Coherent GISAXS Scattering and Phase Retrieval Using DWBA

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Abstract

We have developed a 3 dimensional Coherent Diffraction Imaging algorithm to retrieve phases of diffraction patterns of samples in Grazing Incidence Small Angle X-ray Scattering (GISAXS) experiments. The algorithm interprets the diffraction patterns using the Distorted-Wave Born Approximation (DWBA) instead of the Born Approximation (BA), as in this case the existence of a reflected beam from the substrate causes the diffraction pattern to deviate significantly from the simple Fourier transform of the object. Detailed computer simulations show that the algorithm works. Verification with real experiments is planned.

1 Introduction

Coherent Diffraction Imaging (CDI) with X-Rays has become an important area of synchrotron-based research, with promising applications in materials science, nanoscience, and the biological sciences. The technique can be applied to image objects down to the 10nm length scale. It is based on retrieving the phase information from an oversampled [1] diffraction pattern using iterative methods. Most CDI studies have been carried out in transmission geometry in 2 dimensions (2D) [2]. In such a case, the 2D diffraction pattern is the Fourier transform of the sample normal to the beam direction, according to the Born Approximation (BA).

3 dimensional (3D) CDI in transmission geometry is also possible via tomography by rotating the sample multiple times about an axis normal to the beam during scattering. The first experimental demonstration of 3D CDI was published by Miao et.al [3], in which a 3D diffraction pattern was assembled from a series of 2D diffraction patterns. Later on, Chapman et.al [4] used a similar method to
reconstruct 3D images ab initio. In the same year, Miao et al. [5] succeeded in getting multiple 2D projected images in object space using 2D CDI at different rotation angles and tomographically computing them to a 3D image. For single crystals, 3D CDI was extended to map out the internal strain field inside the crystal [6]. For a review of CDI, see Ref. [7].

However, there are several cases where it is desirable to employ GISAXS, such as where the sample is deposited on an opaque substrate, as in the case of a thin film, or on a liquid surface and to scatter in reflection geometry at low angles of incidence into a 2D detector (Fig. 1). The GISAXS method has become increasingly popular for studying systems such as quantum dots and nanoparticles in thin films, supported catalyst materials, integrated circuits, etc. (For a review, see Ref. [8].)

A problem arises for scattering at angles close to or smaller than the critical angle for total reflection of the substrate, which is how to deal with the problem of the specularly reflected beam from the substrate which interferes coherently with the scattering from the desired object. The standard method of dealing with this is to describe conventional off-specular scattering at grazing incidence has been to use the Distorted-Wave Born Approximation (DWBA) which takes into account the specularly reflected beam [8–10]. In this paper, we address the problem of retrieving the phases of the diffraction patterns using the DWBA rather than the simple BA and applying it to the CDI algorithms.

2 Distorted-Wave Born Approximation

Given the electron density \( f(\mathbf{r}) \) of an object, its Fourier transform \( F(\mathbf{q}) \) is:

\[
F(\mathbf{q}) = \sum_{x=0}^{N_x-1} \sum_{y=0}^{N_y-1} \sum_{z=0}^{N_z-1} f(\mathbf{r}) e^{-2\pi i \left( \frac{q_x}{N_x} x + \frac{q_y}{N_y} y + \frac{q_z}{N_z} z \right)} \quad (1)
\]

where \( \mathbf{r} = (x, y, z) \), the spatial coordinates in object space, and \( \mathbf{q} = (q_x, q_y, q_z) \), the wavevector transfer in reciprocal space. \( \mathbf{q} \) and \( \mathbf{r} \) are discretized and range in each dimension from 0 to \( N_x - 1, N_y - 1, N_z - 1 \), respectively. \( f(\mathbf{r}) \) and \( F(\mathbf{q}) \) are periodic with a period of \( N_x, N_y, N_z \) in the \( x, y, z \) and \( q_x, q_y, q_z \) directions, respectively.

The BA states that the differential cross section is:

\[
\frac{d\sigma}{d\Omega} = r_e^2 |F(\mathbf{q})|^2 \quad (2)
\]

where \( r_e \) is the Thompson scattering length and \( F(\mathbf{q}) \) is also known as the BA form factor.

In reflection geometry, at angles close to or smaller than the critical angle of the substrate, the BA breaks down, as the specularly reflected beam from the substrate interferes coherently with the scattering from the desired object. We assume that the objects to be imaged are contained within a single layer on top of a smooth substrate and introduce the single-layer DWBA diffuse (specular part is omitted because we assume that it is blocked by the beamstop) differential cross
section expression [10]

\[
\left( \frac{d\sigma}{d\Omega} \right)_{\text{diff}} \approx i e^2 |G(q_{||}, \alpha^i, \alpha^f)|^2 \tag{3}
\]

where \( \alpha^i, \alpha^f \) are incident and outgoing angles shown in Fig. 1, and \( q_{||} = (q_x, q_y) \). The DWBA form factor \( G(q_{||}, \alpha^i, \alpha^f) \) is

\[
G(q_{||}, \alpha^i, \alpha^f) = \sum_{m=1}^{4} D^m F(q^m_{||}, q_{||}) \tag{4}
\]

where the \( D^m \) are defined as:

\[
D^1 = T^f T^i, \quad D^2 = R^f R^i, \quad D^3 = T^f R^i, \quad D^4 = R^f T^i \tag{5}
\]

where \( T^i, T^f, R^i, R^f \) are Fresnel transmission and reflection coefficients for reflectivity at the interface between the top layer and substrate. Details about the coefficients can be found in Ref. [11]. \( q^m_{||} \) has been split into components \( q^m_z \) and \( q_{||} \),

\[
q^1_|| = (q_{||}, q^1_z), \quad q^2_|| = (q_{||}, q^2_z), \quad q^3_|| = (q_{||}, q^3_z), \quad q^4_|| = (q_{||}, q^4_z) \tag{6}
\]

and

\[
q^1_z = k^f_z - k^i_z, \quad q^2_z = -k^f_z - k^i_z, \quad q^3_z = -q^2_z, \quad q^4_z = -q^1_z \tag{7}
\]

Note that, the value of \( k^i_z \) is negative, since the vector \( \mathbf{k}^i \) is pointing down.

The complete wavevector transfer [11, 12] from the angles shown in Fig. 1 is defined by the 6 elements of the left-hand-side vector:

\[
\begin{bmatrix}
q_x \\
q_y \\
k^i_z \\
k^f_z \\
k^i_{z,sub} \\
k^f_{z,sub}
\end{bmatrix} = k_0 \begin{bmatrix}
\cos(\alpha^f) \cos(\chi) - \cos(\alpha_i) \\
\sin(\chi) \cos(\alpha^f) \\
-\sqrt{n^2_{top} - \cos^2(\alpha^i)} \\
\sqrt{n^2_{top} - \cos^2(\alpha^i)} \\
-\sqrt{n^2_{sub} - \cos^2(\alpha^i)} \\
\sqrt{n^2_{sub} - \cos^2(\alpha^i)}
\end{bmatrix} \tag{8}
\]

where \( k_0 \) is the incident X-ray wavevector, \( n_{top} \) and \( n_{sub} \) are the complex refractive indices of the top layer and substrate. In this paper, the top layer is air and \( n_{top} = 1 \). \( k^i_{z,sub} \) and \( k^f_{z,sub} \) are the \( z \) component of wavevectors in the substrate, which together with \( k^i_z \) and \( k^f_z \) are used to calculate the Fresnel coefficients in Eq. (5).

Compared to the BA form factor \( F(q) \), the DWBA form factor \( G(q_{||}, \alpha^i, \alpha^f) \) has 4 Fourier transform terms, which correspond to 4 scattering processes. It is interesting to note that the DWBA automatically involves negative values of \( q_z \) in the Fourier transforms of the electron density of the object. These are generally not directly measured in a conventional GISAXS experiment. Details of the 4 scattering processes are illustrated in Fig. (41) in Ref. [8].

Note that, if the sample is a uniform thin film (2D sample), DWBA is not needed. Because \( F(q_x, q_y, q^1_z) = F(q_x, q_y, q^2_z) = F(q_x, q_y, -q^2_z) = F(q_x, q_y, -q^1_z) \) since the Fourier transform of the electron density is independent of \( q_z \), and the DWBA form factor is proportional to the BA form factor. If
the sample has a uniform finite thickness [13], 
$(q_x, q_y)$ and $q_z$ are separable, $F(q_x, q_y, q_z) = F_1(q_x, q_y)F_2(q_z)$. The BA form factor in the $(q_x, q_y)$ plane is independent of that in the $q_z$ direction.

Figure 1: scattering layout.

3 Coherent Diffraction Imaging Algorithm

Several iterative algorithms have been developed to retrieve the phases of the scattering magnitudes $|F(q)|$ based on the principle of oversampling of the scattered intensities of the diffraction patterns [14]. Fig. 2a is an example of the Hybrid-Input-Output (HIO) [15] algorithm. By iterating step 1 to 4 in Fig. 2a, the phases $\Phi(q)$ will converge in a few thousand iterations. Other algorithms, such as Error Reduction (ER) [1,15,16], Relaxed Averaged Alternating Reflection [17] and Difference Map [18] replace the corresponding equations in step 2 [14]. In this paper, we combined the HIO with the ER. Details are given in Section 4.

The algorithm shown in Fig. 2a first needs a random guess of the phases of the scattering magnitudes, and then forces the electron density of the sample $f(r)$ to satisfy the constraints in object space for every iteration. Eventually, the phases of the BA form factor $F(q)$ are retrieved; however, it fails near grazing incidence because of the presence of the reflected wave from the substrate as already discussed in Section 1 and 2. We can use a similar algorithm to retrieve the phase of the DWBA form factor, except for the complication that to relate it to the Fourier transform of the object density, we need to solve for the 4 different terms in Eq. (4).

We proposed a matrix method to solve the more complicated DWBA form factor, which has 4 Fourier transform terms with different $q_z$’s. The matrix method is based on the fact that $q_z^1$ to $q_z^4$ are partially overlapped when we have multiple incident and outgoing angles; for example, $q_z^1(\alpha^i = 0.1deg, \alpha^f = 0.2deg) = q_z^2(\alpha^i = 0.4deg, \alpha^f = 0.1deg)$. Given enough incident and outgoing angle pairs, we can relate the DWBA form factor to the BA form factor (Fourier transform of the electron density) via a matrix, and use the matrix to solve the BA form factor in terms of the DWBA form factor (the form factor is a data array, not a scalar). The iterative algorithm is modified to be in Fig. 2b. Steps 2 and 4 remain the same while steps 1 and 3 are replaced with the combination of Fourier transform and DWBA matrix multiplications.

To solve the matrix $A$ shown in Fig. 2b, which is crucial to this DWBA-CDI algorithm, let us expand Eq. (4):
Figure 2: (a) regular CDI iterative steps [19]. (b) DWBA-CDI iterative steps proposed in this paper. For both figures, S in step 2 is the set of elements inside the support, and n is the iteration number. A value of the feedback parameter of $\beta=0.9$ is used. $|F(q)|$ in a and $|G(q)|$ in b in step 1 are the measured scattering magnitudes in step 0. $|G(q)|$ is short of $|G(q_x, q_y, \alpha^i, \alpha^f)|$.

We assume we have the scattering magnitudes (the DWBA form factor magnitudes in this case) $G(q_x, q_y, k^i_x, k^f_x)$ at equally spaced $q_x, q_y, q^1_z$ to $q^4_z$, which can be achieved through interpolation in the $q_x, q_y$ directions, by rotating the sample azimuthally. We have to choose suitable incident and outgoing angles to equally space $q^1_z$ to $q^4_z$. To be clear, $G(q_x, q_y, \alpha^i, \alpha^f)$ is also written as $G(q_x, q_y, k^i_x, k^f_x)$ in the context; so is $D^m(k^i_x, k^f_x)$ and $D^m(\alpha^i, \alpha^f)$.

According to Eq. (1), $F(q_x, q_y, q_z)$ is discretized and has a period of $N_z$ in the $q_z$ direction. Thus, $\pm q^1_z$ and $\pm q^2_z$ are all in the range of $[0, N_z - 1]$. At fixed $q_x, q_y$, $F(q_x, q_y, \pm q^1_z)$ and $F(q_x, q_y, \pm q^2_z)$ are subsets of $F_{N_z-1}$, defined in Eq. (10). $G$ and $F$ are defined as column vectors:

$$G = D^1(k^i_x, k^f_x)F(q_x, q_y, q^1_z) + D^2(k^i_x, k^f_x)F(q_x, q_y, q^2_z) + D^3(k^i_x, k^f_x)F(q_x, q_y, -q^2_z) + D^4(k^i_x, k^f_x)F(q_x, q_y, -q^1_z)$$

(9)

$$G = \begin{bmatrix} G(q_x, q_y, \alpha^i_1, \alpha^f_1) \\ \vdots \\ G(q_x, q_y, \alpha^i_M, \alpha^f_M) \\ F(q_x, q_y, 0) \\ \vdots \\ F(q_x, q_y, N_z - 1) \end{bmatrix}_{M 	imes N_z + 1}$$

(10)

We have the matrix form of Eq. (9) for each $(q_x, q_y)$ pair:

$$G_{M \times 1} = A_{M \times N_z}F_{N_z \times 1}$$

(11)

$A_{M \times N_z}$ can be calculated by plugging Eq. (9) into Eq. (11). $A_{M \times N_z}$ is independent of $(q_x, q_y)$, while $G$ and $F$ are not. That is, we need to calculate the matrix $A$ only once for all the $(q_x, q_y)$ pairs. Given the magnitudes and phases of $G$, Eq. (11) can then be
inverted via the least square method [20] if \( M \geq N_z \) and matrix \( A \) is non-singular:

\[
F = (A^H A)^{-1} A^H G
\]  

(12)

where \( A^H \) is the conjugate transpose (Hermitian transpose) of matrix \( A \).

Typically, the matrix \( A \) is singular (\(|A^H A| = 0\)), even when \( M \) is greater than \( N_z \). The least square method does not work for Eq. (12). Because at incident and outgoing angles greater than 2 times of the substrate’s critical angle (maximum of incident and outgoing angles have to be large enough to satisfy the condition \( M > N_z \)), \( D^4 \) is too small and \( F(q_x, q_y, -q_z^1) \) can not be calculated accurately via the least square method. We introduce the Friedel’s Law to fix the problem.

The imaginary part of the sample electron density is neglected, since it is much smaller than the real part. The Friedel’s Law states:

\[
F(-q_x, -q_y, -q_z) = F^*(q_x, q_y, q_z)
\]  

(13)

where \( F^* \) is the complex conjugate of \( F \). By the way, the Friedel’s Law is also necessary for 3D diffraction imaging in reflection geometry even using the BA. Because it gives the scattering information in the negative \( q_z \) direction, which BA does not.

Here, let us use the new notation at fixed \( q_x, q_y \):

\[
G^+ = G(q_x, q_y, \alpha_i^i, \alpha_i^f),
\]

\[
G^- = G(-q_x, -q_y, \alpha_i^i, \alpha_i^f)
\]  

(14)

where \( G^+ \) is the same as the \( G \) in Eq. (9) and \( G^- \) in Eq. (10). According to Eq. (9) and Eq. (15), both \( G^+ \) and \( G^- \) are functions of \( Re[F_{N_z}] \) and \( Im[F_{N_z}] \). By calculating the real and imaginary parts of \( G^+ \) and \( G^- \), we get:

\[
\begin{bmatrix}
Re[G^+]
\Im[G^+]
\Re[G^-]
\Im[G^-]
\end{bmatrix}
= \begin{bmatrix}
A_1 & A_2 \\
A_3 & A_4 \\
A_5 & A_6 \\
A_7 & A_8
\end{bmatrix}
\begin{bmatrix}
Re[F] \\
Im[F]
\end{bmatrix}
\]  

(16)

Define:

\[
A = \begin{bmatrix}
A_1 & A_2 \\
A_3 & A_4 \\
A_5 & A_6 \\
A_7 & A_8
\end{bmatrix}_{4M \cdot 2N_z}
\]  

(17)

The matrices \( A_1 \) to \( A_8 \) have size of \( M \cdot N_z \). \( A \) is not singular, if \( M \) is big enough. Details of calculating \( A_1 \) to \( A_8 \) are given in supplementary document.

Finally, given \(|G(q)|\) and retrieving the corresponding phases \( \Phi(q) \) via DWBA-CDI algorithm, \( F(q) \) can be calculated via the least square method.

4 Simulation Result

We chose a sample made up of random solid shapes made of gold, sitting on a Silicon substrate in a certain area, shown in
Fig. 1. Fig. 3a represents the 3D angle-view of the sample. The X-ray energy was chosen as 8.04 keV, with a wavelength of 1.54 Å.

For the simulation, we have assumed that the sample is small enough to always remain inside the footprint of the beam and that the photon flux is uniform across the beam. The diffuse scattering from the Si surface was taken to be negligible. And there is a beam-stop (not shown in the figure) on the detector center, to block the specular scattering from Si surface. The beamstop size in reciprocal space is \(\Delta q_x = \Delta q_y \approx 2 \cdot 10^{-4} \text{Å}^{-1}, \Delta q_z \approx 10^{-3} \text{Å}^{-1}\) (1 pixel size in the \(q_x, q_y, q_z\) directions). The error contributed from the beamstop is very small and neglected here. The contribution of error from the beamstop is well discussed in Ref. [21].

Incident angles were taken to be from 0.02° to 0.65° with a step of 0.02°. The Si critical angle is 0.223°. For each incident angle, the sample is rotated azimuthally in 500 steps from 0° to 180°. After getting 32*500 diffraction patterns, we interpolate them to a 3D diffraction pattern in reciprocal space.

To speed up the program in Fig. 2b, we applied the non-negative constraint (ER) of electron density in step 2 every 60 iterations [14] and update the support every 100 iterations following the Shrink-wrap method [22]. Total number of iterations is 3,000. The reconstructed 3D image is shown in Fig. 3b. In comparison, we also reconstructed the image using the BA, shown in Fig. 3c. We also tried the Oversampling Smoothness algorithm [23], which performs equally well (see supplementary for comparison between HIO-ER which is used in the paper and HIO-OSS).

We added Gaussian noise to the simulated diffraction pattern, defined as:

\[
\text{noise} = \frac{\text{signal}}{\text{SNR}} \times \text{random}
\]

where SNR is the signal-to-noise ratio, we chose \(\text{SNR} = 10\) in the paper; \(\text{random}\) are Gaussian random numbers \(\sim N(0, 1)\). An example of diffraction pattern at incident \(\alpha_i = 0.2^\circ\) is shown in Fig. 3d. The error function is calculated as:

\[
\text{Error} = \sqrt{\frac{\sum_q (|F_s(q)| - |F_r(q)|)^2}{\sum_q (|F_s(q)| + |F_r(q)|)^2}}
\]

where \(|F_s(q)|\) and \(|F_r(q)|\) are the magnitudes of the simulated and reconstructed BA form factors. The reason we use the magnitude of Fourier transform to calculate the error is that translational shift of the whole objects between original and reconstructed images does not affect the error.

In order to find the best reconstruction, we ran the code multiple times and chose the result with the smallest error, which is around 18%.

The reconstructed shapes using the DWBA in Fig. 3b is clear with sharp edges and of the same sizes as the original. The relative distances between shapes remain the same as the original image. The relative locations of the shapes in the z direction are correct. However, reconstruction using the BA is totally a failure; and the iterations do not converge. Note that, people have successfully reconstructed images in reflection geometry...
before [13, 24]; but these have been quasi-2-dimensional. This is the first time that we have shown we can reconstruct detailed structure of the complex objects in the z direction using reflection geometry. Note that, the reconstruction result may have an arbitrary translational shift of the original image in the x and y directions, due to the fact that arbitrary translational shift of the whole objects does not affect the diffraction pattern in these directions. However the substrate acts as a marker in the z direction so there is no arbitrary translation of the image in this direction.

Other than the isolated shapes, we also tested the reconstruction of a single box with a rough top surface, as shown in Fig. 4. The algorithm works for a single rough surface with limitations, i.e the rough surface needs to be smaller than the footprint of the beam, and the roughness needs to be bigger than the resolution in the z direction in object space (which is limited by the largest incident and outgoing angles).

By the definition of discrete Fourier transform Eq. (1), object space resolution (pixel size in object space), the maximum object space size, the reciprocal space resolution (pixel size in the Fourier space), and the maximum reciprocal space size is related as (in the z and qz direction as an example):

$$q_{z,max} \Delta z = \pi$$
$$z_{max} \Delta q_z = 2\pi$$

(20)

where $$q_z \in [-q_{z,max}, q_{z,max}]$$ and $$z \in [0, z_{max}]$$. $$\Delta q_z$$ is the resolution in the $$q_z$$ direction in

Figure 3: 3D angle-view of the a original sample (substrate not shown here), b reconstructed sample using the DWBA and c reconstructed sample using the BA; d A diffraction pattern example of the sample at the incident angle 0.2 degree.
the reciprocal space, $q_{z,\text{max}}$ is the maximum of $q_z$, and the same for the $z$ direction in object space. From the above equation, it is easy to find out that the resolution in the $z$ direction is determined by the maximum of $q_z$, which is determined by Eq. (7) and (8). In our simulation, $n_{\text{top}} = 1$, $q_{z,\text{max}} = k_0 (\sin \alpha_{\text{max}}^i + \sin \alpha_{\text{max}}^f)$. In short, the resolution in the $z$ direction is determined by the largest incident and outgoing angles. In a real experiment, the diffraction intensity at high incident and outgoing angles is too low to be detected, thus limit the $z$ resolution to be around $10\text{nm}$, while the theoretical resolution is about the wavelength of the incident X-ray (typically smaller than $1\text{nm}$ for hard X-rays).

The algorithm is performed via GPU computing. Software: Matlab 2022a, Windows 10. Hardware: Nvidia RTX 2080. Image resolution: 256*256*64 (in pixels). The algorithm takes less than 1 hour.

5 Conclusion

The algorithm succeeds in reconstructing the 3D image at grazing incidences, while regular CDI using BA does not. As we can reach to much smaller incident and outgoing angles, as well as $|q_z^2| < |q_z^1|$, the minimum of $q_z$ in DWBA is much smaller. That is, DWBA is able to reconstruct much thicker sample. BA cannot in principle reconstruct objects thicker than around $5\text{nm}$. The results obtained from computer simulations of the scattering and reconstruction are very encouraging in showing that the method works.

Figure 4: 3D angle-view of the a original box with rough top surface (substrate not shown here) and b reconstructed sample using the DWBA.
Verification with real experiments is planned.

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Supplementary Materials

Derivation of the DWBA Matrix

Here we show how to calculate $A_1 \sim A_8$ from Eq. (9) and (14) in the main paper. Let us first rewrite the Eq. (9) and (14) in the main paper, at fixed $q_x, q_y$:

\begin{align}
G^+ &= D_1 F[q^1_z] + D_2 F[q^2_z] + D_3 F[-q^2_z] + D_4 F[-q^1_z] \\
G^- &= D_1 F^*[q^1_z] + D_2 F^*[q^2_z] + D_3 F^*[q^2_z] + D_4 F^*[q^1_z]
\end{align}

where $G^+, G^-, D_m, F[q^m_z]$ are all functions of incident and outgoing angles $(\alpha^i, \alpha^f)$.

If there are $M$ pairs of $(\alpha^i, \alpha^f)$ at fixed $q_x, q_y$, there are $M$ equations for both $G^+$ and $G^-$. We can rewrite Eq. (1) and (2) in matrix forms. Define $G^+, G^-, D_m F[q^m_z]$ and $D_m F[-q^m_z]$ as vectors of size $M \cdot 1$, corresponding to the number of $(\alpha^i, \alpha^f)$, and $F$ as a vector of size $N_z \cdot 1$, corresponding to all the different $q_z$’s (the number of $q_z$’s $N_z$ is dependent of $M$). It is easy to calculate the matrices $D^m_{\text{mat}}$ and $D^m_{\text{mat}}'$ with sizes of $M \cdot N_z$, such that:

\begin{align}
D^m F[q^m_z] &= D^m_{\text{mat}} F \\
D^m F[-q^m_z] &= D^m_{\text{mat}}' F
\end{align}

Below is an example of calculating the matrix $D^1_{\text{mat}}$. To simplify the example, we choose 4 incident and outgoing angles (see Table. 1). We assume $\alpha^i \geq \alpha^f (q^2_z \geq 0)$; because swapping incident and outgoing angles does not change the DWBA form factor.

From the angle table, we see that there are 10 angle pairs ($M = 10$) and 17 $q_z$’s ($N_z = 17$). The corresponding $D^m F[q^m_z]$ ($m = 1, 2, 3, 4$) can be
Table 1: Angle pairs and corresponding wavetransfers. The incident and outgoing angles, with a unit of $\Delta \alpha$. The wavetransfer $q_z$’s have a unit of $\Delta q = k_0 \sin \Delta \alpha$, where $k_0$ is the incident X-ray beam wavevector.

| $\alpha^i(\Delta \alpha)$ | $\alpha^f(\Delta \alpha)$ | $q_1^z(\Delta q_z)$ | $q_2^z(\Delta q_z)$ | $q_3^z(\Delta q_z)$ | $q_4^z(\Delta q_z)$ |
|--------------------------|--------------------------|--------------------|--------------------|--------------------|--------------------|
| 1                        | 1                        | 2                  | 0                  | 0                  | -2                 |
| 2                        | –                        | –                  | –                  | –                  | –                  |
| 3                        | –                        | –                  | –                  | –                  | –                  |
| 4                        | –                        | –                  | –                  | –                  | –                  |
| 2                        | 1                        | 3                  | 1                  | -1                 | -3                 |
| 2                        | 4                        | 0                  | 0                  | -4                 |                    |
| 3                        | –                        | –                  | –                  | –                  | –                  |
| 4                        | –                        | –                  | –                  | –                  | –                  |
| 3                        | 1                        | 4                  | 2                  | -2                 | -4                 |
| 2                        | 5                        | 1                  | -1                 | -5                 |                    |
| 3                        | 6                        | 0                  | 0                  | -6                 |                    |
| 4                        | –                        | –                  | –                  | –                  | –                  |
| 4                        | 1                        | 5                  | 3                  | -3                 | -5                 |
| 2                        | 6                        | 2                  | -2                 | -6                 |                    |
| 3                        | 7                        | 1                  | -1                 | -7                 |                    |
| 4                        | 8                        | 0                  | 0                  | -8                 |                    |

written as column vectors with sizes of 10 · 1 (corresponding to the 10 rows in the table):
The parameters $D^m = D^m(\alpha^i, \alpha^j)$ are functions of incident and outgoing angles (see Eq. (5) in the main paper for details). $D^1 F[q^1_2]$ can be written as the matrix form (the same applies to $D^2 F[q^2_2], D^3 F[q^3_2]$ and $D^4 F[q^4_2]$):

$$D^1 F[q^1_2] = \begin{bmatrix} D^1(1, 1)F[2] \\ D^1(2, 1)F[3] \\ D^1(2, 2)F[4] \\ D^1(3, 1)F[4] \\ D^1(3, 2)F[5] \\ D^1(3, 3)F[6] \\ D^1(4, 1)F[5] \\ D^1(4, 2)F[6] \\ D^1(4, 3)F[7] \\ D^1(4, 4)F[8] \end{bmatrix} = \mathbf{D}^1_{\text{mat}} \begin{bmatrix} F[8] \\ F[-8] \\ F[-7] \\ \vdots \\ F[0] \\ F[1] \\ \vdots \\ F[8] \end{bmatrix}$$

where the column vector on the right hand side is column vector $\mathbf{F}$ with size of $17 \cdot 1$, and $\mathbf{D}^1_{\text{mat}}$ is a matrix with size of $10 \cdot 17$. It is pretty easy to calculate
the elements of matrix $D_{\text{mat}}^1$:

\[
\begin{align*}
D_{\text{mat}}^1\{1,11\} &= D^1(1,1), \\
D_{\text{mat}}^1\{2,12\} &= D^1(2,1), \\
D_{\text{mat}}^1\{3,13\} &= D^1(2,2), \\
D_{\text{mat}}^1\{4,13\} &= D^1(3,1), \\
D_{\text{mat}}^1\{5,14\} &= D^1(3,2), \\
D_{\text{mat}}^1\{6,15\} &= D^1(3,3), \\
D_{\text{mat}}^1\{7,14\} &= D^1(4,1), \\
D_{\text{mat}}^1\{8,15\} &= D^1(4,2), \\
D_{\text{mat}}^1\{9,16\} &= D^1(4,3), \\
D_{\text{mat}}^1\{10,17\} &= D^1(4,4)
\end{align*}
\] (7)

The rest of the elements in the matrix are zeros. See Eq.(5) in the main paper for getting the parameters on the right hand side of the above equations.

The example can be used to similarly calculate the matrices $D_{\text{mat}}^1$ to $D_{\text{mat}}^4$ and $D_{\text{mat}}^{1'}$ to $D_{\text{mat}}^{4'}$ when $\alpha^i, \alpha^f = 1, 2, 3, 4$. Actually, we cannot use this DWBA matrix $A$ to get the BA form factor; because $M < N_z$ (less equations than unknown variables). In the main paper, we chose $M = 272, N_z = 64$, to guarantee $M > N_z$ and $|A^H A| \neq 0$. In addition, we use fast Fourier transform in the paper, which requires the $q_z$’s to be non-negative. We can assume the Fourier transform of the sample to be periodic in the $q_x, q_y$ and $q_z$ directions.

Next, let us calculating the matrices $A_1$ to $A_8$ from the matrices $D_{\text{mat}}^m$ and $D_{\text{mat}}^{m'}$. The Eq. (1) and (2) can be written in the matrix forms:

\[
\begin{align*}
G^{+}_{M,1} &= D_{\text{mat}}^1 F[q_z]_{N_z,1} + D_{\text{mat}}^2 F[q_z]_{N_z,1} + D_{\text{mat}}^3 F[q_z]_{N_z,1} + D_{\text{mat}}^4 F[q_z]_{N_z,1} \\
G^{-}_{M,1} &= D_{\text{mat}}^{1'} F^*[q_z]_{N_z,1} + D_{\text{mat}}^{2'} F^*[q_z]_{N_z,1} + D_{\text{mat}}^{3'} F^*[q_z]_{N_z,1} + D_{\text{mat}}^{4'} F^*[q_z]_{N_z,1}
\end{align*}
\] (8)

Define:

\[
\begin{align*}
D^+ &= D_{\text{mat}}^1 + D_{\text{mat}}^2 + D_{\text{mat}}^3 + D_{\text{mat}}^4 \\
D^- &= D_{\text{mat}}^{1'} + D_{\text{mat}}^{2'} + D_{\text{mat}}^{3'} + D_{\text{mat}}^{4'}
\end{align*}
\] (10)
$G^+$ and $G^-$ are functions of $Re(F_{N_z \cdot 1})$ and $Im(F_{N_z \cdot 1})$. Next, let us get the real and imaginary part of $G^+$ and $G^-$. We assume $|k^i_z| \geq |k^f_z|$ in the context, as mentioned before in the example ($q^2_z \geq 0$).

\[
Re(G^+) = Re(D^+)Re(F) - Im(D^+)Im(F)
\]

Define:
\[
A_1 = Re(D^+), \quad A_2 = -Im(D^+)
\]

We have:
\[
Re(G^+) = \begin{bmatrix} A_1 & A_2 \end{bmatrix} \begin{bmatrix} Re(F) \\ Im(F) \end{bmatrix}
\]

Similarly,
\[
Im(G^+) = Im(D^+)Re(F) + Re(D^+)Im(F)
\]

\[
A_3 = Im(D^+), \quad A_4 = Re(D^+)
\]

\[
Im(G^+) = \begin{bmatrix} A_3 & A_4 \end{bmatrix} \begin{bmatrix} Re(F) \\ Im(F) \end{bmatrix}
\]

\[
Re(G^-) = Re(D^-)Re(F) + Im(D^-)Im(F)
\]

\[
A_5 = Re(D^-), \quad A_6 = Im(D^-)
\]

\[
Re(G^-) = \begin{bmatrix} A_5 & A_6 \end{bmatrix} \begin{bmatrix} Re(F) \\ Im(F) \end{bmatrix}
\]

\[
Im(G^-) = Im(D^-)Re(F) - Re(D^-)Im(F)
\]

\[
A_7 = Im(D^-), \quad A_8 = -Re(D^-)
\]

\[
Im(G^-) = \begin{bmatrix} A_7 & A_8 \end{bmatrix} \begin{bmatrix} Re(F) \\ Im(F) \end{bmatrix}
\]

Finally, we get the Eq. (16) and (17) in the main paper. To be clear, the real and imaginary part of $G^+$ and $G^-$ have dimensions of $M \cdot 1$. While the real and imaginary part of $F$ have dimensions of $N_z \cdot 1$. $A_1$ to $A_8$ have dimensions of $M \cdot N_z$. 

5
Performance of HIO-OSS and HIO-ER Algorithms

As mentioned in the main paper, the HIO-OSS performs equally as good as HIO-ER; the latter is used in the main paper. We ran the HIO-OSS and HIO-ER algorithms 100 times using the diffraction patterns with $SNR = 5, 10, 20, \infty$ ($SNR = \infty$ means no noise). The error $R_f$ are calculated (see Fig. 1). From the Figures, we learned that the HIO-OSS results have much bigger deviations at different runs at all noise levels. But the minimum $R_f$ is very close to that of HIO-ER result.