ALGORITHMIC RECONSTRUCTION METHODS IN DIFFRACTION MICROSCOPY USING A PRIORI INFORMATION

A DISSERTATION SUBMITTED TO THE DEPARTMENT OF ELECTRICAL ENGINEERING AND THE COMMITTEE ON GRADUATE STUDIES OF STANFORD UNIVERSITY IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

Leili Baghaei Rad
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I certify that I have read this dissertation and that, in my opinion, it is fully adequate in scope and quality as a dissertation for the degree of Doctor of Philosophy.

R Pease, Primary Adviser

I certify that I have read this dissertation and that, in my opinion, it is fully adequate in scope and quality as a dissertation for the degree of Doctor of Philosophy.

Piero Pianetta, Co-Adviser

I certify that I have read this dissertation and that, in my opinion, it is fully adequate in scope and quality as a dissertation for the degree of Doctor of Philosophy.

Jianwei Miao

Approved for the Stanford University Committee on Graduate Studies.

Patricia J. Gumport, Vice Provost Graduate Education

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Abstract

Recent technological and algorithmic advances have enabled lensless imaging techniques, paving the way for high resolution three dimensional analysis. In these methods, a coherent x-ray source illuminates a sample and the amplitude of the far-field diffraction pattern is recorded. Reconstruction of the sample object requires recovery of the missing phase information by exploiting additional side information in conjunction with a computational reconstruction algorithm.

Two different sources of additional information are considered. First, the phase recovery problem is formulated as an optimization problem where knowledge of the form of the object (smoothness, positivity, particular material characteristics) are included as part of the objective or enforced as constraints. This problem is then approximated as a convex problem and solved using existing methods. The second approach assumes that the prior knowledge is in the form of a low resolution image of the sample and uses the wavelet domain to express this information. Experimental limitations include coherence, noise and missing data as well as algorithmic limitations such as data centering, support determination and complex valued reconstructions.

Reconstruction results include those of optical equivalent experiments and of both soft and hard x-ray experiments. In all three settings the two proposed sources of information are successfully used to obtain reconstructions for a variety of objects.

For the application of non-destructively examining the buried metallization pattern of integrated circuits, we employed a coherent beam of 0.17nm X-rays to image a 100 nm metal pattern. The metal layer was fabricated on a 100 micron silicon substrate and buried beneath one micron of silicon dioxide. From the diffraction pattern were able to successfully reconstruct the image of the metal pattern.
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Chapter 1

Introduction

This thesis describes recent work in several distinct areas of algorithmic reconstruction methods for coherent x-ray diffraction microscopy. This field has gained considerable attention as a potential high-resolution microscopy tool for 2- and 3-dimensional nano-scale imaging of both biological and non-biological samples.

Diffractive imaging techniques fundamentally depart from other microscopy imaging techniques by being lensless. Rather than rely on an objective lens to refract and combine the light, diffractive techniques illuminate the sample with a coherent wave and measure the diffraction pattern. This approach is advantageous in that it avoids the aberration and resolution limits of lenses, but it brings about the phase retrieval problem resulting from recording only the intensity, and not also the phase, of the scattered light. This problem can often be solved algorithmically, leading to the term algorithm reconstruction where computer algorithms are used in place of the objective lens.

This thesis is organized as a review of the theory and algorithmic reconstruction techniques of coherent diffraction microscopy. The original contribution of this work has both theoretical and experimental aspects.

Chapter 2 introduces the necessary theory for the discussions present in this thesis. It first covers the physical phenomena occurring in diffraction microscopy and then examines the theoretical aspects of diffraction imaging, in particular, oversampling and over-determination of the of the solution.
Chapter 3 continues by presenting the mathematical formalism of existing algorithmic reconstruction algorithms. This includes the difference map algorithm, a generalized iterative algorithm and a selection of projections that can be used. This includes introduced a new, novel, projection where constraints are expressed in the wavelet domain. The chapter continues by introducing a new formulation of the phase retrieval problem as a convex optimization problem.

Chapter 4 discusses the practical aspects of coherent diffractive imaging. Topics such as noise, missing data and data alignment are considered.

Chapter 5 presents the results of applying the techniques of this thesis to a selection of both real and simulated samples. The experimental results were obtained from a scaled optical experiment and both soft and hard x-ray sources.
Chapter 2

Theoretical Background

This chapter provides an overview of the main theoretical concepts that are necessary to understand the later chapters of this thesis.

2.1 Preliminaries

We first provide a summary of the signal processing tools that are used to describe the diffraction image formation process and later used in the development of new object reconstruction algorithms.

2.1.1 Fourier Transform

The continuous Fourier transform is

\[ \hat{f}(q) = \mathcal{F}f = \int_{-\infty}^{\infty} f(x) e^{-iqx} \, dx \quad (2.1) \]

and the inverse continuous transform is

\[ f(x) = \mathcal{F}^{-1} \hat{f} = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \hat{f}(q) e^{iqx} \, dq. \quad (2.2) \]
CHAPTER 2. THEORETICAL BACKGROUND

We will denote Fourier pairs with a double arrow like

\[ f(x) \leftrightarrow \hat{f}(q) \mathcal{F}f. \]  \hspace{1cm} (2.3)

Throughout this thesis, a quantity as a function of spatial coordinates is said to be in real, or direct, space (domain). The Fourier transform of such a quantity is said to be in Fourier or reciprocal space (domain). Real space coordinates will be expressed with the notation \( x \) for a single dimension or \( r = (x, y, z) \) for three dimensions, and \( q = (q_x, q_y, q_z) \) for Fourier space coordinates.

For square integrable functions \( f(x) \) and \( g(x) \), Parseval’s theorem states

\[ \int_{-\infty}^{\infty} f(x)g(x)dx = \int_{-\infty}^{\infty} \hat{f}(q)\hat{g}(q)dq. \]  \hspace{1cm} (2.4)

From this, the Plancherel theorem (equivalent to Parseval’s theorem) states that

\[ \int_{-\infty}^{\infty} |f(x)|^2dx = \int_{-\infty}^{\infty} |\hat{f}(q)|^2dq. \]  \hspace{1cm} (2.5)

The Plancherel theorem can be interpreted as saying that the Fourier transform preserves the energy of the original quantity.

If \( f(x) \) is a purely real even function, then \( \hat{f}(q) \) is also purely real and even. If \( f(x) \) is purely real and odd then \( \hat{f}(q) \) is purely imaginary and odd.

A shift (translation) in direct space results in a linear phase ramp in Fourier space.

\[ f(x - a) \leftrightarrow e^{-iaq}\hat{f}(q) \hspace{1cm} (2.6) \]

The dual of this also applies. Such a translation in Fourier space may occur, for example, if the center of the diffraction pattern is not precisely known.

\[ e^{2\pi ax}f(x) \leftrightarrow \hat{f}(q - a). \]  \hspace{1cm} (2.7)

The Fourier transform of a real function \( f(x) = f^*(x) \) has the property of inversion
through the origin

\[ \hat{f}(q) = \hat{f}^*(q) \] (2.8)

In diffraction, this arises in Friedel’s Law which states that members of a Friedel pair, \( \hat{f}(q) \) and \( \hat{f}(-q) \), have equal amplitude and opposite phase.

Convolution,

\[ (f \ast g)(x) = \int f(x')g(x - x')dx' \] (2.9)
in one domain corresponds to multiplication in the other. For example, convolution in direct space:

\[ (f \ast g)(x) \Leftrightarrow \hat{f}(q)\hat{g}(q). \] (2.10)

Similarly, for cross-correlation,

\[ (f \circlearrowleft g)(x) = \int f(x')g^*(x' - x)dx' \Leftrightarrow \hat{f}(q)\hat{g}^*(q). \] (2.11)

Of particular note is the autocorrelation,

\[ (f \circlearrowleft f)(x) \Leftrightarrow |\hat{f}(q)|^2 \] (2.12)

As with most signal processing applications the data analysis is performed on a digital computer and so, combined with the spatially sampled detector, the data values are discretized to a digital representation rather than a continuous real number. This necessitates the use of the discrete Fourier transform (DFT),

\[ \tilde{f}_n = \mathcal{F} f = \frac{1}{\sqrt{N}} \sum_{m=0}^{N-1} f_m e^{2\pi imn/N} \] (2.13)

and the corresponding inverse discrete Fourier transform,

\[ f_n = \mathcal{F}^{-1} \tilde{f} = \frac{1}{\sqrt{N}} \sum_{m=0}^{N-1} \tilde{f}_m e^{2\pi imn/N}. \] (2.14)

The discrete Fourier transform of a (assumed periodic) continuous function is
approximated by sampling the function on an appropriately fine grid,

\[ f_m = f(m\Delta x). \]  \hfill (2.15)

Sampling in the spatial domain leads to periodic extension and sampling in the frequency domain,

\[ \tilde{f}(n\Delta q) = \tilde{f}_n, \]  \hfill (2.16)

where the spatial and frequency grid spacings are related as follows:

\[ \Delta x \Delta q = \frac{2\pi}{N}. \]  \hfill (2.17)

A naïve implementation of the DFT requires \( O(N^2) \) operations. In practice one of the fast Fourier transforms is used which is \( O(N \log N) \), such as [4].

As alluded to the sampling of the continuous function must be performed on an appropriately fine grid. For a grid defined by \( \Delta x \) the maximum frequency present in the discrete Fourier transform is given as the Nyquist frequency

\[ q_N = \frac{N\delta q}{2} = \frac{\pi}{\Delta x}. \]  \hfill (2.18)

If the sampling is not sufficiently fine then \( q_N \) is less than the maximum frequency present in the signal and aliasing occurs where those frequency components higher than \( q_m \) are aliased and appear at lower frequencies. If the maximum signal frequency is less than the Nyquist frequency then no aliasing occurs and the signal can be perfectly reconstructed from its samples using the sinc function as

\[ f(x) = \sum_n f_n \sin(x - n\Delta x) \cdot \frac{x - n\Delta x}{x - n\Delta x}. \]  \hfill (2.19)

### 2.1.2 Wavelet Transform

Fourier analysis is based on the complex exponential as a basis function. The complex exponential has a precisely defined frequency but is infinite in spatial extent. The result is that the Fourier transform is excellent at analysing the frequency components
of a signal but with the drawback that it cannot localize the spatial position where that frequency component originates.

One approach to mitigate these limitations is to use a windowed complex exponential basis function but this implies a constant spatial resolution for all frequency components. For many applications a more preferable transform would have reduced spatial resolution but high frequency resolution for low frequency components and good spatial with poor frequency resolution for high frequency components.

These needs lead to considering the alternative where the basis function vary in both 'frequency' and spatial extent. Such a transform is able to provide an excellent time-frequency representation of a signal with time and frequency localization. The continuous wavelet transform provides such an analysis tool where the components are given at a scale, $a > 0$, and translation value, $b \in \mathbb{R}$ by the following transform

$$G_w(a, b) = \frac{1}{\sqrt{a}} \int_{-\infty}^{\infty} g(x) \psi^* \left( \frac{x - b}{a} \right) dx,$$  \hspace{1cm} (2.20)

where $\psi(x)$, known as the mother wavelet, is a continuous function in both spatial and frequency domains. The mother wavelet is a generator of daughter wavelet functions, translated and scaled versions of the mother wavelet function. The inverse transform is

$$g(x) = \int_0^\infty \int_{-\infty}^{\infty} \frac{1}{a^2} G_w(a, b) \frac{1}{\sqrt{|a|}} \tilde{\psi} \left( \frac{x - b}{a} \right) db \, da.$$  \hspace{1cm} (2.21)

where $\tilde{\psi}(x)$ is the dual function of $\psi(x)$. The dual function can be chosen to be $C_{\psi}^{-1} \psi(x)$ with,

$$C_{\psi} = \frac{1}{2} \int_{-\infty}^{\infty} \frac{|\hat{\psi}(\eta)|^2}{|\eta|} \, d\eta,$$  \hspace{1cm} (2.22)

and subject to the admissibility condition, $0 < C_{\psi} < \infty$. This in turn can be shown to imply that the wavelet must integrate to zero, $\psi(0) = 0$. 

\hspace{1cm}
2.1.3 Haar wavelet

The simplest continuous wavelet is the Haar wavelet which consists of a single positive and negative cycle. Figure 2.1 shows the mother wavelet $\psi(x)$ and several of the daughter wavelets, both scaled and shifted.

The discrete Haar wavelet can be represented be the $2 \times 2$ Haar matrix,

$$H_2 = \begin{bmatrix} 1 & 1 \\ 1 & -1 \end{bmatrix}.$$ \hspace{1cm} (2.23)

The Haar wavelet transform can be derived from the Haar matrix. The $4 \times 4$ Haar transform matrix is

$$H_2 = \frac{1}{\sqrt{4}} \begin{bmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & -1 & -1 \\ \sqrt{2} & -\sqrt{2} & 0 & 0 \\ 0 & 0 & \sqrt{2} & -\sqrt{2} \end{bmatrix}.$$ \hspace{1cm} (2.24)

Figure 2.1: Several of the continuous Haar wavelets. The mother wavelet $\psi(x)$ is scaled and shifted.

The 2D wavelet transform can be accomplished by applying 1D transforms in orthogonal directions consecutively as shown in figure 2.2.
Figure 2.2: The 2D wavelet decomposition of an image occurs with down-sampling in both $x$ and $y$ directions consecutively.
Further information about the wavelet transform can be found in many references, including [51].

## 2.2 Scattering of light from inhomogeneous media

In this section we briefly examine the basic elements of the scalar theory of scattering of light. Rather than re-derive the wave equation completely we summarize the analysis of Thibault, 2007 and [50].

Starting from Maxwell’s equations,

\[
\begin{align*}
\nabla \cdot [\epsilon(r)E(r)] &= 0 \\
\nabla \cdot H(r) &= 0 \\
\nabla \times E(r) &= i\omega \mu H(r) \\
\nabla \times H(r) &= -i\omega \epsilon(r)E(r)
\end{align*}
\] (2.25)

where \(\epsilon(r)\) is the electric permittivity and \(\mu\) is the magnetic permeability. Under the assumption that variations in the dielectric medium occur on a length scale greater than the wavelength of the electromagnetic field, i.e.

\[
\nabla \epsilon \ll k|\epsilon|,
\] (2.26)

with wavenumber \(k = 2\pi/\lambda\), then the scalar Helmholtz wave equation can be derived.

\[
\nabla^2 \Psi + k^2 n^2 \Psi = 0,
\] (2.27)

where \(k = \omega/c\) and \(n^2 = c^2 \epsilon \mu\). The Helmholtz equation can only be solved analytically in certain scenarios and must generally be solved by numerical means or approximated.

At x-ray wavelengths, \(n\) is usually expressed in complex form as,

\[
n = 1 - \delta - i\beta
\] (2.28)

where \(\delta\) represents the purely refractive component and \(\beta\) the absorptive component.
Both components are very small ($|\delta + i\beta| \ll 1$) so $n^2$ is well approximated by $1 - 2(\delta + i\beta)$. A consequence of $\delta$ being very small is that x-rays are only very weakly refracted by any matter and a focusing lens would have to be both strongly curved and very thick. However, although $\beta$ is small it is high in comparison and such lens would have high absorption, make such lenses impractical.

### 2.2.1 Diffraction Scattering

For diffraction experiments it is the amplitude of the wave field in a plane transverse to the direction of propagation that is measured. After separating out the transverse ($r_\perp = (x, y)$) and parallel ($z$) components and assuming the back-propagating component is zero, then the well known far-field diffraction equation can be derived,

$$I(u) = |\Psi_{\text{far-field}}(zu)|^2 \propto \frac{1}{1 + u^2} |\tilde{\Psi}(q_\perp = \kappa u)|^2.$$  \hspace{1em} (2.29)

That is, the far-field diffraction pattern is proportional to the absolute value of the Fourier transform of the exit wave field.

### 2.3 Diffraction Imaging

#### 2.3.1 Phase Information Loss

In diffraction imaging experiments it is the intensity of the wave field that is measured. Since the wave field is a complex variable the phase information about the wave field is lost [8, 3]. This information is critical and must be recovered from either redundant information contained within the amplitude information or from an external source [1, 2, 3, 15, 12, 32, 11, 16, 19].

#### 2.3.2 Phase Recovery Techniques

Most coherent diffractive imaging (CDI) methods rely on far-field diffraction data to retrieve the complex-valued projection or three-dimensional density of a sample. Unless the sample is periodic, as is the case in crystallography, finite coherence length
and fixed dimensions of the detector pixels impose a limit on the amount of information carried by the wave for the problem to be tractable [5, 23, 24, 10]. For this reason, all CDI techniques require “oversampling” of the diffraction pattern, i.e. that the region in real-space contributing to the diffraction data does not extend beyond a limit predefined by the experimental conditions. In many experiments, oversampling is achieved with a plane wave incident on an isolated specimen [7]. Despite the successes of this approach [22, 37, 43, 29] the requirement for sufficiently small and isolated samples is an important obstacle to a wider application of the technique [34, 20, 26, 39].
Chapter 3

Reconstruction Algorithms

3.1 Iterative Algorithms

3.1.1 Constraint sets

The phase recovery problem is often posed as a two-constraint satisfaction problem where the first constraint is based on the measured diffraction intensities and the second represents known information in direct space [29, 31, 30, 43, 28, 9, 13, 6, 35, 33, 36, 27, 47]. Formally the two-constraint problem is defined as follows:

Definition 1 (The two-constraint satisfaction problem). Given two constraint sets $C_1$ and $C_2$, find an $x \in C_1 \cap C_2$, i.e. simultaneously satisfying both constraints.

3.1.2 Projections

A key concept to many constraint satisfaction problems is that of taking an element not contained within a constraint set and using this to find one that is. This idea is made more precise with the following definitions.

We assume that there exists an underlying vector space $W$ with the usual vector space axioms:

1. associativity of addition
CHAPTER 3. RECONSTRUCTION ALGORITHMS

Fourier modulus constraint
• enforce the modulus of
the Fourier transform to
‘match’ measurements

Other constraints
• Finite support
• Wavelet coefficients (low res.)
• Smoothness
• Positivity
• Close match to specifications

Look for solution meeting both sets of constraints

Figure 3.1: General phase retrieval constraint problem. The known information forms constraints sets and a solution is sought which simultaneously satisfies both sets of constraints. One constraint set uses the measured Fourier modulus values. The other constraint set can be formed from different forms of a priori information. For example, this might be knowledge of the spatial support of the object or a low-resolution image of the object.

2. commutativity of addition
3. identity of addition
4. inverse elements of addition
5. distributivity of scalar multiplication with respect to vector addition
6. distributivity of scalar multiplication with respect to field addition
7. compatibility of scalar multiplication with field multiplication
8. identity element of scalar multiplication.

On this vector space we can define the general projection as follows:

**Definition 2** (Projection). A projection $P$ is a linear transformation that is idempotent, meaning that $P^2 = P$. In this context this means that once the projection has been applied any subsequent projection (using the same projection) returns the same element.

With $W$ as the underlying vector then the projection operator $P$ induces subspaces $U$ and $V$ as the range and null spaces of $P$ respectively. The following basic properties hold:
1. \( P \) is the identity operator \( I \) on \( U \): \( \forall x \in U : Px = x \)

2. There is the direct sum \( W = U \oplus V \). Every vector \( x \) can be decomposed uniquely as \( x = u + v \) where \( u \in U \) and is given by \( u = Px \), and \( v \in V \) and is given by \( v = x - Px \);

For a vector space equipped with an inner product then an orthogonal projection can be defined as follows:

**Definition 3** (Orthogonal Projection). For a vector space \( W \) equipped with an inner product, a projection is orthogonal if and only if it is self-adjoint, i.e. the associated matrix \( P \) is symmetric relative to an orthonormal basis: \( P = P^T \) (or, in the complex case, \( P = P^* \)). For such a projection the range and null, \( U \) and \( V \) respectively, are orthogonal subspaces of \( W \).

In more practical terms, the orthogonal projection of a vector to a subspace finds the vector that is nearest. That is, given \( x \in W \), then \( u = Px \) is the point in \( U \) that minimizes the distance (as defined by the inner product of \( W \)) \( ||x - u|| \). If the subspace \( U \) is convex then the orthogonal projection is unique while for non-convex subspaces there may exist multiple vectors with the same distance to \( x \).

The orthogonal projection distance can be considered as the error, or amount, by which a vector fails to satisfy a constraint. For a vector \( x \) we say that the constraint set \( C_1 \) is satisfied if \( P_{C_1}x = x \) for the orthogonal projection \( P_{C_1} \) since the error is given by \( ||x - Px|| = 0 \). If the two-constraint satisfaction problem is feasible then all solutions satisfy the condition:

\[
x_{sol} = P_{C_1}x = P_{C_2}x
\]

The determination of suitable projection operators is problem-specific and depends on the constraint spaces that are used. Later, in 3.2, several commonly used projections will be discussed as well as a new projection based on wavelets.
3.1.3 Iterative formulation

In general, the large search space and complicated constraint spaces mean that most algorithms are iterative in nature. Such algorithms repeatedly apply an operator $F$

$$x_{n+1} = F(x_n)$$

until a fixed point is reached where

$$x_{n+1} = F(x_n) = x_n = x^*.$$

The algorithm is then said to converge, however this point may be one of a set of solutions or may not even be a solution at all, depending on the nature of the constraint sets.

3.1.4 Difference map algorithm

The difference map algorithm [29] is defined as:

$$x_{n+1} = x_n + \beta D(x_n). \quad (3.1)$$

where

$$D(x) = y_2(x) - y_1(x), \quad (3.2)$$

and

$$y_1 = P_1[(1 + \gamma_2)P_2(x) - \gamma_2 x], \quad y_2 = P_2[(1 + \gamma_1)P_1(x) - \gamma_1 x]. \quad (3.3)$$

The operators, $P_1$ and $P_2$, are projections and are most often orthogonal projections. The parameters $\beta$, $\gamma_1$ and $\gamma_2$ are complex but usually taken as real and can be chosen as $\pm 1$ to simplify 3.1.

From an initial starting point $x_0$ the difference map is iteratively applied until some termination criteria is met. Assuming for now that the map converges, then
this will occur when a fixed point is reached,

\[ x^* = x^* + \beta D(x) \]  

which requires that \( D(x) = 0 \), i.e.,

\[ y_1(x^*) = y_2(x^*). \]  

(3.5)

The fixed point, \( x^* \), will be a solution to the two-constraint problem since it must lie somewhere in the intersection of the two constraint spaces, \( C_1 \) and \( C_2 \).

The progress of the iterative map can be monitored by observing the difference map error, defined as the norm of the difference between successive iterate values:

\[ \varepsilon_n = ||x_{n+1} - x_n|| \]  

(3.6)

3.2 Projection operators

In this section we present the well known projections for diffraction and direct space as well as introduce wavelet space projections.

3.2.1 Diffraction projections

Modulus projection

Let \( I(q) \) be the measured diffraction intensities.

1. Compute the forward FFT of the current iterate \( \upsilon \):

\[ \hat{\upsilon} = \mathcal{F}_\upsilon \]  

(3.7)

2. Set the Fourier magnitudes to the measured values:

\[ \hat{\upsilon}'(q) = \sqrt{I(q)} \frac{\hat{\upsilon}(q)}{||\hat{\upsilon}(q)||} \]  

(3.8)
3. Compute the inverse FFT of the new Fourier coefficients:

\[ v' = \mathcal{F}^{-1} \tilde{v}' \] (3.9)

Modulus projection with missing data

Let \( M \) be the function indicating which diffraction intensities are present in the measurements.

1. Compute the forward FFT of the current iterate \( v \):

\[ \tilde{v} = \mathcal{F} v \] (3.10)

2. Set the known Fourier magnitudes to the measured values:

\[ \tilde{v}'(q) = \begin{cases} \sqrt{I(q)} \frac{\tilde{v}(q)}{|\tilde{v}(q)|} & \text{if } M(q) = 1 \\ \tilde{v}(q) & \text{if } M(q) = 0 \end{cases} \] (3.11)

3. Compute the inverse FFT of the new Fourier coefficients:

\[ v' = \mathcal{F}^{-1} \tilde{v}' \] (3.12)

Modulus projection with bounds on unknown values

Let \( M \) be the function indicating which diffraction intensities are present in the measurements. Let \( B(q) \) be an upper bound for those \( q \) where \( M(q) = 0 \).

1. Compute the forward FFT of the current iterate \( v \):

\[ \tilde{v} = \mathcal{F} v \] (3.13)
2. Set the known Fourier magnitudes to the measured values and bound the un-
known values:

\[ \tilde{\nu}'(q) = \begin{cases} \sqrt{I(q)} \frac{\tilde{\nu}(q)}{|\tilde{\nu}(q)|} & \text{if } M(q) = 1 \\ \min (B(q), \tilde{\nu}(q)) & \text{if } M(q) = 0 \end{cases} \] (3.14)

3. Compute the inverse FFT of the new Fourier coefficients:

\[ \nu' = \mathcal{F}^{-1} \tilde{\nu}' \] (3.15)

### 3.2.2 Direct space projections

Complementing the projections for Fourier space there is a set of projections that are
used to enforce constraints in direct space. The most basic of these projections is
the support projection where the compact support of the sample is used to set values
outside of the support to zero. This is easily seen to be the orthogonal projection
since it does not affect those values inside the support, only setting those outside the
support to zero, but still respecting the constraint [17].

**Basic support projection**

Let \( S \) be the finite support. The basic finite support projection sets those points
outside the support to zero:

1. \[ \nu'(r) = \begin{cases} \nu(r) & \text{if } r \in S \\ 0 & \text{otherwise} \end{cases} \] (3.16)

**Support projection with threshold**

Let \( T \) be a threshold for \( |\nu(r)| \), then the projection is:

1. \[ \nu'(r) = \begin{cases} \nu(r) & \text{if } |\nu(r)| \geq T \\ \frac{\nu(r)}{|\nu(r)|} & \text{if } \frac{T}{2} < |\nu(r)| < T \\ 0 & \text{otherwise} \end{cases} \] (3.17)
Positivity projection

\[ v'(r) = \begin{cases} v(r) & \text{if } v(r) \geq 0 \\ 0 & \text{otherwise} \end{cases} \] (3.18)

3.2.3 Wavelet Projection

Although much success has been achieved with the finite support constraint, several reasons motivate exploring other types of constraint sets particularly for specific applications:

1. The finite support constraint restricts the method to reconstructing isolated objects [44, 45]. Many applications would benefit from being able to image extended structures.

2. It is often that more detailed spatial information such as low-resolution SEM or x-ray transmission images may be available. Existing algorithms frequently use this to fill in data values that are missing due to a beam stop [23].

3. Rather than precise knowledge of the support the prior information may be more qualitative in nature. For example, it may be that the structure is semi-regular or that it is generally composed of a small number of homogeneous areas.

In this work we introduce the use of a low-resolution version of the real space object as \textit{a priori} information. It explicitly is not assumed that the sample is compactly supported as previously required. The constraint set is now the set of all images which have a low-resolution approximation matching the measured values. It is expected that the resolution could be lower by perhaps a factor of 16 or 32.

This \textit{a priori} data directly provides information about the low frequency components in the Fourier domain, both the phase and magnitude. The magnitude information can prove useful as a beam stop generally obscures the center portion of the image sensor. The low-resolution information sets the approximate structure of the object and resolves phase and translation ambiguities.
The low-resolution constraint may be applied in the spatial domain but can be more readily expressed in a wavelet domain. The Haar wavelet is suitable partly because of its simplicity but also because the structure of the wavelet matches the shape of the expected features the test sample. For other sample types one of the other wavelets may be more suitable. Figure 2.1 shows the continuous Haar mother wavelet and several translated and scaled wavelets. Figure 2.2 illustrates the wavelet decomposition of a two-dimensional image. For the Haar wavelet a single level of decomposition produces four smaller images ($\frac{1}{2}$ the size in each dimension). The first image is the (normalized) approximation sub-image where the sum of each $2 \times 2$ block is computed. The second, third and fourth images are detail sub-images where the difference between sample values horizontally, vertically and diagonally are computed. Additional levels of decomposition can be recursively performed on the sum image. In Figure 2.2 four levels of decomposition are shown with the shaded block indicated the approximation coefficients which will be used as a priori information.

It is assumed that the diffraction data is of size $(n \times n)$ for some $n = 2^k$ where $k$ is a positive integer and that the low-resolution information is of size $(2^l \times 2^l)$ where $l$ is also a positive integer and $l < k$. The approximation coefficients of the $(k-l)$th wavelet transform, $\mathcal{W}_{k-l}$, are then constrained to match the a priori information in wavelet space $X_{W_{meas}}$. Figure 2.2 shows the approximation coefficients for the fourth level decomposition. The constraint set is

$$B^w = \{ x \mid [\mathcal{W}_{k-l}(x)]_{\text{approx.}} = X_{W_{meas}} \}.$$  \hspace{1cm} (3.19)$$

The orthogonal projection is obtained by performing the $(k-l)$ level wavelet transform, setting the approximation coefficients to the low-resolution measurements and then performing the inverse wavelet transform

$$P_{B^w} = \mathcal{W}_{k-l}^{-1} \left([\mathcal{W}_{k-l}(x)]_{\text{detail}}, [X_{W_{meas.}}]_{\text{approx.}}\right).$$ \hspace{1cm} (3.20)

Because the orthogonal projection sets the low-resolution approximation coefficients the change in value is uniformly distributed over all of the sample points which contribute to each coefficient. This leads to the adjustment being performed on blocks
of size $2^{k-1}$ in the projected iterate. This leads to artificial discontinuities between blocks which can be greatly reduced by distributing the required change to values in a smooth non-uniform way across all of the sample points. A non-orthogonal projection can be defined as

$$P_{Bw} = x + \gamma,$$ (3.21)

where $\gamma$ is the solution of the following optimization problem:

$$\text{minimize} \quad ||W_{k-l}(\gamma) - X_{w}^{\text{meas}}||_2 + ||d_{\text{vert.}}||_2 + ||d_{\text{horiz.}}||_2,$$ (3.22)

where $||\cdot||_2$ is the $l_2$ norm and $d_{\text{vert.}}$ and $d_{\text{horiz.}}$ are the difference vectors formed by the pairwise differences of adjacent sample values in the vertical and horizontal directions respectively. The solution of the minimization problem has low-resolution coefficients that match the known data (from the first term) and is smooth in both the vertical and horizontal directions (the second and third terms).

### 3.3 Convex optimization

The optimal solution in the spatial domain has a Fourier magnitude which agrees with the measurements obtained and has certain properties related to the prior information. These additional properties provide information to determine a solution for the unknown phase. The basic phase reconstruction problem has previously been formulated as a convex problem, see [21, 25, 28, 18, 14].

For simplicity in the mathematical formulation we consider only a one dimensional problem. The formulation can be extended to two and three dimensions as necessary. We say that the magnitude of the Fourier transform of the solution agrees with the (noisy) measurements in the least-squares sense. Mathematically this can be expressed as

$$\sum_{j=1}^{n} (|Fx|_j^2 - b_j)^2$$ (3.23)

where

$$x \in \mathbb{C}^n$$ represents the samples of the unknown spatial structure.
CHAPTER 3. RECONSTRUCTION ALGORITHMS

$b \in \mathbb{R}^n$ are the intensity measurements of the diffraction pattern

$F \in \mathbb{C}^{n \times n}$ is the Fourier matrix such that $Fx = \text{DFT}(x)$

The best solution, considering only the measurements, will give the smallest value for expression (3.23).

The prior information from the specification is incorporated by saying that an optimal solution should agree with the specification as much as possible. In other words, the disagreement between the solution and the original specification should be sparse. Mathematically we express this as

$$\sum_{j=1}^{n} |\tilde{x}_j - x_j| \quad (3.24)$$

where

$\tilde{x} \in \mathbb{C}^n$ represents the samples of the specification spatial structure

Furthermore, we assume that the solution is generally smooth. Physically, this corresponds to adjacent samples being highly likely to be the same material. This is expressed as

$$\sum_{j=1}^{n-1} |x_{j+1} - x_j| \quad (3.25)$$

Expressions (3.23 - 3.25) can be combined to form an objective function where we seek the solution that obtains the minimum value for the objective. The relative weights of the three terms can be controlled by the regularization parameters $\sigma$ and $\gamma$. The solution is constrained by requiring that each sample must correspond to the diffraction coefficient of a known material. We call the discrete set of possible values $\mathcal{D}$. The optimization problem can finally be expressed as follows

$$\begin{align*}
\text{minimize} & \quad \sum_{j=1}^{m} (|Fx_j|^2 - b_j)^2 + \sigma \sum_{j=1}^{n} |\tilde{x}_j - x_j| + \gamma \sum_{j=1}^{n-1} |x_{j+1} - x_j| \\
\text{subject to} & \quad x_j \in \mathcal{D}, \quad j = 1, \ldots, n
\end{align*} \quad (3.26)$$
3.3.1 Convex formulation

The problem as expressed is non-convex in both the objective function and in the constraints. The first term of the objective is not convex due the complex modulus. The inner term of the quadratic cannot be guaranteed to be positive or negative and thus the composition is not convex. The constraint that $x \in \mathcal{D}$ is not convex since $\mathcal{D}$ is not a convex set. This can be easily relaxed by replacing $\mathcal{D}$ by the convex hull $\text{conv}\mathcal{D}$. The effects of this relaxation will be discussed when the precise formulation of the solution is given.

In [41] there are several approaches presented for local optimization. In this problem the prior information in the assumed structure provides an excellent initial guess.

We first note that the non-convex part of the problem formulated above is known as the magnitude-squared-least-squares problem. A closely related problem is the magnitude-least-squares problem

$$
\text{minimize } \sum_{j=1}^{m} (|F_jx| - c_j)^2
$$

with $x \in \mathbb{C}^n$. An equivalent problem can be obtained without the complex modulus as

$$
\begin{align*}
\text{minimize } & \sum_{j=1}^{m} (F_jx - c_j z_j)^2 \\
\text{subject to } & |z_j| = 1, \quad j = 1, \ldots, n
\end{align*}
$$

with $x \in \mathbb{C}^n$ and $z \in \mathbb{C}^n$. The unit-modulus variables $z_j$ are introduced to represent the phases of the $F_jx$ terms. This new formulation can be used for a variable exchange method where first $x$ is minimized for a fixed $z$ and then $z$ is computed from the phases of $F_jx$. With no regularization terms the solution for $x$ is a linear least squares problem. With the additional terms it a standard convex problem. Using
this method and assuming no prior known values we have the following

\[
\text{minimize} \quad \sum_{j=1}^{m} (w_j(F_j x - c_j z_j))^2 + \gamma \sum_{j=1}^{n-1} |x_{j+1} - x_j| + \sigma \sum_{j=1}^{n} |\tilde{x}_j - x_j| \\
\text{subject to} \quad x_j \in \text{conv} D, \quad j = 1, \ldots, n \\
|z_j| = 1, \quad j = 1, \ldots, n
\]  

(3.29)

where \(c_j = \sqrt{b_j}\) and the optimization is only over \(x\).

The convex optimization problem in equation (3.29) is in standard form and can be solved using existing commercial or open source solvers. The CVX Matlab based modeling system was used to solve the examples given in this paper [48, 49].

### 3.3.2 Heuristics

This formulation is appropriate for a general matrix \(F\). In the problem considered \(F\) is actually the Fourier matrix where \(F_{jk} = e^{-\frac{2\pi i j k}{n}}\) and the value \(F_j x\) is the \(j\)th Fourier coefficient for \(x\). We take the convention of \(i = \sqrt{-1}\). For the structures considered in this problem the distribution of energy in the Fourier domain is roughly known. Generally most of the energy is concentrated at lower frequencies with less at higher frequencies. Intuitively it makes sense to weight the lower frequency components more, leading to them being optimized first and establishing the general structure.

As the sequential optimization progresses the reconstruction is refined and the higher frequency components are optimized. Experimentally, weighting the terms was found to have a considerable effect and lead to faster convergence. In addition it was experimentally observed that the weighting modified the problem sufficiently such that in some cases the correct solution was obtained whereas without weighting a local minimum existed that was not the global minimum. A simple quadratic weighting is used where \(w_j = (-1 + \frac{j}{n})^2 + 0.5\).

A further heuristic can be added to improve the speed of convergence. After each iteration the history of each \(x_j\) can be examined. As each \(x_j\) converges to its optimal value it generally does this by following the line segment connecting the initial value and the final value. This can be detected by performing a least squares linear fit
to some number of the previous values. If the fit is determined to be good and
the projected line is close to another value \( d_j \in \mathcal{D} \) then \( x_j \) can be set as \( d_j \). This
is performed only when the objective value is decreasing slowly. Since this abrupt
change can disrupt the optimization following the value being modified it is held
constant for several iterations.

The algorithm is then:

1. Choose a tolerance \( \epsilon > 0 \) and \( \epsilon_{\text{jump}} > 0 \).
2. Take the initial \( x \) to be the assumed diffraction values, \( \tilde{x} \).
3. Compute the phase (argument) terms, \( z_j = e^{i \arg(A_jx)} \), \( j = 1, \ldots, n \).
4. Solve the convex problem in equation (3.29).
5. If objective decrement is less than \( \epsilon_{\text{jump}} \) perform a least squares linear fit to the
   history of each value. If the fit for \( x_j \) is good and the line extends to a known
   value \( d_j \in \mathcal{D} \) then set \( x_j = d_j \).
6. Repeat steps (3) to (5) until the decrease in objective is less than the tolerance
   \( \epsilon \).

### 3.3.3 Scaling to 3D datasets

The computational feasibility of solving the convex optimization problem is a major
hurdle to its usage. In this section we briefly examine how the structure of the
phase retrieval problem in particular can be exploited through the use of oracle based
methods to reduce the necessary computation.

A general optimization problem can be written as

\[
\min \{ g(x) \mid x \in \mathbb{X} \}.
\]

An oracle based solver for such problems typically only needs first order information.
For example the OBOE\(^1\) solver requires the following at each step:

\(^1\)https://projects.coin-or.org/OBOE
$x$ feasible: value of $g(x)$ and an element of the subdifferential of $g$ at $x$.

$x$ infeasible: hyperplane separating $x$ from the feasible set.

To illustrate this we will consider the 1D version of the variable exchange problem with a simple finite support constraint. The objective and constraint set are:

$$g(x) = \sum_{k=0}^{N-1} \left( |X_k|^2 - (b_k z_k)^2 \right)^2$$

$$X = \{ x \mid x_k = 0 : k \in \bar{S} \},$$

where $X_k$ is the $k$th coefficient of the discrete Fourier transform of $x$ and $\bar{S}$ is the complement of the finite support set $S$. The 1D DFT can be written as an $N \times N$ matrix multiplication $X = W x$ where $W$ is the Vandermonde matrix (up to normalization) of the $N$th root of unity, $e^{-2\pi i N}$. The objective is then written as

$$g(x) = \sum_{k=0}^{N-1} \left( |w_k^H x|^2 - b_k \right)^2,$$

where $w_k = W_k^H$ are the Hermitian transposes of the rows of the Fourier matrix $W$. The objective is a function of the complex vector $x \in \mathbb{C}^n$ but it can be rewritten as a function of a real-valued vector in $\mathbb{R}^{2n}$. We can split each vector into real and imaginary parts giving $x_R, x_I, w_k R$, and $w_k I$ allowing us to write

$$|w_k^H x|^2 = \begin{bmatrix} x_R \end{bmatrix}^T \begin{bmatrix} w_k R & -w_k I \\ w_k I & w_k R \end{bmatrix} \begin{bmatrix} w_k^T R & w_k^T I \\ -w_k^T I & w_k^T R \end{bmatrix} \begin{bmatrix} x_R \\ x_I \end{bmatrix}.$$ 

This can be simplified by introducing $W$ and $u$ as

$$W_k = \begin{bmatrix} w_k R & -w_k I \\ w_k I & w_k R \end{bmatrix} \quad \text{(3.30)}$$

$$u = \begin{bmatrix} x_R \\ x_I \end{bmatrix} \quad \text{(3.31)}$$
The objective function is now
\[
g(u) = \sum_{k=0}^{N-1} (u^T W_k W_k^T u - (b_k z_k)^2)^2
\]
\[
= \sum_{k=0}^{N-1} g_k(u)^2
\] (3.32)

For \(x\) feasible an element of the subdifferential must be found but since each \(g_k(u)\) is differentiable the subdifferential \(\partial g(u)\) is simply \(\{\nabla g(u)\}\) which is given by
\[
\nabla g(u) = 2 \sum_{k=0}^{N-1} g_k(u) \nabla g_k(u)
\]
\[
= 4 \sum_{k=0}^{N-1} (u^T W_k W_k^T u - (b_k z_k)^2) W_k W_k^T u.
\] (3.35)

We now turn to the case when \(x\) is infeasible and a separating hyperplane must be computed. The finite support constraint requires that certain components of \(x\) be zero. For each of these components considered separately a separating hyperplane is simply \(\text{sgn}(x_k R) x_k R \leq \epsilon x_k R\) with \(\epsilon\) some small positive real number. A separating hyperplane for all of the components can be constructed using indicator functions as
\[
\left[ 1^T x_k \in S \cdot \text{sgn}(x_R)^T \right] \left[ 1^T x_k \in S \cdot \text{sgn}(x_I)^T \right] \leq \epsilon \min\{x_k R, x_k I : x_k \in \bar{S}\},
\]
where \(\cdot\) denotes element-wise multiplication.

The principal benefit of an oracle based solver is that the structure of the DFT matrix can be exploited. In the above formulation the matrix \(W\) has been used to compute the DFT requiring \(O(N^2)\) operations. This DFT computation is used for evaluating the objective and also to find the gradient where, for example, \(W_k^T u\) gives the real and imaginary parts of the \(k\)th DFT coefficient of \(x\). Any computation involving the DFT can be significantly accelerated by employing a FFT algorithm to exploit the structure, achieving \(O(N \log N)\) complexity. For large and/or multidimensional problems the savings can be considerable.
Chapter 4

Working with Experimental Data

In this chapter we consider experimental diffraction microscopy and how it is different from the theory presented in Chapter 3. We look first at experimental limitations, including inherent limitations due to noise and coherence, followed by more practical limitations such as missing data and detector dynamic range. In light of these limitations we then look at steps required to prepare experimental data for use as input to a reconstruction algorithm.

4.1 Experiment limitations

4.1.1 Noise

Diffraction-based imaging techniques are based on the measurement of particle flux at discrete detector positions over a finite exposure time. The measurement is the count of (or related to through some transfer function) the discrete particles arriving randomly over time and exhibits statistical fluctuations. These fluctuations, commonly known as ‘shot noise’, provide an upper bound on the detection signal-to-noise that is independent of the detection mechanism. We first discuss shot noise before examining other noise sources, both true random noise and also systematic errors that can be considered as forms of noise.

Photon arrival at a detector site is a stochastic process. Individual arrivals occur
continuously and independently of each other and therefore the process is a Poisson process. The probability distribution of the total count, $N(t)$, is a Poisson distribution. The number of arrivals in an interval between time $a$ and $b$ is $N(b) - N(a)$ and also has a Poisson distribution. The process is characterized by its rate parameter $\lambda$ which is the expected number of arrivals that occur per unit time. For a *homogeneous* process the rate parameter is constant in time but, in general, for a non-homogeneous process where the rate may vary over time, the generalized rate is the expected number of arrivals over the time interval,

$$\lambda_{a,b} = \int_a^b \lambda(t) \, dt.$$  

In diffractogram measurements it is usual for the intensity to be measured and, assuming a monochromatic source, the rate can be found from the time averaged intensity,

$$\lambda_I = \frac{1}{I_0} \int_a^b I(t) \, dt,$$

where $I_0$ is the energy of a single particle. Finally, the particle count distribution is,

$$f(n) = \frac{e^{\lambda_I} \lambda_I^n}{n!}, \quad (4.1)$$

where $n$ is the particle count.

For a given count, $n_m$, the expected measured intensity is $n_m I_0$, with measurement error $\delta n = n^{1/2} I_0$. The error can be reduced by increasing the measurement time and thus the particle count. It is worth noting that this noise component varies on a per-pixel basis and depends inverse quadratically on the particle count.

The particle count can also be undesirably increased due to other sources, including radiation and heat. In general, these sources will be static in time, after possibly a suitable warm-up period, and can be eliminated by first measuring the signal obtained under the same experimental conditions but with the source removed. The resulting measurement can then simply be subtracted from the actual measurements.

CCD detectors introduce an additional source of noise at data readout (caused by
CHAPTER 4. WORKING WITH EXPERIMENTAL DATA

the signal amplifiers and other electronics). This gives an apparent additional count, \( n_r \), which contributes to the count and is (generally, again after a suitable warm-up period) not time or signal dependent. The signal-to-noise ratio can therefore be written as,

\[
S/N = \sqrt{t} \frac{\sqrt{F_m}}{\sqrt{1 + F_b/F_m + n_r/(F_m t)}},
\]

(4.2)

where \( t \) is the measurement time, \( F_m \) is the measurement particle detection rate and \( F_b \) is the background rate. Longer exposures increase the signal count resulting in higher signal-to-noise ratios, but exposure time is bounded by the dynamic range and well saturation level of the CCD detector.

4.1.2 Systematic errors

In contrast to random errors, systematic errors can arise which could, in principle, be removed before measurement if they are known. A common cause of systematic error is the presence of additional, undesirable, scatterers. These may be in the form of dust particles or imperfections in sample substrates or base material. The measured diffraction pattern is then the sum of the diffraction pattern from the sample and the contaminant. If the scatterer is not accounted for the constraints may make the reconstruction infeasible or, at least, of lower quality. For example, for the common constraint of a finite support, if the scatterer is outside of the assumed sample finite support then this would potentially lead to the reconstruction algorithm failing to converge. If the support was extended to include the scatterer then the reconstruction would successfully recover both the sample and the scatterer, assuming oversampling conditions are still met.

4.1.3 Source Coherence

Source coherence is essential to achieving good diffractograms and subsequent reconstructions. Indeed, the sign of good coherence is the ability to form interference patterns with good contrast. In this section we briefly introduce spatial and temporal coherence before commenting on their effects on the quality of diffraction information.
Temporal coherence measures the correlation between the phase of the light wave at longitudinally separated points (along the direction of propagation). Temporal coherence is determined by the monochromaticity of the source. If a source emits waves of wavelength $\lambda \pm \delta \lambda$ then the waves will constructively and destructively interfere over some path delay due to the different wavelengths. The delay over which the waves significantly interfere is called the coherence time $\tau_c$ and the coherence length is the distance the wave travels in $\tau_c$.

Spatial coherence considers the correlation between the phase of the light at transversely separated points (transverse to the direction of propagation). Spatial coherence is determined by collimation and tells us how uniform the phase of the wavefront is.

Poor temporal coherence has the result of limiting the usable extent of the diffraction pattern, reducing the maximum resolution. Poor spatial coherence affects the entire diffraction pattern, possibly rendering reconstruction impossible.

### 4.1.4 Missing Data

Measured diffraction patterns are often incomplete due to experimental limitations. For both x-ray and optical laser experiments the undiffracted photon flux is very high and can damage, or at least saturate, the detector. Saturated detector wells lead to non-linearities in the count and can also lead to charge spill over into adjacent wells, both significantly degrade the measurement. Although only the center, undiffracted, part of the beam must be blocked due to physical construction considerations the beam stop is often a rectangular shape and blocks an entire quadrant (more precisely, slightly more than a quadrant).

The finite dimensions of the detector mean that only a fraction of the scattered photons are counted. Photons scattered by more than some angle are lost, giving rise to the maximum possible spatial resolution. In experiments of 3D samples only a small number of discrete 2D slices of the full 3D diffraction pattern are recorded.

In both classes of missing data it does not necessarily lead to problems in reconstruction. In the case of a missing quadrant and centro-symmetry the information
can be found in another quadrant. Otherwise, if the missing data is bounded in size (typically less than the speckle size) then the necessary information will be contained in redundant data in the remainder of the oversampled diffraction pattern or obtained from the constraints.

Reconstruction difficulties can occur for some situations. Iterative algorithms employing a spatial support constraint do not cope well if large regions of low frequency information are absent since it is this information that is used to determine the object’s basic shape. This can be understood by noting that if the missing regions are larger than the speckle size then there are functions well localized in both Fourier and direct space which meet the constraints. Although functions perfectly localized in both domains are impossible, there are functions in which the power is less than the noise power.

4.1.5 Dynamic Range

Diffractograms can exhibit an extremely high range of values, often differing by six or more orders of magnitude, and pose a significant challenge to the successful capture of such information. The range of values is referred to as the dynamic range, the difference between the largest and smallest data values. In practice, the dynamic range of a CCD is much less than that present in the diffractogram. For example, the Sony ICX429AK Exview CCD (used in the scaled optical experiment of Chapter 5) has a full-well capacity of at least 70,000 electrons and readout noise of less than 12 electrons RMS, leading to a maximum dynamic range of $70,000/12 = 5,833$ or approximately 12.5 bits. This limited dynamic range necessitates the combining of multiple exposures covering the desired range. Figure 4.1 demonstrates the use of different exposure times to capture a range wider than that of the detector.

4.2 Data Preparation

The true test of a successful experiment lies in the quality of the reconstructed image. However, obtaining this image requires analyzing and processing the acquired
measurements. Although no amount of data analysis can correct poorly obtained measurements, good data quality may prove useless if not analyzed well. Hence these pre-reconstruction procedures play a crucial role in lens-less imaging, and here we give them a fair amount of attention.

### 4.2.1 Background subtraction

In order to minimize the amount of stray scattering and instrument readout noise, we start by subtracting the background. This step is performed independently for each set of experimental conditions (exposure time, beam stop position and possibly sample rotation). If the background exposure shorter in duration than the measurement exposure then it must be rescaled. For measured intensities, $I_M$, and background intensity, $I_B$ the rescaling factor, $\alpha$ can be found by minimizing the difference between $I_M$ and $I_B$ over a region $S$ located behind the beam stop. Figure 4.2 illustrates the improvement in contrast after the background is subtracted.

### 4.2.2 Data centering

The position of the center of the measured diffraction pattern is not precisely known but may be essential knowledge for the reconstruction. If the center position is not
measured then it must be deduced from the data itself.

From the fundamental properties of the Fourier transform, the translation of a diffraction pattern manifests as a linear phase ramp in the direct space image. If a simple finite support constraint is used then this phase ramp has no effect on the projection and thus on the convergence of iterative algorithms. Complications arise when more complicated constraints are required. In an optically thick specimen the exit wave is expected to have values confined to one half of the complex plane (the imaginary part of the refractive index is always negative) but the phase ramp can interfere with this.

In the absence of a beam stop and detector saturation the center pixel could be identified as the brightest. Instead, the center must be recovered using other information. For centro-symmetric data\(^1\), the center may be found by inducing this symmetry as follows: choose a region in the vicinity of the center, and compare it with possible match regions to find the best match. Figure 4.3 illustrates this process.

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\(^1\)This is the case in most of our data as we deal with high energy x-rays.
Figure 4.3: The center of centro-symmetric data can be found by comparing different regions which would be symmetric for some choice of center position. The position that produces the minimum difference between regions is selected as the center.
4.2.3 Recovering Missing Information

The center position can be used to recover a missing quadrant. Once the center is determined, the missing quadrant data can be recovered by flipping the respective second quadrant.
Chapter 5

Reconstructions and Simulations

5.1 Simulation

Simulation was used to test both of the new approaches presented in Chapter 3, utilizing low resolution information through enforcing wavelet coefficients and the new formulation of the phase recovery problem as a convex optimization problem. The quality of the reconstructions was also compared to results obtained using standard reconstruction algorithms such as the hybrid input-output and difference map algorithms.

5.1.1 Wavelet domain constraints

The SEM image of Figure 5.1(a) was assumed as the exit surface wave of the diffracting object. The image values were real and positive. This wave was propagated into the far field by the discrete Fourier transform. The amplitudes of the Fourier coefficients were retained and used as the simulated diffraction measurement data for both reconstruction algorithms.

The low-resolution data to be used as a priori information, shown in Figure 5.1(b), was obtained by first transforming the SEM image of Figure 5.1(a) to the wavelet domain with four levels of decomposition. All of the detail coefficients were then set to zero and the image was reconstructed. This has the effect of replacing each $16 \times 16$
block of the original image with the average of the block.

The low-resolution information of 5.1(b) was used in the wavelet constraint and projection of equation 3.20. Figure 5.1(c) shows the reconstruction after 500 iterations. The successful reconstruction illustrates that even without a finite support the low-resolution information is sufficient. For comparison Figure 5.1(d) shows the result of 500 iterations of the hybrid input-output algorithm where the same diffraction data was used with the minimal rectangular support which covered the structure. The support was not refined over time with a dynamic support such as the “shrink wrap” algorithm [32]. The second copy of the structure is due to a phase ambiguity. In both cases the algorithms were initialized with random phases and both reliably converged to the results as shown across many initializations.

Low-resolution sample information is indeed sufficient to allow reconstruction from Fourier magnitude data. This indicates that it may be worthwhile to examine a range of different possible constraints to augment or supplant the existing widespread use of a finite spatial support constraint. These early results are qualitative only and no direct quantitative comparison to existing reconstruction methods has been performed. This is the subject of ongoing research.

This section demonstrated a successful reconstruction from simulated diffraction data without requiring a finite spatial support. Additional information in the form of a low-resolution image of the diffracting sample was used. This information formed a second constraint space which was expressed in the Haar wavelet domain. A suitable projection operator was given such that the framework of the Difference Map algorithm could be applied to reconstruct the sample. The reconstruction was successful and qualitatively was similar to the reconstruction from the well-studied HIO algorithm.

5.1.2 Convex optimization formulation

A reference sample was used with a single base value and two sections of a different value. A set of four possible values was used to form $\mathcal{D}$. These values do not represent
Figure 5.1: Iterative phase reconstruction where constraints are enforced in the Fourier domain and in the wavelet domain. (a) shows the original sample from which diffraction measurements are obtained (b) shows the low-resolution approximation used for the wavelet constraint (c) shows the reconstruction using constraints in wavelet space while (d) shows a reconstruction using a basic finite support constraint (the support is not modified as the algorithm progresses—as such the phase ambiguity is not resolved).
actual diffraction coefficients but serve to illustrate the behaviour of the algorithm.

\[
\beta = [0.39, 0.18, 0.28, 0.22] \\
\delta = [0.24, 0.11, 0.13, 0.22] \\
D_j = 1 - \beta_j - i\delta_j, \quad j = 1, \ldots, 4
\]

A variety of modifications were then made to this reference sample. In the following figures it should be noted that in most cases the actual values are not visible since the reconstruction perfectly matches the actual values. Also, note that the complex magnitudes are shown and thus the value indicated is used to indicate \(d_j \in D\). Figure 5.2 changes the width of the two components and adds a further two components of different widths.

Figure 5.2: In this example the widths of both existing structures are changed and two additional structures are added. The samples forming the structure are considered purely as samples with no associated physical scale. For (a) the upper graph shows the specification structure, the mid graph shows the actual structure and the lower graph shows the reconstruction at the final step. The reconstruction closely matches the actual structure. In (b) the objective value is seen to undergo two steps. These steps occur when the algorithm correctly escapes local minima.
In the previous example the actual values were perfectly reconstructed and convergence was generally good with between 20 and 45 iterations required. Figure 5.3 is an example where the correct reconstruction was not obtained. However, since all phase information has been lost and the modification is symmetric this solution is equally valid. The convergence of the optimization is initially very slow, most likely due to the phase ambiguity. This can be accelerated by an additional step of the algorithm where a guess is made of the correct value. The effect of this is shown by a temporary increase in the objective before it converges.

Figure 5.3: This example illustrates the ambiguity present with no phase information. The samples forming the structure are considered purely as samples with no associated physical scale. The initial structure shown in the top graph of (a) has two identical structures. The mid graph shows the modified structure where one width is increased. The reconstruction at the final step detects the change in width but for the incorrect structure. Due to the symmetry and loss of phase information this solution is entirely consistent with the measurements and the prior information. The solid objective curve shows where a guess on the correct value (step 5 of the algorithm) leads to faster convergence.
5.2 Scaled Optical Experiment

Experiments were performed using an optical diffraction experimental setup. The wavelength of the source (helium-neon) laser, 632.8 nm is considerably greater than hard x-rays but other experimental conditions were scaled to make the experiment comparable [46, 42]. We begin this section by detailing the experimental conditions before demonstrating reconstructions of scaled objects using the algorithms of Chapter 3.

A helium-neon (HeNe) laser was used with wavelength 632.8 nm. These low cost lasers offer a highly coherent beam with a coherence length exceeding 30 cm and are well collimated (typical divergence 1 to 2.5 mR). The beam is narrow but because of the collimation it can be expanded through a beam expander to a suitable diameter. Figure 5.5 illustrates the optical pathway of the experimental setup.

The scaling factor of the optical experiment is the ratio of the HeNe laser wavelength to the x-ray wavelength,

\[
\frac{\lambda_{\text{HeNe}}}{\lambda_{\text{x-ray}}} = \frac{632.8 \text{nm}}{1.4 \text{Å}} = 4520.
\]

To properly scale the experiment the sample structures must be scaled by this scaling factor, resulting in structures that are of the order of 150µm. To achieve the necessary indices of refraction microfluidic channel structures were formed on a PDMS
substrate.

The detector is a high dynamic range CCD sensor. The CCD detector has 752×580 pixels of size 8.2\( \mu m \times 8.4\mu m \) with full-well capacity of 70,000 electrons. The camera has a 16 bit ADC but with a read out noise of 12 electrons RMS the maximum dynamic range is approximately 5,833 (rather than 2\( ^{15} \)). The CCD sensor includes anti-blooming circuitry to minimize the effect of excessive incident light but this leads to a highly non-linear response and so saturation of the sensor was avoided.

Figure 5.6 shows the results of several reconstructions. Each of the structures imaged are isolated structures of approximately 300\( \times \)300\( \mu m \). The constructions correctly recover the outline of each structure. In effect the high pass version of the structure has been recovered.

The optical experiment demonstrated many of the difficulties that would be encountered in a x-ray diffraction experiment. Some of the points noted include the following

- Sensor dynamic range: Although quite high (the ADC is 16 bits) the dynamic range of the sensor is limited by non-linearities as it approaches saturation. To ensure that the response was linear only sensor values less than 2\( ^{15} \) were considered. Multiple exposures and multiple exposure times were combined to achieve the best possible signal to noise ratio and dynamic range.

- Beam stop: The center portion of the diffraction pattern is typically an order of magnitude more intense than the rest of the diffraction pattern. Normally a beam stop is used to block this part of the signal. In this experiment a ND filter was used to reduce the overall intensity of the diffraction pattern. This prevents the sensor from being overloaded but requires many exposures to recover the signal to noise ratio (and is still not optimum as the sensor system noise floor is a lower limit).

- Dark frame: For each exposure time used several dark frames were also obtained. Although the image sensor was cooled it was not cooled uniformly and the dark frame showed a distinct brightness in the lower portion of the frame.
Figure 5.5: Experimental setup. The narrow coherent laser source is expanded to a wider beam before illuminating the sample. A lens focuses the diffraction pattern on to the CCD image sensor. Rather than use a beam stop a ND filter was used to attenuate so that the CCD was not overloaded.

Figure 5.6: Reconstructions of various isolated structures. Notice that the high frequency has been reconstructed correctly but that the low frequency information is absent. In these reconstructions the central part of the diffraction data had saturated the detector and was removed. During the reconstruction these missing values were allowed to float and thus the low frequency components of the samples were not accurately recovered.
5.3 Soft X-ray Experiment

Both algorithms presented in Chapter 3 were tested on soft x-ray experimental data. The experiments were conducted at the Stanford Synchrotron Radiation Laboratory using 1.38 nm soft x-rays. The fabricated samples were simple, isolated structures approximately 100$\mu$m $\times$ 100$\mu$m. The experiment setup and reconstructions are shown in Figure 5.8 and Figure 5.9.

5.4 Hard X-ray Experiment

To explore the use of x-ray diffraction microscopy for buried structures on thick substrates several experiments were conducted using hard x-rays (0.18 nm or 7 KeV photons). This work was undertaken at the SPring8 facility in Harima Science Park City, Hyogo Prefecture, Japan. A photo of the experimental setup is shown in figure 5.10.

The samples that were prepared were fabricated as a single copper layer (100 nm high, 50 nm wide) on a 700 um silicon substrate. After fabricating the features a SEM was taken before the structures were buried under 1 um of SiO$_2$. An exemplary sample and its reconstruction are shown in figure 5.11.
Figure 5.8: The experiment was performed at Stanford Synchrotron Radiation Laboratory (SSRL) BL13-3 with 1.38 nm wavelength x-rays. A $1041 \times 1041$ pixel CCD with pixel size $20 \mu m$ was used to record the diffraction pattern for a $150 \times 75 \mu m$ area. Exposure times of one second and 0.1 seconds were used for the high quality and low quality information respectively.

(a) specification  (b) fabricated sample  (c) reconstruction

Figure 5.9: A fabricated sample (b) was imaged using the setup of Figure 5.8 and compared to a slightly different specification (a). The reconstruction (c) successfully recovers the added segment. The sample is approximately $2 \mu m \times 2 \mu m$ and the reconstruction is at $50 nm$. The reconstruction shown was achieved in 40 steps.
Figure 5.10: Setup photo
Figure 5.11: Sub-figure (a) shows a SEM micrograph of the sample before the SiO$_2$ was applied. The samples consisted of a single layer of copper wire (100 nm high and 50 nm wide) fabricated on a 700 um thick silicon substrate. Sub-figure (b) shows the successful reconstruction of the buried sample.
Chapter 6

Conclusion

Diffractive imaging of non-periodic objects has rapidly advanced over the past decade and, as the field matures, different lines of research are now just beginning to converge to a common understanding of the theoretical underpinnings of the phase reconstruction problem. Yet, there is still much to do, both in theoretical understanding and in overcoming the challenges presented on the experimental side. The use of diffractive imaging has been demonstrated in numerous fields and, particularly with new advances in coherent sources, it promises to expand into new directions.

In this thesis we have examined the recent progress of the algorithmic phase retrieval aspect of diffractive imaging. We began with a brief summary of the physical processes involved in light diffraction microscopy before describing the dominant family of methods of phase retrieval: iterative algorithms. We described the origins of the additional information necessary in existing algorithms with the requirement that the object has a finite spatial support. This is severely constraining as a key future application of coherent x-ray diffraction microscopy is inspection of fabricated integrated circuits. Such objects are spatially constrained but are much larger than the field of view of any source/detector and require modifications to existing algorithms and/or new algorithms. We have also discussed some of the technical challenges in experimental diffraction microscopy and provided guidance on experimental setup and data preparation.

One of the most promising future applications of diffraction microscopy is the
imaging of single macromolecules by an x-ray free electron laser. The very short pulse of the laser are destructive but many diffractograms of identical molecules can be obtained and assembled to form a complete dataset suitable for reconstruction [38, 40]. This assembly is extremely challenging as the individual diffractograms are both noisy and of random orientation but early results have already shown successful reconstructions.
Bibliography


BIBLIOGRAPHY


