of the discrepancy, indicating that the diffraction patterns with intensity integration and a lower linear oversampling ratio

TABLE I. Quantitative discrepancy between Figs. 2(a)–2(d). The bold values correspond to the largest errors.

	Fig. 2(a)	Fig. 2(b)	Fig. 2(c)	Fig. 2(d)
Fig. 2(a)	0	**5.62%**	**5.51%**	**6.02%**
Fig. 2(b)	**5.62%**	0	3.60%	3.33%
Fig. 2(c)	**5.51%**	3.60%	0	3.90%
Fig. 2(d)	**6.02%**	3.33%	3.90%	0

caused the largest error in phase retrieval. With exact sampling of diffraction patterns, the quality of phase retrieval is independent of the linear oversampling ratio. These experimental observations are in a good agreement with the simulation results shown in Fig. 1.

When the linear oversampling ratio $\geqslant 2$, exactly oversampled diffraction patterns can be directly obtained from measured data through deconvolution. Our computer simulations have shown that exact sampling of diffraction patterns significantly improves the quality of phase retrieval and in some cases the errors can be reduced as much as 20%. By using the experimental data from a GaN nanoparticle, we have demonstrated that the images reconstructed from exactly oversampled diffraction patterns are distinctively better than those obtained from intensity-integrated patterns. Furthermore, the exact sampling scheme works as well for a lower oversampling ratio as for a higher oversampling ratio. The former requires less incident coherent flux than the latter to achieve the same signal-to-noise ratio of the diffraction intensity. This is especially important in imaging biological samples where radiation damage is a critical issue.[12,17,25] As x-ray diffraction microscopy is currently under rapid development, we anticipate that the ability to perform accurate phase retrieval will broaden its applications to the areas where small electron density variations are critical to the understanding of the structures with the functions.

J.M. thanks S. Sinha for a stimulating discussion on exact sampling of diffraction patterns. This work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences (DE-FG02-06ER46276) and the U.S. National Science Foundation, Division of Materials Research (DMR-0520894). Use of the RIKEN beamline (BL29XUL) at SPring-8 was supported by RIKEN.

*Email address: miao@physics.ucla.edu

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