We present the framework for convergent beam Bragg ptychography, and, using simulations, we demonstrate that nanocrystals can be ptychographically reconstructed from highly convergent x-ray Bragg diffraction. The ptychographic iteration engine is extended to three dimensions and shown to successfully reconstruct a simulated nanocrystal using overlapping raster scans with a defocused curved beam, the diameter of which matches the crystal size. This object reconstruction strategy can serve as the basis for coherent diffraction imaging experiments at coherent scanning nanoprobe x-ray sources. © 2011 Optical Society of America

Various lensless coherent x-ray diffraction imaging (CXDI) techniques have been developed that image micron and nanometer scale structural features in samples using measured coherent speckle [1,2]. To overcome the phase problem, image reconstruction in CXDI is done iteratively and relies on a priori knowledge about the sample, the beam, or the relative spatial relationship between the two [3,4]. Plane wave Bragg CXDI can image isolated nanocrystals with strain sensitivity [1], while x-ray ptychography [5–7] and Fresnel CXDI [3,9] can reconstruct the density of continuously structured samples. However, reconstructing three-dimensional (3D) densities and strain fields from densely arranged self-similar nanocrystals is an outstanding problem with many practical applications in nanoscience. Here, we show that Bragg ptychography employing a focused beam can determine 3D reconstructions of complex density in nanocrystals. We show that the speckle contrast can be maximized by using a defocused conical beam to fully illuminate the nanocrystal. Finally, we demonstrate that data collected under such conditions can be reconstructed using an adaptation of the ptychographic iterative engine (PIE) [5,10].

The focusing behavior of an ideal hard x-ray zone plate optic can be calculated by defining a phase shift in the plane of the zone plate:

$$\phi_{zp} = -k \sqrt{x_{zp}^2 + y_{zp}^2 + f^2 - f}.$$  \hspace{1cm} (1)

Here, \(x_{zp}\) and \(y_{zp}\) are coordinates in the plane of the zone plate, the focal distance \(f\) is defined by the zone plate diameter \((d_{zp})\) and minimum zone width \(\Delta_{zp}^{\text{min}}\):

$$f = d_{zp} \times \Delta_{zp}^{\text{min}} / \lambda,$$

and the wave vector \(k = 2\pi / \lambda\). The focused wave amplitude at any point downstream of the zone plate \(r_{pt}\) can then be determined by wave summation using a set of zone plate phases \(\phi_{zp}\):

$$A_{pt} = \sum_{j=1}^{N_{zp}} \exp \left( i k \|r_{zp} - r_{pt}\| + \phi_{zp} \right) / \|r_{zp} - r_{pt}\|.$$  \hspace{1cm} (2)

where \(r_{zp} = (x_{zp}, y_{zp}, z_{zp})\). From these equations we calculate focused beam amplitudes within a model nanocrystal by establishing a set of emitter positions in the zone plate plane, a focal length, and a set of voxels that define a nanocrystal volume. In this work, we modeled a zone plate with 54,000 emitters based on the zone plate parameters of the Center for Nanoscale Materials/Advanced Photon Source hard x-ray nanoprobe which produces a 40 nm focal spot with a convergence angle of 0.3°, given by \(d_{zp} = 133 \mu m, \Delta_{zp}^{\text{min}} = 24 \text{ nm at } \lambda = 1.2 \AA\). The sample was a 580 nm faceted model crystal with uniform real density made of 22,963 scattering points (voxels) on a perfect cubic lattice that was tilted with respect to the beam axis by 12.5° to mimic a specular Bragg geometry, shown in Fig. 1(a).

In the absence of a focusing optic, coherent plane wave diffraction can be calculated by Fourier transforming (FT) the crystal density, as shown in Fig. 1(a). Using the zone plate equations, we can simulate the interaction of a focused or defocused wavefront with a nanocrystal. As seen in Fig. 1(b), the focusing of the illuminating wavefront near the focus introduces nonuniform localized amplitudes and concentric phase wraps within the crystal volume (visualized as an isosurface and colored cut plane within the crystal). In this case, the Bragg diffraction is given by the Fourier transform of the beam profile times crystal density, a quantity that depends on illumination. By moving the crystal farther from focus [Figs. 1(b) and 1(c)], the beam wavefront spreads, filling the nanocrystal more uniformly and introducing more pronounced speckles in the diffracted intensity.

We have determined from a series of simulations that the sensitivity of diffracted speckle to variations in beam position or crystal orientation is maximized at a defocus where the beam diameter matches the crystal size. This condition spreads the beam and maximizes the number...
of phase wraps (∼3 for this beam geometry) within the bounds of the crystal, a condition found to improve two-dimensional (2D) Fresnel CXDI performance [9,11]. In Fig. 2, the three speckle volumes shown were generated by displacing a 540 nm diameter beam by 90 nm and by rotating the crystal 30° about the vertical axis. The noticeable changes in speckle due to small relative movements are severely muted when the focused beam diameter is much smaller or larger than the crystal and is absent in the case of plane wave illumination. With proper defocus, therefore, beam raster scans are possibly similar to those required for ptychographic object reconstruction.

The PIE requires overlapping raster positions and a known beam wavefront to iteratively reconstruct an object density and to phase the corresponding diffraction patterns [3]—an approach that we extended to use multiple projections of 3D objects. This algorithm, PIE3D, uses 3D FTs to simultaneously phase diffraction patterns generated with both translational and rotational diversity (grid scans and sample rotations). With a fixed beam diameter of 540 nm and a step size of 200 nm in x and y, diffraction grids (denoted by their total number of translations T) were done at two rotations (R) where Δθ_R = 90° [Fig. 3(a)]. With noise-free diffraction,
Fig. 3(b) shows that the PIE3D performance dramatically improves for a given grid by introducing this rotational diversity. In terms of translations, reconstruction quality improved (lowest $\chi^2_{\text{mod}}$ error metric) as $T$ increases, constraining a given projection in a manner analogous to 2D ptychography [12]. Snapshots of the crystal during phasing reveal accurate 3D reconstruction of both the object density and its flat internal phase without the use of an object-bounding support. Reconstructions were repeated under this condition ($9T \times 2R$), while varying both $\Delta \theta_R$ and the beam defocus [Fig. 4(a)]. The final object reconstruction quality was highest when $\Delta \theta_R \approx 90^\circ$ and when the defocused beam matched the crystal size and maximized phase curvature in the sample. This optimized focused beam Bragg ptychography geometry ($9T \times 2R$, $\Delta \theta_R = 90^\circ$) can be compared to plane wave reconstructions of the same crystal. Phasing of noisy diffraction was carried out with the PIE3D and with standard plane wave reconstruction algorithms [3] given a perfect support (i.e., support = object) [Fig. 4(b)]. A comparison of the $\chi^2_{\text{obj}}$ object-space error metric shows that the PIE3D reconstruction quality at a given noise level is equivalent to the corresponding plane wave phasing and does not require any object support.

Experimental implementation of coherent focused beam Bragg diffraction measurements require a hard x-ray nanoprobe diffractometer with exceptional sample and beam stability as well as a method to ensure positional and orientational registry throughout the scan [13]. Beyond the measurement itself, phasing experimental data presents additional challenges including incomplete knowledge of the beam wavefront and reconstruction artifacts from refraction effects. Fortunately, probe recovery has successfully been implemented in 2D experimental ptychography [6,10], refraction corrections to 3D plane wave reconstructions have been developed [14], and these concepts can be adapted to 3D focused beam Bragg ptychography to address new challenges in crystalline nanoscience imaging.

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References