Inversion of x-ray diffuse scattering to images prepared objects


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The inversion of far-field scattered intensity to a near-field wave field (image) is demonstrated using soft-x-ray scattering from a random cluster of gold balls. The only a priori information assumed about the scatterer is compact support (an isolated object), a condition enforced by using an atomic force microscope to assist in the preparation of the object. The object support function is obtained from the autocorrelation function of the object. X-ray images of 50-nm-diam gold balls are obtained by this method with 10-nm resolution. The method may be applied to other radiations and to nanostructures and macromolecules which cannot be crystallized. Other favorable types of prepared objects are considered.

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The inversion problem of coherent scattering—the reconstruction of a single-scattering potential from measurements of scattered intensity in the far field—has occupied physicists for over a century and arises in fields as varied as optics, radar, x-ray crystallography, medical tomographic imaging, holography, electron microscopy, and particle scattering generally. The scientific payoff from a solution to this problem is understood to be considerable, in view of the lensless imaging capability it would provide for the various radiations (such as coherent atom beams, neutrons, and x rays) for which no lenses exist. Where imperfect lenses do exist, such a diffraactive imaging method would offer the possibility of diffraction-limited image reconstruction without aberrations. This promise of high-resolution imaging using particles whose interaction may reduce radiation damage has stimulated a burst of recent activity (see Ref. 1 for a review). Here we are concerned with x-ray diffraction, where, for nonperiodic structures such as inorganic nanostructures and macromolecules which cannot be crystallized, the established methods of crystallography cannot be used to solve the phase problem, yet an urgent need exists for tomographic imaging of these individual nanostructures in controlled environments for greater thicknesses, and with less radiation damage, than is possible by other techniques such as cryomicroscopy. In this paper we describe the inversion of experimental soft-x-ray transmission diffraction patterns to images of an isolated object at 10-nm resolution, using only the diffraction data.

The possibility of solving the x-ray phase problem for an isolated object was first suggested by Sayre in 1952, who pointed out that Bragg diffraction undersamples the autocorrelation function of the object (see also Refs. 3 and 4). An iterative phase-retrieval method, successful with simulated data, first appeared in 1972, followed by important theoretical advances due to Fiddy, Bates, and others (see Ref. 14 for a review). The iterative oversampling algorithm was greatly improved through the introduction of feedback by Fienup in 1982, and perhaps the earliest inversions of experimental data (using laser-light scattering) are those reported in Refs. 7 and 8. A significant breakthrough occurred in 1999 (Ref. 9) with the reconstruction of a two-dimensional nonperiodic x-ray image at 0.1-μm resolution from diffraction data and a low-resolution image of the object. Subsequent work has produced nanocrystal images at submicrometer resolution using hard x rays and striking tomographic images at higher resolution using zone-plate x-ray images to provide the low-resolution data. Images have also been obtained by inversion of experimental coherent electron diffraction patterns and further laser-light images. The hybrid input-output (HiO) algorithm used Fourier transforms iteratively between real and reciprocal space, imposing known information at each step. In our work, part of this known information is the unit transmissivity of the transparent silicon membrane (on which our object is mounted) outside the boundary (support) of the isolated object. This support defines a Nyquist or Shannon sampling frequency for the diffraction pattern. In order to extract phase information from the diffraction pattern and avoid loss of information, the diffracted intensity is sampled at this Nyquist frequency (twice the Bragg frequency, e.g., two samples per period in the Young’s fringe intensity generated by points at opposite sides of the support). This leads to twofold oversampling of the diffracted wave amplitude (e.g., four samples per period in the Young’s fringe wave function) and, hence, to a zero-padded border around the object, consistent with the requirement that the diffraction data contain no contributions from outside the support. In this way the number of (nonlinear) Fourier equations linking real and reciprocal space become equal to the number of unknown amplitudes and phases (see Ref. 11 for references). An extensive review of iterative methods and an analysis of their convergence in terms of projections onto convex and nonconvex sets can be found in Ref. 14.

All previous x-ray work has required an additional conventional image (usually using a zone-plate lens or scanning electron microscope (SEM)) either to supply the low-order spatial frequencies of the object obscured by the beam stop or to provide the support mask for the reconstruction. In this paper we report the x-ray image reconstructions which are based on the diffraction pattern of an isolated nonperiodic object. This is important for the application of the method to radiations for which no lenses exist for secondary imaging (such as hard x rays) and may provide a more accurate sup-
port than an image formed with a different particle interaction. It also results in a simpler, more self-contained imaging technique.

Our samples consist of monolayer clusters of about 40 randomly positioned gold balls, $d = 50$ nm diameter, lying on a 75-$\mu$m square silicon nitride membrane. Since the HiO algorithm assumes a compact, isolated object, an atomic force microscope was used to form isolated clusters. The use of the atomic force microscope was primarily to remove gold balls from the membrane in all but the region of interest. This was performed by first imaging in tapping mode to determine the location of the tip relative to the sample, followed by scanning in contact mode to scrape away material. The cluster was illuminated by the coherent soft-x-ray beam ($\lambda = 2.1$ nm, $E/\Delta E = 500$) generated by an undulator and zone-plate monochromator at the Advanced Light Source (ALS) storage ring at Lawrence Berkeley National Laboratory (Beam Line 9.0.1). Transmission speckle patterns were recorded on a nude 1024×1024 charge-coupled-device (CCD) camera (16 bits, pixel size $d_p = 24 \mu$m). The width of the reconstructed region $W = \lambda / \theta_p$ = 10 $\mu$m is 4 times the area of the isolated cluster (due to the oversampling of the diffraction pattern) and is less than the spatial coherence width of the beam. (Here $\theta_p = d_p / L$ is the angle subtended by the first-order CCD pixel at the sample, with $L = 105$ mm the working distance from sample to detector.) A detailed description and diagram of our experimental apparatus is given elsewhere.

Figure 1 shows the speckle diffraction pattern obtained from a cluster of gold balls. The pattern was compiled from data taken at several exposure times in order to increase the dynamic range of the measurement. (Total exposure time 3 h.) Diffraction patterns could also be recorded with and without absorption filters, so that the loss of data in the high-intensity central region is minimized. The pattern shows the Airy’s disk-like pattern from a single ball modulated by interference between different balls. Using the known refractive index for gold, calculations show a first minimum in the diffraction pattern from one such phase-object ball of diameter $d$ at a scattering angle $\theta$ where $\sin \theta \lambda = 1.394 / d = 0.059$ rad. A mathematical description of the HiO algorithm we use has been given elsewhere. In brief, a set of random phases is assigned initially and an image obtained by Fourier transform. This is set to the known pixel values (e.g., the transmissivity of the supporting membrane) outside the support of the object (modified by feedback). No sign constraint was applied to the image wave field. A second transform gives a new estimate of the diffraction pattern, in which amplitudes are replaced with experimental values. The algorithm contains one adjustable parameter, the feedback $f$, which we set to 0.9, and an error metric $E$ is used which measures agreement between the current image estimate and

FIG. 1. Soft x-ray transmission speckle pattern at 588 eV from cluster of $d = 50$ nm gold balls. The first minimum in the pattern occurs at $\sin \theta \lambda = 1.394 / d$.

FIG. 2. Autocorrelation (Patterson) function obtained from Fig. 1, showing two ellipses used to define the acentric support. (The pattern is centric.)

FIG. 3. Comparison of reconstructed soft-x-ray image (right) and SEM images of gold ball clusters (left). Each ball has a diameter of 50 nm.
the known form of the image outside the support. Iterations continue until $E$ is small. Solutions independent of the choice of initial phases are accepted.

The repeated use of the support bears a close similarity to the method of solvent flattening or density modification in x-ray crystallography. Missing data due to the beam stop and its supports are left unconstrained during the iterations. The image is somewhat similar to the x-ray dark-field mode; however, the missing low spatial frequencies are supplied to some extent by the known pixel values outside the object support. The central 50 x 50 missing pixels are mainly due to a point-projection shadow image of the Si3N window supporting the sample. The convergence of this algorithm has been studied in detail and the uniqueness of solutions for $E = 0$ established. Rather than rely on a low-resolution secondary imaging method to obtain an estimate of the support, we have obtained an estimate of the object support from the support of its autocorrelation. Figure 2 shows the autocorrelation function, which is the Fourier transform of the diffracted intensity distribution. It is defined in real space and provides, among other things, a map of all interball vectors. Since the object consists of two large clusters, the autocorrelation function consists of three major clusters in a centrosymmetric pattern. To generate the object support, two ellipses where drawn centered on the central clusters and one side cluster, about half the size of the respective clusters. (Autocorrelation magnifies laterally by 2.) More elaborate and general procedures are given elsewhere. As the algorithm progresses, successively tighter supports can be imposed manually, greatly reducing the total iteration time. Figure 3 shows the final result after 53 iterations of the HiO algorithm. The image is compared with a scanning electron microscope image of the same gold balls and is seen to be in excellent agreement. Although a strict object-independent definition of resolution is not possible for this type of phase-contrast microscopy, our experimental conditions generate 5–6 pixels across each ball diameter, giving a resolution of about 10 nm. This is consistent with the size of the diffraction pattern in Fig. 1.

We note that, for an object consisting of a weakly scattering unknown region such as a cell, plus a single gold ball positioned nearby using the AFM, the autocorrelation function would include a faithful image of the cell, limited in resolution by the size of the ball. (The heavy-atom method of crystallography and the technique of Fourier transform holography are closely related.) This image might then be used directly as the support.

In summary, we find that the ability to make "prepared objects" using an AFM provides just the a priori information needed to solve the phase problem. This capability can be combined with the use of the autocorrelation function of an isolated object in order to estimate its support. We have succeeded in reconstructing the image of a nonperiodic object from its far-field scattered intensity alone. Extensions of the method would, for example, allow a single smaller gold nanoball to be placed near an unknown object (such as a cell). Then the autocorrelation function includes a convolution of the ball and object and so gives a direct image of the object, which may be used to provide an improved estimate of the support.

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