Dynamical and geometric effects in ptychographic diffractive imaging.

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Abstract. The principle of the ptychographical method is to collect a number of high-angle Fraunhofer diffraction patterns from an object as it is moved relative to a substantially localised illuminating beam. The many diffraction patterns are then used to solve the phase problem in order to synthesise a computational lens than can achieve much higher resolution than any of the lens or aperture components used to generate the illumination function. Although ptychography has now been demonstrated using visible and hard X-ray photons, there remain many unanswered questions relating to the scattering approximations involved in ptychography. We examine the breakdown of the kinematical and 2D approximations.

1. Introduction

Ptychographical imaging has been demonstrated using visible light [1] and hard X-rays [2]. The latter result is the first demonstration of iterative phase retrieval ptychography using an atomic scale wavelength of radiation for which it difficult to manufacture a good quality lens. Indeed, the demonstration used a conventional zone plate lens as a test object function; by resolving the outermost zones, the method was shown to be capable of matching the resolution of the best conventional imaging systems for short wavelength (8keV) photons.

A number of difficulties present themselves when we attempt to extend ptychographical imaging to the electron microscope. Firstly, electron beams are far from perfectly coherent, even from a cold field emission gun: all phase-retrieval methods require very good coherence. Secondly, the large dynamic range in the diffraction plane is hard to accommodate experimentally. Thirdly, for electrons, dynamical (multiple) scattering is strong. The first two of these issues can be tackled by improving the experimental configuration, and is the subject of current research. In this paper, we briefly consider the fundamental limitation of the geometric and multiple scattering constraints on the ptychographical principle.

2. The multiplicative approximation

We can regard ptychography as a deconvolution process [3]. In real space we have an illumination function $P(r)$, where $r$ is a two-dimensional vector and $P(r-R)$ is this illumination function shifted by a vector $R$. The illumination is incident upon an object which in all the literature to date has been described by a two-dimensional transmission function $O(x,y)$. We first assume that the far field diffracted intensity we measure is a Fourier transform of the product $P(r-R).O(x,y)$ (i.e. a convolution),
where $P(\mathbf{r}-\mathbf{R})$ is estimated in the plane of the object, which for convenience we will take as the plane $z=0$. The scattered intensity we will call $I(\mathbf{k}_s, \mathbf{R})$, where $\mathbf{k}_s$ is a vector corresponding to the direction and wavenumber of the scattered radiation. We can map $I(\mathbf{k}_s, \mathbf{R})$ into rectilinear coordinates $I(u, v, R_x, R_y)$, where

$$u = \sin \beta_x / \lambda \quad \text{and} \quad v = \sin \beta_y / \lambda, \quad (1)$$

where $\beta_x$ is the angle between the $y$-$z$ plane and the plane parallel to $\mathbf{k}_s$ which contains the line $x=z=0$, and similarly for $\beta_y$. In the 2D thin object approximation, ptychography mathematically solves for $O(x,y)$, where we would hope that

$$\int dz z y x V i e y x O (2)$$

where $\int V(x, y, z) dz$ is the integral of the atomic potential in the $z$-direction (parallel to the optic axis) and $\sigma$ is a scattering constant. In fact, both $O(\mathbf{r})$ and $P(\mathbf{r})$ are three-dimensional and $O(\mathbf{r})$ is in general strongly scattering. In what follows, $\mathbf{r}$ will therefore be taken as three-dimensional, with components $x, y$ and $z$, with $z$ lying along the optic axis and $O(\mathbf{r})$ containing the plane $z=0$. We assume that intensity is recorded over a hemisphere, of radius large enough to satisfy the Fraunhofer condition given the size of the illuminated area of object, and centered on $\mathbf{r}=0$. The coordinates over this hemisphere are as defined in equ.s (1).

3. Dynamical scattering

Consider Fig. 1, where we represent a one-dimensional travelling wave passing over a potential well.

In the absence of the well, the magnitude of the k-vector remains constant, represented by the dotted wave. The solid line represents the wave that passes through the potential. In the weak phase approximation, we assume the scattered wave (that has passed through the potential) is only very slightly altered in phase relative to the unscattered wave, represented in the phasor diagram Fig. 2a. In a (hypothetical) one-dimensional multi-slice calculation, much larger phase changes can be accommodated by representing the potential well as a series of thin wells, each of which incrementally alters the relative phase of the scattered wave, giving a final exit wave loosely of the form

$$\psi_{exit} = A e^{i\sigma V_1} e^{i\sigma V_2} e^{i\sigma V_3} \ldots = A e^{i\sigma \int V(z) dz} \quad (3)$$

where $V_1$, $V_2$, $V_3$, etc are the inner potentials of each one-dimensional value of the potential at points equally spaced points along the $z$-axis, as illustrated in Fig. 2b, and $A$ is the incident amplitude. Of course, in a real three-dimensional object, we must propagate the wave laterally between slices. However, in the case of very low resolution images (where the lateral propagation throughout the
whole thickness of the object is much smaller than the lateral resolution element), we can say that the relative phase of the exit wave is indeed accurately modelled by eqn. 2. This is not a weak phase approximation, it is the WKB approximation. The consequence is that we would expect ptychography, like low-resolution holography, to give a good representation of the phase change induced by the integrated atomic potential through the thickness of the object. Similarly, if it were possible to create a strongly scattering object which was infinitesimally thin, then by taking the natural logarithm of the exit wave value we could measure directly the (strong) atomic potential, notwithstanding phase wrapping. In this very limited sense, ptychography provides a correct image, even in the presence of dynamical scattering. The breakdown of the first Born approximation for a thin object can be thought of as having to account for the higher order terms in $e^{i\theta} = 1 + i\phi - \phi^2 ...$ etc. By the convolution theorem, these higher order terms represent multiple scattering in the Fraunhofer plane [4].

4. Geometric 3D phase changes

Of course, even in the kinematical approximation further complications arise from the depth (z-direction) separation of the scattering potential (atoms). How is ptychography affected by the resulting phase changes in the scattered radiation?

Consider first the conventional microscope arrangement in Fig. 3a. Over our notional detector sphere, each scattered vector $k_s$ has an amplitude and phase resulting from the integral over the entire volume of the object. A range of these complex values are integrated by the lens to form one point in the image plane. This surface can be represented in reciprocal space by the Ewald sphere labelled $M$ in Fig. 3b. According to the Fourier shift theorem, a displacement of the object along the z-axis introduces a phase ramp over reciprocal space in the $w$-direction (where we label reciprocal-space coordinates $u, v, w$, corresponding to the real space coordinates $x, y, z$ and where the Ewald sphere maps on $u, v$ according to eqns (1)). This phase ramp maps onto the $u, v$ coordinates according to

$$\varphi(u, v) = \frac{2\pi\Delta z}{\lambda} \left(1 - \cos \left(\lambda \left(u^2 + v^2\right)^{1/2}\right)\right)$$

which in the small angle approximation reduces to the usual parabolic defocus term. It is well known that in the conventional microscope shifting the object in the z-direction simply brings different layers of the object into focus. This is because the integral over surface $M$ gives large sums for scattering centres that are coincident with the focal plane. However, ptychography does not solve for $M$. Because the incident radiation must be localized, if only to fulfill the sampling condition in reciprocal space, there are a range of incident-beam vectors. Scattering in any particular direction $k_s$ consists of a sum over the curved surface marked $S$ in Fig. 3b. Furthermore, the mechanism of reconstruction imposes further phase changes in the $u, v$ direction (corresponding to movement of the illumination probe through $R$) in order to construct an image. What we can say, however, is that reconstructions which are likely most comparable to a real lens image will occur when the illumination function has a relatively low ranges of incident angles associated with it [5].
5. Conclusions

At very small scattering angles (that is, low resolution) ptychography should give a good representation of the exit wavefield, even in the presence of dynamical scattering. In other words, it is wrong to suppose that the domain of its usefulness is only within the weak phase grating approximation (as has been reported in some of the literature). In this sense, the method is potentially much more powerful than conventional imaging. Its ability to map phase changes over large fields of view [1] will certainly have applications in the measurement of inner potentials and magnetic fields. In the case of using near-parallel illumination (a large illumination area) on a weakly scattering object, ptychography should in theory solve for the phase and amplitude of the exit wavefield over the Ewald sphere. However, the main interest of the method is in resolution improvement beyond the usual coherence envelope constraints. In these circumstances, high angles of scatter must be processed. By moving to higher resolution, dynamical scattering and geometric effects will dominate. However, conventional imaging is also seriously compromised by such effects (although in electron microscopy it is normal to assume the projection approximation). We note that the total volume of reciprocal space that can contribute to the ptychographical data set (part of which is labelled \(D\) in Fig 3b) is somewhat larger than in the conventional image (surface \(S\)). Exactly how to exploit these data will be the subject of further work.

![Diagram of reciprocal space and illumination](image)

Figure 3: a) In a conventional microscope, scattered waves represented by a sum of plane wave each of k-vector \(k_s\), are incident upon the imaging lens. b) This can be represented in reciprocal space as surface \(M\): the Ewald sphere (grossly exaggerated). Ptychography constructs an image from an integral over surface \(S\), determined by the range of incident plane waves making up the illumination function. The total extent of reciprocal space contributing to the image lies between the two dotted semi-circles, part of which has been shaded and labelled \(D\).

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