Hybrid real- and reciprocal-space full-field imaging with coherent illumination

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Abstract
We present a novel diffractive imaging method that harnesses a low-resolution real-space image to guide the phase retrieval. A computational algorithm is developed to utilise such prior knowledge as a real-space constraint in the iterative phase retrieval procedure. Numerical simulations and proof-of-concept experiments are carried out, demonstrating our method’s capability of reconstructing high-resolution details that are otherwise inaccessible with traditional phasing algorithms. With the present method, we formulate a conceptual design for the coherent imaging experiments at a next-generation x-ray light source.

Supplementary material for this article is available online

Keywords: coherent diffraction imaging, iterative phase retrieval algorithms, x-ray imaging

(Some figures may appear in colour only in the online journal)

1. Introduction

Phase retrieval, the procedure in which the phase is recovered from the measured intensity pattern(s), is crucial for diffraction-based imaging approaches and has been extensively studied in the past decades. Following Gerchberg and Saxton’s pioneering work [1], an improved version of the iterative phase retrieval algorithm, namely hybrid input-output (HIO), was presented by Fienup and is widely used by researchers in this field ever since [2–5]. HIO not only exhibits an elegant mathematical formulation, but also demonstrates superior versatility and robustness against the stagnation problem, which is often a significant issue when utilising the other precedent algorithms. Typical iterative phase retrieval algorithms like HIO start with inversely Fourier transforming the observed magnitude profile in the reciprocal-space (supplemented with a set of random phases as the initial guess) to the real-space, where some constraints are applied to update the real-space image. A forward Fourier transform is then executed to bring the data back to the reciprocal-space, where the Fourier modulus is replaced by the measured value and the newly calculated phase is kept. Such consecutive inverse and forward Fourier transformations are repeated until it reaches a convergence or until a meaningful real-space image is obtained.

X-ray coherent diffraction imaging (XCDI), in which the sample is illuminated by a coherent x-ray light source and the resulting far-field diffraction pattern is phased to recover the image, has been regarded as a potentially groundbreaking development in x-ray microscopy and as one of the major motivations for the development of x-ray free electron lasers (X-FEL) [6–10]. XCDI relies on numerical algorithms like
HIO to execute the phase retrieval, which yields the real-space images. To this end, several alternatives and modifications to the original HIO have been proposed and widely studied, such as MHIIO [11], HPR [12], RAAR [13], shrink-wrap [14], GHIO [15] and OSS [16]. In addition to the algorithm developments, modified experimental schemes for XCDI have been proposed for achieving better spatial resolution and improved robustness of phase retrieval [17]. Examples include key-hole holography [18], in-flight holography [19] and the approaches using a metallic template [20], gold nanoclusters [17, 21, 22], or a block-reference [23].

Despite all the advancements, phase retrieval in XCDI is still one of the major obstacles that hinders the broad application of XCDI. It is nontrivial to construct a robust phase retrieval workflow that eliminates the vulnerability to common sources of image imperfection, e.g. missing data and quantum noise. Missing low-q data in the diffraction pattern is often caused by a beam stop that blocks the direct beam to protect the detector. Quantum noise is inevitable in diffractive imaging and it largely depends on the brilliance of the light source. Although these adversities could potentially be alleviated by the proposed and ongoing major upgrades of the x-ray facilities and beamlines, here we demonstrate an alternative approach that is effective and practical.

In this work, we demonstrate a hybrid approach, in which we image the same sample with two different modalities: a coherent diffraction imaging apparatus provides a diffraction pattern and a direct imaging setup gives a real-space image, where the latter intrinsically has a lower image resolution due to the limited numerical aperture of the objective lens. We develop a phase retrieval algorithm that uses the low-resolution (LR) image as a real space constraint and reconstructs high resolution details from a diffraction pattern recorded from the same object. The contribution of this paper is three-fold: (i) we provide a phase retrieval algorithm that takes advantage of the LR image; (ii) we provide an experimental proof-of-concept with a He–Ne laser-based table-top setup; and (iii) we conceive a novel conceptual design of an experimental setup that allows the collection of diffraction patterns and LR images simultaneously at an X-FEL beamline.

2. Principle

The present approach uses an LR real-space image of the sample as a priori information to guide the high-resolution image reconstruction. Such LR images can be obtained from a variety of sources and can be acquired concurrently or consecutively, experimentally or computationally. To take advantage of such LR images for the phase retrieval, we have developed a modified phase retrieval algorithm, which works as follows. A random phase component is assigned to the square-root of the experimentally measured diffraction intensity, making the input \( F^{(0)}(k) \). The input is then inversely Fourier-transformed to the real-space, where two constraints are applied. In addition to the conventional support constraint, which is based on the estimated shape of the non-zero region and the non-negativity, here we also enforce an LR constraint. The modified real-space constraint reads

\[
\rho^{(t)}(r) = \begin{cases} 
\rho^{(0)}(r) + \gamma \left[ g(r) - \rho^{(0)}(r) \right], & r \in S \\
\rho^{(t-1)}(r) - \beta \rho^{(0)}(r), & r \notin S 
\end{cases}
\]  

(1)

where \( \rho^{(0)}(r) \) is the inverse Fourier transform of the updated Fourier modulus \( F^{(t-1)}(k) \) (see figure 1), \( g(r) \) is the LR prior, \( S \) is the real space support, \( \beta \) is the feedback parameter, typically set to 0.9 as discussed in [2], and \( \gamma \) is defined as follows.

The basic idea behind our approach is that there shall be a consistency between the input real-space LR prior and the low-spatial-frequency component of the final high-resolution image. This consistency is, subsequently, utilised as an additional constraint that guides the update of the real-space image throughout the iterations. To execute the idea discussed above, we introduce a new parameter \( \gamma \) that determines the strength to which the LR constraint is enforced. This parameter should be adaptive. A strong enforcement is desired during the early iterations for an effective guidance using the LR. This LR constraint, however, should eventually vanish as one would anticipate achieving high resolution in the final result with high-spatial-frequency information that is not available in the LR. Intuitively, we can connect \( \gamma \) to a figure of merit (FOM) that
evaluates the image quality on-the-fly. One example is the normalised Fourier space residual function

\[ E_F = \frac{\sum_{k \notin U} |F(k)| - |F(k)^{\text{obs}}|}{\sum_{k \in U} |F(k)^{\text{obs}}|^2} \]  

(2)

where \( U \) denotes the missing data region and \( F(k) \) is the Fourier transform of the on the fly image right before the real-space constraints. As one can see, if the calculated Fourier modulus is consistent with the measured amplitude, \( E_F \) will be close to zero. The exact form of the FOM is not crucial and other FOM functions might be used, as long as the output is in \([0, 1]\) and can monotonically reflect the on-the-fly image quality. In the rest of this paper, the value of \( \gamma \) will be determined at each step by \( E_F \), i.e. \( \gamma = E_F \). After the real space constraints are applied, the Fourier magnitude constraint follows to ensure consistency with the experimental observation:

\[ F^{(i)}(k) = \begin{cases} |F^{(i)}(k)| \exp j\phi^{(i)}(k), & k \in U \\ |F^{(i)}(k)| \exp j\phi^{(i)}(k), & k \notin U \end{cases} \]  

(3)

where \( |F^{(i)}(k)| \) is the square-root of the experimentally measured diffraction intensity, \( |F^{(i)}(k)| \) and \( \phi^{(i)}(k) \) are the magnitude and phase of the Fourier transform of \( \rho^{(i)}(r) \), respectively. For a more intuitive presentation, we depict in figure 1 the workflow of the herein developed LR-guided (LRG) phase retrieval algorithm.

3. Simulation

We cropped an image (‘Two macaws’) from the Kodak Lossless True Color Image Suite and resampled to \( 255 \times 255 \) pixels (figure 2(a)), then zero-padded to \( 1275 \times 1275 \) pixels to simulate \( 5 \times \) oversampling along each dimension. We assume the photon density of \( 1 \times 10^7/\text{pixel} \) at the centre of the diffraction pattern and use Poisson statistics to simulate the quantum noise. At the centre of the diffraction pattern \( 11 \times 11 \) pixels were removed to mimic the missing centre with \( \eta = 1 \), i.e. missing 1 wave component. Formally, the number of missing waves is formulated as \( \eta = (D - 1)/(2\xi) \), where \( D \) is the number of pixels along each dimension of the missing data region and \( \xi \) is the oversampling ratio, as defined by Miao et al in [24].

An exact support was used in our simulations. The LR image, shown in figure 2(b), was generated by applying a Gaussian kernel in the real-space, with standard deviation \( \sigma = 5 \) along each dimension. For each trial, each phase retrieval algorithm started from the same random phase set and ran until a convergence criterion or the maximum of 1000 iterations is met. The best reconstruction result with each algorithm out of the 25 independent trials was selected for the following discussion.

The reconstruction result using our LRG method (figure 2(d)) clearly reproduces more details than the LR image (figure 2(b)) by means of direct visual assessment. To further quantify the improvement, we use the real-space R-factor function

\[ E_R = \frac{\sum_{r \in S} |\rho(r) - \rho(r)^{(\text{model})}|}{\sum_{r \in S} \rho(r)^{(\text{model})}} \]  

(4)

to evaluate the quality of image \( \rho(r) \) in real-space against the model \( \rho(r)^{(\text{model})} \), which indicates that the LRG result

\[ \text{Figure 2. Simulation results. (a) Ground truth. (b) The LR image. (c) Reconstruction with HIO, } \eta = 1. \text{ (d) Reconstruction with our LR-guided method, with } \eta = 1. \text{ (e) Real space residual (} E_R \text{) versus iteration number (} \eta = 1). \text{ Each trace represents an independent reconstruction. The bold blue and orange trace correspond to reconstruction with the best } E_R \text{ with HIO and LRG, respectively. Only one LRG trace is visible as other traces have similar trends. (f), (g) Reconstruction results with HIO and LRG, respectively, with } \eta = 3. \]
(ER = 0.088) is superior to the HIO result (ER = 0.120). Figure 2(e) shows the ER versus iteration number trace for each independent reconstruction with each algorithm when η = 1. Unambiguously, our method not only converges much faster than HIO, but also is shown to be insensitive to the initial phase guess, i.e. one would only need to run one trial with LRG instead of tens or hundreds of (see supplementary material). Note that the noticeable artefacts in the reconstruction results (e.g. in figures 2(d) and (g)) are in part due to the quantum noise added to the diffraction pattern; we observe decreased ER when the photon flux increases (see supplementary material).

It is of practical importance to evaluate the scenario of more missing data in the centre of the diffraction pattern, which is a common issue in this field and could be restricted by a number of practical constraints. For this purpose, we further increased the area of the missing centre, from η = 1 to η = 3, and conducted the phase retrieval under this condition. The HIO result, as shown in figure 2(f), again failed to faithfully represent the ground truth; whereas our result, as shown in figure 2(g), clearly demonstrates superior quality and the ER value stays almost the same as the case of η = 1.

Intuitively, one would expect that the larger the missing data area, the more difficult to faithfully reconstruct the real-space image. Miao and colleagues suggested that some structural information in the diffraction pattern is permanently lost if η > 1 even if an exactly precise support is used [24]. However, our result in figure 3(a) can show that even with η = 4 our method can deliver recognisable results with ER < 0.2 from a photon limited diffraction pattern shown in figure 3(b). We also tested our algorithm using a number of different LR images with different spatial resolutions as the real-space constraint (figure 3(c)). Strikingly, figure 3(a) reveals that even a dramatically blurred LR image (σ = 20, right in figure 3(c)) still facilitates the LRG approach to deliver a meaningful reconstruction (with ER = 0.177 at η = 4). Figure 3(d) shows close-up views of the centre area in figure 3(b), where different degrees of missing data are exemplified. Remarkably, because LRG allows a larger missing area, for an XCDI experiment, whose theoretical resolution is determined by the geometry of the set-up, one can consider having a larger η to trade for a higher possible resolution at the detector edge.

4. Experiments and results

To provide an experimental proof-of-concept, we performed table-top optical experiments, in which we acquired diffraction and LR images in a serial manner. We used a setup with two bi-convex lenses to relay the image of a 200 µm diameter pinhole onto the sample with unity magnification, as shown in figure 4(a). We imaged one of the pattern groups on a 1951 USAF resolution target (Thorlabs) as described below. An air-cooled CCD camera (Starlight Xpress SXVR-H694) was placed approximately 32 mm downstream of the sample to record diffraction patterns. We took ten images with each of the following exposure times: 100, 1000 and 10 000 ms. Averaged images of each time exposure group were subtracted with
a dark image and then assembled to a high dynamic range diffraction image of 2201 × 2201 pixels. After we had done the measurement of the diffraction patterns, a bi-convex was then inserted between the sample and the detector, as shown in figure 4(b), to capture LR images. The LR image is the average of ten images with exposure time of 1 ms and subtracted with the same dark image. To properly register the LR image, we aligned the peaks in its Fourier transform with the diffraction pattern at \( k_y = 0 \) and \( k_x = 0 \). We then re-scaled the pixel intensity of the LR image by finding a scaling factor \( s \) that minimises the \( L^{-1} \) residual between the measured diffraction pattern and the Fourier transform of the LR image, i.e. minimize \( \sum |F(k) - s \text{FT}\{g(r)\}(k)| \).

We imaged the 114 group on the 1951 USAF resolution target, as shown in figure 5(a). The number 114 indicates that the arrangement of the vertical and horizontal lines is 114 cycles per mm, equivalent to 8.77 \( \mu \)m per cycle. Figures 5(b) and (c) show the LR image we collected with the lens mode depicted in figure 4(b) and its Fourier transform, respectively. We conducted the phase retrieval using the pattern we collected in the diffraction mode (figure 4(a)) with HIO and our LRG algorithm. Figures 5(d) and (e) show unambiguously that our reconstruction result (e) outperforms the HIO result (d) and is capable of correctly resolving the fine horizontal and vertical lines, which are hardly visible in the LR image (figure 5(b)). In contrast, the HIO result is not recognisable, and, in addition, it has the horizontal and vertical lines in a highly distorted arrangement. It is also clear, by comparing figures 5(c) and (f), the Fourier transform of the LR image and the diffraction pattern, that the LR image only has information within a relatively narrow bandwidth. We point out here that in the experiments, we purposely saturated the detector to mimic the realistic experimental conditions, in which the low-\( q \) data is not accessible, as shown in figure 5(f) inset.

5. A proposed X-FEL experimental setup

Our demonstrations in the numerical simulation and the proof-of-concept laser experiment are encouraging. The present imaging method is expected to be directly adaptable in x-ray imaging experiments at x-ray light source facilities. To further explore such a possibility, we simulate a Mimivirus imaging experiment in an X-FEL setup [25, 26]. Figure 6(a) shows a schematic of the proposed diffraction imaging setup, where a focusing element (e.g. a Fresnel zone-plate) is placed in front of the first detector and is used to capture the low-\( q \) component that would otherwise be dumped or blocked. The rear detector records the LR real-space image concurrently with the acquisition of the x-ray diffraction pattern.

Figure 6(b) shows the simulated diffraction pattern that would be recorded by the front detector, e.g. a Rayonix MX340-XFEL detector [27], one of the detectors utilised at LCLS that has 1920 × 1920 pixels with pixel size of 177 \( \mu \)m (4 × 4 binning mode) and a circular hole at its centre to allow the intense x-ray beam to pass through. The shaded region at the centre in figure 6(b) is where diffraction patterns are not recorded because of the circular hole. We assume an x-ray wavelength of 1.03 nm (1.2 keV) and the distance between the sample and the front of the detector to be 1.65 m so the full-period resolution at the edge of the front detector is 10 nm. The zone-plate images the sample onto the rear detector [28, 29]. The electron density profile of the viral capsid is derived from the published Cryo-EM map data (EMD-5039) [30]. We assume the incoming photon flux density \( 2 \times 10^{12} \) photons/\( \mu \)m²/pulse and use Poisson statistics to simulate the quantum noise. Figures 6(c) and (d) show the simulated image that would be collected by the rear detector and its Fourier transform, respectively. Note the image from the rear detector can be registered to the diffraction pattern by the method we
Figure 5. (a) Illustration of the group 114 on a 1951 USAF resolution target. (b) The LR image measured using the diffraction mode depicted in figure 4(b). (c) Fourier transform of (b) (logarithmic scale). (d) Reconstruction using the HIO algorithm. (e) Reconstruction result of our algorithm. (f) Experimentally measured diffraction pattern (logarithmic scale). Inset: zoom-in of the centre area, where the saturated (white) region is treated as missing data during the phase retrieval.

Figure 6. Simulated Mimivirus hybrid imaging experiment. (a) Proposed imaging set-up (not drawn to scale). (b) Simulated diffraction pattern of the mimivirus at the front detector in (a). The shaded circle marks the missing data area. (c) Fourier intensity of the LR image, i.e. (e). (d) Model derived from the projection of the published EM data [30]. (e) LR image, as if the real image recorded by the rear detector in (a). (f) Result with HIO. (g) Reconstruction with our LRG method, starting from the same random phase as that used for HIO. (h) Comparison of line profiles in the model (d), the LR image (e), and LRG reconstruction result (g). (i) Comparison of Fourier spectra at \( k_y = 0 \). The shaded region indicates the missing data area corresponding to the circle in (b).
described in the previous section to ensure that they are consistent with each other. To simulate limited resolution caused by the limited numerical aperture and the radius of the outermost ring of the zone plate (as shown in figure 6(a)), we low-pass filter the ground-truth image in the Fourier space with bandwidth $\sigma_F = 0.02 \text{ px}^{-1}$ (equivalent to a point spread function in real-space with full width at half maximum of 65 nm), then perform an inverse Fourier transform to obtain the real-space image as shown in figure 6(e). We use a circular loose support and fixed set of random phases for phase retrieval.

Figures 6(f) and (g) show the reconstruction results by using HIO and LRG, respectively. The LRG result, with error $E_R = 0.0345$, not only outperforms the traditional method, but also retrieves details that are apparently not available in the LR image ($E_R = 0.147$, figure 6(e)). To further highlight the capability of our method in recovering the missing low-q data, we compare the line profiles of the LR image and our reconstruction result (figure 6(h)). Our LRG algorithm is capable of recovering the Fourier intensity in the missing centre by incorporating the LR image and reconstructing high resolution details, as shown in figure 6(i).

6. Discussion

Our method makes use of an LR real-space image to deliver a successful image reconstruction. At one extreme, if the a priori information barely provides the shape of the sample, it is similar to running the HIO with a tight support and an advanced phase retrieval capable of generating an estimated support. On the other hand, if the a priori information is as good as the ground truth would be, then the algorithm would converge in a few steps without providing much improvement. Our algorithm leverages an FOM function to assess the reconstruction quality on-the-fly and, thus, can adaptively control the strength that the LR constraint is applied. As we demonstrated in figure 3(a), even a considerably coarse LR image can be used to facilitate the phase retrieval, especially when a substantial area of low resolution data in diffraction patterns is lost.

We emphasise that the use of low resolution real-space images for XCDI had been previously demonstrated. Pioneers of XCDI used the Fourier transform of a low resolution image as a patch to circumvent the missing centre [11], although later, researchers have also learned that the missing intensities at the beam stop region can be set as variables and solved by the phase retrieval algorithms [24]. Additionally, electron microscopists have also used the Fourier transform of a LR image to enhance the electron diffraction images [31]. Unlike other work, here we use the LR image in real-space and use it throughout the iterations to guide the phase retrieval, not just as the initial guess. The advantage of our approach is twofold: leveraging the LR image itself, rather than its Fourier transform, largely reduces the burden of stitching and rescaling in the reciprocal space; our adaptive constraint ensures that the a priori information will not be washed away until the algorithm has significantly converged. Beyond X-FEL and electron microscopy, our method can potentially be applied to other instruments with little modification, such as a multipass microscope, where real and diffractive images are readily available from its separate ports [32].

7. Conclusion

In this paper we demonstrated a novel coherent diffraction imaging approach that takes advantage of a real-space LR image as the a priori information to enhance the quality and robustness of the phase retrieval. Simulation and proof-of-concept experimental results show that the reconstructed images by our approach are superior to the results by the conventional phasing method. Importantly, our method works even in considerably adverse conditions, i.e. large missing centre and limited photon fluxes and can in turn pave ways toward XCDI with higher resolution as the missing wave constraint can be traded off. The present approach could potentially find applications at synchrotron radiation and X-FEL beamlines, providing a robust, high-resolution diffraction imaging solution, owing to its invulnerability to the missing data.

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