

High resolution electron exit wave reconstruction from a diffraction pattern using Gaussian basis decomposition

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Abstract. We describe an algorithm to reconstruct the electron exit wave of a weak-phase object from single diffraction pattern. The algorithm uses analytic formulations describing the diffraction intensities through a representation of the object exit wave in a Gaussian basis. The reconstruction is achieved by solving an overdetermined system of non-linear equations using an easily parallelisable global multi-start search with Levenberg-Marquard optimisation and analytic derivatives.

1. Introduction

Restoration of the exit wave from a diffraction pattern (diffractive imaging or ptychography) has been recognised as a promising technique in electron microscopy capable of providing diffraction resolution limited data and eliminating the need for an objective lens [1]. In addition, recording a single diffraction pattern (DP) requires a considerably lower radiation dose compared to recording a ptychographic series of diffraction patterns or a focal or tilt series of images which is particularly important for radiation sensitive materials and for atomic resolution tomography [2,3].

Currently, the most common algorithm used for restoration of the exit wave from a DP is an iterative numerical algorithm based on the original ideas due to Gerchberg and Saxton [4]. However, in general the restoration is an ill-posed problem of finding a $2n$ -parameter complex wave function from n pixel diffraction data, which in principle can lead to multiple solutions. To avoid this problem the solution space is usually downscaled so that the complex exit wave sought has the same total number of pixels as the input DP. An alternative approach is to obtain DPs from overlapping regions in a ptychographic scheme to provide additional redundancy for the successful restoration [5].

In the present work we test a direct approach for converting a single DP into the corresponding exit wave using a representation of the exit wave in a Gaussian basis. This approach does not require scaling, greatly reduces the number of variables needed to find the solution and allows analytic calculation of the DP and derivatives needed for the restoration process.

2. The exit wave in a Gaussian basis

2.1. The diffracted wave in one dimension

For simplicity of derivation we first consider the exit wave with unity amplitude and phase shift $\phi(x)$ in one dimension as:

$$\Psi(x) = \exp(i\phi(x)) \quad (1)$$



In case of coherent plane wave illumination the Fraunhofer diffracted wave, $D(k)$, is a Fourier Transform (FT) of $\Psi(x)$:

$$D(k) = \int_{-\infty}^{+\infty} \exp(i\phi(x)) \exp(-i2\pi kx) dx \quad (2)$$

Using the Euler identity the diffracted wave can be rewritten as:

$$D(k) = \int_{-\infty}^{+\infty} (\cos(\phi(x)) + i \sin(\phi(x))) \exp(-i2\pi kx) dx = \int_{-\infty}^{+\infty} \cos(\phi(x)) \exp(-i2\pi kx) dx + i \int_{-\infty}^{+\infty} \sin(\phi(x)) \exp(-i2\pi kx) dx \quad (3)$$

We now represent $\phi(x)$ as a sum of Gaussian functions:

$$\phi(x) = \sum_n a_n \exp\left(-\frac{1}{2}(x-b_n)^2 c_n^{-2}\right) = \sum_n G_n(a_n, b_n, c_n, x) \quad (4)$$

The FT of a Gaussian function in its general form is:

$$\int_{-\infty}^{+\infty} a \exp\left(-\frac{1}{2}(x-b)^2 c^{-2}\right) \exp(-i2\pi kx) dx = \cos(2\pi kb) a c \sqrt{2\pi} \exp(-2\pi^2 k^2 c^2) - i \sin(2\pi kb) a c \sqrt{2\pi} \exp(-2\pi^2 k^2 c^2) = \cos G_{1t}(k) - i \sin G_{1t}(k) \quad (5)$$

For simplicity of derivation, we consider the case with $n=2$ where $\phi(x) = G_1 + G_2$:

$$D(k) = \int_{-\infty}^{+\infty} \cos(G_1 + G_2) \exp(-i2\pi kx) dx + i \int_{-\infty}^{+\infty} \sin(G_1 + G_2) \exp(-i2\pi kx) dx = \text{Re}(k) + i \text{Im}(k) \quad (6)$$

The observed diffracted intensity is then calculated as $|D(k)|^2 = \text{Re}(k)^2 + \text{Im}(k)^2$.

Using identities for the sine and cosine of a sum of angles the real part of $D(k)$ becomes:

$$\text{Re}(k) = \int_{-\infty}^{+\infty} \cos(G_1) \cos(G_2) \exp(-i2\pi kx) dx - \int_{-\infty}^{+\infty} \sin(G_1) \sin(G_2) \exp(-i2\pi kx) dx \quad (7)$$

The imaginary part of the $D(k)$ is then:

$$\text{Im}(k) = \int_{-\infty}^{+\infty} \sin(G_1) \cos(G_2) \exp(-i2\pi kx) dx + \int_{-\infty}^{+\infty} \cos(G_1) \sin(G_2) \exp(-i2\pi kx) dx \quad (8)$$

Using first order approximations for the sine and cosine function which are valid for small phase shifts (small a coefficients in the basis Gaussians) and small overlap between the basis Gaussians ($|b_1 - b_2| > c_1$ or c_2) $\sin(G(x)) \approx G(x)$ and $\cos(G(x)) \approx 1$. Ignoring the delta function at $k=0$ it can be shown that the first order approximation of the FT in this case is:

$$D(k) \approx \sin G_{1t}(k) + \sin G_{2t}(k) + i \cos G_{1t}(k) + i \cos G_{2t}(k) \quad (9)$$

The FT of the exit wave represented as a sum of n Gaussians to first order can be then easily calculated as:

$$D(k) \approx \sum_n \sin G_{nt}(k) + i \sum_n \cos G_{nt}(k) \quad (10)$$

2.2. The diffracted wave in two dimensions

A symmetric Gaussian function in two dimensions has the general form

$$G(a, b_x, b_y, c, x, y) = a \exp\left(-\frac{1}{2}((x-b_x)^2 + (y-b_y)^2) c^{-2}\right) \quad (11)$$

Using the expressions obtained for the FT of a general Gaussian in 1D the FT in 2D is:

$$\begin{aligned}
& \int_{-\infty-\infty}^{+\infty+\infty} a \exp\left(-\frac{1}{2}((x-b_x)^2 + (y-b_y)^2)c^{-2}\right) \exp(-i2\pi k_x x) \exp(-i2\pi k_y y) dx dy \\
&= ac^2 2\pi \exp(-2\pi^2 c^2 (k_x^2 + k_y^2)) (\cos(2\pi k_x b_x) \cos(2\pi k_y b_y) - \sin(2\pi k_x b_x) \sin(2\pi k_y b_y)) \\
&\quad - i \sin(2\pi k_x b_x) \cos(2\pi k_y b_y) - i \cos(2\pi k_x b_x) \sin(2\pi k_y b_y) = \text{Re}(k_x, k_y) + i \text{Im}(k_x, k_y) \\
&\quad = \cos \cos G_{It}(k_x, k_y) + i \sin \cos G_{It}(k_x, k_y) \tag{12}
\end{aligned}$$

By analogy to the first order approximation for the DP as a sum of 1D Gaussians the DP of an exit wave represented as a sum of 2D Gaussians in a first-order approximation can then be written as:

$$D(k_x, k_y) = \text{Re}(k_x, k_y) + i \text{Im}(k_x, k_y) \approx \sum_n \sin \cos G_{nt}(k_x, k_y) + i \sum_n \cos \cos G_{nt}(k_x, k_y) \tag{13}$$

The observed DP intensities can then be rewritten as

$$\begin{aligned}
D^2(k_x, k_y) &= \sum_n \cos \cos G_{nt}(k_x, k_y)^2 + \sum_n (\cos \cos G_{nt}(k_x, k_y) \sum_{m \neq n} \cos \cos G_{mt}(k_x, k_y)) \\
&\quad + \sum_n \sin \cos G_{nt}(k_x, k_y)^2 + \sum_n (\sin \cos G_{nt}(k_x, k_y) \sum_{m \neq n} \sin \cos G_{mt}(k_x, k_y)) \tag{14}
\end{aligned}$$

The general expression for the partial derivative of the diffraction pattern intensities computed using the above first-order approximation with the general parameter p ($p = a_n, b_{xn}, b_{yn}, c_n$) of an individual contributing basis Gaussian n in the exit wave is therefore:

$$\begin{aligned}
\frac{\partial D^2(k_x, k_y)}{\partial p_n} &= \frac{\partial \cos \cos G_{nt}(k_x, k_y)^2}{\partial p_n} + 2 \frac{\partial \cos \cos G_{nt}(k_x, k_y)}{\partial p_n} \sum_{m \neq n} \cos \cos G_{mt}(k_x, k_y) \\
&\quad + \frac{\partial \sin \cos G_{nt}(k_x, k_y)^2}{\partial p_n} + 2 \frac{\partial \sin \cos G_{nt}(k_x, k_y)}{\partial p_n} \sum_{m \neq n} \sin \cos G_{mt}(k_x, k_y) \tag{15}
\end{aligned}$$

In practice, the accuracy of the simulated DP can be increased by using higher order approximations for the sine and cosine functions in addition to more Gaussian functions in the basis to represent the exit wave. A straightforward, although slow, implementation of this strategy is to use a numerical FT to compute the DP from an exit wave represented as a sum of Gaussians and also to use numerical derivatives. In this approach the exit wave can be restored by solving an overdetermined system of non-linear equations using an appropriate algorithm.

3. DP simulation and exit wave restoration

To test the approach, an exit wave from a model of a 0.41 nm diameter carbon nanotube fragment with a total of 60 atoms was simulated at 80 kV using the multislice algorithm [6]. The noiseless DP intensities computed from the FT of the exit wave and weighted to remove the central beam were subsequently used as input for the reconstruction code with the Levenbergh-Marquardt optimisation method implemented in MATLAB to solve the system of non-linear equations. These reconstructions are shown in Figure 1.

4. Results and discussion

The solution of the non-linear equations often depends on the initial approximation of the function and it was found that in the present case a scan of initial values is required to provide a high quality restoration. This final restoration shows good agreement with the input exit wave as shown in Figure 2. The computations of the first order approximation with analytic derivatives work well to quickly refine the initial estimate of the exit wave. However, for a better quantitative agreement higher order approximation and larger basis sets with multiple Gaussians were needed. In this particular example we have found that two Gaussian functions per peak were adequate to obtain the restoration shown in Figures 1 and 2, although more Gaussians are needed to reproduce the phase shifts more accurately.

It is well known that Gaussians are appropriate for descriptions of the atomic potential, and therefore, for modelling exit wave shifts. A sum of a number of differently shaped Gaussians is often satisfactory to produce meaningful results that require fitting of the atomic potential as, for example, in quantum chemistry. Moreover, the method described here can be extended to a general object where the amplitude of the exit wave is not unity and other functions, such as Lorentzians, which have analytical FTs and better reflect the behaviour of atomic electron density in the vicinity of the nucleus, can be used to better represent the exit wave.

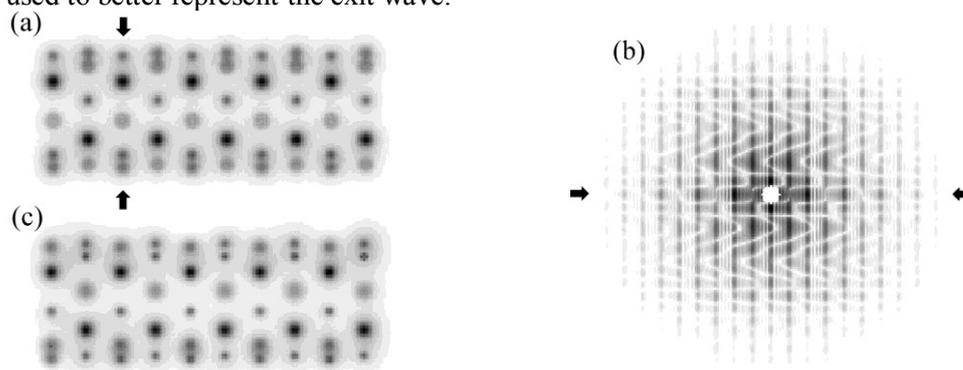


Figure 1. Original exit wave phase (a), logarithm of the square root of the corresponding input diffraction pattern (b) and the restored exit wave phase (c) all shown in inverted contrast to improve presentation.

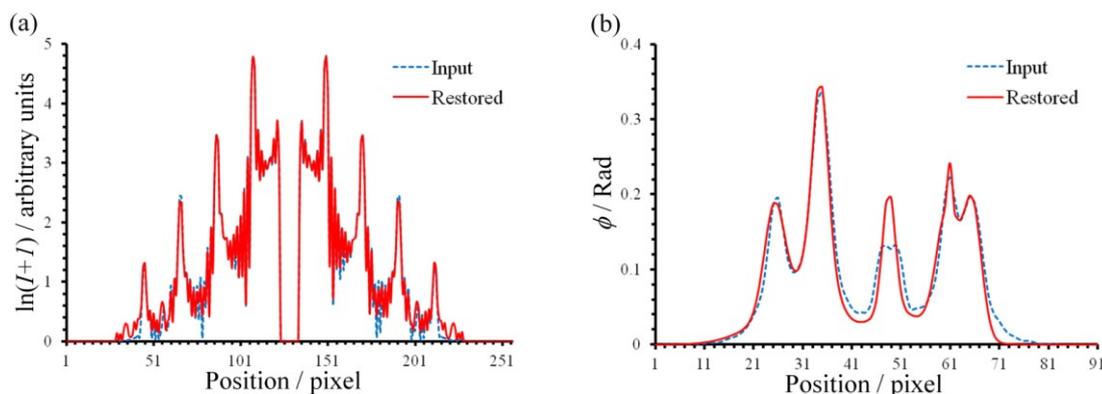


Figure 2. (a) Comparison between the input and restored diffraction patterns intensities, I , along the line defined by the arrows in Figure 1(b); (b) comparison between the input and restored exit wave phases, ϕ , along the line defined by the arrows in Figure 1(a).

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6. References

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