

XUV Lensless Imaging with Spatially Shaped High-Harmonic Beams



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A thesis submitted for the degree of

Doctor of Philosophy

Trinity Term 2019

Abstract

This thesis presents two novel advances in the field of high harmonic generation along with a demonstration of application to the lensless imaging technique known as Fourier Transform Holography. Also presented are images obtained with the technique of coherent diffractive imaging.

The first advance is that of an increase in harmonic brightness through the manipulation of the spatial phase of the driver laser. The transverse driver intensity in the focal plane of a fixed lens was shaped from Gaussian to supergaussian whilst the spot size was concomitantly increased. Within the range of experimental values, the increasing supergaussian order enabled the harmonic source size to increase at a faster rate than the driver spot size. This changed the scaling relationship between the detected harmonic flux and driver spot size from w_0^2 , to $w_0^{2.3 \pm 1.72}$. Without changing the focussing optic, the brightness of the harmonic beam was increased by a factor of 5 ± 0.65 .

The second advance is that of the generation of multiple harmonic sources through the manipulation of the spatial phase of the driver laser. The intensity distribution of the driver beam was spatially shaped to form a user defined distribution of foci with programmable positions and relative energies. For two foci, experimental results are presented showing the generation of two independent but mutually coherent harmonic beams. Using these two harmonic beams, a Fourier Transform Holography target comprising two distinct features was illuminated. It is shown that under comparable conditions, distributing the illumination to match the shape of the target yielded an image with a superior signal to noise ratio.

Acknowledgments

My immeasurable gratitude goes first to Dr David Lloyd for instilling in me the analytical and objective mindset required to find the most efficient and effective solution to any challenge. This is a mentality I will carry with me for the rest of my life, for which I am extremely thankful.

I would also like to express my enormous appreciation for the lessons of Dr Kevin O’Keeffe who gave me the knowledge, ability and confidence to unashamedly call myself an experimental physicist.

The value of the advice and guidance provided by Prof. Simon Hooker cannot be overstated. Without it I would have been lost, lacking the motivation and understanding required to complete my work. With it, I have achieved what I would have considered impossible four years ago.

Additional thanks go to

- Dr Jason Brown for instruction in the use of scanning electron microscopes and focussed ion beams.
- Dr David Sharp for instruction in the use of atomic force microscopes.
- Dr Enrico Salvati in Prof. Korsunsky’s group for spending an unreasonable amount of time with me trying to engineer impossible things.
- The rest of the Hooker group for keeping me entertained over the last four years.

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Acronyms

$\mathcal{D}$ detector domain. [9](#), [14](#), [18](#), [22–26](#), [29](#), [31–33](#), [35](#), [39](#), [70](#), [95](#), [107](#), [109](#), [121](#), [122](#)

$\mathcal{O}$ object domain. [9](#), [14](#), [17](#), [18](#), [21–25](#), [28–35](#), [70](#), [71](#), [106](#), [107](#), [109](#), [110](#), [114](#)

ASM angular spectrum method. [18](#), [21](#), [22](#), [111](#), [116](#), [139](#)

BPP beam parameter product. [103](#), [105](#), [110](#), [114](#), [119](#), [122](#), [124](#), [126–128](#), [130](#)

CCD charge coupled device. [22](#), [65](#), [68](#), [70–72](#), [92](#), [93](#), [97](#), [106](#), [107](#), [109](#), [135](#), [140](#)

CCF complex coherence function. [26](#)

CDI coherent diffractive imaging. [3](#), [5](#), [24–29](#), [33](#), [34](#), [36](#), [37](#), [61–63](#), [94](#), [97](#), [99](#), [100](#),
[102](#), [147](#)

ER error reduction. [31](#), [32](#)

ESW exit surface wave. [22](#), [23](#), [25](#), [28–30](#), [97](#), [98](#)

FIB focussed ion beam. [72–75](#), [95](#), [99](#)

FoV field of view. [28](#)

FRC Fourier ring correlation. [38](#), [39](#), [98](#), [101](#), [147](#)

FTH Fourier transform holography. [1](#), [3](#), [5](#), [24–26](#), [34](#), [36](#), [37](#), [62](#), [63](#), [99](#), [100](#), [102](#),
[133–135](#), [138](#), [141](#), [145](#), [147](#), [148](#)

GS Gerchberg-Saxton. [29](#), [30](#)

HDR high dynamic range. [37](#), [96–98](#)

HHG high-harmonic generation. [2–5](#), [41–46](#), [51](#), [57–59](#), [62](#), [63](#), [65](#), [67](#), [93](#), [103–105](#),
[107](#), [113](#), [131–134](#), [142](#), [145](#), [146](#), [148–150](#)

HIO hybrid input-output. [31](#), [32](#), [97](#)

IPRA iterative phase retrieval algorithm. [1](#), [25](#), [26](#), [28](#), [29](#), [33](#), [36](#), [37](#), [61](#), [62](#), [97](#), [98](#),
[149](#)

LCoS liquid crystal on silicon. [67](#), [78](#), [79](#), [83](#), [146](#)

LCs liquid crystals. [76–79](#), [83](#), [85](#), [87](#), [88](#)

NA numerical aperture. [8](#), [9](#), [37](#), [101](#)

PAN parallel aligned nematic. [67](#), [76](#), [77](#), [84](#)

RoI region of interest. [28](#), [89](#), [90](#)

S:N signal to noise ratio. [3](#), [9](#), [31](#), [36–38](#), [62](#), [98](#), [104](#), [133](#), [142](#), [145](#), [148](#)

SLM spatial light modulator. [4](#), [5](#), [64](#), [65](#), [67](#), [68](#), [75–86](#), [88–92](#), [94](#), [104](#), [113](#), [115–117](#),
[130](#), [132](#), [134](#), [136](#), [137](#), [146](#), [147](#), [149](#)

XFEL x-ray free electron laser. [2](#), [4](#), [41](#), [61](#), [62](#), [103](#)

XUV extreme ultraviolet. [3](#), [4](#), [41](#), [69–71](#), [102–104](#), [130](#), [133](#), [134](#), [141–143](#), [146](#), [147](#),
[150](#)

Chapter 1

Introduction

1.1 Motivation



LENSLESS imaging is an approach to satisfying the need for ever finer resolution images requiring illuminating radiation in short wavelength spectral regions that cannot be efficiently manipulated by traditional optics. An overview of some of the common lensless imaging modalities is given by Chapman in reference [1]. As the wavelength of the illumination decreases into the ultraviolet, traditional optics are dispensed with, and diffraction takes the place of refraction. In addressing one issue however, another arises: The phase problem (explored in section 2.2.5). This can be solved through the application of an [iterative phase retrieval algorithm \(IPRA\)](#), reviews of which are given by Fienup in reference [2] and Marchesini in reference [3]. An alternate solution to the phase problem is holography, which is considered in this thesis only in its implementation as [Fourier transform holography \(FTH\)](#). With the simultaneous illumination of the object and a reference aperture, [FTH](#) encodes the lost object phases into the interference fringes detected by the detector. This enables a single Fourier transform to reveal the cross-correlation of the reference and object. In the case where the reference is a point like feature, this cross-correlation can be considered an image of the object.

CHAPTER 1. INTRODUCTION

1.1. MOTIVATION

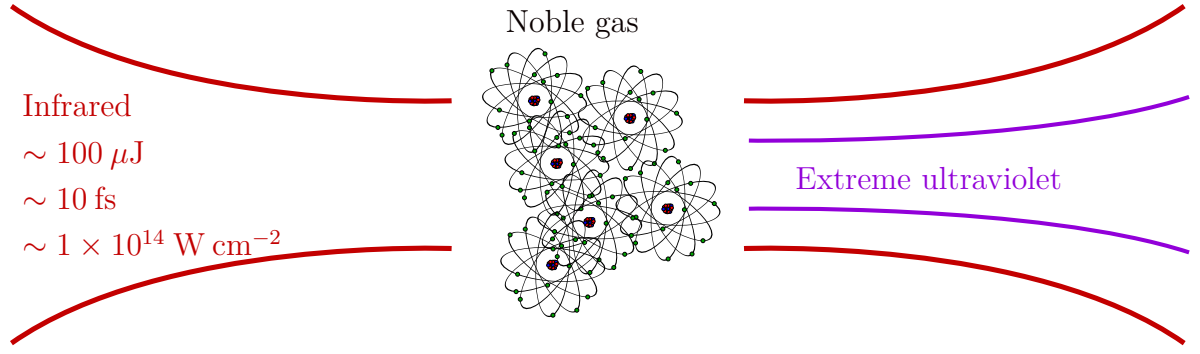


Figure 1.1: A schematic diagram of the HHG process. An infrared driver beam (solid red lines) of focal peak intensity $\approx 1 \times 10^{14} \text{ W cm}^{-2}$ interacts with a noble gas to produce a comb of discrete harmonics (solid violet lines) extending from the ultraviolet to the soft x-ray.

The brightest sources of short wavelength radiation are the latest generations of synchrotrons and [x-ray free electron laser \(XFEL\)](#)s. However, these facilities require an enormous amount of space and funding for both their construction and maintenance; the construction cost of the Diamond synchrotron (Oxfordshire, UK) which covers $\approx 66000 \text{ m}^2$ was £260 million [4]. In addition, although the discoveries that are performed at such facilities are groundbreaking, the acceptance rate of user proposals at the Diamond synchrotron is approximately 73% [5], implying that the science behind a non-negligible number of proposals is either delayed or goes undiscovered. An alternative source of short wavelength radiation that is accessible to small research facility scale laboratories can be found in [high-harmonic generation \(HHG\)](#), schematically depicted in figure 1.1. The cost of constructing a beamline capable of producing high harmonics is of the order of 0.1% that of a synchrotron, whilst the physical area required to house it is less than $\approx 10 \text{ m}^2$. Implicitly, this involves a compromise between affordability and photon flux which motivates the search for methods that increase the flux and brightness of harmonic sources.

The author's original objective upon starting his doctoral work was to increase the coherence of a high-harmonic beam by spatially shaping the transverse spatial intensity

CHAPTER 1. INTRODUCTION

1.2. SCOPE OF THIS THESIS

profile of the driving beam. In particular it was anticipated that changing the transverse profile of the driver from a Gaussian to a top-hat function would minimise the variation of the intensity dependent dipole phase (explored in section 3.3.3) and thus improve the harmonic coherence [6]. Unfortunately the experiments designed to measure the harmonic coherence yielded a null result. This was likely due to the small value of the expected change compared to the experimental uncertainties in the measurements. Factors such as the instability of the shaped profile, pointing error in the driver beam and low [signal to noise ratio \(S:N\)](#) of detected data combined to obscure any change in coherence caused by the dipole phase. This conclusion made a study of the harmonic coherence within this thesis somewhat unnecessary; an observation that marked the end of an approximately 18 months long experimental campaign. Subsequently, the author's work changed direction toward novel methods of manipulating harmonic radiation through spatially shaping the driver beam.

It was initially anticipated that after improving the coherence of the harmonic beam, the resolution of images recovered with coherent imaging techniques such as [coherent diffractive imaging \(CDI\)](#) would improve. This required an [extreme ultraviolet \(XUV\)](#) lensless microscope capable of performing [CDI](#), which the author constructed and optimised over the period of several years. In spite of the aforementioned null result, the author built and used this microscope to perform [CDI](#) and [FTH](#), constituting the first experimental demonstration of lensless imaging in the [XUV](#) realised within the author's research group. The results of the early imaging experiments will be presented in this thesis as a record of the author's first few years of doctoral work.

1.2 Scope of this Thesis

This thesis is based on work performed by the author to increase the utility of the non-linear optical process of [HHG](#). Two distinct approaches were taken that have separately

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formed two bodies of work that comprise chapters 6 and 7. The later chapter has been published by Optics Express as reference [7].

Chapter 6 describes a method which improves the scaling of harmonic flux with the driver spot size from a power of 2 to a power of 2.3 ± 1.72 . The uncertainties in this measure encompass the improvement in scaling predicted by supporting simulations. This enhancement is achieved by spatially shaping the focal intensity of the fundamental beam with a phase-only [spatial light modulator \(SLM\)](#) at the focus of a fixed lens. The altered scaling leads to an increase in brightness without the need to change the focussing optic, lending a new degree of flexibility to tabletop [HHG](#). This also contributes to the demand for high photon flux of [XUV](#) light required by many applications ranging from imaging to seeding [XFELs](#).

Chapter 7 describes a method to independently control the locations and relative powers of multiple decoupled harmonic beams. The utility of this approach is illustrated by performing a first of its kind holography experiment with two decoupled harmonic beams. Having optimally distributed the harmonic illumination, for a given incident harmonic flux, an image with a superior signal to noise ratio is recovered than could be achieved by a single harmonic beam of the same total harmonic flux. This newfound flexibility will facilitate a multitude of new experiments and enable the use of much shorter acquisition times.

As a precursor to the aforementioned advances, chapter 5 presents the first demonstration of lensless imaging in the author's research group, along with incidental observations considered relevant.

The remaining chapters are organised as follows; Chapter 2 outlines the theoretical background behind both modalities of lensless imaging demonstrated by the author -

CDI and FTH along with methods for determining the resolution of a lensless imaging system. Chapter 3 contains the theoretical background to HHG. Chapter 4 describes the constituent components of the experimental beamline used in this thesis, along with the operating principle behind the SLM. Finally, chapter 8 contains conclusions and an outlook to possible future work.

1.3 Role of the Author

All work presented in this thesis was performed and analysed by the author under the supervision of Prof. Simon Hooker. Post doctoral research associate Dr David Lloyd and graduate student Mr Florian Wiegandt provided advice and guidance throughout. Dr Kevin O’Keeffe also provided invaluable advice and guidance throughout, although Dr O’Keeffe relocated to Swansea University for the latter two years of the author’s time as a graduate student.

1.4 Publications

Below are a list of publications of which the author is either a co-author or first author. First author publications will be listed in bold.

- D. J. Treacher, D. T. Lloyd, F. Wiegandt, K. O’Keeffe, and S. M. Hooker
“Optimised XUV holography using Spatially Shaped High Harmonic Beams”. *Optics Express*, **27**, 20 (2019) - doi: [10.1364/OE.27.029016](https://doi.org/10.1364/OE.27.029016)
- D. J. Treacher, D. T. Lloyd, F. Wiegandt, K. O’Keeffe, and S. M. Hooker
“Increasing the Brightness of Harmonic XUV Radiation with Spatially-Tailored Driver Beams”. *In Progress*
- M. M. Mang, D. T. Lloyd, P. N. Anderson, D. Treacher, A. S. Wyatt, S. M. Hooker, I. A. Walmsley, and K. O’Keeffe “Spatially resolved common-path high-order har-

monic interferometry”. *Optics Letters*, **43**, 21 (2018) - doi: [10.1364/OL.43.005275](https://doi.org/10.1364/OL.43.005275)

- Florian Wiegandt, Patrick N. Anderson, Fei Yu, Daniel J. Treacher, David T. Lloyd, Peter J. Mosley, Simon M. Hooker, and Ian A. Walmsley “Quasi-phase-matched high-harmonic generation in gas-filled hollow-core photonic crystal fiber”. *Optica*, **6**, 4 (2019) - doi: [10.1364/OPTICA.6.000442](https://doi.org/10.1364/OPTICA.6.000442)

1.5 Conference Contributions

Below are a list of conference contributions of which the author is either a co-author or first author. First author contributions will be listed in bold.

- Patrick N. Anderson, Florian Wiegandt, Daniel J. Treacher, Matthias M. Mang, Ilaria Gianani, Andrea Schiavi, David T. Lloyd, Kevin O’Keeffe, Simon M. Hooker, Ian A. Walmsley “Blind digital holographic microscopy”. *Proceedings Volume 10127, Practical Holography XXXI: Materials and Applications; 101270H* (2017) - doi: [10.1117/12.2250317](https://doi.org/10.1117/12.2250317)
- David T. Lloyd, Kevin O’Keeffe, Adam S. Wyatt, Patrick N. Anderson, Daniel Treacher, Simon M. Hooker “Combined visible and near-infrared OPA for wavelength scaling experiments in strong-field physics”. *Proceedings Volume 10088, Nonlinear Frequency Generation and Conversion: Materials and Devices XVI; 1008815* (2017) - doi: [10.1117/12.2250775](https://doi.org/10.1117/12.2250775)
- Patrick N. Anderson, Florian Wiegandt, Fei Yu, Daniel J. Treacher, David T. Lloyd, Peter J. Mosley, Simon M. Hooker, Ian A. Walmsley “Multimode quasi-phase-matching of high-order harmonic generation in gas-filled photonic crystal fibers”. *Lasers and Electro-Optics Europe (CLEO EUROPE/EQEC), Conference on and European Quantum Electronics Conference* (2017) - doi: [10.1109/CLEOE-EQEC.2017.8086804](https://doi.org/10.1109/CLEOE-EQEC.2017.8086804)
- Daniel Treacher, David T. Lloyd, Kevin O’Keeffe, Patrick N. Anderson, Simon

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M. Hooker “**Improving the resolution obtained in lensless imaging with spatially shaped high-order harmonics**”. *Lasers and Electro-Optics Europe (CLEO EUROPE/EQEC), Conference on and European Quantum Electronics Conference* (2017) - doi: [10.1109/CLEOE-EQEC.2017.8086546](https://doi.org/10.1109/CLEOE-EQEC.2017.8086546)

Chapter 2

Imaging Theory



MAGING is integral to how we understand the world around us. From galaxies in deep space to the atomic structure of everyday materials, the extraction and representation of information in a visual manner is unparalleled in providing insight into our surroundings. Throughout history a steady advance has been made from the first attempts to build primitive lenses [8], to a full mathematical description of the behaviour of light [9]. This has culminated today in equipment that can afford the user images with near atomic scale resolution.

2.1 Motivation

In 1873, Ernst Abbe proposed that the minimum achievable resolution, d , in a microscope is a function of both the illumination geometry and wavelength λ [10]:

$$d = \frac{\lambda}{2n \sin(\theta)}. \quad (2.1)$$

Here n is the local refractive index and θ is half the imaging system acceptance angle. The product $n \sin(\theta)$ gives the [numerical aperture \(NA\)](#) of the system. This limit originates from the diffraction of light and details the radius of the zeroth diffraction

order in the Airy disk produced when imaging a point source. Given the form of equation 2.1, the resolution limit in an imaging system can be reduced by either increasing the NA, or decreasing λ . The NA can be maximised by bringing the imaging lens and object very close together, but is intrinsically limited to approximately unity*. However, the use of shorter imaging wavelengths is complicated by the low quality and high absorption of traditional refractive optics in this spectral range. This limitation can be circumvented by the removal of the lens from the imaging system such that in place of an image, a diffraction pattern will be detected instead. Under appropriate conditions, numerical techniques can invert the detected diffraction pattern into an image with a resolution now limited only by the NA, S:N of the pattern itself and the radiation tolerance of the object [12].

2.2 Imaging

A single lens imaging system comprises an object in the object domain ($\mathcal{O}$) domain, a lens, and an image in the image domain I . As the illumination leaving the object transitions through the lens, the phase is changed only by the addition of the phase curvature characterised by the focal length. In the focal plane, this results in an image that is easily recognisable as representative of the object. In order to access a smaller resolution size, the NA is increased to its limit and the illuminating wavelength is decreased. However, lenses and optics for use with shorter wavelengths are typically inefficient and not readily available. As a result, the lens is removed and the physical process of refraction is replaced by diffraction. It is the diffracted intensity that is subsequently detected in the detector domain ($\mathcal{D}$) domain. Consequently, a mathematical framework within which diffraction can be considered is required. For the purposes of this thesis, the framework of scalar diffraction theory is adequate.

*This limit can be exceeded in wet optical microscopy using immersion oils into which the lens and sample are immersed giving an NA up to approximately 1.6 [11]

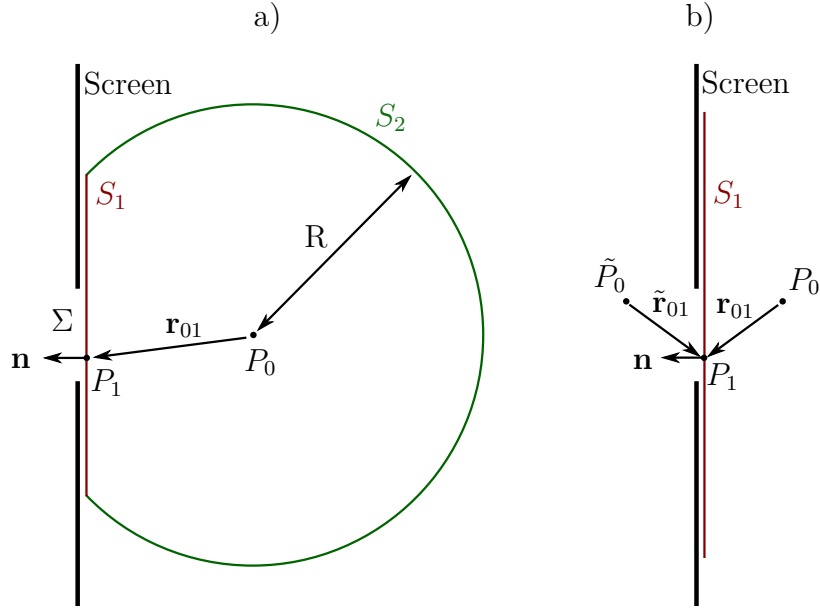


Figure 2.1: Geometries employed to describe scalar diffraction. (a) shows an aperture Σ embedded within a planar screen. The surfaces S_1 and S_2 encompass the volume V described in the text. (b) shows a zoomed view of the aperture. Figure replicated from reference [13].

2.2.1 Scalar Diffraction Theory

Following the analysis presented by Goodman [13], in order to find the complex field U at a point P_0 , downstream of an aperture Σ as shown in figure 2.1 a), we start by invoking Green's theorem [14]. This theorem states that if $U(P)$ and $G(P)$ are two complex valued functions of position and their first and second partial derivatives are single valued and continuous within and on a closed surface S surrounding a volume V , then

$$\iiint_V (U \nabla^2 G - G \nabla^2 U) dv = \iint_S \left(U \frac{\partial G}{\partial \mathbf{n}} - G \frac{\partial U}{\partial \mathbf{n}} \right) ds. \quad (2.2)$$

Here, the outward pointing unit normal is denoted $\mathbf{n}$. In order to find a tractable expression for the desired field U , an ansatz of a spherical wave expanding about the point P_0 is made for G such that its value at point P_1 is

$$G(P_1) = \frac{\exp(ik_0 \mathbf{r}_{01})}{\mathbf{r}_{01}}, \quad (2.3)$$

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where $\mathbf{r}_{01}$ is the vector pointing from P_0 to P_1 and k_0 is the wavenumber equal to $2\pi/\lambda$. It can be shown that under simplifications rigorously explained in reference [13], equation 2.2 combined with the spherical wave ansatz yields the integral theorem of Helmholtz and Kirchhoff:

$$U(P_0) = \frac{1}{4\pi} \iint_{S=S_1+S_2} \left(\frac{\partial U(P_1)}{\partial \mathbf{n}} G - U(P_1) \frac{\partial G}{\partial \mathbf{n}} \right) ds. \quad (2.4)$$

In order to calculate this integral, the surface S is chosen as the sum of the red plane S_1 and the green spherical cap S_2 of radius R as shown in figure 2.1. Considering the second term on the right hand side of equation 2.4, it can be shown through the properties of the directional derivative [15] that

$$\frac{\partial G}{\partial \mathbf{n}} = \nabla_{\mathbf{r}} G \cdot \mathbf{n} = \cos(\mathbf{n}, \mathbf{r}_{01}) \left(ik_0 - \frac{1}{\mathbf{r}_{01}} \right) \frac{\exp(ik_0 \mathbf{r}_{01})}{\mathbf{r}_{01}}. \quad (2.5)$$

where $\cos(\mathbf{n}, \mathbf{r}_{01})$ is the cosine of the angle θ_{01} between the outward pointing unit normal $\mathbf{n}$ and the vector $\mathbf{r}_{01}$ connecting P_0 to P_1 in figure 2.1.

Upon the surface S_2 , where $\mathbf{r}_{01} = R$, equation 2.5 can be approximated by $ik_0 G$ when R becomes large. This enables the integral in equation 2.4 over S_2 to be written as

$$\iint_{S_2} \left(G \frac{\partial U}{\partial n} - ik_0 U G \right) ds = \int_{\Omega} G \left(\frac{\partial U}{\partial n} - ik_0 U \right) R^2 d\omega, \quad (2.6)$$

where the right hand side has changed variable from surface area to solid angle using the definition $ds/R^2 = d\Omega$ [16], where Ω is the angle subtended by S_2 at P_0 . In order to ensure that equation 2.6 has only a unique solution describing outgoing waves (travelling toward $R = \infty$), a condition known as the Sommerfeld radiation condition [17, 18] is imposed upon U :

$$\lim_{R \rightarrow \infty} R \left(\frac{\partial U}{\partial n} - ik_0 U \right) = 0. \quad (2.7)$$

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When this is satisfied, the outgoing field U can be assumed to decay at the same rate as a diverging spherical wave, thus the corresponding integral over S_2 tends to zero at large R . This enables the integral in equation 2.4 to be considered only over S_1 .

To compute the remaining integral over S_1 in equation 2.4, typically Kirchhoff boundary conditions comprising two assumptions are adopted. First, within the aperture Σ embedded within an opaque screen, the field U and its partial derivative $\partial U/\partial n$ are the same as they would be if there were no screen. Secondly, over S_1 that lies behind the screen, both U and $\partial U/\partial n$ are exactly zero. However, it can be shown that for the latter condition to be true, the function U must have a value of zero everywhere revealing a lack of self consistency in these conditions [19].

An alternate route to solving the integral over S_1 that remains self consistent is found by making a different ansatz for G comprising the addition of two point sources at mirrored locations either side of S_1 as shown in figure 2.1 b). The new ansatz can be written as

$$G^\pm(P_1) = \frac{\exp(ik_0\mathbf{r}_{01})}{\mathbf{r}_{01}} \pm \frac{\exp(ik_0\tilde{\mathbf{r}}_{01})}{\tilde{\mathbf{r}}_{01}} \quad (2.8)$$

where $\tilde{\mathbf{r}}_{01}$ is the position vector of point $\tilde{P}_0$. The $\pm$ between the two terms corresponds to the equally valid ansätze of two point sources, radiating either 0 or π radians out of phase. These are known as the positive and negative ‘alternate Green’s functions’ respectively.

The normal derivative of the positive Green’s function vanishes over Σ when we take $\mathbf{r}_{01} = \tilde{\mathbf{r}}_{01}$ and $\cos(\mathbf{n}, \mathbf{r}_{01}) = -\cos(\mathbf{n}, \tilde{\mathbf{r}}_{01})$, revealing the corresponding expression for U^+ from equation 2.4:

$$U^+(P_0) = \frac{1}{4\pi} \iint_{\Sigma} \frac{\partial U(P_1)}{\partial n} G^+ ds. \quad (2.9)$$

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The negative Green's function itself vanishes on Σ such that

$$U^-(P_0) = \frac{-1}{4\pi} \iint_{\Sigma} U(P_1) \frac{\partial G^-}{\partial n} ds. \quad (2.10)$$

When both equations described by 2.8 are substituted into equations 2.9 and 2.10, we find that the complex fields can be described as a function of the original ansatz in equation 2.3:

$$U^+(P_0) = \frac{1}{2\pi} \iint_{\Sigma} \left[\frac{\partial U(P_1)}{\partial \mathbf{n}} \frac{\exp(ik_0 \mathbf{r}_{01})}{\mathbf{r}_{01}} \right] ds, \quad (2.11)$$

and

$$U^-(P_0) = \frac{1}{4\pi} \iint_{\Sigma} \left[U(P_1) \cos(\mathbf{n}, \mathbf{r}_{01}) \left(ik_0 - \frac{1}{\mathbf{r}_{01}} \right) \frac{\exp(ik_0 \mathbf{r}_{01})}{\mathbf{r}_{01}} \right] ds \quad (2.12a)$$

$$= \frac{1}{i\lambda} \iint_{\Sigma} \left[U(P_1) \cos(\mathbf{n}, \mathbf{r}_{01}) \frac{\exp(ik_0 \mathbf{r}_{01})}{\mathbf{r}_{01}} \right] ds, \quad (2.12b)$$

where equation 2.12a has been simplified by assuming that $\mathbf{r}_{01} \gg \lambda$, allowing the second term within the integral to be dropped. When the aperture Σ is illuminated from the opposite side to P_0 by spherical waves emanating from a point source P_2 located at position $\mathbf{r}_{21}$,

$$U(P_1) = \frac{\exp(ik_0 \mathbf{r}_{21})}{\mathbf{r}_{21}}. \quad (2.13)$$

When this source term is substituted into equation 2.11 or 2.12b we find the two forms of the Rayleigh-Sommerfeld diffraction formula:

$$U^+(P_0) = \frac{-1}{i\lambda} \iint_{\Sigma} \left\{ \cos(\mathbf{n}, \mathbf{r}_{21}) \frac{\exp[ik_0(\mathbf{r}_{01} + \mathbf{r}_{21})]}{\mathbf{r}_{01}\mathbf{r}_{21}} \right\} ds \quad (2.14a)$$

$$U^-(P_0) = \frac{1}{i\lambda} \iint_{\Sigma} \left\{ \cos(\mathbf{n}, \mathbf{r}_{01}) \frac{\exp[ik_0(\mathbf{r}_{01} + \mathbf{r}_{21})]}{\mathbf{r}_{01}\mathbf{r}_{21}} \right\} ds. \quad (2.14b)$$

At this stage, three different Ansätze have been inspected as possible solutions to

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equation 2.4; Kirchhoff's G and Sommerfeld's positive and negative $G^\pm$. It can be shown that the corresponding solutions, $U(P_0)$, $U^+(P_0)$, and $U^-(P_0)$ differ only by what is known as the obliquity factor ψ . When $\mathbf{r}_{21} = \infty$ and the illumination comprises normally incident plane waves, this factor is defined in each case as

$$\psi = \begin{cases} \frac{1}{2} [1 + \cos(\mathbf{n}, \mathbf{r}_{01})] = \frac{1}{2} [1 + \cos(\theta_{01})] & \text{in } U(P_0) \\ 1 & \text{in } U^+(P_0) \\ \cos(\mathbf{n}, \mathbf{r}_{01}) = \cos(\theta_{01}) & \text{in } U^-(P_0), \end{cases} \quad (2.15)$$

where θ_{01} is the angle between $\mathbf{n}$ and $\mathbf{r}_{01}$.

When only small angles are considered, ψ tends to unity in all three cases in equation 2.15. Consequently, we progress with this analysis using $U^-(P_0)$ as representative of all three cases in the small angle approximation. Accordingly, a final substitution of $\cos(\theta_{01}) = z/\mathbf{r}_{01}$ into equation 2.12b gives an expression for the field $P_0 \in (X, Y)$ detected at a point within the $\mathcal{D}$ domain, as a function of the field in the $\mathcal{O}$ domain within the aperture $\Sigma \in (x, y)$. This is known as the Huygens-Fresnel principle:

$$U(X, Y) = \frac{z}{i\lambda} \iint_{\Sigma} U(x, y) \frac{\exp(ik_0 \mathbf{r}_{01})}{\mathbf{r}_{01}^2} dx dy. \quad (2.16)$$

where z is the longitudinal distance separating the $\mathcal{O}$ and $\mathcal{D}$ domains, and

$$\mathbf{r}_{01} = \sqrt{z^2 + (X - x)^2 + (Y - y)^2}. \quad (2.17)$$

Figure 2.2 shows a schematic diagram of the diffraction geometry being considered. From the Huygens-Fresnel principle in equation 2.16, simplifications can be made that enable the mathematical representation of diffraction to be separated into different regimes according to the relative sizes of z and the aperture Σ . We discriminate therefore

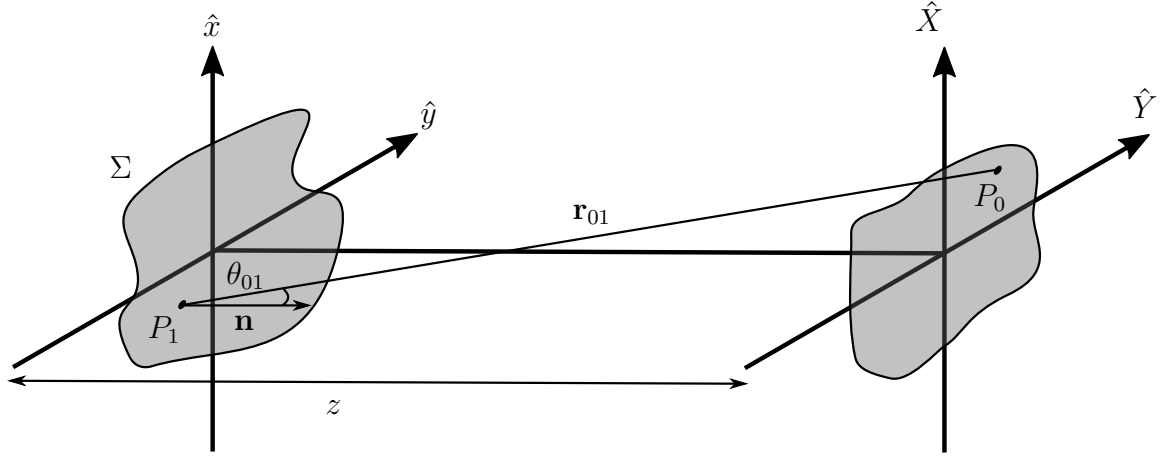


Figure 2.2: Schematic diagram of the diffraction from a point P_1 within an aperture Σ in the $\mathcal{O}$ domain described by coordinates (x, y) being detected at point P_0 in the $\mathcal{D}$ domain described by coordinates (X, Y) a distance z downstream.

between situations where z is much less than the aperture size, approximately the aperture size, or much larger than the aperture size. These are known as the geometrical optics, Fresnel and Fraunhofer regimes respectively and differ in the approximations applied to equation 2.17, as explored below.

2.2.2 Fresnel Diffraction

When only small angles are considered such that $(X - x)^2$ and $(Y - y)^2$ are much smaller in magnitude than z , the $\mathbf{r}_{01}$ in the denominator of equation 2.16 can typically be approximated by z . However, a higher degree of accuracy is required in the approximation of $\mathbf{r}_{01}$ that appears in the exponential since this quantity oscillates with a period of λ and is therefore sensitive to within fractions of a wavelength. Consequently, a binomial expansion of equation 2.17 up to the first order is required:

$$\mathbf{r}_{01} \approx z \left[1 + \frac{1}{2} \left(\frac{X - x}{z} \right)^2 + \frac{1}{2} \left(\frac{Y - y}{z} \right)^2 \right]. \quad (2.18)$$

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Substituting this into equation 2.16 gives

$$U(X, Y) = \frac{e^{ik_0 z}}{i\lambda z} \iint_{-\infty}^{\infty} U(x, y) \exp\left(\frac{ik_0}{2z} [(X - x)^2 + (Y - y)^2]\right) dx dy, \quad (2.19)$$

which can be interpreted as the convolution integral

$$U(X, Y) = \iint_{-\infty}^{\infty} U(x, y) h(X - x, Y - y) dx dy. \quad (2.20)$$

where

$$h(X, Y) = \frac{e^{ik_0 z}}{i\lambda z} \exp\left[\frac{ik_0}{2z} (X^2 + Y^2)\right] \quad (2.21)$$

is known as the free space propagation function. By expanding the exponent in equation 2.19 we can write

$$U(X, Y) = \frac{e^{ik_0 z}}{i\lambda z} e^{\frac{ik_0}{2z} (X^2 + Y^2)} \iint_{-\infty}^{\infty} \left\{ U(x, y) \exp\left[\frac{ik_0}{2z} (x^2 + y^2)\right] \right\} \exp\left[-i\frac{2\pi}{\lambda z} (Xx + Yy)\right] dx dy, \quad (2.22)$$

which is the Fourier transform of

$$U(x, y) \exp\left[\frac{ik_0}{2z} (x^2 + y^2)\right], \quad (2.23)$$

evaluated at spatial frequencies of $k_{x,y} = \frac{(x,y)}{\lambda z}$, having taken the definition of the continuous Fourier transform as

$$\text{FT}[f(x, y)] = F(k_x, k_y) = \iint_{-\infty}^{\infty} f(x, y) \exp[-2\pi i(k_x x + k_y y)] dx dy. \quad (2.24)$$

Equation 2.22 is said to describe the ‘Fresnel’ regime of diffraction.

2.2.3 Fraunhofer Diffraction

A further approximation can be made by considering Fresnel diffraction at a distance

$$z \gg \frac{k_0}{2}(x^2 + y^2) \quad (2.25)$$

In this case, the variation in the quadratic phase factor in expression 2.23 across the aperture can be neglected and the diffracted field $U(X, Y)$ can be taken as proportional to the Fourier transform of only the input field $U(x, y)$ multiplied by some pre-factors. This is known as Fraunhofer diffraction:

$$U(X, Y) = \frac{e^{ik_0 z}}{i\lambda z} e^{\frac{ik_0}{2z}(X^2 + Y^2)} \iint_{-\infty}^{\infty} U(x, y) \exp\left(-i\frac{2\pi}{\lambda z}[Xx + Yy]\right) dx dy, \quad (2.26)$$

The field dependent phase in the prefactor of equation 2.26 can be removed when the fields $U(X, Y)$ and $U(x, y)$ are considered in the back and forward focal planes of a lens. In this 2-f focal geometry, as described in more detail in section 4.4.1, equation 2.26 describes, to within a multiplicative constant, an exact Fourier transform relationship, again with transform variables $k_{x,y} = \frac{(x,y)}{\lambda z}$. When considered on an $N \times M$ discrete grid, this relationship can be written as the discrete Fourier transform

$$\text{DFT}[f(n, m)] = F(p, q) = \sum_{m=-\frac{M}{2}}^{\frac{M}{2}-1} \sum_{n=-\frac{N}{2}}^{\frac{N}{2}-1} f(n, m) \exp\left(-2\pi i \left[\frac{pm}{M} + \frac{qn}{N}\right]\right) \quad (2.27)$$

where the indices m and p vary according to

$$-\frac{M}{2} \leq \begin{Bmatrix} m \\ p \end{Bmatrix} \leq \frac{M}{2} - 1, \quad (2.28)$$

with an equivalent range for n and q limited by N . In this discrete case, the sampling periods dx and dy in the $\mathcal{O}$ domain are related [20] to the sampling periods dX and dY

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in the $\mathcal{D}$ domain by

$$dX = \frac{\lambda z}{N dx} = \frac{\lambda z}{L_x} \quad (2.29a)$$

$$dY = \frac{\lambda z}{M dy} = \frac{\lambda z}{L_y}, \quad (2.29b)$$

where $L_{x,y}$ are the physical lengths of the computational grid in x and y respectively.

The distinction between the Fresnel and Fraunhofer regimes of diffraction can be parameterised by the Fresnel number $\mathcal{F}$ [13]

$$\mathcal{F} > \frac{a^2}{\lambda z}, \quad (2.30)$$

which describes whether the diffracted phase curvature a distance z downstream of an aperture of characteristic size a is negligible or whether it must be accounted for. According to a convention, $\mathcal{F} \leq 0.1$ is defined as the Fraunhofer regime and $\mathcal{F} \geq 1$ as the Fresnel regime. For $0.1 < \mathcal{F} < 1$, one can use either a combination of the aforementioned methods, or an alternate technique such as the [angular spectrum method \(ASM\)](#) [20].

2.2.4 Angular Spectrum Method

The [ASM](#) considers the process of scalar diffraction in the frequency domain of the complex fields in the $\mathcal{O}$ and $\mathcal{D}$ planes. The approximations inherent in Fresnel (equation 2.18) and Fraunhofer diffraction (equation 2.25) are not made in [ASM](#), enabling results with a wider range of validity to be found. The compromise in using [ASM](#) is that the $\mathcal{O}$ and $\mathcal{D}$ domain resolution sizes are the same. Ergo, unless the two fields are of approximately the same transverse size, vast array sizes are required to ensure no aliasing of the signal in one or both domains.

The complex field in the $\mathcal{O}$ domain where $z = 0$ can be expressed as its Fourier

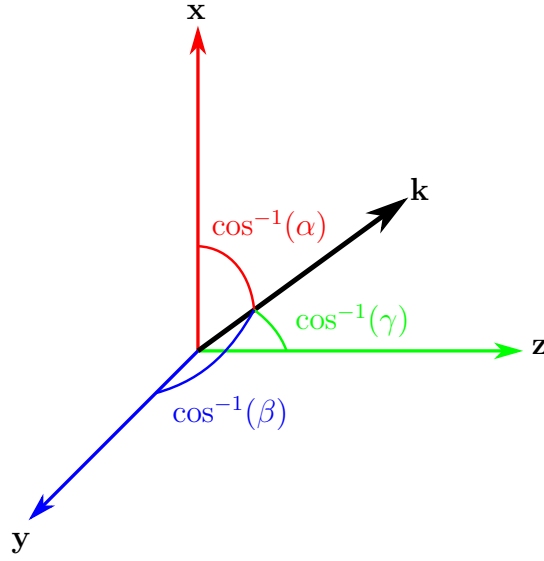


Figure 2.3: The wavevector $\mathbf{k}$ propagating in Cartesian coordinates with direction cosines α , γ and β .

transform

$$\text{FT}[U(x, y, 0)] = \hat{U}(f_x, f_y, 0) = \iint_{-\infty}^{+\infty} U(x, y, 0) \exp(-i2\pi[f_x x + f_y y]) dx dy. \quad (2.31)$$

The complex exponential in the integral of equation 2.31 exhibits a form reminiscent of a plane wave:

$$U_{\text{plane}}(x, y, z) = e^{i\mathbf{k} \cdot \mathbf{r}} = \exp\left[i\frac{2\pi}{\lambda}(\alpha x + \beta y + \gamma z)\right], \quad (2.32)$$

where we have taken $\mathbf{k} = k_0(\alpha\mathbf{x} + \beta\mathbf{y} + \gamma\mathbf{z})$ and $\mathbf{r} = x\mathbf{x} + y\mathbf{y} + z\mathbf{z}$. The angles α , β and γ , shown in figure 2.3, represent the direction cosines of the wavevector $\mathbf{k}$.

By comparing equations 2.32 and 2.31 it can be seen that the complex exponential in the integral of equation 2.31 describes a plane wave propagating in a direction specified

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by

$$\alpha = \lambda f_x \quad (2.33a)$$

$$\beta = \lambda f_y \quad (2.33b)$$

$$\gamma = \sqrt{1 - (\lambda f_x)^2 - (\lambda f_y)^2} \quad (2.33c)$$

We subsequently define

$$\hat{U}(f_x, f_y, 0) = \hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, 0\right) = \iint_{-\infty}^{+\infty} U(x, y, 0) \exp\left(-i2\pi \left[\frac{\alpha}{\lambda}x + \frac{\beta}{\lambda}y\right]\right) dx dy \quad (2.34)$$

as the angular spectrum of $U(x, y, 0)$.

At a distance z further downstream, the angular spectrum can be written as

$$\hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, z\right) = \iint_{-\infty}^{+\infty} U(x, y, z) \exp\left(-i2\pi \left[\frac{\alpha}{\lambda}x + \frac{\beta}{\lambda}y\right]\right) dx dy, \quad (2.35)$$

where its conjugate form

$$U(x, y, z) = \iint_{-\infty}^{+\infty} \hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, z\right) \exp\left(i2\pi \left[\frac{\alpha}{\lambda}x + \frac{\beta}{\lambda}y\right]\right) d\frac{\alpha}{\lambda} d\frac{\beta}{\lambda} \quad (2.36)$$

must satisfy the Helmholtz equation

$$\nabla^2 U(x, y, z) + k_0^2 U(x, y, z) = 0. \quad (2.37)$$

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Substituting equation 2.36 into 2.37 gives:

$$\nabla^2 U(x, y, z) + k_0^2 U(x, y, z) = 0 \quad (2.38a)$$

$$\left(\frac{d^2}{dx^2} + \frac{d^2}{dy^2} + \frac{d^2}{dz^2} \right) \hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, z\right) + k_0^2 \hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, z\right) = 0 \quad (2.38b)$$

$$\left[-\left(\frac{2\pi}{\lambda}\right)^2 \alpha^2 - \left(\frac{2\pi}{\lambda}\right)^2 \beta^2 + \frac{d^2}{dz^2} \right] \hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, z\right) + \left(\frac{2\pi}{\lambda}\right)^2 \hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, z\right) = 0 \quad (2.38c)$$

$$\frac{d^2}{dz^2} \hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, z\right) + \left(\frac{2\pi}{\lambda}\right)^2 (1 - \alpha^2 - \beta^2) \hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, z\right) = 0. \quad (2.38d)$$

A solution to the second order differential equation 2.38d is of the form

$$\hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, z\right) = \hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, 0\right) \exp\left(i \frac{2\pi z}{\lambda} \sqrt{1 - \alpha^2 - \beta^2}\right). \quad (2.39)$$

Taking an inverse Fourier transform of this angular spectrum enables us to find the counterpart complex field at a distance z from the $\mathcal{O}$ domain as

$$U(x, y, z) = \iint_{-\infty}^{+\infty} \hat{U}\left(\frac{\alpha}{\lambda}, \frac{\beta}{\lambda}, 0\right) \exp\left(i \frac{2\pi z}{\lambda} \sqrt{1 - \alpha^2 - \beta^2}\right) \exp\left(i 2\pi \left[\frac{\alpha}{\lambda} x + \frac{\beta}{\lambda} y\right]\right) d\frac{\alpha}{\lambda} d\frac{\beta}{\lambda}. \quad (2.40)$$

Equation 2.40 reveals that we can express the complex field at a distance z downstream of the $\mathcal{O}$ domain as a function of the angular spectrum within the $\mathcal{O}$ domain, and a spatial frequency transfer function of the form

$$H(f_x, f_y, z) = \begin{cases} \exp\left[i 2\pi \frac{z}{\lambda} \sqrt{1 - (\lambda f_x)^2 - (\lambda f_y)^2}\right] & \sqrt{f_x^2 + f_y^2} < \frac{1}{\lambda} \\ 0 & \text{otherwise.} \end{cases} \quad (2.41)$$

Here, we have used relations 2.33 to move between the direction cosines and spatial frequencies. Finally, a compact expression describing ASM propagation incorporating equations 2.41, 2.40 and 2.36 can be written as

$$U(x, y, z) = \text{FT}^{-1} \{ \text{FT} [U(x, y, 0)] H(f_x, f_y, z) \}. \quad (2.42)$$

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The form of the equation 2.42 is responsible for its alternate identification as either the ‘convolution method’ or ‘double Fourier transform method’.

A common problem encountered when using ASM is that of aliasing caused by under-sampling. From equation 2.42, it can be seen that the convolution of the two functions $U(x, y, 0)$ and $H(f_x, f_y, z)$ is required in order to calculate $U(x, y, z)$. If the maximum spatial frequencies of $U(x, y, 0)$ and $H(f_x, f_y, z)$ are limited to k_u and k_H respectively, the spatial frequency limit of their convolution is limited to $k_u + k_H$ [20]. This finite limit of constituent spatial frequencies is known as a band limit. If a sampling rate appropriate for either $U(x, y, 0)$ or $H(f_x, f_y, z)$ is applied to their convolution, aliasing may still occur. Both constituent functions must be sampled at the appropriate rate for the higher band limit of their convolution: $2(k_u + k_H)$.

2.2.5 The Phase Problem

The complex field $f(x)$ leaving the rear surface of the object or aperture in the $\mathcal{O}$ domain is known as the exit surface wave (ESW). It describes the rear surface of the object or aperture to within three trivial operations; translation, rotation and complex conjugation [21]. However, the charge coupled device (CCD) detector in the $\mathcal{D}$ domain is sensitive only to the magnitude squared of the incident complex field, $|F(X)|^2$, which lacks any phase information. Assuming the $\mathcal{D}$ domain resides in the far field of the $\mathcal{O}$ domain ($\mathcal{F} \leq 0.1$), then the complex field detected by the CCD can be represented by the discrete Fourier transform of the ESW. Without loss of generality, on a one dimensional detector of grid length N this is

$$|F(X)| = \left| \sum_{x=0}^{N-1} f(x) \exp \left(-2\pi i \frac{Xx}{N} \right) \right|. \quad (2.43)$$

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Consequently, to recover $f(x)$ from $|F(X)|$, $2N$ unknowns (amplitude and phase) in the $\mathcal{O}$ domain must be solved for with only N knowns (amplitudes) in the $\mathcal{D}$ domain. This ill posed inversion problem is known as the ‘phase problem’ [22]. Some progress can be made by making certain assumptions about the nature of the diffracting object. If $f(x)$ is real, or equivalently, has no phase (absorptive) component, then the Fourier transform will exhibit central symmetry, reducing the number of unknowns to N and the number of equations described by equation 2.43 to $N/2$. Evidently, further constraints are necessary in order to recover $f(x)$.

To solve the phase problem and recover $f(x)$, it is required that either the number of known quantities is increased or the number of unknown quantities is decreased. The latter can be achieved by using a priori information about the object $f(x)$, for example, a region within the object where the transmission is already accurately known. However, access to this information is not always possible. Instead, to increase the number of equations describing the detected diffraction, the sampling density can be increased such that the diffraction pattern is ‘oversampled’. Equations 2.29a and 2.29b relating the $\mathcal{O}$ and $\mathcal{D}$ domain pixel sizes, dx and dX respectively, reveal that if dX is reduced, then the $\mathcal{O}$ domain physical size L_x increases. Given that the object $f(x)$ has a fixed physical size, this approach generates a new region of $\mathcal{O}$ domain space surrounding the object. This region, denoted by ‘S’, is known as the support and necessarily contributes zero signal to the detected diffraction, ergo the number of known valued pixels in the $\mathcal{O}$ domain has been increased.

The degree of oversampling is denoted by σ and must be > 2 for a one dimensional object and $> \sqrt{2}$ in each dimension for a two dimensional object in order for the detected diffraction pattern to contain sufficient information to recover the ESF. Experimentally, σ specifies the oversampling relative to the finest interference fringe, or equivalently, the highest spatial frequency, within the detected diffraction pattern. This highest frequency

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is known as the Nyquist frequency ν_N , which for an object of characteristic diameter a is given by

$$\nu_N = \frac{a}{\lambda z}. \quad (2.44)$$

Following the argument above, in order to achieve the minimum oversampling criterion ($\sigma = 2$) the diffraction pattern in the $\mathcal{D}$ domain must be sampled with a period of $2\nu_N$ [23, 24]. From equation 2.29a, this implies that the longitudinal separation, z , of the $\mathcal{O}$ and $\mathcal{D}$ domains must satisfy

$$z = \frac{2a dX}{\lambda} \quad (2.45)$$

where dX is the pixel size in the $\mathcal{D}$ domain and the factor of 2 accounts for two pixels per finest fringe spacing. Exchanging the factor of 2 for σ , a more general expression the degree of oversampling is given by

$$\sigma = \frac{\lambda z}{a dX}. \quad (2.46)$$

2.3 Lensless Imaging

Two methods for processing the oversampled data gathered in the manner described in section 2.2.5 will be explored in this thesis. These methods are both modalities of ‘lensless imaging’, known as **CDI** and **FTH** [25, 26]. However, the acronym **CDI** is commonly used as an umbrella term referring to a multitude of variations upon the specific implementation outlined in this thesis. A comprehensive review of these variants can be found in reference [27]. For the remainder of this thesis, the term **CDI** will be used to represent the specific variant of lensless imaging schematically depicted in figure 2.4.

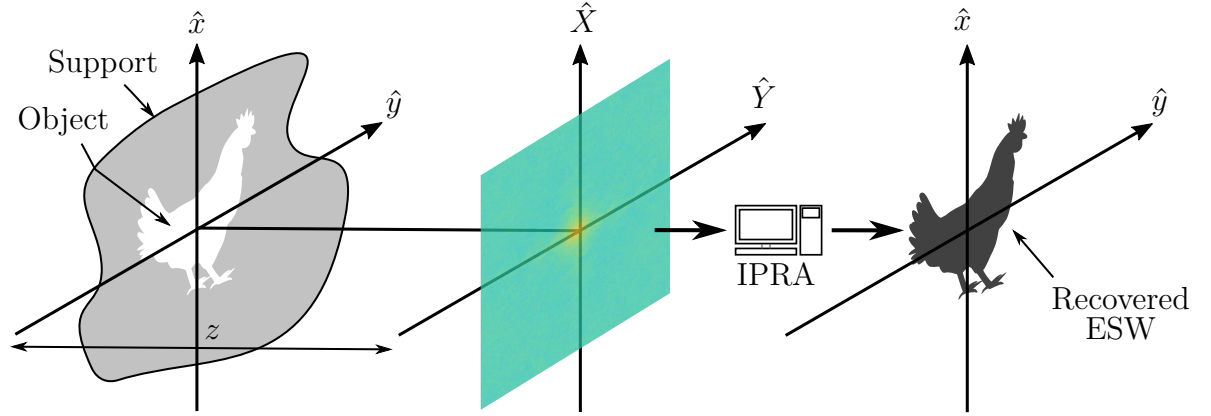


Figure 2.4: Schematic diagram showing the experimental setup of a CDI experiment.

In **CDI**, the lost phases of the **ESW** are recovered by an **IPRA** that exploits a priori knowledge about the spatial extent of the object and support to iteratively calculate estimations for the **ESW** phases. Ideally the **IPRA** is convergent and is said to have converged when a user defined error metric drops below some threshold. With **FTH**, the **ESW** is combined with a reference wave, both in the $\mathcal{O}$ domain. Upon propagation, the **ESW** and reference wave interfere and produce a hologram that is detected in the $\mathcal{D}$ domain. The lost **ESW** phases are encoded into the detected interference fringes. An inverse Fourier transform of the hologram can be analytically expressed as the convolution of the **ESW** with the reference aperture, typically a pinhole, that provided the reference wave.

2.3.1 Coherent Diffractive Imaging

A typical **CDI** experiment is shown schematically in figure 2.4. It comprises an object and support in the $(x, y) \in \mathcal{O}$ domain, a diffraction pattern in the $(X, Y) \in \mathcal{D}$ domain and the **ESW** reconstructed in the $(x, y) \in \mathcal{O}$ domain by an **IPRA**. A number of factors must be considered in order for this implementation of **CDI** to be successful: the illuminating beam must be at least partially coherent and have a known bandwidth: the object or illumination must be spatially constrained to exist within a region of zero transmission,

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and an [IPRA](#) must converge giving a self consistent solution to the phase problem. Each of these considerations will be described in more detail below.

Coherence of the Illumination

A high degree of spatial coherence in the beam illuminating the object enables high visibility diffraction fringes to be observed in the [D](#) domain. Fringe visibility, ν , is commonly parameterised by

$$\nu = \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}} \quad (2.47)$$

where $I_{\max}$ and $I_{\min}$ are the maximum and minimum intensities observed within the fringe pattern. A direct relation [\[28\]](#) between ν and the [complex coherence function \(CCF\)](#), μ of the illuminating beam measured between two points x_1 and x_2 located on a plane transverse to the beam propagation direction can be written as

$$\nu = \frac{2\sqrt{I^{(1)}I^{(2)}}}{I^{(1)} + I^{(2)}} |\mu(x_1, x_2)|. \quad (2.48)$$

Here, $I^{(j)}$ is the intensity reaching the [D](#) domain from point x_j alone. The e^{-2} width of the [CCF](#) is taken to be the transverse coherence length of the beam[\[29\]](#) that can also be defined as [\[30\]](#)

$$L_c^\perp = \frac{\lambda z_s}{2\pi D}. \quad (2.49)$$

Here, D is the size of the source of the illumination and z_s is the distance between the source and the object. Experimentally, for an object of size a , sufficient coherence is achieved for a [CDI](#) experiment when $L_c^\perp \geq 2a$ [\[31\]](#). This is equivalent to the width of the autocorrelation of the object.

With modifications to the [IPRA](#), [CDI](#) can be driven by totally incoherent [\[32\]](#) or partially coherent radiation [\[33–36\]](#). With adjustments to the experimental geometry, [FTH](#) can also be performed with incoherent illumination [\[37, 38\]](#). Nevertheless, throughout this

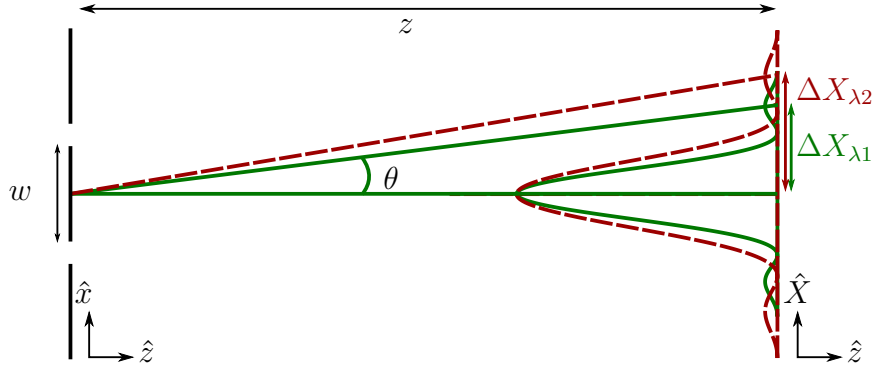


Figure 2.5: Schematic diagram showing the diffraction from a pair of pinholes spaced by w into the far field at a distance z . The fringe spacings ΔX_λ of the first maxima are shown for two different wavelengths.

thesis, unless otherwise specified, coherent illumination is assumed.

Bandwith of the Illumination

In the simplest implementation of [CDI](#), the illumination is monochromatic. This ensures that all fringes in the detected diffraction pattern can be attributed to a known wavelength. For example, in figure [2.5](#), it can be shown that within the small angle approximation, the spacing of the m^{th} maxima from the optical axis in the far field of a double pinhole spaced by w is

$$\Delta X = \frac{m \lambda z}{w}. \quad (2.50)$$

The linear proportionality between ΔX and λ implies that a broadband source produces diffraction that can be considered as the addition of the intensities of many diffraction patterns with slightly different fringe spacings. For certain intrinsically broadband sources, the spectral filtering of a narrow bandwidth results in an inefficient use of available photons. This has motivated progress in the field of polychromatic lensless imaging [\[39, 40\]](#) and the development of complex supporting algorithms that can cater for the use of a much larger illuminating bandwidth [\[41\]](#). In the remainder of this thesis, unless otherwise specified, monochromatic illumination will be assumed.

Spatially Isolated Object

For [CDI](#), the object must be spatially isolated in order to provide a physical analogue to the support region. This can be achieved by embedding the object within an opaque screen or coating such that the [ESW](#) has a fixed spatial extent. Alternatively, this spatial restriction can instead be imposed upon the illuminating beam by placing an aperture of known dimensions in the beam upstream of the $\mathcal{O}$ domain, creating a finite diverging beam that produces illumination of an accurately known spatial extent. This is the premise of keyhole [CDI](#) [\[42\]](#).

The [field of view \(FoV\)](#) available in a [CDI](#) experiment is equal to the illuminating beam size in the $\mathcal{O}$ domain. Implicitly, when the illumination of the object is spatially restricted, so is the [FoV](#). A modality of lensless imaging known as ptychography [\[43–45\]](#), which can be considered as an extension of keyhole [CDI](#), removes the isolated object constraint. Experimentally, diffraction patterns are gathered as the object is scanned over a transverse grid downstream of the pinhole. At each grid point k , there is a known overlap between the illuminated region of the object at points k and $k-1$. Each measured diffraction pattern therefore contains a contribution that is shared with the previous pattern, representing a redundancy of information that can assist the convergence of an appropriate [IPRA](#). In this case the [FoV](#) is limited only by the computational resources available in the experiment.

By spatially isolating the object, the spatial frequency content of the diffraction pattern is bandlimited. Equation [2.44](#) reveals that a limit must be set on a in order to bandlimit the diffraction to a spatial frequency of $\nu_N \text{ m}^{-1}$. This limit is created by masking the object being imaged with a screen, coating or aperture that is opaque everywhere except for a region of diameter a centred on the [region of interest \(RoI\)](#) within the object.

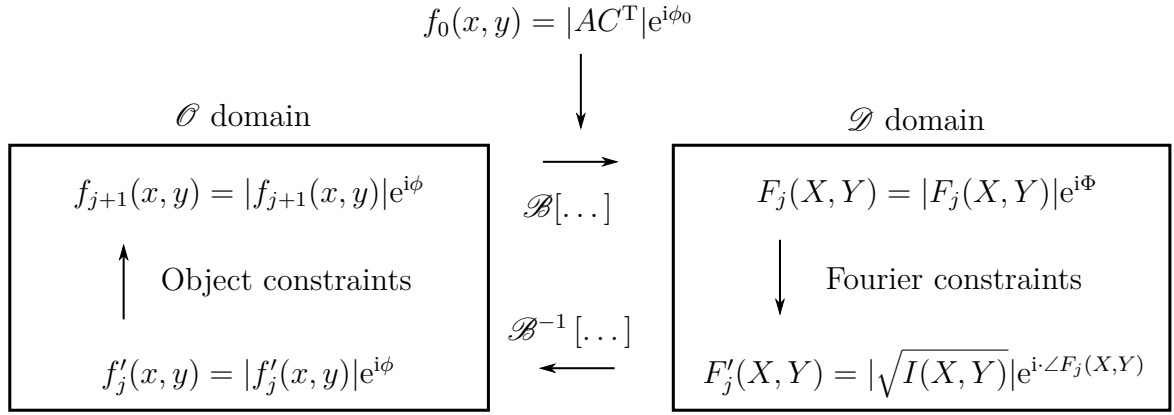


Figure 2.6: Flow chart of the GS phase retrieval algorithm. The two rectangular boxes represent the $\mathcal{O}$ and $\mathcal{D}$ domains, which are related by an invertible propagator, $\mathcal{B}$. Within each domain constraints are applied that drive the algorithm to converge upon a self consistent solution.

Iterative Phase Retrieval Algorithm

The [IPRA](#) used to recover the lost [ESW](#) phase can comprise one or more algorithms that each implement different constraints in the $\mathcal{O}$ and $\mathcal{D}$ domains [\[46\]](#). One of the earliest and most conceptually simple of these algorithms was proposed in 1972 by Gerchberg and Saxton [\[47\]](#). Although modern [CDI](#) algorithms have expanded greatly upon their founding work, the [Gerchberg-Saxton \(GS\)](#) algorithm illustrated in figure 2.6 can often be found as the underlying framework.

In figure 2.6, the [GS](#) algorithm is separated into the $\mathcal{O}$ and $\mathcal{D}$ domains which are connected by an invertible propagator $\mathcal{B}$ that transforms the complex field from one domain to the other. $\mathcal{B}$ is used to represent a propagator that is appropriate for the diffractive regime that relates the $\mathcal{O}$ and $\mathcal{D}$ domains (Fresnel or Fraunhofer diffraction). It can be further generalised to account for situations where there is a loss of energy between the two domains due to either diffraction or obstruction by a beam block for example. In this case, an additional loop is used to calculate the form of the non-unitary propagator necessary for the algorithm to converge [\[48\]](#). In this thesis $\mathcal{B}$ is equivalent

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to a Fourier transform unless otherwise specified.

The first step of the [GS](#) algorithm shown at the top of figure [2.6](#) is an initial guess $f_0(x, y)$ of the [ESW](#) in the $\mathcal{O}$ domain. This guess comprises an amplitude distribution that contains a region of unity within which the object resides and is zero elsewhere. This region, known as the ‘mask’, can be found by exploiting a priori information about the object such as a low resolution image. However, in situations where such additional information is not available, the Wiener-Khinchin theorem [\[49\]](#) may be used to relate the detected data to the autocorrelation of the object itself thereby providing information about the spatial extent of the object: The theorem states that the Fourier transform of the modulus squared of a function is equal to the autocorrelation of the function itself. The autocorrelation is at least twice as large as the object [\[31\]](#), such that when appropriately thresholded, the mask containing the full spatial extent of the object is found. The correlation operation is described [\[50\]](#) analytically by equation [2.51a](#).

$$\text{Correlation: } f(x) \star g(x) = \int f(\rho) g^*(x + \rho) d\rho \quad (2.51a)$$

$$\text{Convolution: } f(x) \otimes g(x) = \int f(\rho) g(x - \rho) d\rho. \quad (2.51b)$$

Equation [2.51b](#) describes the convolution operation which can be considered a conjugated spatially inverted correlation that is functionally equivalent for real symmetric signals. In this thesis, the mask(s) were calculated using the Wiener-Khinchin theorem giving the autocorrelation denoted $AC(x, y)$ according to equation [2.52a](#):

$$AC(x, y) = \text{FT} [I(X, Y)] \quad (2.52a)$$

$$AC^T(x, y) = \begin{cases} 1 & \forall(x, y) > T \\ 0 & \forall(x, y) < T \end{cases} \quad (2.52b)$$

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Here, T is a threshold applied to the autocorrelation, such that when exceeded, the value of the mask at the corresponding location is set equal to 1, and zero otherwise. The specific value of T is empirically determined based on the **S:N** achieved in the data as well as the nature of the object itself: The sharpness of the edges in the autocorrelation are proportional to the sharpness of the edges in the object. The phase distribution ϕ_0 in the initial guess $f_0(x, y)$ can be taken as constant, or randomised between empirically determined limits.

The initial guess $f_0(x, y)$ is Fourier transformed into the $\mathcal{D}$ domain giving the complex field $F(X, Y)$. At this point, the phase content of the field contains no information about the object as it is the transform of the initial guess ϕ_0 . Within the $\mathcal{D}$ domain, the Fourier constraints are applied that introduce information from the data into the algorithm. The most common constrain consists of replacing the magnitude of $F(X, Y)$ with the square root of the detected intensities $I(X, Y)$ creating $F'(X, Y) = |\sqrt{I(X, Y)}|e^{i\Phi}$. The phase is left unchanged representing the continuity of information around the loop.

Once transformed back into the $\mathcal{O}$ domain, a priori information about the object is introduced based on the support, S , taken as the (x, y) values where $AC^T = 1$. This is facilitated by the **error reduction (ER)** and **hybrid input-output (HIO)** approaches developed by Fienup [51], which are summarised in equations 2.53 and 2.54 respectively.

$$f_{j+1}(x, y) = \begin{cases} f'_j(x, y) & \forall (x, y) \in S \\ 0 & \forall (x, y) \notin S \end{cases} \quad (2.53)$$

$$f_{j+1}(x, y) = \begin{cases} f_j(x, y) & \forall (x, y) \in S \\ f_j(x, y) - \beta f'_j(x, y) & \forall (x, y) \notin S. \end{cases} \quad (2.54)$$

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Here β represents an empirically determined regularisation parameter that is used to optimise the convergence of the algorithm [2]. The support region S can be adjusted within each iteration through a technique called shrinkwrapping [52]. This recalculates the size of the support by convolving it with a Gaussian function of decreasing size, decreasing the support area until it very closely matches the true object size. Additional constraints can be applied to leverage more a priori information about the object, as described in references [2, 53, 54].

Having applied equations 2.53 and or 2.54 as the $\mathcal{O}$ domain constraints, the current iterant is again transformed into the $\mathcal{D}$ domain where the Fourier constraints are reapplied. In each subsequent iteration, the convergence to a self consistent set of phases is parameterised by an error metric

$$E_j = \sqrt{\frac{\sum_{X,Y} |F_j(X,Y) - F'_j(X,Y)|^2}{N^2}} \quad (2.55)$$

that quantifies the difference in the j^{th} iteration between the current solution, $F'_j(X,Y)$ and that of the prior iteration $F_j(X,Y)$ [2]. When the normalised difference between these two possible solutions drops below a user defined value, the algorithm is said to have converged. The phase $\Phi = \angle F'_j(X,Y)$ is then taken as an estimate of the solution to the phase problem.

Typically this error should decrease monotonically and tend asymptotically toward the correct solution. However, there are cases such as HIO where no mathematical proof of convergence has yet been developed [55]. In spite of this, it is still widely employed in tandem with ER due to its ability to escape local minima. Stagnation can occur if the $\mathcal{O}$ and or $\mathcal{D}$ domains are non-convex, as shown in figure 2.7. In this case, traditional convex minimisation approaches may lead to convergence upon local minima, as illustrated

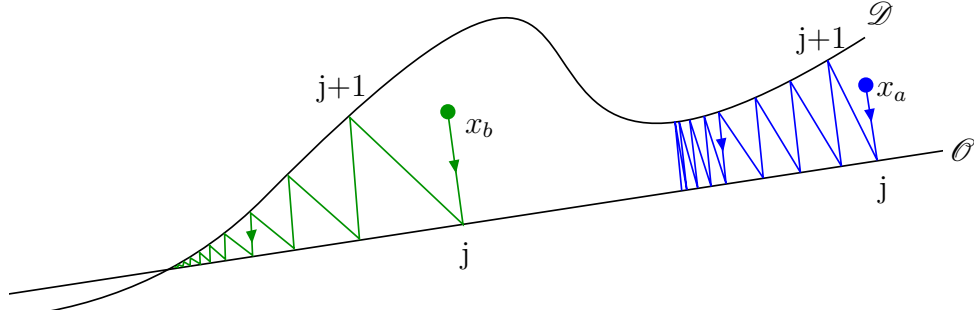


Figure 2.7: Sketch of the $\mathcal{O}$ and $\mathcal{D}$ domains represented as two sets (solid black lines). The starting points x_a and x_b represent initial guesses in the 0th iteration of an IPRA. With each application of the propagator $\mathcal{B}$, the current solution is projected from one set onto the other. The green trace represents successful convergence to a global minima, whereas the blue trace shows stagnation in a local minimum.

by the blue trace in figure 2.7. To build confidence that the output of an IPRA is the global solution, the IPRA is run multiple times with randomised initial seeds to confirm that the output consistently converges upon the same solution.

Having verified and extracted the phase provided by the IPRA, an estimate to the solution of the phase problem has been found. However, this recovered solution is subject to certain ‘trivial characteristics’ identified by Bates in reference [21]: The recovered phase cannot be distinguished from its complex conjugate or a translated and or rotated version of itself. This results in an undesirable situation where twin images are recovered simultaneously, superimposed upon each-other. This is because solution $f(x, y)$ and its conjugate mirror $f^*(-x, -y)$ have the same Fourier modulus. In reference [56] Fienup suggests a method for suppressing an emerging twin image by using a support constraint with no inversional symmetry for some number of iterations until one of the two images becomes dominant.

The $\mathcal{O}$ and $\mathcal{D}$ domain constraints can be extended to incorporate more information about the object if it is available which enhances the probability of convergence of the IPRA. This enables CDI to be used as a highly flexible and effective tool in imaging a

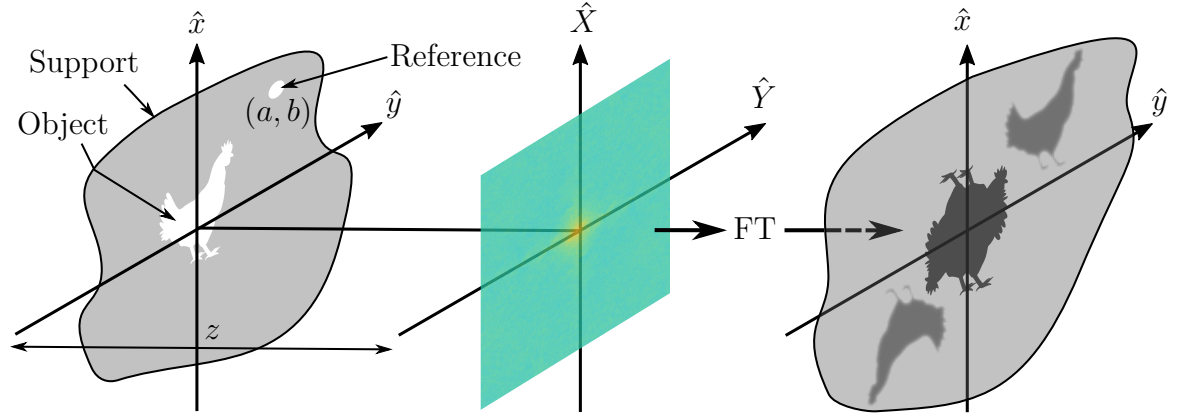


Figure 2.8: Schematic diagram showing the experimental setup in FTH.

wide variety of samples ranging from inorganic [57] to organic [58] in a non destructive fashion.

2.3.2 Fourier Transform Holography

Holography was developed by Dennis Gabor in 1948, for which he won the 1971 Nobel prize [59, 60]. The flexibility in how the reference wave is introduced and combined with the object wave has led to a wide range of modalities including FTH [61–64]. Holography is predicated upon the presence of a well defined reference wave that interferes with the light reflected or transmitted by an object. These waves must be mutually coherent to ensure fringes are formed [65].

Theory

With respect to their experimental implementation, FTH is very similar to CDI, but with the addition of a reference feature, shown schematically in figure 2.8 at a position (a, b) relative to the object and optical axis. The reference feature exists within the same plane as the object in the $\mathcal{O}$ domain, and typically manifests as a vanishingly small pinhole through the opaque coating or mask that spatially isolates the object. A pinhole is often used as it provides a well defined point source that emits a well defined

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spherical wave. Mathematically, the pinhole acts as a function that samples the object such that the finest resolution will be achieved when the pinhole most closely resembles a delta function, $\delta(x, y)$. The field from the object, $u_o(x, y)$, and the reference feature, $\delta(x - a, y - b)$ interfere upon propagation to the $\mathcal{D}$ domain. The measured data, $I(X, Y)$, comprises a hologram that encodes the unknown object phases into the interference fringes.

The complete complex field in the $\mathcal{O}$ domain is denoted by

$$u(x, y) = u_o(x, y) + \delta(x - a, y - b), \quad (2.56)$$

where $u_o(x, y)$ and $\delta(x - a, y - b)$ represent the fields from the object and the point like reference respectively. By invoking the Wiener-Khinchin theorem, the Fourier transform of the detected hologram $I(X, Y)$ can be taken to reveal

$$\text{FT}\{I(X, Y)\} = \text{FT}\{|U(X, Y)|^2\} \quad (2.57a)$$

$$= u(x, y) \star u(x, y) \quad (2.57b)$$

$$= u(x, y) \otimes u^*(x, y) \quad (2.57c)$$

$$= [\delta(x - a, y - b) \otimes \delta^*(x - a, y - b)] + [u_o(x, y) \otimes u_o^*(x, y)] \quad (2.57d)$$

$$+ [u_o(x, y) \otimes \delta^*(x - a, y - b)] + [u_o(x, y)^* \otimes \delta(x - a, y - b)].$$

In order to find equation 2.57c from 2.57b, we have taken the correlation ($\star$) operation to be equivalent to convolution ($\otimes$) when applied to a complex conjugate [50] as described in section 2.3.1. Equation 2.57d is found by directly substituting equation 2.56 into 2.57c and observing the distributive property of convolution.

Equation 2.57d comprises four terms. The first two terms correspond to the auto-correlations of the object and reference with themselves and lie on the optical axis. The

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last two terms represent the cross-correlation between the object and the reference along with a complex conjugate. In the remainder of this thesis, these cross-correlations will be referred to as the ‘image’ of the object and its complex conjugate for brevity.

In order to avoid an overlap between the on-axis autocorrelations and the off-axis images, the reference hole must be placed a minimum of 1.5 times the object diameter away from the centre of the object itself. This is known as the holographic separation condition. Multiple images can be recovered if multiple reference features are incorporated in the object design [66]. As each of the N references produces a complex conjugate pair of images, this approach yields $2N$ images which can be averaged together to improve the S:N. In this case, an analogous separation requirement exists between adjacent reference holes.

The resolution of an image recovered in FTH is limited to approximately 70% of the corresponding reference aperture diameter [26]. A compromise must be made therefore between a small pinhole that affords the finest resolution and a pinhole large enough to permit detectable levels of flux. For pinhole references the flux transmitted by the reference feature should remain comparable to that transmitted through the object [67]. Efforts to accommodate both of these conflicting requirements simultaneously have motivated the development of extended arbitrary apertures or edges to generate reference waves [62]. A common implementation of this is ‘Holography with Extended Reference by Autocorrelation Linear Differential Operation’ (HERALDO) [68]. The resolution in HERALDO is a function of the sharpness of the reference feature edge. This exchanges the engineering challenge of creating a vanishingly small pinhole with the easier task of creating a larger aperture with a very sharp edge.

FTH can act as an unambiguous corroboration of CDI with only a minor change in experimental geometry. A combination of both techniques can be achieved by providing the FTH image to a CDI IPRA as an initial guess of the support. This enables the

IPRA to refine the resolution down to the limit set by both the illuminating wavelength and NA of the detector, not the pinhole size [69].

2.4 Image Quality

Common metrics of image quality are the intensity contrast [70] and resolution. Both are measured according to a variety of definitions, some of which are explored here.

2.4.1 Signal to Noise Ratio

The intensity contrast between the image itself and the background is intrinsically linked [49] to the S:N of the image defined as [71]

$$\text{S:N} = \frac{\mu_{\text{sig}}}{\sigma_{\text{bg}}}. \quad (2.58)$$

Here μ_{sig} and σ_{bg} are the mean of the image signal and standard deviation of the background respectively. The contrast in an image recovered through lensless imaging is proportional to the visibility of the interference fringes. In FTH, this is maximised when the amplitudes of the object and reference waves are comparable [26]. In CDI the detected fringe S:N is a function of the proportion of flux transmitted or reflected by the object. It can be enhanced by averaging together multiple exposures of different lengths into one high dynamic range (HDR) dataset.

2.4.2 90-10 Resolution

A commonly employed method of quantifying image resolution is the ‘90-10’ measurement [72]. This describes the spatial distance perpendicular to an edge within an image that separates points of 10% and 90% of the peak value along that line. However, this method characterises the resolution of an image based on an implicitly large feature,

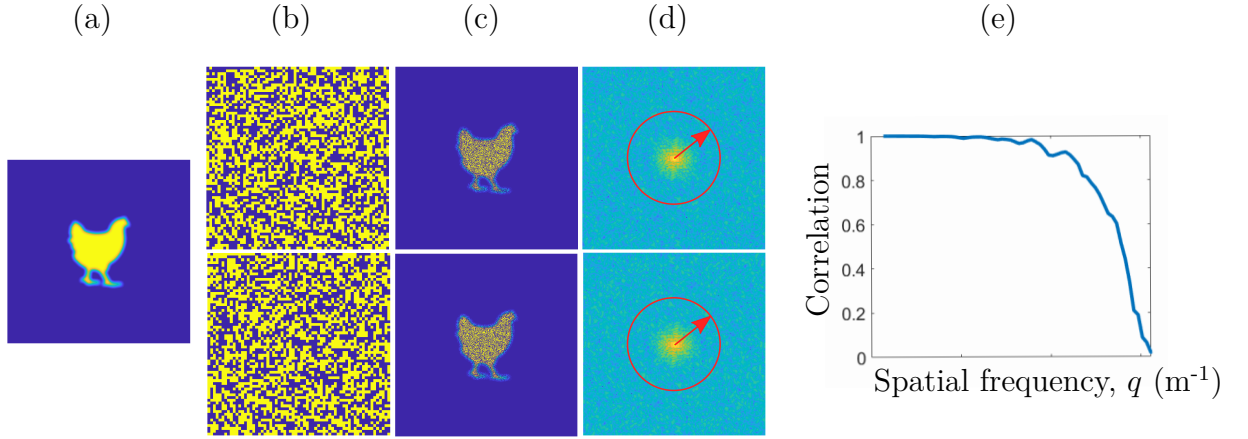


Figure 2.9: The steps involved in calculating a FRC curve. Schematic adapted from reference [74]. Detail of each step is given in the text.

not the smallest resolved component directly. It is also influenced by the illuminating phase, coherence and S:N in an image [73], which has motivated the development of approaches to quantifying resolution in terms of resolvable spatial frequencies of an imaging system [73].

The ease of implementation of this method makes it a useful tool in quickly estimating the resolution achieved in an image although it is prudent to be aware of the aforementioned drawbacks.

2.4.3 Fourier Ring Correlation Resolution

Fourier ring correlation (FRC) is a method of quantifying image resolution in Fourier space [74]. This involves splitting detected data into two subimages in Fourier space and computing their correlation within multiple annuli of incrementally increasing radius. The steps involved in calculating a FRC curve are shown in figure 2.9.

In figure 2.9, an example image is shown in column (a). A random binary mask

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of 50% occupancy and its compliment, both shown in column (b) are then used to separate the image into two subimages, shown in column (c) that are then Fourier transformed in the $\mathcal{D}$ domain, shown in column (d). The correlation is then computed between data points located in each Fourier transform within an annulus, shown in red in column (d). The correlation between the two sets, A and B , of N values within the annuli defined by

$$\rho(A, B) = \frac{1}{N-1} \sum_{i=1}^N \left(\frac{A_i - \mu_A}{\sigma_A} \right) \left(\frac{B_i - \mu_B}{\sigma_B} \right) \quad (2.59)$$

is recorded as the radius of the annulus is increased. Here, μ denotes the mean and σ denotes the standard deviation. For small radii the correlation will be high signifying the low spatial frequency content of both subimages present near the optical axis. At larger radii, the higher spatial frequency information will eventually drop below the noise threshold, signifying the spatial frequency beyond which there is no significant correlation between the two subimages. The spatial frequency values are calculated according to

$$q = \frac{2 \sin(\frac{\theta}{2})}{\lambda}. \quad (2.60)$$

where θ is the angle subtended at the object by the annulus in the $\mathcal{D}$ domain [75].

The curve of correlation vs spatial frequency shown in column (e) of figure 2.9 fully expresses the resolution of the image in column (a). It is common for a somewhat arbitrary threshold (e.g. 0.5) to be applied to this curve that relates to a certain perceived information content within the data. This figure is then quoted as the image resolution. However, as noted in reference [76], "There is no scientific justification for the use of fixed [FRC] threshold criteria ...". For the remainder of this thesis therefore, a threshold of $T = 0.17$ is taken only to provide a self consistent comparative metric between images recovered by the same imaging beam line. The author therefore does not intend for the threshold and corresponding quoted resolution(s) to be comparable

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to other imaging systems.

Chapter 3

High Harmonic Generation



High harmonic generation driven by tabletop laser systems has emerged as an accessible method for generating highly coherent radiation in the [XUV](#) and soft x-ray spectral regions [\[77\]](#). The high spatial and temporal coherence of [HHG](#) sources coupled with their sub-femtosecond pulse durations has made [HHG](#) an attractive source for a range of applications including the real-time observation of ultrafast dynamics in gases [\[78\]](#) and solids [\[79\]](#), the metrology of quantum-tunnelling [\[80\]](#), element and depth sensitive imaging [\[81, 82\]](#), and lensless imaging [\[83, 84\]](#).

3.1 Motivation

[HHG](#) provides access to short wavelength radiation without the cost, time and spatial constraints associated with larger facilities such as [XFELs](#) [\[85, 86\]](#) or synchrotrons. These advantages come at the cost of a limited conversion efficiency that typically restricts harmonic sources to below μW of average power [\[87, 88\]](#). Flux intensive applications such as imaging motivate the search for methods that either enhance the intrinsic properties of the beam such as brightness [\[89\]](#) or optimally distribute the harmonic radiation upon the target to minimise losses.

Several approaches have been adopted to increase the conversion efficiency, including the use of phase-matching [90], quasi-phase matching [91, 92], high-repetition rate driver lasers [93], driver wavefront manipulation [94, 95] and high pulse energy drivers in a loose focussing geometry [96].

However, as will be explored in chapters 6 and 7, scope remains for the manipulation and enhancement of the harmonic radiation in order to optimally utilise all harmonic photons in a given experiment.

3.2 The Three Step Model

The theoretical treatment of HHG can range from the semi-classical approach of Corkum [97] to the quantum mechanical approach of Lewenstein [98]. The former approach based on the ‘Three Step Model’ is considered sufficient to explain the findings presented in this thesis.

A short overview of the Three Step Model will now be presented, followed by a more detailed treatment of each step in sections 3.2.1 to 3.2.3. Corkum’s three step model describes the constituent processes involved in HHG: First, an electron is ionised from an atom by a driving laser field in the non-perturbative regime. Second, the electron propagates through the continuum accelerated by the electric field of the driving laser field. Third, the electron recombines with the ion and a harmonic photon is emitted with an energy equal to the sum of the atomic binding energy and the kinetic energy of the recombining electron. It will be shown that the possible energies of harmonic photons generated in a gas comprise well defined peaks at odd integer multiples of the driver frequency. A schematic illustration of these three steps is shown in figure 3.1.

In each of the three steps in Corkum’s model, an assumption is made in order to

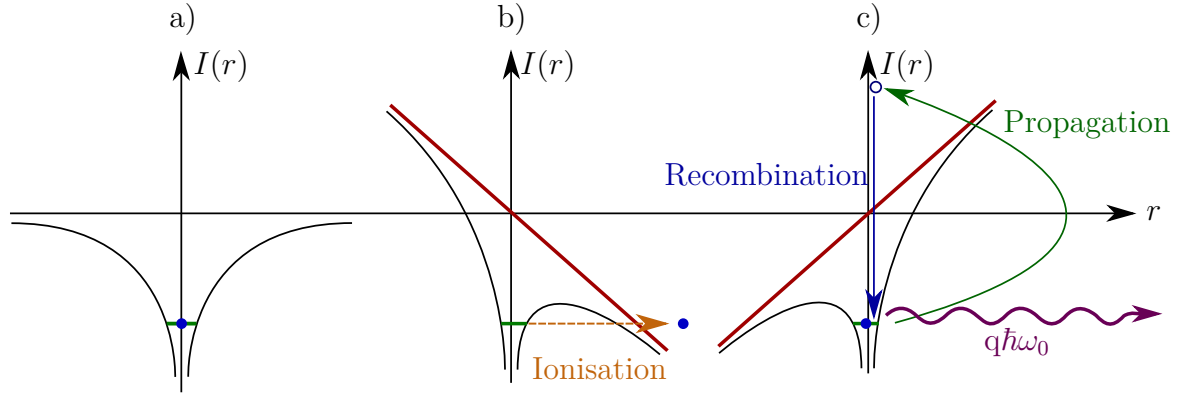


Figure 3.1: A schematic diagram showing the basic steps of the three step model. The axes represent potential I as a function of radial distance r . a) An electron (blue) in a bound state (green) within an atomic potential (black). b) A driving laser field (red) enables tunnel ionisation. c) The ionised electron propagates through the continuum and can recombine with the host ion resulting in the emission of a harmonic photon.

maintain the validity of the semi-classical treatment of HHG [28]. These assumptions are:

1. Upon ionisation, the electron appears in the continuum with zero velocity.
2. Having been accelerated through the continuum by the driver laser electric field, the electron returns to the ion.
3. Upon recombination with the ion, the kinetic energy of the returning electron combined with the binding energy of the atom is released as a harmonic photon.

Their origins lie in the conclusions drawn from a saddle point analysis[99] of the frequency dependent dipole moment [100, 101].

3.2.1 Ionisation

Figure 3.1 a) shows a schematic diagram of a single electron in a bound state within an atomic potential. Figure 3.1 b) shows how in the non-perturbative regime, the presence of a sufficiently intense driver laser field warps the atomic potential and facilitates the tunnel ionisation of the electron into the continuum. This is the first step of the three

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step model of HHG. The particular regime of ionisation involved can be inferred from the Keldysh parameter [102] defined as

$$\gamma = \frac{\omega_0}{q_e E_0} \sqrt{2I_p m_e}, \quad (3.1)$$

where ω_0 and E_0 are the driving laser frequency and electric field magnitude, q_e is the electron charge, I_p is the ionisation potential of the atomic species being considered and m_e is the classical electron mass.

Tunnel ionisation dominates when $\gamma < 1$, whilst multiphoton ionisation dominates when $\gamma > 1$. This distinction can be understood by considering the optimal conditions required for these processes to occur: Tunnel ionisation requires the atomic potential to be reduced. As the driving laser field oscillates sinusoidally, such a reduction is only possible when the magnitude of the driving laser field is near its maximum. This occurs during only two short time windows within the whole cycle. These time windows are larger when the driver field is slowly varying, or when ω_0 is small, implying $\gamma < 1$. Conversely, multiphoton ionisation is predicated upon the simultaneous absorption of many driver photons. This process is optimised when the incident photons are of higher energy such that fewer will be required in order to ionise an electron. This situation occurs when ω_0 is large implying $\gamma > 1$.

A popular method of calculating the theoretical ionisation rate of gases in high intensity laser fields is with the Ammosov-Delone-Krainov (ADK) rate [103], shown below in equation 3.2 with the Coulomb correction required to account for nearby ionic potentials [104]:

$$W_{\text{ADK}} = A_{n^*} \omega_p \left(\frac{4\omega_p}{\omega_t} \right)^{2n^*-1} \exp \left(-\frac{4\omega_p}{\omega_t} \right), \quad (3.2)$$

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Here,

$$\begin{aligned} A_{n^*} &= \frac{2^{2n^*}}{n^* \Gamma(n^* + 1) \Gamma(n^*)} & \omega_p &= \frac{I_p}{\hbar} \\ \omega_t &= \frac{q_e E(t)}{\sqrt{2m_e I_p}} & n^* &= Z \sqrt{\frac{H}{I_p}}. \end{aligned} \quad (3.3)$$

where Z is the atomic number of the atom after ionisation and H is the ionisation potential of hydrogen. It should be noted however that the same expression popularised by ADK in 1986 had been established for hydrogen in 1966 by Perelomov, Popov and Terent'ev [105]. The ADK ionisation rate as described above is valid only in the region where $\gamma \ll 1$ and assumes the driver intensity to be quasi-static, neglecting its time dependence. A more general expression that accounts for these shortcomings is that of the Yudin rate W_{Yudin} [106] which is valid for any value of γ .

Depending on the choice of ionisation equation, W_{ADK} or W_{Yudin} , the degree of ionisation at time t is

$$\eta(t) = 1 - \exp \left[- \int_{-\infty}^t W(t) dt \right] \quad (3.4)$$

3.2.2 Propagation

Figure 3.1 c) shows the propagation of the electron through the continuum and back to the location of the ion in accordance with assumption 2. In keeping with the semi-classical description of HHG, this motion of the electron in the continuum can be described by the classical equations of motion in the electric field of the driving laser. In this case, the force experienced by an electron in a sinusoidally varying external field, linearly polarised in x , is

$$m_e \ddot{x}(t) = -q_e E_0 \cos(\omega_0 t), \quad (3.5)$$

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where t is the time coordinate. The velocity of the electron is found by integrating equation 3.5 between an ionisation time t_0 and time t :

$$\dot{x}(t, t_0) = \int_{t_0}^t \ddot{x}(t) dt = \frac{-q_e E_0}{m_e \omega_0} [\sin(\omega_0 t) - \sin(\omega_0 t_0)]. \quad (3.6)$$

Note that $\dot{x}(t_0, t_0) = 0$, which comprises assumption 1. Equation 3.6 can be integrated again to find the position of the electron at a given time t :

$$x(t, t_0) = \int_{t_0}^t \dot{x}(t, t_0) dt = \frac{q_e E_0}{m_e \omega_0^2} [\cos(\omega_0 t) - \cos(\omega_0 t_0) + \omega_0(t - t_0) \sin(\omega_0 t_0)]. \quad (3.7)$$

During propagation, the transverse spreading of the electron wavepacket becomes a non-negligible quantum mechanical effect. The velocity of this spreading, $v_\perp$, can be expressed in atomic units as [107, 108]

$$v_\perp = \frac{\sqrt{3E_0}}{(2I_p)^{0.25}} \quad (3.8)$$

such that the wavepacket transverse dimension upon recollision with the ion is

$$x_\perp = v_\perp \frac{2\pi}{\omega_0}. \quad (3.9)$$

For the example of an 800 nm ($\omega_0 = 0.057$ a.u.) driver laser with a peak intensity of 4×10^{14} W cm⁻² ($E_0 = 0.004$ a.u.) producing HHG in argon with $I_p = 15.76$ eV ($I_p = 0.58$ a.u.), equation 3.9 reveals that the transverse size of the electronic wavepacket upon recombination has increased by a factor of ~ 10 . Accounting for both dimensions transverse to the wavepacket propagation direction, this spreading becomes a factor of $\sim 10^2 = 100$. This implies a reduction in the recombination probability by the same factor and thus a large decrease in HHG yield. Although wavepacket diffusion is an inherently quantum effect, it is accepted that the longitudinal motion of the wavepacket can be adequately described by the classical equations of motion shown in equations 3.5

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to 3.7 [109]

Following the above, it can be understood that the harmonic yield is strongly coupled to the polarisation of the driving laser, as any transverse component of the driver polarisation steers the electron wavepacket away from the ion [109]. As a result, in the recombination stage of the three step model, the electron wavepacket in an elliptically polarised driver field would be spatially displaced from the ionic wavefunction with which it tries to recombine. Compounded by the aforementioned transverse diffusion of the electron wavepacket, the probability of recombination is reduced further as this displacement increases until the limiting case of a circularly polarised driver where the electron wavepacket ‘orbits’ the host ion with a vanishingly small chance to recombine. Conversely, for a linearly polarised driver the electrons that fail to recombine upon their first return to the ion have a chance, albeit very low, to recombine at subsequent encounters with the ion as they oscillate in the same plane [110, 111].

3.2.3 Recombination

The kinetic energy E_{KE} of an electron at a time t can be calculated from equation 3.6 as

$$\begin{aligned} E_{ke}(t, t_0) &= \frac{q_e^2 E_0^2}{2m_e \omega_0^2} [\sin(\omega_0 t) - \sin(\omega_0 t_0)]^2 \\ &= 2U_p [\sin(\omega_0 t) - \sin(\omega_0 t_0)]^2, \end{aligned} \quad (3.10)$$

where U_p is the ponderomotive energy given by

$$U_p = \frac{q_e^2 E_0^2}{4m_e \omega_0^2} = 9.3 \times 10^{-14} I_0 \left[\text{W cm}^{-2} \right] \lambda^2 \left[\mu\text{m} \right]. \quad (3.11)$$

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This form of U_p can be found by considering the oscillatory motion of the electron in the external driver field of frequency ω_0 . The time averaged energy of such a particle is

$$U = \frac{m_e \omega_0^2}{2} \langle x(t)^2 \rangle \quad (3.12)$$

where the angled brackets denote a time average. Substituting equation 3.7 into the above recovers equation 3.11.

The driver phases upon recombination, $\omega_0 t_r$, are the nontrivial roots of $x(t, t_0) = 0$. These are found from equation 3.7 as

$$\cos(\omega_0 t_r) - \cos(\omega_0 t_0) + \omega_0(t_r - t_0) \sin(\omega_0 t_0) = 0. \quad (3.13)$$

This allows the kinetic energy of the electron upon recombination to be found with equation 3.14:

$$E_{ke}(t_r, t_0) = 2U_p [\sin(\omega_0 t_r) - \sin(\omega_0 t_0)]^2. \quad (3.14)$$

For $0 < \omega_0 t_0 < \pi/2$, the ionised electron is carried through the continuum on a path that returns and intersects with the ion at least once. If $\pi/2 < \omega_0 t_0 < \pi$ however, the electron never returns to the ion and is instead lost to the continuum. Thus, only half of the ionised electrons ever return to the vicinity of the ion. Each of the trajectories undertaken by electrons with $0 < \omega_0 t_0 < \pi/2$ have a corresponding driver phase at the point of recombination that is found by solving equations 3.13 and 3.14. The kinetic energy of these trajectories found from equation 3.14 takes a maximum value of $3.17 U_p$ [100] when $\omega_0 t_0 = 17^\circ$ and $\omega_0 t_r = 255^\circ$.

For kinetic energies less than $3.17 U_p$, there exists a pair of trajectories that satisfies equation 3.14. The two trajectories are distinguished by the lengths of the paths travelled by the electron in the continuum. The shorter path is described by ionisation

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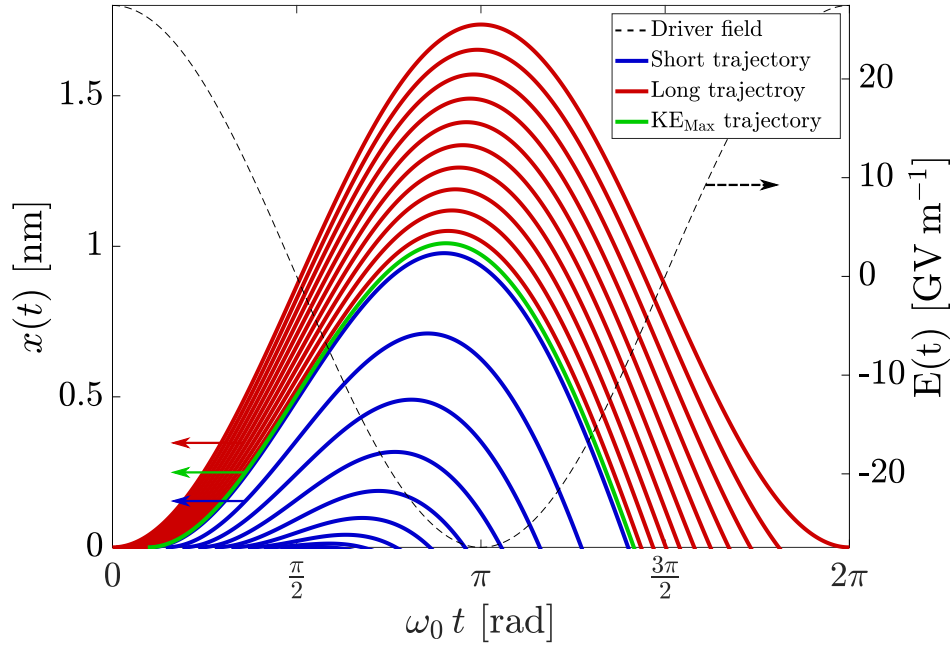


Figure 3.2: The displacements of 10 long (solid red lines) and 10 short (solid blue lines) electron trajectories ionised at driver field phases of $0^\circ < \omega_0 t_0 < 16^\circ$ and $18^\circ < \omega_0 t_0 < 90^\circ$ respectively. The trajectory with the maximum kinetic energy is shown in green where $\omega_0 t_0 = 17.1^\circ$ and $\omega_0 t_r = 255^\circ$. The driving laser parameters used are $\lambda = 800$ nm and a peak intensity of 1×10^{14} W cm $^{-2}$.

at $17.1^\circ < \omega_0 t_0 < 90^\circ$ and recombination at $90^\circ < \omega_0 t_r < 255^\circ$ whilst the longer is described by ionisation at $0^\circ < \omega_0 t_0 < 17.1^\circ$ and recombination at $255^\circ < \omega_0 t_r < 360^\circ$. These are known as the short and long trajectories respectively.

Figure 3.2 shows the electron displacement $x(t)$ as it follows one of 10 short or 10 long trajectories, denoted by the blue and red curves respectively. The single trajectory in this half of the driver cycle that provides the maximum kinetic energy upon recombination has been highlighted in green and it delineates the short and long paths. The dashed curve corresponds to the electric field of a 800 nm driver with a peak intensity of 1×10^{14} W cm $^{-2}$.

Given that a pair of driver phase values $(\omega_0 t_0, \omega_0 t_r)$ satisfies equations 3.13 and 3.14, additional solutions of the form $(\omega_0 t_0 + m\pi, \omega_0 t_r + m\pi)$ must also be valid, where m

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is a positive integer. This implies that the harmonics are emitted as a train of pulses separated by half the driving laser period, T . The time dependent harmonic intensity $I_q(t)$ can therefore be described by a Dirac comb [112] in time, which holds a Fourier transform relationship with its frequency space representation [113], $I_q(\omega)$:

$$I_q(t) \propto \sum_n^{\infty} \delta\left(t - n\frac{T}{2}\right) \quad (3.15a)$$

$\Downarrow$ FT

$$I_q(\omega) \propto 2\omega_0 \sum_n^{\infty} \delta(\omega - 2n\omega_0). \quad (3.15b)$$

Equation 3.15b describes a harmonic spectrum comprising discrete components separated by twice the driver frequency.

When generated in a monatomic gaseous medium the harmonic orders are produced at odd integer multiples of the driving laser frequency. This is a consequence of the inversion symmetry of the nonlinear process: The induced polarization of a centrosymmetric atom must be an odd function of the driver electric field, resulting in only odd harmonic orders.

Assumption 3 in section 3.2 specified that the total energy of the recombining electron, comprising E_{ke} and the ionisation potential I_p of the ion, is released as a harmonic photon. Accordingly, the maximum harmonic photon energy $E_{q,\max}$ is set by the maximum electron kinetic energy upon recombination $E_{ke,\max} = 3.17 U_p$ [100, 114] such that

$$E_{q,\max} = I_p + 3.17 U_p \quad (3.16a)$$

$$q_{\max} = \frac{1}{\hbar\omega_0} (I_p + 3.17 U_p). \quad (3.16b)$$

Equation 3.16b is found from 3.16a by observed that the energy of a photon in the q^{th} harmonic order is q times that of the driver photon energy.

3.3 Harmonic Beam Properties

In chapter 6 HHG driven by a driver beam with a supergaussian transverse profile will be discussed. It is therefore useful to consider the harmonic generation process with a generalised driver profile described by

$$I_{\text{IR}}^{\perp}(r) = I_0 \cdot \exp \left[-2 \left(\frac{r}{w_0} \right)^n \right], \quad (3.17)$$

where n is the supergaussian order, r is the transverse radial coordinate, w_0 is the e^{-2} spot size and I_0 is the peak intensity. Implicitly, when $n = 2$, equation 3.17 describes a Gaussian intensity profile.

The complex field of a harmonic source of order q in the generation plane can be approximately expressed through the simple dipole model [115] as

$$E_q^{\perp} \propto |E_{\text{IR}}^{\perp}|^{q_{\text{eff}}} \cdot \exp [-i(q\Phi_{\text{IR}} + \Phi_{\text{dipole}})]. \quad (3.18)$$

Here, $E_{\text{IR}}^{\perp}$ is the transverse amplitude profile of the driver beam, q_{eff} is the effective non-linearity of the HHG interaction, Φ_{dipole} is the dipole phase and Φ_{IR} comprises both the transverse driver and geometric phase.

Through the simple dipole model, this implies that the harmonic intensity in the source plane takes the form

$$I_q^{\perp}(r) \propto \exp \left[-2 q_{\text{eff}} \left(\frac{r}{w_0} \right)^n \right], \quad (3.19)$$

where the driver spot size and harmonic source size w_q are related [6, 116] through

$$w_q = \frac{w_0}{\sqrt[n]{q_{\text{eff}}}}. \quad (3.20)$$

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The harmonic phase terms in equation 6.8 will now be considered along with conditions under which their magnitudes can be manipulated and or neglected.

3.3.1 Transverse Driver Phase

The transverse phase of the driver beam in the harmonic generation plane is imprinted onto the harmonic field and multiplied by the harmonic order q :

$$\Phi_{\text{IR}}^{\perp} = q \frac{\pi r^2}{\lambda R(z)}. \quad (3.21)$$

Here, $R(z)$ is the radius of curvature of the Gaussian driver beam given by

$$R(z) = z \left[1 + \left(\frac{\pi w_0^2}{\lambda z} \right)^2 \right]. \quad (3.22)$$

The magnitude of this term can be adjusted by changing the driver spot size in the generation medium or the position of the focus relative to the generating medium. However, doubling w_0 quadruples the beam area and therefore the pulse energy required to support the same peak intensity. This approach to manipulating the driver phase is therefore subject to the available pulse energy.

In equation 3.22, the focus is located at $z = 0$, enabling the transverse driver phase component of the harmonic field to be neglected if the generating medium is located in the focal plane. This situation can be achieved by using a generating medium with thickness much less than the Rayleigh range of the driver beam such that the transverse variation of the phase is minimised over the longitudinal distance occupied by the generating medium. In all experiments presented within this thesis, the generating medium thickness was less than $900 \mu\text{m}$, whilst the driver beam Rayleigh range was greater than 4 mm . In section 6.1, the procedure for locating the generating medium exactly at focus will be described.

3.3.2 Geometric Phase

A consequence of focussing radiation is a phase advance along the propagation axis. In the case of a Gaussian beam, this phase advance is known as the Gouy phase [117] and can be shown analytically [118] to be

$$\Phi_{\text{geo}}^{\parallel} = -\tan^{-1} \left(\frac{z\lambda}{\pi w_0^2} \right). \quad (3.23)$$

The form of equation 3.23 shows that the phase monotonically decreases from $\frac{\pi}{2}$ to $-\frac{\pi}{2}$ as z varies from $-\infty$ to ∞ with the driver focus located at $z = 0$.

Manipulation of the form of the geometric phase of the driver beam has been reported in reference [119] where two foci longitudinally separated in space were shifted in relative phase. This shift introduced inflection points into the geometric phase such that its variation was no longer monotonic with z . A similar behaviour was observed in reference [120] where two contiguous radial regions of a single collimated beam were shifted in relative phase. Upon focussing, the geometric phase was again observed to develop inflection points as the phase difference between the two regions was varied.

The absolute value of the geometric phase is dependent upon the mode of the focussed beam. For higher order transverse electromagnetic and Laguerre Gaussian modes, the value of the geometric phase advance is multiplied by a factor dependent on the sum of their respective mode indices [121, 122]:

$$\Phi_{\text{geo}}^{\parallel} = \begin{cases} -(n + m + 1) \tan^{-1} \left(\frac{z\lambda}{\pi w_0^2} \right) & \text{for TEM mode } I_{n,m} \\ (2p + |l| + 1) \tan^{-1} \left(\frac{z\lambda}{\pi w_0^2} \right) & \text{for LG mode } I_{p,l} \end{cases} \quad (3.24)$$

If the longitudinal dimension of the generating medium is much less than the Rayleigh range of the driver, it can be assumed that the variation of the geometric phase across

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the generating region is negligible. This allows the value of the geometric phase to be assumed constant within the generating region.

3.3.3 Dipole Phase

The dipole phase is accrued by the electron during the time it spends in the continuum in the propagation step of the three step model. It can be thought of as the phase difference between the recombining electron wavepacket and ionic wavefunction. In the simple dipole model based on the strong field approximation, the dipole phase can be approximated by [123]

$$\Phi_{\text{Dipole}} \propto U_p(t_r - t_0) \approx \alpha_q^j |E_{\text{IR}}^\perp|^2. \quad (3.25)$$

where α_q^j is a proportionality factor assumed constant for a given harmonic order q and trajectory j . In order to calculate α_q^j , a quantum mechanical expression of the time dependent dipole moment $D(t)$ is required. In reference [98] this is written as

$$D(t) = i \int_0^{t_r} \int^{\mathbf{P}} \left[E_{\text{IR}} \cos(t_0) d_x(\mathbf{p} - \mathbf{A}(t_0)) \cdot d_x^*(\mathbf{p} - \mathbf{A}(t_r)) e^{-iS(\mathbf{p}, t_r, t_0)} + c.c \right] d^3\mathbf{p} dt_0 \quad (3.26)$$

where $E_{\text{IR}} = \frac{\partial \mathbf{A}}{\partial t}$, $\mathbf{A}(t) = (E_{\text{IR}} \sin(t), 0, 0)$ is the vector potential of the driver field, $\mathbf{p}$ is the canonical momentum of the electron, d_x is the dipole transition matrix component parallel to the x dimension, $c.c$ represents the complex conjugate terms and

$$S(\mathbf{p}, t_r, t_0) = \int_{t_0}^{t_r} \left(\frac{[\mathbf{p} - \mathbf{A}(t'')]^2}{2} + I_p \right) dt'' \quad (3.27)$$

is the semi-classical action of the electron. The constituent terms of equation 3.26 can be understood as follows; $E_{\text{IR}} \cos(t_0) d_x(\mathbf{p} - \mathbf{A}(t_0))$ is the probability amplitude for the electron to be ionised into the continuum at time t_0 with canonical momentum $\mathbf{p}$. The electron accrues a phase $e^{-iS(\mathbf{p}, t_r, t_0)}$ during the time $t_r - t_0$ spent in the continuum after

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which it recombines with the host ion with a probability amplitude $d_x^*(\mathbf{p} - \mathbf{A}(t_r))$.

The primary contributions to the momentum integral in equation 3.26 are from the stationary points of the classical action: Rapid oscillations in the exponential term will cancel out for $t_r - t_0$ greater than one driver cycle meaning that the dominant contributions to the momenta integral occur when $S(\mathbf{p}, t_r, t_0)$ is minimised. This is known as the principle of stationary action [124]:

$$\nabla_{\mathbf{p}} S(\mathbf{p}, t_r, t_0) = 0. \quad (3.28)$$

As the action describes the path taken by the electron, equation 3.28 can be interpreted as the trajectory undertaken by the electron between ionisation and recombination times t_0 and t_r that results in the return to the same spatial location from which it was ionised.

The partial derivatives resulting from the time integral in equation 3.26 give two expressions that when combined with equation 3.28 can be solved [123] to reveal

$$S = I_p \tau + U_p \tau - \frac{U_p}{\omega_0^2 \tau} \sin^2 \left(\frac{\omega_0 \tau}{2} \right) - \frac{U_p}{\omega_0} \cos(2\omega_0 t_r - \omega_0 \tau) \cdot \left[\sin(\omega_0 \tau) - 4 \frac{\sin^2 \left(\frac{\omega_0 \tau}{2} \right)}{\omega_0 \tau} \right], \quad (3.29)$$

where $\tau = t_r - t_0$. From equation 3.29, the assumed constant trajectory coefficient α_q^j can be found by [123]

$$\frac{\partial S}{\partial t} = \alpha = \frac{q_e^2}{2\omega_0^3 m_e c \epsilon_0 \hbar} \left[\omega_0 t - \frac{4}{\omega_0 t} \sin^2 \left(\frac{\omega_0 t}{2} \right) - \cos(2\omega_0 T - \omega_0 t) \cdot \left(\sin(\omega_0 t) - \frac{4 \sin \left(\frac{\omega_0 t}{2} \right)}{\omega_0 t} \right) \right], \quad (3.30)$$

The parameter α in units of $\text{cm}^2 \text{W}^{-1}$ can be found by solving this with the parameters associated with a certain harmonic order and trajectory.

3.3.4 Dispersion

Assuming harmonic generation takes place within a gaseous medium, both the neutral atoms and free electrons will contribute to the dispersion of the propagating harmonic field. These contributions manifest as the wavevector mismatch $\Delta \mathbf{k}_{\text{at}}$ and $\Delta \mathbf{k}_{\text{plasma}}$ respectively:

$$\Delta \mathbf{k}_{\text{at}} = \frac{2\pi q}{\lambda_0} \left[n(\lambda_0) - n\left(\frac{\lambda_0}{q}\right) \right] (1 - \eta) \frac{P}{P_{\text{atm}}}, \quad (3.31a)$$

$$\Delta \mathbf{k}_{\text{plasma}} \approx -N_e r_e \lambda_0 q. \quad (3.31b)$$

Here, λ_0 is the central wavelength of the driver beam, $n(\lambda_0)$ is the refractive index as a function of wavelength, η is the fraction of ionised atoms, P is the backing pressure of the gas cell, P_{atm} is atmospheric pressure, N_e is the density of free electrons in the interaction region and r_e is the classical radius of the electron.

3.3.5 Phase Matching

The longitudinal distance over which harmonics constructively interfere is a function of the mismatch between the driver and harmonic wavevectors. A metric that describes the distance over which the harmonics remain coherent is known as the coherence length:

$$L_c = \frac{\pi}{|\Delta \mathbf{k}_q|}, \quad (3.32)$$

where

$$\Delta \mathbf{k}_q = \mathbf{k}(q\omega_0) - q\mathbf{k}(\omega_0) = \Delta \mathbf{k}_{\text{geo}} + \Delta \mathbf{k}_{\text{at}} + \Delta \mathbf{k}_{\text{plasma}} + \Delta \mathbf{k}_{\text{Dipole}}. \quad (3.33)$$

Here, $\Delta \mathbf{k}_q$ is the wavevector mismatch between the q^{th} harmonic wavevector $\mathbf{k}(q\omega_0)$ and driver wavevector $q\mathbf{k}(\omega_0)$ [125]. Phase matching is achieved if $|\Delta \mathbf{k}_q| = 0$; the

harmonic emission produced in each sequential driver cycle constructively interferes with harmonics generated earlier in the medium resulting in a coherent build-up of amplitude [126]. If $|\Delta \mathbf{k}_q| \neq 0$ however, a phase slip will be accumulated as the harmonics propagate. If this phase slip reaches π radians, the harmonic intensity will oscillate with the propagation distance z .

A nontrivial solution to $|\Delta \mathbf{k}_q| = 0$ is possible when the constituent wavevectors cancel out. This is typically achieved by manipulating the pressure P and the location of the generating medium, changing $\Delta \mathbf{k}_{\text{at}}$ and $\Delta \mathbf{k}_{\text{geo}}$. The sign of the $\mathbf{k}_{\text{geo}}$ is negative for $z < 0$ and positive for $z > 0$ [127] enabling large changes to the phase matching conditions to be made. As P is increase from 0 mbar, the detected harmonic flux increases linearly until the phase matching pressure is reached, at which point, the linear increase in harmonic flux rolls off and negligible further gains are made. A simple test to establish whether harmonic generation is phase matched therefore, is to vary the pressure and observe the trend in detected flux.

In reference [128], the conditions surrounding on-axis and off-axis phase matching are explored. For the purposes of this thesis, only the on-axis harmonics produced by the short trajectory electronic excursions are considered: The longer trajectory harmonics are more divergent and are consequently neglected as they fall outside the acceptance aperture of the imaging system described in this thesis.

3.4 Macroscopic Properties

3.4.1 Spectrum

A typical [HHG](#) spectrum produced in a gaseous medium comprises three distinct regions; an initial decrease in harmonic intensity with harmonic order q , a plateau of

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harmonics with roughly constant intensity and an abrupt cut-off above which the harmonic intensity drops to zero. In figure 3.3 a), these three regions are denoted i), ii) and iii), separated by the blue dashed lines. The harmonics in region i) are produced by the anharmonic radiation from electrons that remain within bound states. Their motion remains within the perturbative regime. In region ii) however, the intensities of increasing harmonic orders remains roughly constant. This occurs when the electrons are able to tunnel into the continuum and therefore produce harmonics with at least the ionisation potential of the atom from which they were ionised in addition to the kinetic energy they receive from the driver laser. The plateau extends until the cutoff region iii) where the harmonic intensity rapidly drops. The cutoff occurs at the point when the recombining electron has the maximum kinetic energy available from the driver laser. Equation 3.16b in section 3.2.3 showed that this corresponded to $3.17U_p$.

As $U_p \propto I\lambda^2$, the plateau, and number of harmonic orders can be extended by either increasing the driver intensity in the generation plane, or using a driver with a longer wavelength. Increasing the driver intensity has two advantages: The scaling of U_p extends the plateau, and the increased intensity enables HHG to occur in ions with a higher I_p [129]. However, this increases the number of free electrons in equation 3.31b, making it more difficult to phase match the harmonics. In addition, the transverse variation of the free electron content, and therefore local refractive index, acts to defocus the harmonic beam, decreasing the overall brightness [130].

For intensities above approximately $1 \times 10^{17} \text{ Wcm}^{-2}$, the magnetic field of the driver can no longer be neglected. The oscillation of a charged particle in a magnetic field B is subject to the $E \wedge B$ drift that causes the centre of the oscillation to drift in an orthogonal direction to both E and B . As the driver intensity increases further, the magnitude of this drift increases until the overlap between the recombining electron and ionic wavefunctions drops to zero.

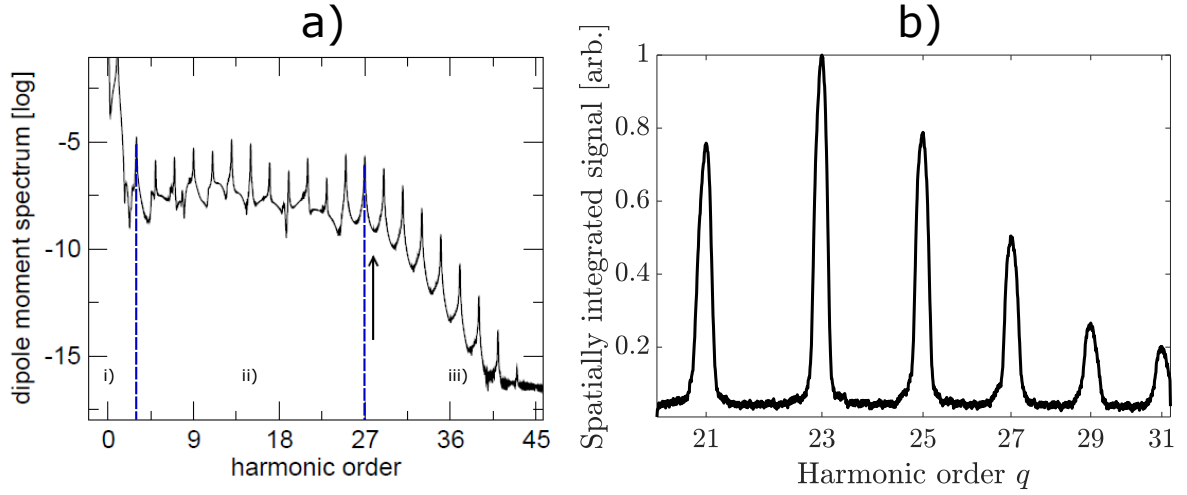


Figure 3.3: a) Simulated high harmonic spectrum produced in a model atom representative of xenon by an 800 nm driving laser of peak focal intensity $1.5 \times 10^{14} \text{ W cm}^{-2}$. The arrow marks the location of the cut-off order according to equation 3.16b. Plot adapted from reference [132]. b) Experimental spectra obtained by the author using a 800 nm Gaussian driver beam of peak intensity $4.7 \times 10^{14} \text{ W cm}^{-2}$ and pulse length of 35 fs focussed into 75 mbar of argon with an exposure of 5 seconds.

It has been shown that using both the driver and high order harmonics simultaneously to driving a second stage HHG process can increase the intensity of the generated harmonics in the plateau by up to 10 orders of magnitude [131].

Experimentally, harmonic spectra are modulated by the reabsorption of the harmonics in the generating medium, IR filters, and the acceptance aperture of the detection system. These factors obscure the three characteristic regions of the simulated harmonic spectrum resulting in the experimental spectra such as that measured by the author, shown in figure 3.3 b).

3.4.2 Beam Profile

An example of the harmonic fluence profile of the 25th harmonic order detected by the author is shown in figure 3.4 a). The lineouts in figure 3.4 b) confirm the Gaussian

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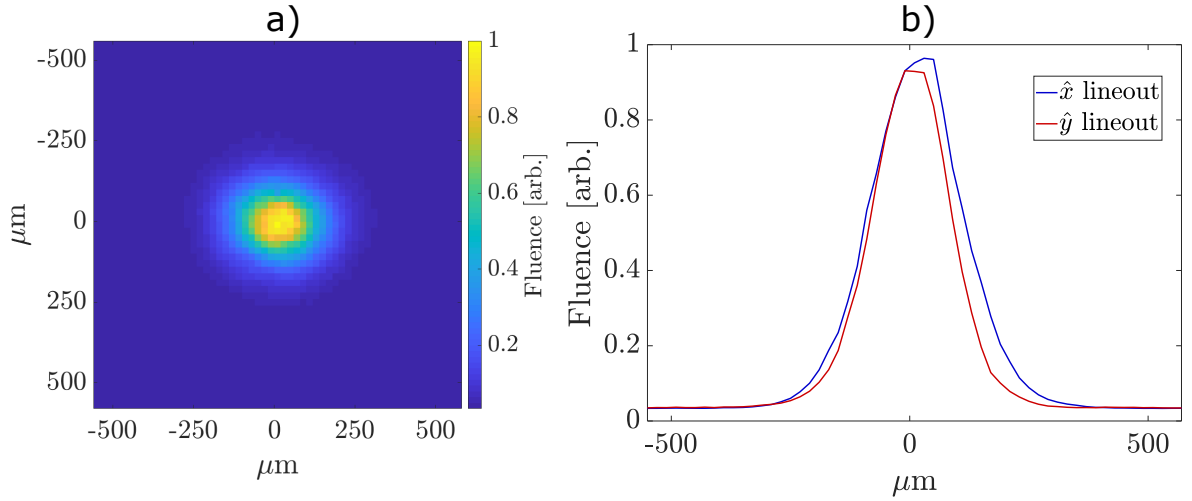


Figure 3.4: a) Experimentally observed transverse fluence profile of the 25th harmonic order obtained using a 800 nm Gaussian driver beam of peak intensity $4.7 \times 10^{14} \text{ W cm}^{-2}$ and pulse length of 35 fs focussed into 75 mbar of argon with an exposure of 0.1 seconds. b) Lineouts of the profile shown in a) along the $\hat{x}$ (blue) and $\hat{y}$ (red) dimensions.

profile of the harmonic beam and reveal a small degree of ellipticity. This is likely due to the astigmatism introduced into the harmonic beam upon reflection from an off-axis focussing mirror shown later in figure 4.2.

The harmonic beam has a very high degree of spatial coherence due to its near Gaussian profile described by equation 3.18 and generation mechanism [133]. This is vital for its application to techniques such as lensless imaging, which become far more complex when applied to the case of partial coherence [33]. A number of techniques exists for characterising the coherence of a beam such as through the inspection of the visibility of the interference fringes observed downstream of the illumination of a pair of apertures such as slits or pinholes [134]. However, this only permits the quantification of the coherence between the two points of the beam transmitted by the apertures. Ideally, a surface describing the coherence of the beam over an entire transverse plane would be calculated. Due to the experimental complexity of such a measurement and the expected cylindrical symmetry of the beam, a technique called ‘SCanning Interference Measurement for Integrated Transverse Analysis of Radiation’ (SCIMITAR) [135] can

be used to characterise the coherence along a complete line orthogonal to the beam propagation direction.

3.5 State of the Art

Each modality of lensless imaging has found applications in a wide range of research and industrial pursuits. This flexibility has driven the development each modality at different rates, resulting in a wide field, rich with varied results and records.

In 2014, ptychographic [CDI](#) was performed at the at the Advanced Light Source synchrotron (Lawrence Berkeley National Lab) by Shapiro et al. [[136](#)]. Components of a test object containing features with widths down to 5 nm were successfully resolved using 3 nm wavelength radiation. Also in 2014, Rui et al. [[137](#)] performed single shot [CDI](#) at the SACLA [XFEL](#) (Japan), imaging individual gold nanocrystals with a resolution of ≈ 5.5 nm using 2.3 Å wavelength radiation. These results comprise the finest resolution images retrieved by [CDI](#) to date.

The resolution of images recovered through harmonic driven [CDI](#) has recently breached the wavelength limit. In 2017, Gardner et al. [[138](#)] ptychographically imaged a Fresnel zone plate using 13.5 nm harmonic radiation, recovering an image with a resolution of 12.6 nm after processing their data with a novel [IPRA](#). By providing a single image of the unscattered beam as a priori information to the [IPRA](#), the estimation of the imaging beam could be highly constrained, increasing the likelihood that the algorithm would converge to a global minimum.

The elemental discrimination accessible with harmonic driven ptychography has been demonstrated by Shanblatt et al. [[139](#)] in 2016 who used 29 nm harmonic radiation to image buried copper structures at the interface between aluminium and silicon oxide.

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3.5. STATE OF THE ART

The resulting diffraction limited images with resolutions of 162 nm were obtained using [CDI](#) in reflection geometry in a non destructive manner. Also in 2016, Kim et al. [[140](#)] used a high-harmonic source to perform lithography, generating features with a < 200 nm pitch. These results have solidified the applications of both [HHG](#) and lensless imaging in a contemporary industrial setting.

The limitations of [CDI](#) with respect to coherence are also being challenged. In 2017, Classen et al. [[141](#)] developed a technique that exploits the intensity correlations between incoherently scattered x-ray radiation to image the full three dimensional structure of the scattering object. The extension of lensless imaging into single shot attosecond imaging requires a broadband treatment such as that used by Huijts et al. [[142](#)] in 2019. Their approach involves the numerical monochromatisation of the broadband data by a regularised inversion of a matrix with spectrally dependent entries. The subsequent experimental results indicate resolutions comparable and superior to monochromatic [CDI](#) under otherwise similar conditions.

In 2018, Tadesse et al. imaged a binary transmissive aperture (comprising three letters) with [FTH](#) driven by a harmonic with a wavelength of 18.1 nm. The detected data was then passed through an [IPRA](#) which refined the resolution of the recovered image down to 34 nm, currently the finest resolution ever achieved by either a harmonic, synchrotron or [XFEL](#) source in this mode of imaging. The reference aperture(s) used in the sample plane were only 50 nm in diameter. Although this restricted the detected flux, a high brightness source and multiple references were used to increase the [S:N](#) of the final image.

The combination of [CDI](#) and [FTH](#) was suggested in 2012 by Latychevskaia et al. [[143](#)] largely in the context of using an image recovered through [FTH](#) to seed an [IPRA](#). However, the introduction of a pinhole or reference feature into or near the object plane can be experimentally challenging and inconvenient, a fact that has reinforced

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the separation between [CDI](#) and [FTH](#). However, whilst imaging nanoparticles in 2018, Gorkhover et al. [[144](#)] used nanoclusters in the object plane as a source of scatter that provided the reference wave. With this technique, images with resolutions limited only by photon flux to 20 nm were recovered.

Further improvements in the resolution of images recovered from harmonic driven lensless imaging are predicated upon improvements in the efficiency and wavelength limit of [HHG](#). In 2012, experimental measurements were made by Popmintchev et al. [[145](#)] of a harmonic source with a near keV bandwidth that, appropriately compressed, could support transform limited pulse durations of 2.5 as. Further, under phase matching conditions in 10s of atmospheres of helium, the 5001st harmonic order of a $3.9\text{ }\mu\text{m}$ driver beam was generated, corresponding to a wavelength of $7.8\text{ }\text{\AA}$. However, the photon yield at these ultrahigh harmonic orders is very low, even under phase matching conditions.

In the future, the optimisation of ultrahigh harmonic orders and lensless imaging techniques may lead to sub-nanometre scale resolutions accessible to small research facility scale laboratories.

Chapter 4

Beamline Overview

4.1 Laser Specifications



IN this chapter an overview of the laser system and beamline used in this thesis is provided. The Ti:Sapphire laser system comprised an oscillator, pump and regenerative amplifier. The parameters of each component are summarised in table 4.1. Also within this chapter are details about the fabrication of samples used in the lensless imaging experiments described in chapter 5, and the SLM used to spatially shape the driver pulses.

The output mode quality of the regenerative amplifier can be parametrised by the M^2 metric [146]. For the amplifier used to gather all results presented in this thesis, the M^2 parameter was 1.5 according to a manufacturer supplied specifications sheet. However, due to aberrations within the amplifier optics and environmental effects such as temperature gradients and air currents, the experimentally achievable M^2 was likely between $1.7 < M^2 < 1.9$. If this value becomes much larger than 1, the low beam quality compromises certain assumptions made during experimentation. For example, the minimum size of the driver focal spot will be larger than that of a Gaussian beam resulting in a lower peak intensity at focus, reducing the number of harmonic orders

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Table 4.1: Laser system components

Laser	Property	Value
Oscillator (MaiTai, Spectra-Physics)	Wavelength	780–820 nm
	Average power	240 mW
	Pulse energy	2.8 nJ
	Repetition rate	84 MHz
	Pulse length	35 fs
Pump (Empower, Spectra-Physics)	Wavelength	527 nm
	Average power	20 W
	Pulse energy	20 mJ
	Repetition rate	1–10 kHz
	Pulse length	10 ns
Regenerative amplifier (Spitfire Pro, Spectra-Physics)	Wavelength	780–820 nm
	Average power	4 W
	Pulse energy	4 mJ
	Repetition rate	1 kHz
	Pulse length	35 fs
	e^{-2} intensity radius	4.3 mm

that can be generated in [HHG](#). Further, the phase masks imprinted onto the beam in chapters [6](#) and [7](#) using a [SLM](#) assume the transverse intensity profile of the driver to be radially symmetric. Any deviation from this will therefore result in a degradation of the achieved shaping and subsequent experimentation.

4.2 Beamline Layout

The diagram in figure [4.1](#) illustrates schematically the path taken by the driver beam from the amplifier to the focal plane of the IR focussing lens. The driver pulses passed through a mechanical shutter (ThorLabs, SC10) which prevented laser pulses from reaching the target chamber when not required and was synchronised to the readout rate of the [CCD](#) such that no pulses landed on the chip between acquisitions. The shutter also served to minimized the thermal load on the [SLM](#).

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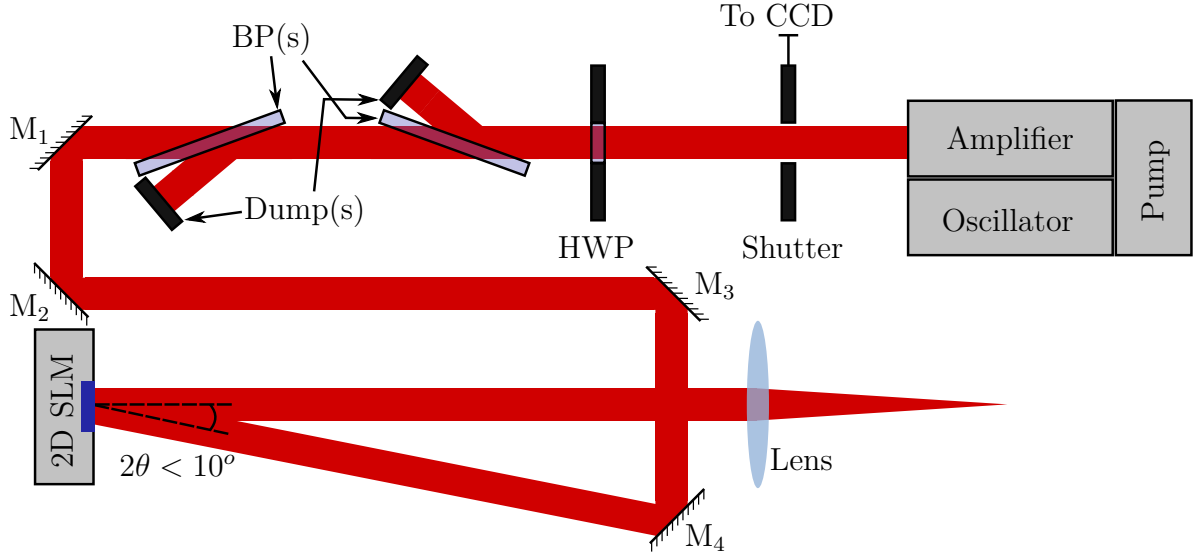


Figure 4.1: A schematic diagram of the driving laser beamline. HWP denotes a half-wave plate, BP(s) are Brewster plates, M_{1-4} are dielectric mirrors and SLM is the spatial light modulator.

A pair of half-wave plates (Thorlabs, zero order for use between 780–820 nm) were used to rotate the linear polarisation of the laser pulses. In conjunction with the Brewster plates (Eksma Optics broadband 750–850 nm thin film laser polarizers), this allowed the pulse energy of the driving pulses to be adjusted to within approximately $5 \mu\text{J}$. At a Brewster's angle of 70° these plates transmitted P-polarisation (P-pol) without losses, each with an extinction ratio of 200:1. This angle was determined experimentally through inspecting the transmitted power of P-pol light through a cubic polarizer. The rejected light from the Brewster plates was disposed of in two beam dumps. The second Brewster plate was placed at the complimentary angle to the first in order to minimise the amount of spatial chirp introduced into the beam that would compromise the achievable peak intensity.

All mirrors (Thorlabs BB2-E03 for use at 750–1100 nm) were broadband dielectrics. When a beam comprising a mix of S-pol and P-pol is reflected from a dielectric mirror

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with a non-zero angle of incidence, the S and P polarisations will experience different phase shifts [147]. Consequently, a pure S-pol or P-pol state must be established before reflection from any dielectric mirrors in order to maintain a pure polarisation state. This motivated the installation of the Brewster plates as they represented a good compromise between extinction ratio, damage threshold and price.

The SLM (Hamamatsu model X10468-02) was an 8-bit electronically addressed parallel aligned nematic (PAN) liquid crystal on silicon (LCoS) phase-only SLM. The SLM active area comprised 792×600 square pixels of side $20 \mu\text{m}$ with a fill factor of 98%. A phase mask with the device calibration and residual aberration corrections was displayed on the SLM at all times. The operational principle of this device will be explored in section 4.4. The IR focussing lens (Eksma Optics FemtoLine BBAR lens 700–900 nm) was a plano convex UV fused silica 25.4 mm diameter lens of focal length $f_{\text{IR}} = 50 \text{ cm}$. The driver beam was subsequently focussed into a nickel gas cell where HHG took place.

During propagation through materials such as the focussing lens, nonlinear phase can be accrued which creates distortions in the beam focus. The length over which this becomes significant is known as the nonlinear length L_{nl} and can be expressed as [148]

$$L_{\text{nl}} = \frac{c A \tau}{E_0 n_2 \omega_0} \quad (4.1)$$

where c is the speed of light, A is the beam area in the relevant plane, τ is the pulse duration, E_0 is the pulse energy, n_2 is the nonlinear refractive index and ω_0 is the central frequency of the pulse. For UV fused silica, the refractive index at 800 nm ($\omega_0 = 2.36 \times 10^{15} \text{ rad s}^{-1}$) is $n = 1.45$, corresponding to a nonlinear index of $n_2 = 27.3 \times 10^{-21} \text{ m}^2 \text{ W}^{-1}$ [149]. Ergo for a $300 \mu\text{J}$ pulse of 35 fs duration and 4.3 mm e^{-2} radius, the nonlinear length is 3.16 mm. Although this is longer than the 1.2 mm thickness of the focussing lens used in all experiments presented within this thesis, it is

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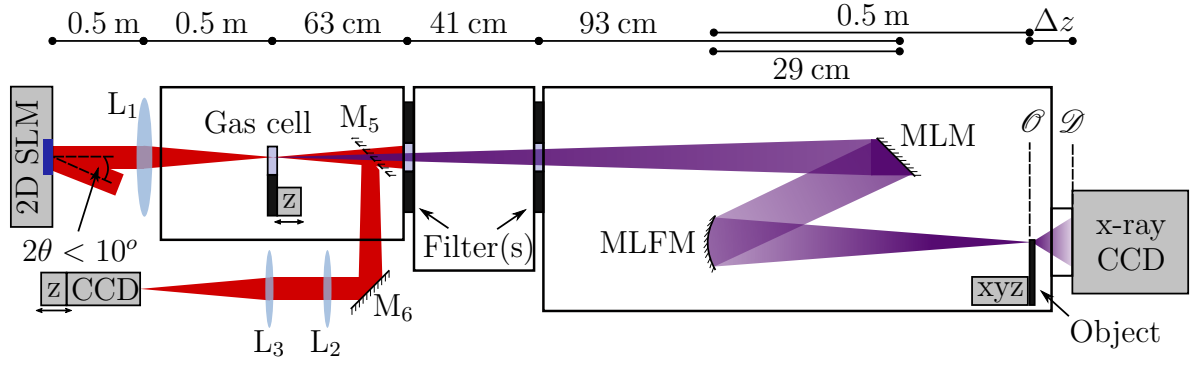


Figure 4.2: Schematic of the harmonic beamline. L_1 is the driver focussing lens, MLM is a multilayer XUV mirror, MLFM is a multilayer XUV focussing mirror. $\mathcal{O}$ is the object domain and $\mathcal{D}$ is the detector domain, separated by a distance of Δz . Z and XYZ are linear and three axis stages, M_5 and M_6 are 90/10 beamsplitters, the former of which is removable. L_2 is a $f = 50$ cm collimating lens and L_3 is a $f = 50$ cm refocussing lens. The three large boxes denote the first, second and third vacuum chambers.

shorter than the thickness of the Brewster plates in figure 4.1. As a result, nonlinear effects may be present in the beam focus, although given the results gathered, they were not considered deleterious. In principle, these could be corrected for by displaying the appropriate phase upon the SLM.

The diagram in figure 4.2 illustrates schematically the path taken by the harmonic beam after its generation by the focussed driver. The harmonics were generated within a nickel gas cell with typical internal thicknesses of between $300 - 900 \mu\text{m}$. The gas cell was typically backed with between $30 - 150$ mbar of argon (Ar) depending on the requirements of the experiment. The cell was mounted on a linear translation stage with 25 mm of travel which enabled the cell to be translated longitudinally relative to the IR focus.

Approximately 30 cm downstream of the gas cell, a 90/10 beamsplitter M_5 , mounted on a flip mount, could be inserted into the driver beam. This deflected the driver through a unitary magnification 4-f reimaging line onto a CCD webcam with 1024×1024 square pixels of side $2.2 \mu\text{m}$. The CCD was mounted on a linear translation stage with 50 mm

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of travel, enabling a caustic of the reimaged driver focal volume to be obtained.

When M_5 was removed from the beamline, all generated harmonics propagated 63 cm to the first of two aluminium (Al) filters separated longitudinally by 41 cm. Each filter comprised two 200 nm thick unsupported aluminium layers (Lebow sandwich filter) with a clear aperture of 10 mm diameter. Two layers were required to eliminate any transmission through nanoscale pinholes that appear in the layers during the manufacture process. The two filters eliminated the IR driver whilst transmitting all propagated harmonics with wavelengths longer than the aluminium L-edge at 17 nm [150]. The second filter was mounted within an aluminium cylinder plugged into the beampipe connecting the second and last vacuum chambers. This cylinder is schematically shown in figure 4.3 and served as a light filter eliminating all ambient contaminant radiation from the experiment.

The first of a pair of multilayer XUV optics (Fraunhofer IWS) was located 93 cm downstream of the last filter. These optics comprised a planar multilayer mirror, MLM, and a multilayer focussing mirror, MLFM. These optics were designed to reflect radiation of a central wavelength of 32 nm with a bandwidth of 1.6 nm. This corresponds to the 25th harmonic of the 800 nm driver beam. Both optics were boron carbide on silicon (B_4C/Si) multilayers, designed for an angle of incidence of 7° .

In the XUV spectral range, the flatness of reflective optics is limited by the flatness of the interfacial surfaces that comprise the multilayer coating. Modern XUV optics used in advanced light sources can exhibit a surface flatness down to the order of nanometres [151]. However, for optical components commercially accessible to small research facility scale laboratories, achievable surface flatness values are inferior. In this case, the optics are typically optically polished, giving a surface roughness of less than one wavelength.

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As the surface roughness approaches and exceeds the incident radiation wavelength, the reflectivity of the optic will become severely compromised. Further, a random phase shift proportional to the optic surface topology will be imprinted onto the harmonic beam, compromising the focal spot, brightness and the retrieved phase in a lensless image reconstruction. The achieved surface flatness in a planar reflector will be higher than that achieved in the focussing mirror due to the complexity introduced by multilayer deposition onto a curved surface. However, by considering the highly Gaussian form of the harmonic beam shown in figure 3.4 a), it can be qualitatively inferred that the surface flatness of the XUV optics used in all experiments presented in this thesis lies within acceptable bounds.

At the focal plane of the MLFM a sample or edge was mounted on an optically encoded three axis stage (SmarAct SLC-1780). During data acquisition the optical encoder was deactivated in order to prevent contamination of the data.

At a distance Δz of typically between 3 – 7 cm downstream of the $\mathcal{O}$ domain, one of two x-ray CCDs detected the harmonic beam in the $\mathcal{D}$ domain. These CCDs were an ANDOR iKon-L with an e2v manufactured chip that comprised 2048×2048 square pixels of side $13.5 \mu\text{m}$ with a fill factor of 100%, and a Princeton Instruments PIXIS-XO 400B with an e2v manufactured chip containing 1340×400 square pixels of side $20 \mu\text{m}$ with a fill factor of 100%. To reduce the thermal current in the detector during data acquisition, the CCD was cooled to between -40 and -50 degrees. According to manufacturer supplied specification sheets, this corresponded to between 0.001–0.003 electrons per pixel per second. The first two vacuum chambers housing the harmonic radiation were maintained at approximately 10^{-3} mbar. The last chamber supporting the CCD was held at 10^{-6} mbar. Two cameras were used due to an intermittent readout fault with the ANDOR camera that prevented any data from being gathered for periods of time varying from 1 week up to several months. This fault was

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never successfully diagnosed or fixed despite being replicated by ANDOR in their factory.

The attenuation of the harmonic beam between the primary and secondary source planes was caused by reabsorption in the harmonic generation chamber, the filters and the **XUV** optics. Each filter transmitted 39% [150] and each **XUV** optic reflected 26% of the 25th harmonic order. At a typical ambient argon gas pressure of 5×10^{-3} mbar, 91% of the 25th harmonic order was transmitted over 1.04 m from the gas cell to the second filter [150]. Hence approximately 1% of the generated harmonic reached the **O** domain.

Several additional factors were considered to ensure that gathered data was of the highest quality. These included ambient light filtration and preventing laser pulses from reaching the **CCD** between acquisitions. Figure 4.3 shows the cylindrical housing containing the second aluminium filter. This assembly was inserted into a beampipe connecting the last two vacuum chambers in order to prevent all ambient light from entering the imaging chamber.

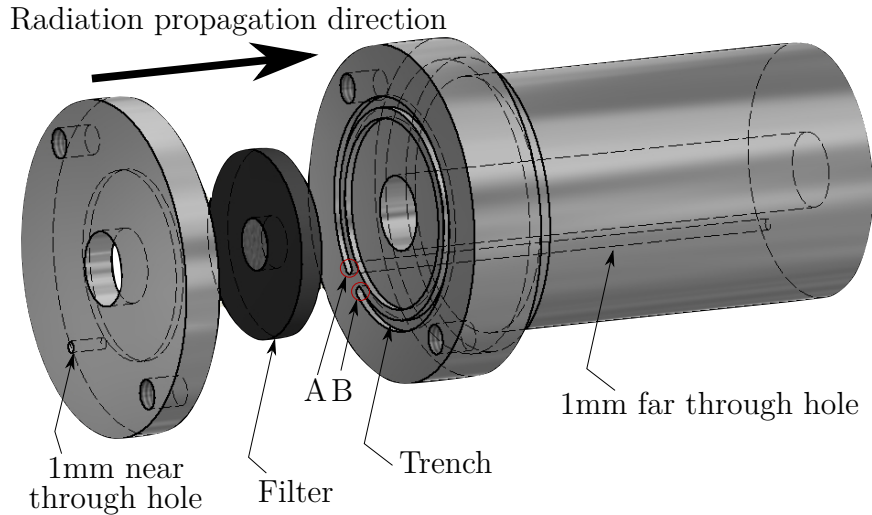


Figure 4.3: Schematic of the filter mount.

Once this filter mount was in place, a route was required to enable pressure equalisation to minimise mechanical stress on the filter membrane. This was achieved with the 1 mm

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4.3. SAMPLE FABRICATION

near and far through holes shown in figure 4.3. These holes were not aligned azimuthally and hence they did not provide a clear sight-line for contaminant light to enter the imaging chamber. Once assembled, the near through hole terminates at location ‘B’ whilst the far through hole starts at location ‘A’. These two locations are connected only by the trench of depth 1 mm that is confined to a plane orthogonal to the optical axis. Gas diffusing down a pressure gradient across the filter mount can travel around this trench but optical radiation will not be able to follow such a path.

The turbo pumps attached to the imaging chamber were connected to roughing pumps by translucent pipes. It was established that ambient light transmitted through these and into the imaging chamber (around the blades of the turbo pump) downstream of the filter mount caused an undesirable background signal. Consequently, shielding was applied to these pipes to remove this contaminant.

The mechanical shutter located in the driver beam path in figure 4.1 was synchronised to the transistor-transistor-logic (TTL) pulses produced by the x-ray CCD detectors during acquisition. These pulses were timed such that the shutter was closed during the chip cleaning and readout stages, but open during the acquisition stage. This prevented harmonic pulses from being detected by the chip whilst the prior acquisition was being read out. Without the shutter, this would manifest as a continuous stripe of signal parallel to the CCD readout direction with a width equal to that of the harmonic beam.

4.3 Sample Fabrication

The samples used in this thesis comprised a $200 \times 200 \times 0.05 \mu\text{m}$ silicon nitride (Si_3N_4) membrane supported within a $5 \times 5 \times 0.525\text{mm}$ silicon wafer (Silson Ltd). This was coated with gold of 125 nm thickness through thermal evaporation deposition. An aperture was then milled through both the gold and Si_3N_4 substrate with a gallium-based focussed ion

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4.3. SAMPLE FABRICATION

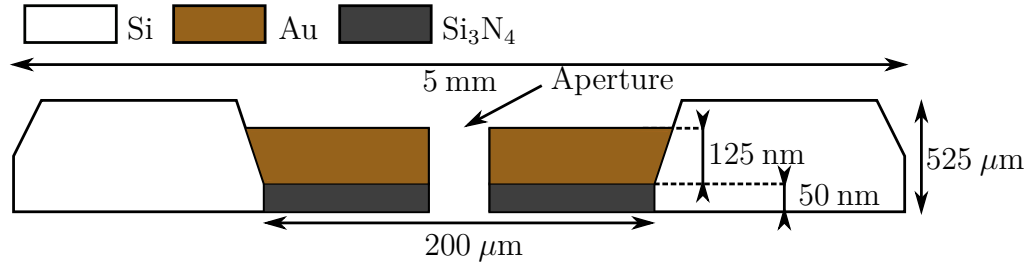


Figure 4.4: A cross section of a sample, comprising the silicon nitride substrate, gold coating, aperture and supporting silicon wafer. Not to scale.

beam (FIB) (FEI company) to act as an object. The construction of all samples imaged in this thesis was performed by the author on a FIB located in the University of Oxford, Department of Physics. A schematic of a sample cross section is shown in figure 4.4.

The naming convention used henceforth shall be as follows: ‘Sample’ refers to the entire coated substrate and wafer assembly. ‘Aperture’ refers to a hole milled through the substrate that is to be imaged. ‘Object’ refers to an imaging target that can be an aperture in the context of the author’s experiments, or an abstract item in the context of imaging theory. ‘Reference’ refers to a hole milled through the substrate that provides a well defined reference wave for holographic imaging. Unless otherwise specified, the term reference refers to a pinhole.

An electron micrograph of a set of apertures and references is shown in figure 4.5 below. The black regions correspond to areas where the transmission is unity and the grey regions represent undisturbed gold coating. The FIB parameters used during milling were a beam current of 46 pA at 30 keV over the course of 2 minutes per aperture, and 4 seconds per reference. The large transmissive rectangle in the upper left is an orientation feature used to easily locate and align the sample with respect to the harmonic beam. Due to its larger size, this alignment hole was milled at a higher beam current of 0.9 nA at 30 KeV for 5 minutes. However, due to the elasticity of the underlying membrane, small ‘filaments’ remain in the corners and around the edges of the orientation feature.

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4.3. SAMPLE FABRICATION

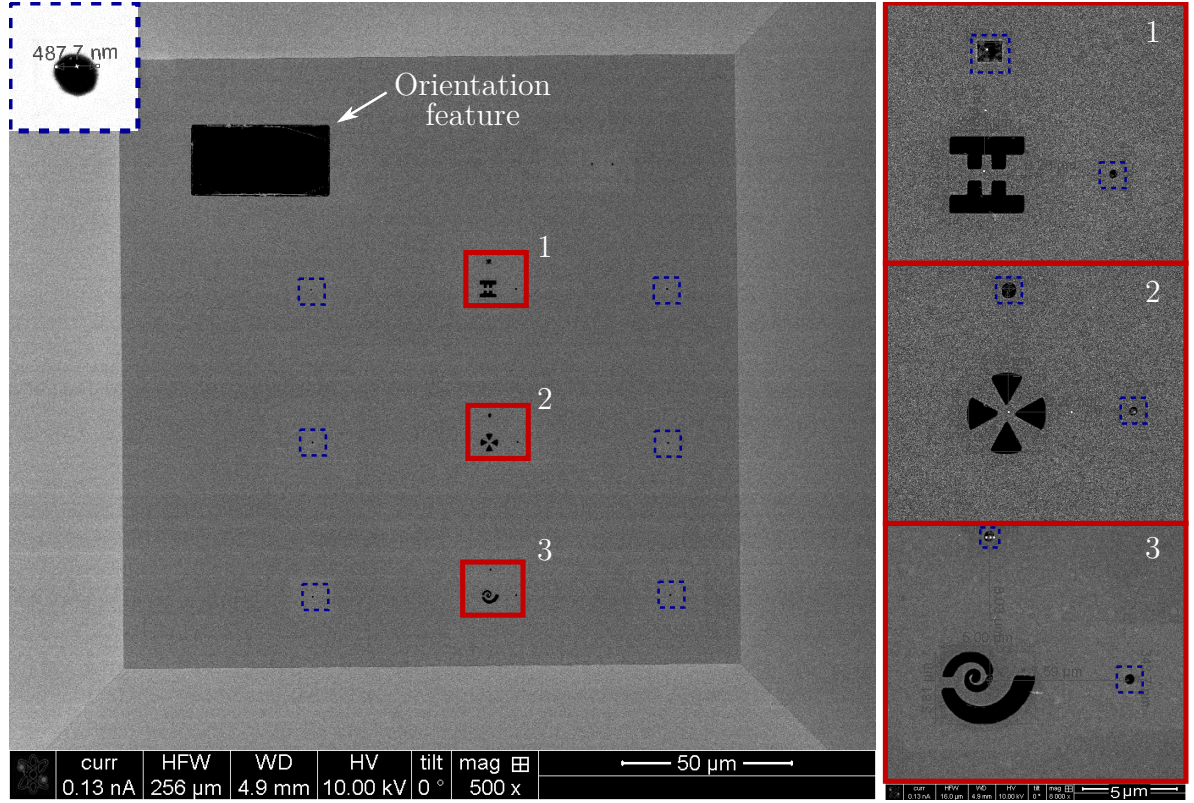


Figure 4.5: An electron beam image of a sample containing three apertures labelled 1, 2 and 3 along with their accompanying references. Each blue box outlines a holographic reference feature, a representative high contrast view of which is shown in the upper left inset. A zoomed view of each of the apertures within the labelled red boxes is shown on the right hand side. Note the 50 μm and 5 μm scale bars.

The inset image in the upper left of figure 4.5 is a high contrast image of one of the holography reference pinholes, showing that no substrate remains within the reference.

The substrate acts as a supportive layer for the Au. The materials and their thicknesses were chosen for their structural integrity and opacity at 32 nm. A 50 nm thick layer of Si_3N_4 and 125 nm thick layer of Au transmits 12.8% and $7.1 \times 10^{-6}\%$ of the 32 nm imaging radiation respectively [150], although ideally, no Si_3N_4 would remain in within the aperture(s) once milling was complete. During the milling process, the FIB is scanned across the area to be removed, spending 1 μs at each constituent pixel. Once complete, the 5 mm square supporting wafer is mounted on the SmarAct XYZ stage

and inserted into the harmonic focus.

The grain size, or minimum characteristic particulate size, in a metallic coating deposited by thermal evaporation is proportional to the deposition time and inversely proportional to the particulate energy upon deposition. Optimising these deposition parameters enables the minimisation of the grain size. When milling a pattern with a [FIB](#), the highest resolution is attainable in materials with a grain size comparable to the diameter of the [FIB](#) focus (≈ 10 nm). The grain size of gold produced in the evaporation deposition process was approximately 15–20 nm in diameter. Under optimal deposition conditions, the grain size of platinum can be 10 nm or less, enabling higher resolution patterns to be milled with the [FIB](#). However, all attempts at milling through platinum coated substrates failed due to substrate collapsing from within the supporting silicon wafer. Subsequently, all samples within this thesis were constructed from gold coated substrates.

4.4 Spatial Light Modulation

The term [SLM](#) describes any device that spatially modulates (shapes) the phase and or intensity of light. Intensity modulation involves the spatially structured loss of energy in the radiation travelling through the modulator. Although energy inefficient, the ease with which this technique is implemented has resulted in its widespread use in many optical systems such as the very earliest projectors (Kircher’s ‘steganographic mirror’ [[152](#)] constructed in 1645) and photographic film [[153](#)].

Phase modulation involves the introduction of a spatially structured phase delay into the radiation travelling through the modulator. This approach is more energy efficient and enables the user to imprint an arbitrary phase onto the illumination in a highly flexible manner.

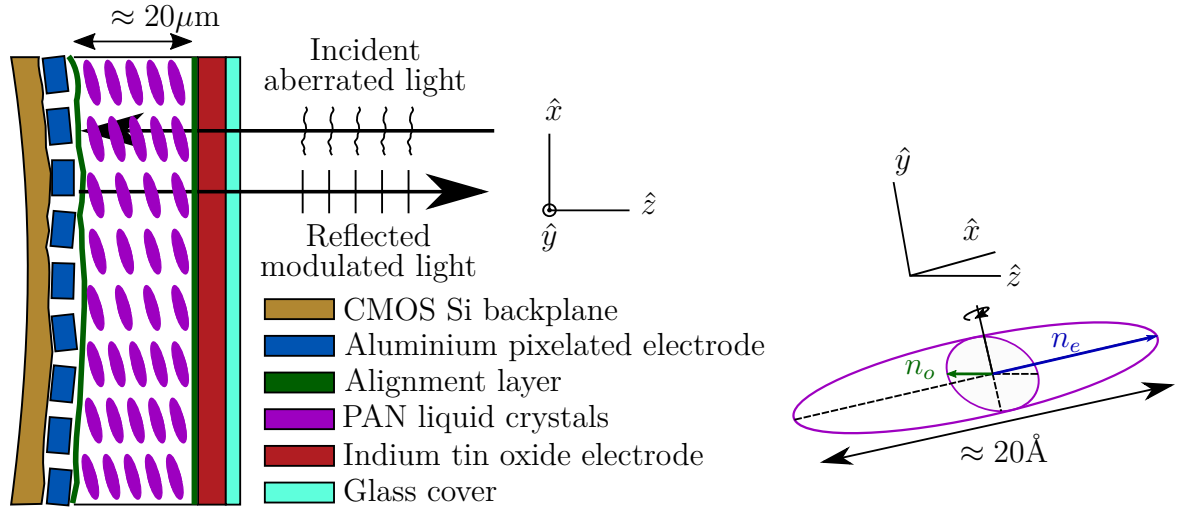


Figure 4.6: Illustration of the main components of a reflective LCoS SLM accompanied by a schematic of an individual liquid crystal with the ordinary n_o and extraordinary n_e axes labelled.

Phase-only SLM operating principle

A schematic diagram of a reflective phase-only SLM is shown on the left hand side of figure 4.6. The illumination impinging upon the SLM passes without attenuation through a layer of liquid crystals (LCs) held between two electrodes. The illumination then reflects from the silicon backplane, transitioning through the LCs a second time before exiting the device. A schematic diagram of a single liquid crystal is shown on the right hand side of figure 4.6. In this thesis, the LCs contained within the SLM are parallel aligned nematic (PAN). This type of liquid crystal is uniaxially birefringent, meaning that there is one preferred axis that dictates the anisotropic optical response known as the extraordinary axis, orthogonal to the ordinary axis. Incident illumination polarised with the electric field coincident with the extraordinary axis of the LCs experiences a refractive index n_e . By exploiting the electrically controlled birefringence [154] of the PAN LCs, their angle of rotation in the $x - z$ plane and thus n_e , can be manipulated by the potential difference V between the electrodes. In figure 4.6, the LCs are constrained to rotate in the $x - z$ plane by virtue of the alignment layers and the liquid crystal type. This means that the ordinary refractive index n_o remains constant regardless of the

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potential difference between the electrodes.

There are a range of different kinds of LCs, each of which exhibit certain self ordering properties. The common types are; nematic, smectic and cholesteric. Nematic LCs exhibit only directional ordering, smectic LCs are both directionally and positionally ordered, whilst cholesteric, are directionally ordered but helically twisted from layer to layer [155–158]. In the case of a phase-only SLM, PAN LCs are commonly used because they do not alter the incident polarization [159] and their lack of positional ordering prevents them from stratifying into layers with high and low densities of LCs. The two alignment layers either side of the LCs act to reduce the disruption in self ordering of the crystals at a liquid solid interface[160].

The rear electrode of the SLM is discretised into a two dimensional array, each element of which can support a different voltage V . Consequently, the phase delay that can be imprinted onto the incident beam is encoded by a two dimensional grid of voltages that can be updated at a rate limited only by the ~ 10 millisecond switching time of the device. The desired array of phase delays is typically specified by the user as a bitmap of a certain bitdepth dependent on the device control unit. The values within the bitmap are interpreted by a control unit that then ‘displays’ the appropriate voltage distribution upon the SLM. The minimum size of the pixels is set by the crosstalk between them: Below a certain size the electric field of one pixel starts to interfere with the field of the adjacent pixel [161]. This will cause a smoothing of the design phase features such that discontinuities are insufficiently represented.

As a result of the pixelated nature of the electrodes, the fill factor of the SLM used in this thesis was 98%. Consequently, upon illumination, the SLM chip acts as a two dimensional diffraction grating creating an array of diffracted orders in the focal plane. The diffractive efficiency of the device is then quantified as the ratio of energy that

exists within the zeroth order and the incident illumination.

Phase-only SLM calibration

During the construction of a LCoS SLM the silicon backplane becomes warped which forces the LCs into a layer of non-uniform thickness. [162, 163]. As the phase delay suffered by the beam is proportional to the thickness of the LCs through which it propagates, the warping introduces a phase variation across the device, even with no applied voltage. This variation can be mitigated by displaying a calibration phase mask that modifies the voltages applied to the electrode to ensure a planar phase response is elicited from the device at a particular design wavelength. The calibration masks are calculated by iteratively adjusting a seed mask whilst optimising the interferometrically detected wavefront. The particular calibration mask used in this thesis for $\lambda_0 = 800\text{nm}$ is shown in figure 4.7.

Once the calibration mask is applied, the SLM acts a planar mirror. However, the phase modulation of the device is not necessarily linear with the applied voltage. In practise, the voltage is expressed as an integer with a bit length that specifies the achievable

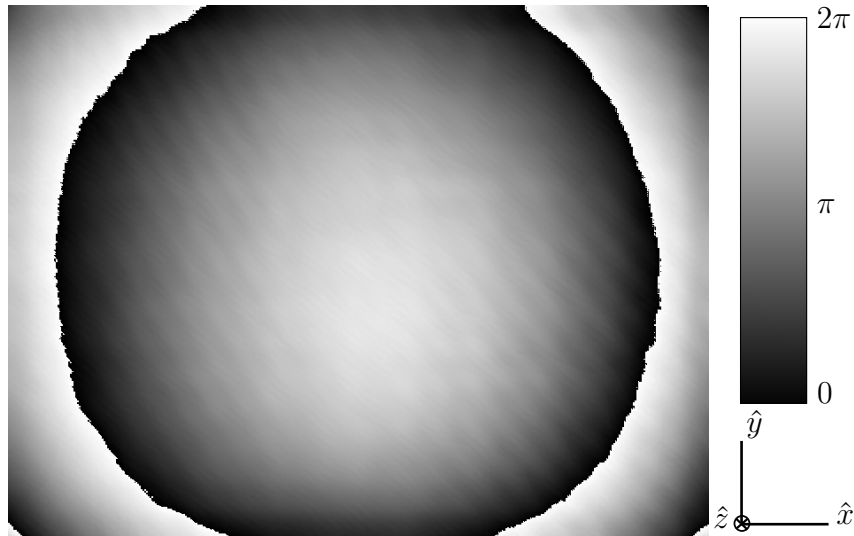


Figure 4.7: Calibration mask used for $\lambda_0 = 800\text{ nm}$ illumination of the SLM. The roughly circular discontinuity is a phase wrap where phase delays of 0 and 2π radians are equivalent.

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phase resolution. In order to ensure that the phase delay is linear with voltage, a manufacturer supplied look-up-table is typically provided. This table comprises a set of calibration figures that adjusts the voltages corresponding to the user's desired phase delay to voltages that provide that delay having accounting for any non-linearity. The look-up-tables are calculated by a procedure outlined in section 4.4.3.

The aforementioned considerations all assume monochromatic illumination. SLMs are produced for a specific design wavelength based on the coating applied to the reflective components. However, the bandwidth of the laser pulses used in this thesis was approximately 55 nm centred on the central wavelength of 800 nm. Any phase mask created for an incident wavelength of exactly 800 nm will therefore be sub-optimal at the extrema of the incident bandwidth. This issue is particularly prevalent when the SLM deflects the illumination away from the optical axis. This adds spatial chirp into the already temporally chirped beam resulting in a reduced peak intensity at focus. Analytical treatment of this effect along with other space time couplings have been developed in references [164–166], although these are outside the remit of this thesis.

With respect to the use of ultrafast laser pulses, a significant drawback of a LCoS SLM is that of the limited damage threshold. As the thermal load experienced by the LCs increases, their temperature increases leading to a reduction in their birefringence. In extreme cases they will boil, undergoing an irreversible chemical change although this issue can be eased with liquid cooling. Further, the LCs can break down when exposed to excessive ultraviolet light. This issue is compounded by the increasing absorption coefficient of LCs at wavelengths below 350 nm. Recent developments of SLMs with components constructed from new materials provide hope that higher damage threshold SLMs may soon become available [167]. However, regrettably, device damage thresholds are often quoted for pulse lengths down to only nanosecond durations where extrapolation to femtosecond durations is prevented by the emergence of different, less

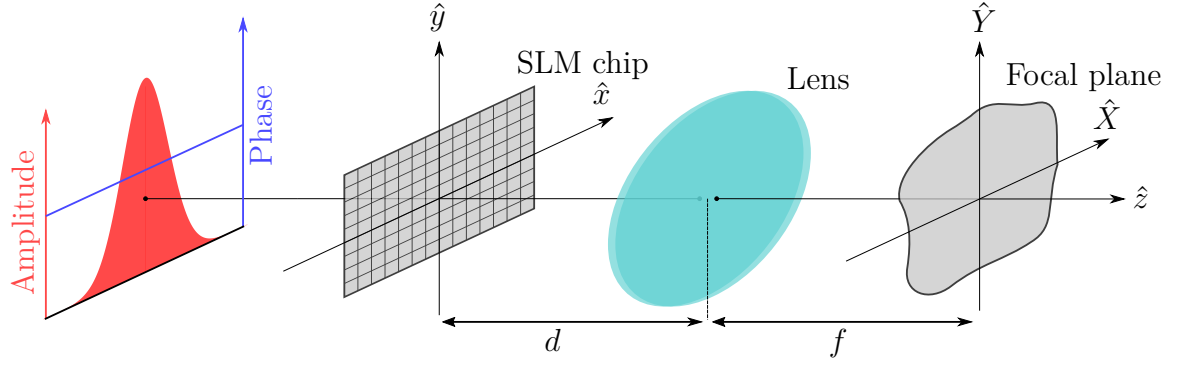


Figure 4.8: A schematic diagram showing a collimated Gaussian beam impinging upon a SLM chip. A distance d separates the SLM chip from a lens with a corresponding focal plane a distance f farther downstream.

understood, damage mechanisms.

4.4.1 Experimental Geometry

The experimental geometry relating the illuminating beam, SLM, lens and the focal plane of the lens is shown in figure 4.8. The beam impinging upon the SLM is assumed to comprise a Gaussian amplitude $|f(x, y)|$ and a collimated (constant) phase $\psi(x, y)$. This field is described by

$$f(x, y) = |f(x, y)|e^{i\psi(x, y)}. \quad (4.2)$$

The SLM chip imprints a phase upon the incident beam equivalent to a complex transmission function denoted $T_{\text{slm}}(x, y)$, such that the field leaving the SLM chip is $f(x, y) \cdot T_{\text{slm}}(x, y)$. This field propagates a distance d downstream until it encounters a lens of focal length f with a transmission function [13] given by

$$T_{\text{lens}}(x, y) = \exp \left[\frac{-ik}{2f} (x^2 + y^2) \right], \quad (4.3)$$

The field is then focussed into the focal plane a distance f farther downstream.

If the distance d is vanishingly small, then the associated propagation can be neglected,

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in which case the field leaving the lens can be written as

$$f'(x, y) = f(x, y) \cdot T_{\text{slm}}(x, y) \cdot T_{\text{lens}}(x, y), \quad (4.4)$$

The field at the focus of the lens can now be found by using the Fresnel diffraction formula from equation 2.22, repeated here in equation 4.5a. Explicitly substituting equations 4.4 and 4.3 into equation 4.5a yields equation 4.5b as follows

$$U(X, Y) = \frac{e^{\left[\frac{ik}{2z}(X^2+Y^2)\right]}}{i\lambda z} \iint_{-\infty}^{\infty} f'(x, y) \exp\left[\frac{ik}{2z}(x^2 + y^2)\right] \exp\left[-i\frac{2\pi}{\lambda z}(Xx + Yy)\right] dx dy \quad (4.5a)$$

$$U(X, Y) = \frac{e^{\left[\frac{ik}{2f}(X^2+Y^2)\right]}}{i\lambda f} \iint_{-\infty}^{\infty} f(x, y) T_{\text{slm}}(x, y) \exp\left[-i\frac{2\pi}{\lambda f}(Xx + Yy)\right] dx dy. \quad (4.5b)$$

In moving from equation 4.5a to 4.5b we have observed that $z = f$ and therefore the quadratic phase within the integral, and that of the T_{lens} have cancelled. Equation 4.5b consequently corresponds to an expression for the field at the focus of the lens proportional a Fourier transform of field leaving the SLM. The prefactor comprising a field depended quadratic phase prevents this from being an exact Fourier transform relationship.

When the distance d is not negligible however, an additional step is required in the above analysis to account for the free space propagation of the field between the SLM and the lens. It is now mathematically convenient to express this analysis in terms of spatial frequencies $\hat{f}(f_X, f_Y) = \text{FT}[f(x, y)]$, in which case the free space propagator, repeated here from equation 2.21, can be written as

$$h(X, Y, z) = \frac{e^{ikz}}{i\lambda z} \exp\left[\frac{ik}{2z}(X^2 + Y^2)\right], \text{ and} \quad (4.6a)$$

$$\hat{h}(f_X, f_Y, z) = e^{ikz} \exp\left[-i\pi\lambda z(f_X^2 + f_Y^2)\right]. \quad (4.6b)$$

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Consequently, the field arriving at the lens after propagating a distance d can be written as

$$\hat{f}'(f_x, f_y) = \text{FT} [f(x, y) \cdot T_{\text{slm}}(x, y)] \cdot \hat{h}(f_X, f_Y, d). \quad (4.7)$$

In order to find the corresponding field in the focal plane, equation 4.5b can be rewritten as

$$U(X, Y) = \frac{e^{\left[\frac{ik}{2f}(X^2+Y^2)\right]}}{i\lambda f} \cdot \hat{f}'(f_x, f_y). \quad (4.8)$$

Now substituting equation 4.7 into equation 4.8 we find

$$\begin{aligned} U(X, Y) &= \frac{e^{\left[\frac{ik}{2f}(X^2+Y^2)\right]} e^{ikd} e^{[-i\pi\lambda d(f_X^2+f_Y^2)]}}{i\lambda f} \\ &\times \iint_{-\infty}^{\infty} f(x, y) T_{\text{slm}}(x, y) \exp \left[-i\frac{2\pi}{\lambda f} (Xx + Yy) \right] dx dy. \end{aligned} \quad (4.9)$$

When the prefactor is evaluated at spatial frequencies of $(f_X, f_Y) = (\frac{X}{\lambda f}, \frac{Y}{\lambda f})$, a simplification can be made that reveals

$$\begin{aligned} U(X, Y) &= \frac{e^{ikd}}{i\lambda f} \exp \left[\frac{ik}{2f} \left(1 - \frac{d}{f} \right) (X^2 + Y^2) \right] \\ &\times \iint_{-\infty}^{\infty} f(x, y) T_{\text{slm}}(x, y) \exp \left[-i\frac{2\pi}{\lambda f} (Xx + Yy) \right] dx dy. \end{aligned} \quad (4.10)$$

From equation 4.10 it is clear that when $d = f$, the prefactor to the integral comprises only a constant phase factor and there is no field dependent quadratic phase. Ergo, the focal plane of the lens is now an exact Fourier transform of the field leaving the SLM chip. Taking $d = f$, equation 4.10 can be inverted to find the form of the SLM transmission function $T_{\text{slm}}(x, y)$ that generates the complex field $U(X, Y)$ at focus when illuminated by an incident field $f(x, y)$:

$$T_{\text{slm}}(x, y) = \frac{i\lambda f}{f(x, y)} e^{-ikd} \text{FT}^{-1} [U(X, Y)]. \quad (4.11)$$

The user is free to specify the desired form of $U(X, Y)$ which entirely specifies the complex transmission function $T_{\text{slm}}(x, y)$ according to equation 4.11. As a result of the above analysis, in all experiments presented in this thesis, a 2-f relationship was maintained between the SLM, the 50 cm focal lens and the focal plane. This simplifies the calculations for finding the appropriate phase masks that elicits the desired response.

Generally speaking, the form of $T_{\text{slm}}(x, y)$ may include both amplitude and phase modulation. We will now consider the behaviour of a phase-only SLM with respect to the modulation of phase, intensity and both phase and intensity.

4.4.2 Phase-only Modulation

Phase-only modulation of incident illumination can be achieved with a LCoS SLM in an energy efficient manner without altering the polarisation. The mathematical description of this is now outlined.

For a liquid crystal layer thickness L , the phase delay imparted to a beam of design wavelength λ_0 with an electric field polarised parallel to the extraordinary axis of the LCs is

$$\delta(V) = \frac{2\pi}{\lambda_0} 2L n_e(V), \quad (4.12)$$

where the extraordinary refractive index has been written as an explicit function of applied voltage V . The birefringence $\Delta n(V) = n_e(V) - n_o$ describes the relative phase delay between the extraordinary and ordinary waves. Accordingly, when $n_e(V) = n_o$, no birefringence is present and the SLM is said to act as a planar mirror. Henceforth, the extraordinary axis of the LCs and hence ‘working direction’ of the SLM is parallel to $\hat{x}$ unless otherwise specified.

The Jones formalism [168] enables the field incident to the SLM to be considered

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as a linear combination of horizontal and vertical components with amplitudes E_x and E_y respectively:

$$\mathbf{E}^{\text{in}} = \begin{bmatrix} E_x^{\text{in}} \\ E_y^{\text{in}} \end{bmatrix}, \quad (4.13)$$

where for a field linearly polarised in the $\hat{x}$ dimension, $E_y^{\text{in}} = 0$. The Jones matrix for a phase-only **PAN SLM** can be written using equation 4.12 as

$$T_{\text{SLM}} = \begin{bmatrix} e^{ikLn_e} & 0 \\ 0 & e^{ikLn_o} \end{bmatrix} = e^{ikLn_o} \begin{bmatrix} e^{ikL\Delta n} & 0 \\ 0 & 1 \end{bmatrix}, \quad (4.14)$$

where the voltage dependence of the birefringence Δn has been dropped for brevity. This enables the field leaving the **SLM** to be expressed as

$$\mathbf{E}^{\text{out}} = \begin{bmatrix} E_x^{\text{out}} \\ E_y^{\text{out}} \end{bmatrix} = T_{\text{SLM}} \mathbf{E}^{\text{in}} = e^{ikLn_o} \begin{bmatrix} e^{ikL\Delta n} & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} E_x^{\text{in}} \\ E_y^{\text{in}} \end{bmatrix} = e^{ikLn_o} (E_x^{\text{in}} e^{ikL\Delta n} + E_y^{\text{in}}). \quad (4.15)$$

The intensity of the output field in equation 4.15 is

$$I = \mathbf{E}^\dagger \mathbf{E} \quad (4.16a)$$

$$= (E_x^{\text{in}})^2 + (E_y^{\text{in}})^2 = I_{\text{in}}, \quad (4.16b)$$

where $\dagger$ denotes the complex transpose. Clearly, the incident polarization remains unchanged if $E_y^{\text{in}} = 0$ or $E_x^{\text{in}} = 0$ and the energy is conserved.

4.4.3 Intensity-only Modulation

A phase-only **SLM** can also be used to modulate the intensity of incident illumination. This is achieved by manipulating the polarisation in combination with a half-wave plate and a polariser as described below.

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When both E_y^{in} and $E_x^{\text{in}} \neq 0$, then a fixed phase delay is imparted into the beam in the $\hat{y}$ polarisation component parallel to the ordinary axis of the [LCs](#), whilst the $\hat{x}$ component receives the voltage dependent phase shift as described above. Ergo, the [SLM](#) now manipulates the output polarisation state.

Consider the case of an incident beam linearly polarised in $\hat{x}$. This beam travels through a half-wave plate with a fast axis at an angle of 22.5° to $\hat{x}$ such that the outbound linear polarisation is at an angle of 45° to both $\hat{x}$ and $\hat{y}$. This field is reflected from a [SLM](#) and subsequently directed through a linear polariser with a transmission axis parallel to the polarisation incident upon the [SLM](#). Mathematically this can be represented as

$$\mathbf{E}^{\text{out}} = T_{\text{Pol}}^{+45^\circ} T_{\text{SLM}} T_{\text{HPW}}(+22.5^\circ) \mathbf{E}^{\text{in}}, \quad (4.17)$$

where the Jones matrix of a half-wave plate that rotates the axis of polarisation by 2θ from $\hat{x}$ is

$$T_{\text{HPW}}(\theta) = \begin{bmatrix} \cos^2(\theta) - \sin^2(\theta) & 2 \cos(\theta) \sin(\theta) \\ 2 \cos(\theta) \sin(\theta) & \sin^2(\theta) - \cos^2(\theta) \end{bmatrix}, \quad (4.18)$$

and the Jones matrix of a linear polariser with an axis of transmission at $\pm 45^\circ$ to $\hat{x}$ is

$$T_{\text{Pol}}^{\pm 45^\circ} = \frac{1}{2} \begin{bmatrix} 1 & \pm 1 \\ \pm 1 & 1 \end{bmatrix}. \quad (4.19)$$

Hence, taking $kL\Delta n = \delta$ as the phase value displayed on the [SLM](#), the output field is written as

$$\mathbf{E}^{\text{out}} = \frac{e^{ikLn_o}}{2\sqrt{2}} \begin{bmatrix} 1 & 1 \\ 1 & 1 \end{bmatrix} \begin{bmatrix} e^{i\delta} & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 1 & 1 \\ 1 & -1 \end{bmatrix} \begin{bmatrix} 1 \\ 0 \end{bmatrix} = \frac{e^{ikLn_o}}{2\sqrt{2}} \begin{bmatrix} e^{i\delta} + 1 \\ e^{i\delta} + 1 \end{bmatrix}, \quad (4.20)$$

where the outbound intensity found through equation 4.16a is

$$I = \frac{1}{2} [1 + \cos(\delta)] , \quad (4.21)$$

Now the transverse intensity profile of the output beam depends on the phase displayed on the SLM. By measuring $I(\delta)$ a look-up-table can be formed which enables the linearity of the phase response to be accurately calibrated.

4.4.4 Intensity and Phase Modulation

Ideally, both the amplitude and phase of the illumination leaving the SLM could be specified. This would allow the complex field to be specified at any transverse plane downstream of the SLM. Three methods for achieving such a level of control will now be outlined.

Intensity modulating SLM reimaged onto a phase modulating SLM

One method of achieving this control, described by Zhu et al. in reference [169], makes use of two phase-only SLMs. The first is used to modulate the intensity of the incident illumination as described in section 4.4.3, whilst the second is used to modulate only the phase as describe in section 4.4.2. A 4-f relationship is established between the two SLM chips such that the first is reimaged onto the second. The result is the full specification of the complex field in the transverse plane coincident with the second SLM chip.

In order to produce a complex field composed of a user specified amplitude $A(x, y)$ and phase $B(x, y)$ leaving the second SLM, reference [169] gives the first and second SLM

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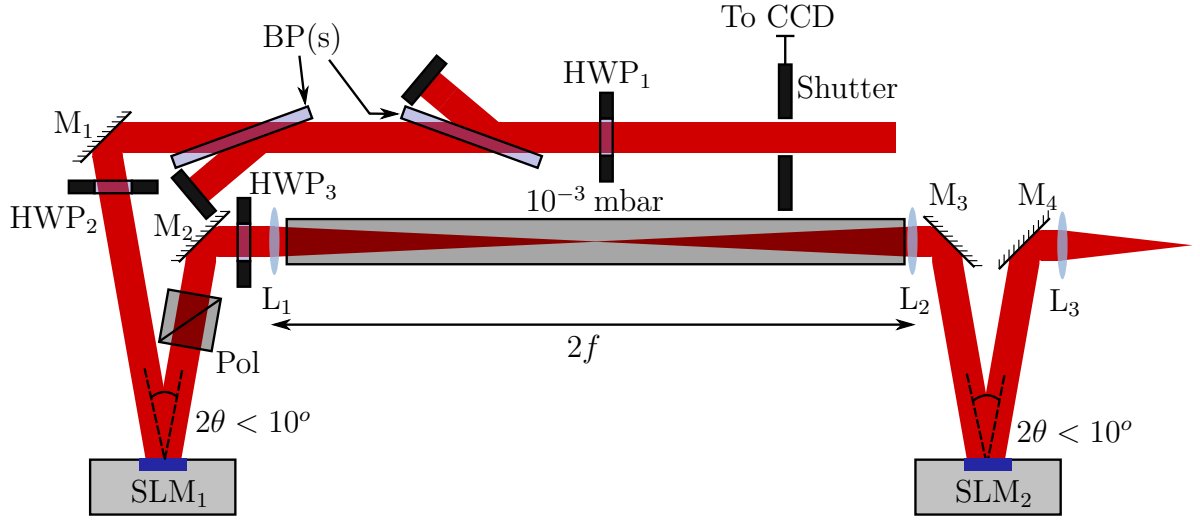


Figure 4.9: A schematic diagram showing the beamline constructed by the author that employed two SLMs to specify both the intensity and phase of the field leaving the second SLM. Pol represents a polariser that was oriented at 45° to the x axis.

phase distributions as

$$\delta_1(x, y) = 2 \cos^{-1} [A(x, y)] \quad (4.22a)$$

$$\delta_2(x, y) = B(x, y) - \frac{1}{2} \delta_1(x, y) \quad (4.22b)$$

respectively. This approach was implemented by the author with the experimental setup schematically illustrated in figure 4.9. The combination of HWP₂, SLM₁ and Pol modulated the intensity of the illumination through the method detailed in 4.4.3. HWP₃ rotated the polarisation back into the working direction of the LCs, which enabled SLM₂ to act as a phase-only modulator. The four 30 cm focal lengths that comprised the 4-f reimaging system were located between SLM₁ and L₁, L₁ and its back focal plane, L₂ and its front focal plane, and L₂ and SLM₂. A vacuum pipe represented by the shaded grey rectangle was installed between L₁ and L₂ to prevent ionisation at the focus between L₁ and L₂ from distorting the spectral phase. This pipe was held at 10^{-3} mbar during operation.

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A subtlety of using a phase-only SLM as an intensity modulator is the applicability of the calibration mask. When the incident polarisation comprises components parallel and perpendicular to the working direction of the SLM, the calibration mask will imprint the corrective phase only onto the parallel component, resulting in an inhomogeneous polarisation state. Ergo, the calibration mask is no longer applicable, and instead must be implemented in a conjugate plane when the incident polarisation is entirely parallel to the rotation plane of the LCs. In figure 4.9, the 4-f relationship between the two SLM chips ensured that the field was identical in both SLM planes. This enabled the calibration masks for both SLM₁ and SLM₂ to be displayed on SLM₂, after HWP₃ had rotated the polarisation back into the rotation plane of the LCs.

By specifying the desired field at the focus of L₃, the field required as an input to L₃ and thus the field required in the plane of SLM₂ could be calculated. However, in all experiments presented within this thesis, the phase mask produced by equation 4.22a exhibited a typical energy efficiency of between approximately 1% and 5%. Consequently, this approach was not pursued by the author.

Two SLMs acting as aspheric lenses

An alternative method that achieves full field control is described by Hoffnagle et al. in reference [170], based on a patent belonging to Kreuzer described in reference [171]. Two phase-only SLMs are used as a pair of programmable aspheric lenses where the first SLM redistributes the energy onto the second SLM, which then restores a planar phase to the complex field. The first and second aspheric lens surfaces are described by

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equations 4.23a and 4.23b

$$z(r) = \int_0^r \left\{ (n^2 - 1) + \left[\frac{(n-1)d}{h(x) - x} \right]^2 \right\}^{-0.5} dx \quad (4.23a)$$

$$Z(R) = \int_0^R \left\{ (n^2 - 1) + \left[\frac{(n-1)d}{h^{-1}(x) - x} \right]^2 \right\}^{-0.5} dx, \quad (4.23b)$$

where r and R are the transverse radii at which the incident ray enters the first lens and exits the second lens, n is the refractive index and d is the distance between the two lens surfaces. $h(x)$ is known as the ray-tracing function and describes the relationship between the radii r and R as $R = h(r)$.

The form of $h(r)$ is found by solving the statement of energy conservation relating the input intensity $f(x)$ and the desired output intensity $g(x)$ from the optical system:

$$\int_0^r f(x)x dx = \int_0^R g(x)x dx. \quad (4.24)$$

The phase distributions corresponding to $z(r)$ and $Z(R)$ can then be displayed on the SLMs to generate $g(x)$ with a flat phase in the plane of the second SLM chip. This method was not pursued by the author as experimentally simpler alternatives based on a single phase-only SLM alluded to in chapters 7 and 6 produced the required focal driver intensity profiles adequately.

One SLM controlling both intensity and phase within a RoI

Liu et al. in reference [172] describe a method to achieve a certain degree of control over both the intensity and phase of the field at the focus of a Fourier transforming lens if only a single SLM is available. In this method, the shaping domain at the focus of the lens is decomposed into an on-axis RoI denoted by S_p and an off-axis region that can be ignored, denoted by S_q . A transmission function T is displayed upon the SLM

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that comprises a high frequency phase modulation that deflects a structured portion of the incident energy away from the [RoI](#).

Within S_p , the target beam profile is $U_p(X, Y)$, whilst the remaining (neglected) profile $U_q(X, Y)$ exists within S_q . The complex field leaving the [SLM](#) plane is $u(x, y) = |u_0(x, y)|e^{i\psi} T$ composed of an incident amplitude $u_0(x, y)$ and phase ψ . This can be expressed as the inverse transforms of the two focal regions

$$u(x, y) \propto \text{FT}^{-1}[U_p(X, Y)] + \text{FT}^{-1}[U_q(X, Y)] = P(x, y) + Q(x, y). \quad (4.25)$$

Since $U_p(X, Y)$, and therefore the amplitude of $P(x, y)$, are both known and exist within the [RoI](#) close to the optical axis, the constituent spatial frequencies of $P(x, y)$ are low. Conversely, the spatial frequencies contained within $Q(x, y)$ are much higher as they correspond to the off-axis domain S_q . Consequently, $P(x, y)$ is specified by the target beam profile, leaving $Q(x, y)$ responsible for the deflection of incident energy away from the [RoI](#). This is achieved by combining $Q(x, y)$ with a high spatial frequency modulation, $\phi(x, y)$:

$$Q(x, y) = Q_0(x, y)e^{i\phi(x, y)} \quad (4.26)$$

The value of $Q_0(x, y)$ can be found by solving the equation

$$|u(x, y)|^2 = |u_0(x, y)e^{i\psi} T|^2 = |P(x, y) + Q(x, y)|^2, \quad (4.27)$$

which expands to

$$Q_0^2 + (Pe^{-i\phi} + P^*e^{i\phi})Q_0 + (PP^* - u_0^2) = 0 \quad (4.28)$$

having dropped the explicit (x, y) dependence for brevity and used $|T|^2 = 1$. Solving the above quadratic equation for Q_0 enables the full complex field leaving the [SLM](#) to

CHAPTER 4. BEAMLINE OVERVIEW

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be expressed as

$$|u_0|e^{i\psi} T = P + Q_0 e^{i\phi} = P + \left\{ -\Re [P e^{-i\phi}] \pm \sqrt{\Re [P e^{-i\phi}]^2 - (|P|^2 - |u_0|^2)} \right\} e^{i\phi}, \quad (4.29)$$

where the form of T can then be found.

The flexibility in the choice of ϕ enables the optimisation of the diffractive efficiency. In spite of this, for all experiments presented within this thesis, the required phase mask produced by equation 4.29 proved highly inefficient and this method was consequently not pursued by the author.

4.4.5 Aberration Correction

A common use for a phase-only SLM is to correct for optical aberrations within the illuminating beam [173, 174]. These aberrations are most elegantly described by a set of orthogonal polynomials defined on the unit circle with radial and azimuthal coordinates ρ and ϕ , known as Zernike polynomials [175]:

$$Z_n^m(\rho, \phi) = R_n^m(\rho) \cos(m\phi) \quad (4.30a)$$

$$Z_n^{-m}(\rho, \phi) = R_n^m(\rho) \sin(m\phi). \quad (4.30b)$$

The radial and azimuthal indices n and m are positive integers where $n \geq m$. $R_n^m(\rho)$ is the radial polynomial generating function

$$R_n^m(\rho) = \sum_{k=0}^{\frac{n-m}{2}} \frac{(-1)^k (n-k)!}{k! \left(\frac{n+m}{2} - k\right)! \left(\frac{n-m}{2} - k\right)!} \rho^{n-2k}. \quad (4.31)$$

Table 4.2 shows the first ten Zernike polynomials, normalised to an area of π over the unit circle. Of note, Z_0^0 details the absolute value of the detected phase which in the majority of applications is an inconsequential arbitrary offset. $Z_1^{\pm 1}$ are terms

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Table 4.2: Zernike polynomials

n	m	$Z_n^m(\rho, \phi)$	Aberration
0	0	1	Piston
1	-1	$2\rho \sin(\phi)$	Tilt
1	1	$2\rho \cos(\phi)$	Tip
2	-2	$\sqrt{6}\rho^2 \sin(2\phi)$	Oblique astigmatism
2	0	$\sqrt{3}(2\rho^2 - 1)$	Defocus
2	2	$\sqrt{6}\rho^2 \cos(2\phi)$	Vertical astigmatism
3	-3	$\sqrt{8}\rho^3 \sin(3\phi)$	Vertical trefoil
3	-1	$\sqrt{8}(3\rho^3 - 2\rho) \sin(\phi)$	Vertical coma
3	1	$\sqrt{8}(3\rho^3 - 2\rho) \cos(\phi)$	Horizontal coma
3	3	$\sqrt{8}\rho^3 \cos(3\phi)$	Oblique trefoil

that describe the average slope of the phase profile. $Z_2^{\pm 2}$ can be caused by an off-axis reflection from a spherical optic and results in a disparity between the longitudinal position of the focus in the vertical and horizontal dimensions. $Z_3^{\pm 1}$ is caused when a beam enters a lens at an angle to the optical axis.

As shown in figure 4.2, a beamsplitter could be inserted into the beamline downstream of the driver focus to deflect the driver beam into a 4-f unitary magnification reimaging setup. By inspecting the reimaged driver focus with a CCD, the aberrations within the beam could be estimated. Experimentally, this was achieved by scanning the CCD longitudinally and observing the evolution of the beam profile through the focal volume. Software written by the author enabled the specification of up to three Zernike polynomials each with a user defined magnitude that could be displayed upon the SLM. By adjusting these magnitudes whilst observing the beam profile on the CCD, the aberration content of the reimaged beam could be reduced. This procedure was repeated before each experimental run. However, despite careful alignment, it is possible that a component of the required aberration correction was caused by the reflection from and propagation through M₅₋₆ and L₂₋₃ respectively in figure 4.2, which would result in additional aberrations within the driver primary focus in the gas cell. Regardless, the

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aberrations most common in the produced corrective mask shown in figure 4.10 were Z_2^2 , Z_2^{-2} and Z_3^{-1} .

Ideally, the unobscured harmonic beam detected on the x-ray CCD would be used to best minimise the aberrations within the primary focus of the driver. However, as the harmonic beam is reflected from a curved focussing mirror at a non-zero angle of incidence, astigmatism is unavoidably imprinted into the harmonic beam between the generation and detection planes. It is theoretically possible to pre-correct this aberration by introducing the same astigmatism of opposite sign and appropriate magnitude into the driver beam prior to the HHG interaction. However, the extreme non-linearity of the HHG process makes the direct control of the harmonic wavefront complex [176]. Additionally, the deliberate introduction of aberrations into the driver primary focus can prevent the desired driver focal spatial distribution from being achieved. For these reasons this was not attempted.

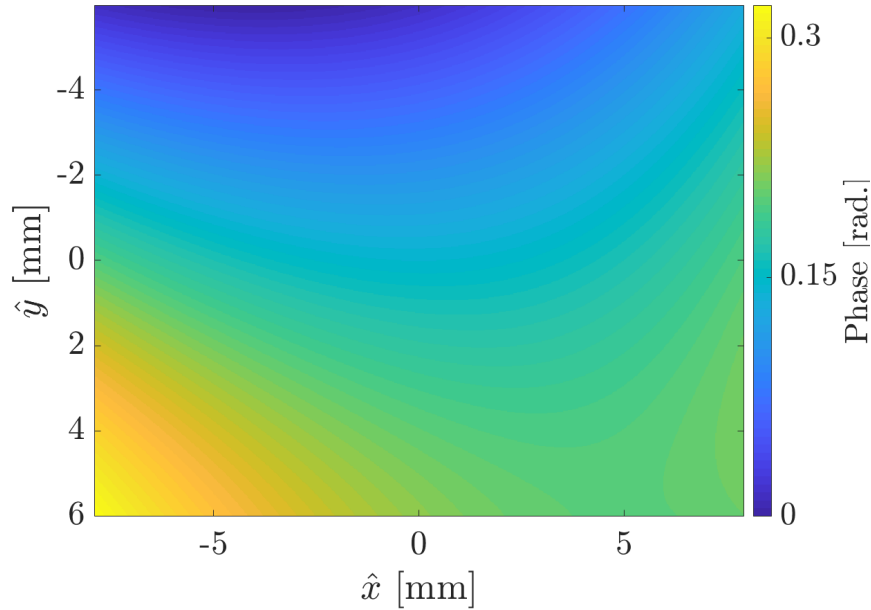


Figure 4.10: An example of a mask displayed on the SLM in order to correct the aberrations intrinsic to the driver beam. The constituent polynomials used are Z_2^2 , Z_2^{-2} and Z_3^{-1} with empirically determined magnitudes of 0.4, -0.3 and -0.1 respectively.

Chapter 5

Early Imaging Results

5.1 Coherent Diffractive Imaging



CHRONOLOGICALLY CDI was the first lensless imaging technique performed by the author. A schematic diagram of the beamline used at that time is shown in figure 5.1. The two differences between this beamline and that used to gather the results shown in chapters 6 and 7 is the use of a dielectric mirror in place of the SLM and the absence of a reimaging line in the harmonic beampath. The radiation illuminating the object was 32 nm in wavelength and the pixel size of the detector was 13.5 μm . More detail can be found in section 4.2.

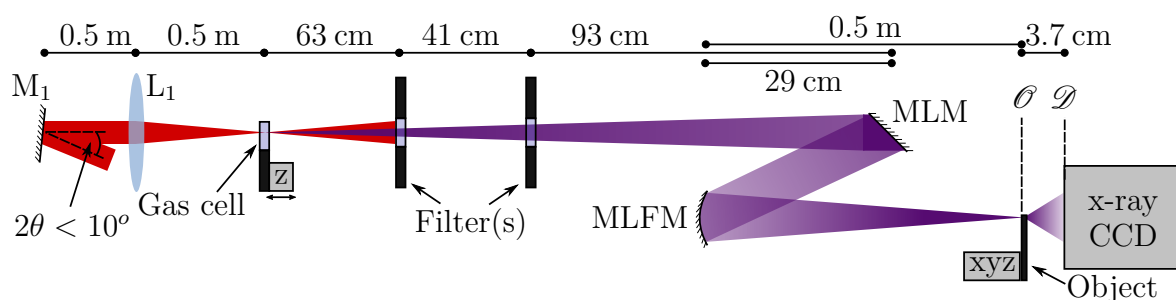


Figure 5.1: A schematic diagram of the beamline used to gather the early imaging results presented in this chapter. A summary of the components was presented in section 4.2.

5.1.1 Object

The object, shown in figure 5.2 a), comprised a triangular obstruction of side length $3.4\text{ }\mu\text{m}$ held within a transmissive circle of diameter $7\text{ }\mu\text{m}$, constructed by the author using a FIB. The first attempt at fabricating this object involved milling through only the opaque gold coating, leaving the transmissive substrate untouched as shown in figure 5.2 b). However the FIB milling process did not remove the gold coating uniformly, but instead resulted in the inhomogeneous clumps of gold as seen in the electron micrograph. As the milling process continued, the remaining gold clumps decreased in size, but not before the more fragile substrate ruptured, twisting the triangular obstruction out of the substrate plane as seen in figure 5.2 c). Consequently, it was decided to incorporate supporting arms of width $\sim 500\text{ nm}$ at the corners of the triangular obstruction as shown in figure 5.2 a), whilst intentionally milling through both the remaining coating and substrate. Both figures 5.2 b) and 5.2 c) were taken from an angle of 52° to the normal of the plane of the object which accounts for the elliptical appearance of the circular feature.

The elasticity of the substrate resulted in small ‘filaments’ of silicon nitride persisting in the corners and around the edges of the main triangular feature in figure 5.2 a). Their approximate widths are in the range of $100 - 150\text{ nm}$.

5.1.2 Data

Figure 5.3 a) shows the calculated intensity of the diffraction pattern which would be produced by uniform illumination of the object shown in the inset of figure 5.2 a) with 32 nm radiation and a propagation distance of 3.7 cm to the discrete $\mathcal{D}$ domain comprising a grid of 2048×2048 square pixels of width $13.5\text{ }\mu\text{m}$. This corresponds to an oversampling ratio of 12.5 according to equation 2.46, having taken the diameter of the feature in figure 5.2a) to be $7\text{ }\mu\text{m}$. Geometrically, the numerical aperture of this experiment was 0.34 corresponding to a diffraction limited theoretical resolution

CHAPTER 5. EARLY IMAGING RESULTS

5.1. COHERENT DIFFRACTIVE IMAGING

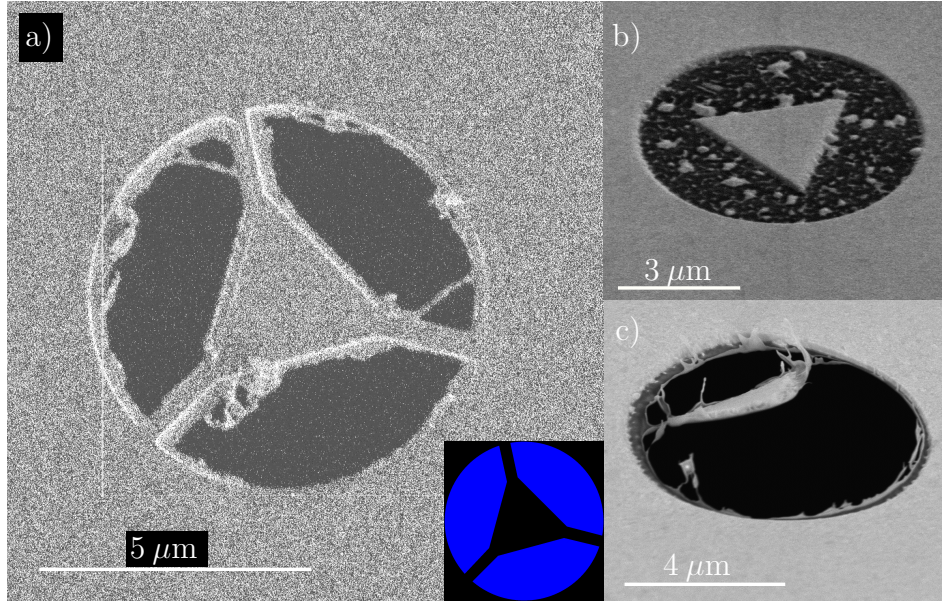


Figure 5.2: a) An ion beam image of the first object imaged in this work with CDI. The faint vertical dashed line is a distance measure performed in the focussed ion beam software. Light grey signifies the gold coating and black is empty space. The lower right inset is the milling pattern read by the FIB. b) Electron beam image of the partially complete first attempt at fabrication. Light grey signifies the gold coating and black is the silicon nitride substrate. c) Electron beam image of the object shown in b) once the substrate had ruptured. Light grey signifies the gold coating and black is empty space.

of 47.3 nm. The spatial frequency axes with dimensions of inverse length have been calculated according to equation 5.1 [75]:

$$q = \frac{2 \sin(\frac{\theta}{2})}{\lambda}. \quad (5.1)$$

Figure 5.3 b) shows a HDR [177] image comprising 5 experimentally observed diffraction patterns with exposure times of 0.5, 1.0, 1.5 and 2.0 seconds at a count rate of approximately 35-45 thousand per second. The images were combined using the method described in reference [75]. This was required to address the limited dynamic range of the 16-bit detector: The on-axis signal reached the saturation limit of the detector before an appreciable amount of off-axis signal had been collected. The disparities between the simulated and observed diffracted intensities in figure 5.3 are largely relegated to the regions of high spatial frequency, corresponding to the fine structure of the object itself.

CHAPTER 5. EARLY IMAGING RESULTS

5.1. COHERENT DIFFRACTIVE IMAGING

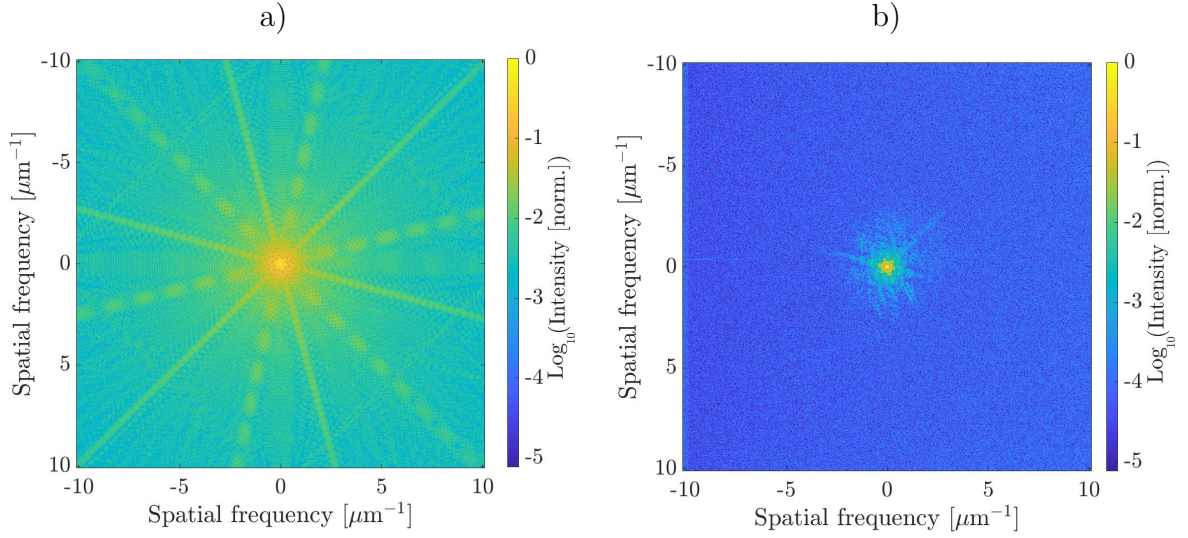


Figure 5.3: a) Simulated intensity corresponding to the illumination of the design aperture shown in the inset of figure 5.2 a), presented on a log scale. b) Experimentally observed intensity corresponding to the illumination of the aperture shown in figure 5.2 a), presented on a log scale.

Even with HDR, the signal levels in these regions are still relatively low as the longest possible exposure time was limited by the damage threshold of the detector. For the CCD used to acquire these data, the chip had a damage threshold of approximately 4 times the saturation limit. More detail on the CCD chips used in this thesis is given in section 4.2.

5.1.3 Reconstruction

The CDI algorithm described in section 2.3.1 was applied to the data shown in figure 5.3 b). The IPRA consisted of 1000 iterations of the HIO constraint described by equation 2.54 with a β parameter of 0.95. The shrinkwrapping technique was applied every 10 iterations to drive the support size closer to the true object size. The intensity of the recovered ESW is shown in figure 5.4 a), next to the electron micrograph of the object shown in figure 5.4 b).

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5.2. FOURIER TRANSFORM HOLOGRAPHY

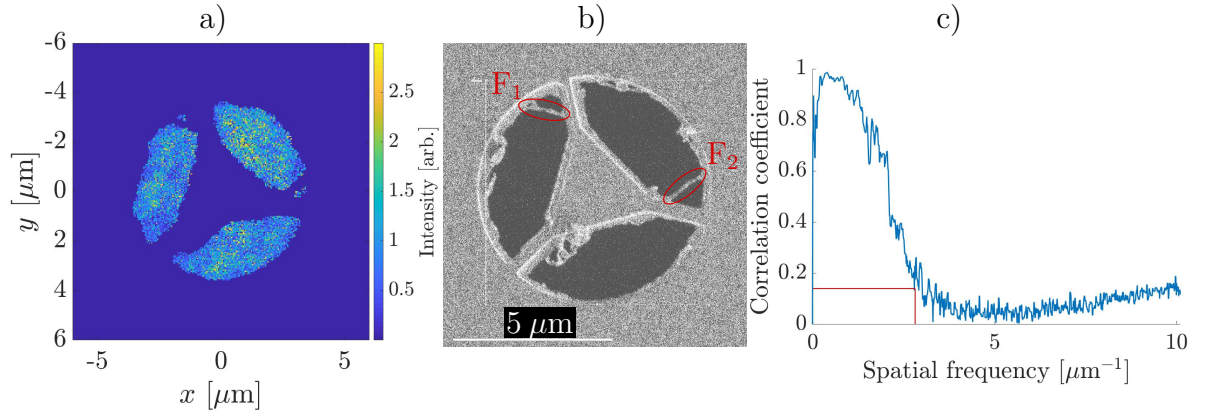


Figure 5.4: a) Recovered intensity of the ESW of the object shown again in b). c) The calculated FRC curve for the image shown in a), giving a correlation coefficient at the 0.17 threshold of $2.91 \mu\text{m}^{-1}$ corresponding to a resolution of approximately 340 nm.

The ESW clearly shows the two small filaments of coated substrate at locations F_1 and F_2 in the electron micrograph, indicative of a resolution of the order of 100 nm. In figure 5.4 c), the FRC curve used to quantify the image resolution is shown along with the threshold of 0.17 alluded to in section 2.4.3. This corresponds to a significant presence of signal above the noise floor that extends to spatial frequencies of $2.91 \mu\text{m}^{-1}$ with corresponding resolution of approximately 340 nm, or just over 10 wavelengths. This resolution is lower than the theoretically achievable diffraction limit due to the short exposure times comprising the HDR data. This was a consequence of the large amount of transmissive area in the object allowing a very strong zero order component of the beam to saturate the chip. This saturation led to bleeding of the counts between pixels, contaminating the data to an unusable state for acquisitions in excess of 2 seconds.

5.2 Fourier Transform Holography

The use of an IPRA with data of a limited S:N, such as those shown in figure 5.3 b), is associated with an unreliable convergence and stagnation in local minima. The simple

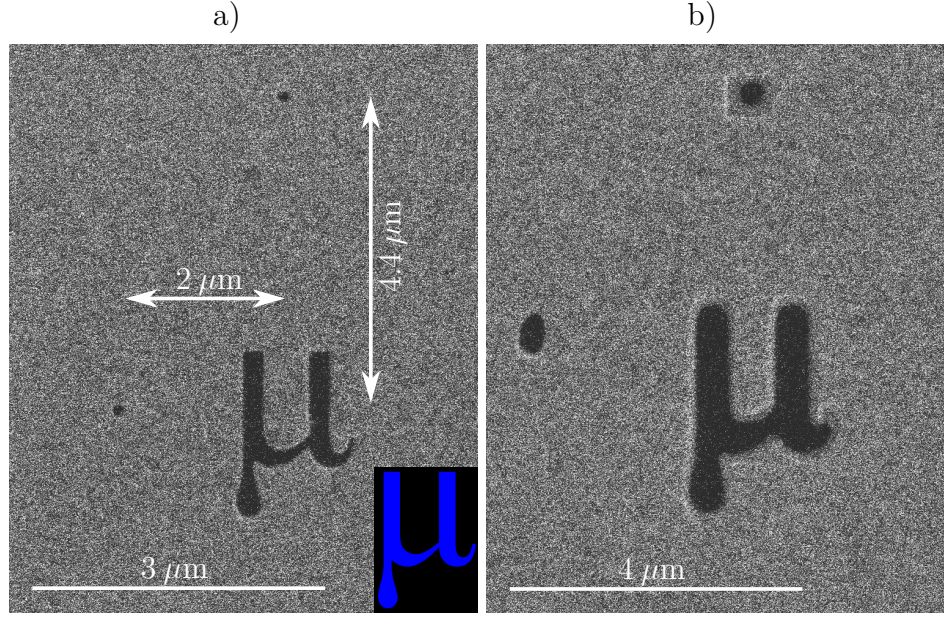


Figure 5.5: a) An ion beam image taken of the first object the author imaged with FTH. There are two reference holes, one located vertically above and another located horizontally to the left. The distances denote the separation of the references from the centre of the object. The lower right inset is the milling pattern read by the FIB. b) An ion beam image of an earlier version of the object shown in a).

analytical recovery of an image* of the object offered by FTH (see equation 2.57d) coupled with its experimental similarity to CDI prompted the author to seek to image objects by FTH.

5.2.1 Object

The object used in the first FTH experiment was an aperture shaped as the Greek letter ‘ μ ’ of dimensions $3 \times 2 \mu\text{m}$, as shown in figure 5.5 a). An earlier version of this object is shown in figure 5.5 b) which displays evidence of an excessive FIB milling time in the badly defined edges and large reference features.

The size and largely opaque nature of this object was chosen to minimise the high

*The author reiterates here that the cross-correlation between the object and a reference feature in a FTH experiment will be referred to as the ‘image’ for brevity.

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intensity contrast between the zero order on-axis component of the detected data and the remainder of the signal, enabling longer exposure times to be accessed without saturating the detector. Two reference holes, approximately 100 – 150 nm in diameter, were milled at distances that satisfy the holographic separation condition. As stated in section 2.3.2, the reference size is proportional to the resolution of the recovered image.

5.2.2 Data

Figure 5.6 shows the simulated and experimentally observed intensity patterns according to the illumination of the FTH aperture and both reference holes, shown in figure 5.5 a). The data presented in figure 5.6 b) reveals that signal persists into the higher spatial frequency regions, as compared to the counterpart CDI data in figure 5.3, indicative of fine structure content. This difference is attributed to the nature of the objects being imaged: The CDI aperture comprised a much greater transmissive area which contributed a much stronger zero order component of the beam to the data than was the case with the FTH aperture. This meant that much longer exposure times could be used in the FTH experiment without reaching the saturation limit of the detector, enabling more high spatial frequency content to be captured. The dominant features of the simulated and experimentally observed patterns in figure 5.6 are the horizontal interference fringes along $Y = 0 \mu\text{m}^{-1}$. The periodicity of these horizontal fringes corresponds to a pair of sources separated by $(660 \pm 34) \text{ nm}$ which closely matches the separation of the two vertical slots in the ‘ μ ’ aperture established in the electron micrograph of 680 nm.

5.2.3 Reconstruction

In accordance with equation 2.57d in section 2.3.2, the object plane is retrieved by taking a single inverse Fourier transform of the measured data. A zoomed view of this plane is shown below in figure 5.7 a). In figure 5.7 b), a zoomed view of the image labelled H_2

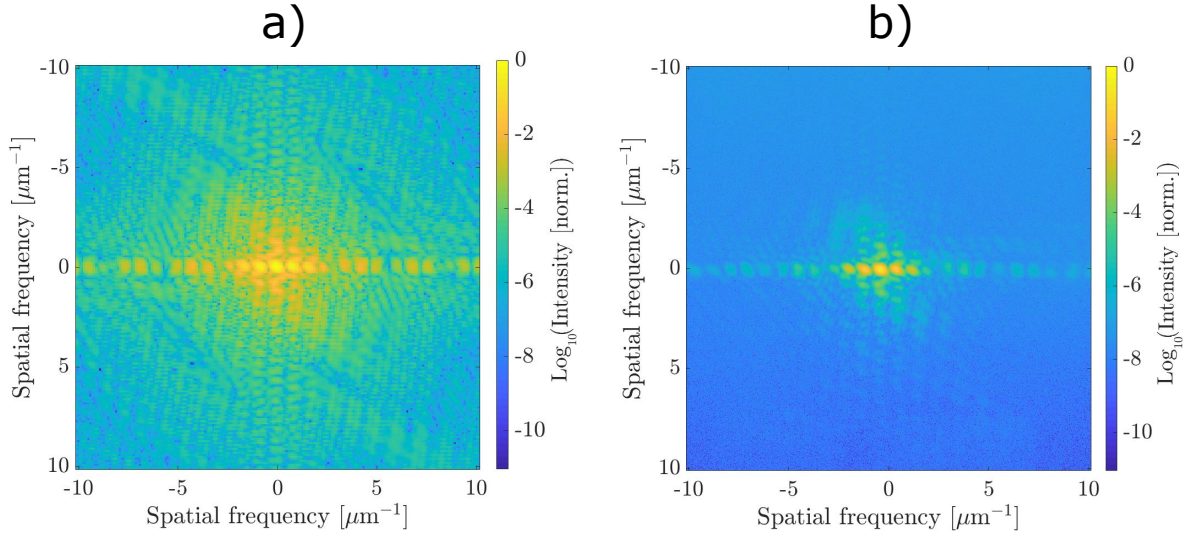


Figure 5.6: a) Simulated intensity corresponding to the illumination of the design aperture shown in the inset of figure 5.5 a). b) Experimentally observed intensity corresponding to the illumination of the aperture shown in figure 5.5 a) for an exposure of 30 seconds. Both images are presented on a log scale.

is shown revealing a gradient in the image intensity indicative of off-axis illumination. Regardless, the similarity with the design aperture shown in the inset of figure 5.7 a) is clear. All regions of the ‘ μ ’ shaped aperture in figure 5.7 b) show some amount of signal, indicative of a resolution at least as small as the narrowest 100 nm scale features within the aperture. Figure 5.7 c) shows the FRC curve corresponding to the image shown in figure 5.7 b) which remains above the threshold value for all spatial frequencies captured in the data. This implies that the resolution of figure 5.7 b) is limited by the NA of the imaging system, which is corroborated by the clear persistence of signal to the edges of the chip in figure 5.6 b). The distance between the sample and chip was $z = (4.1 \pm 0.1)$ cm, corresponding to a resolution limited to (98 ± 2) nm, calculated from equation 5.1.

The transverse spatial locations of both H_1 and H_2 closely match those of the two reference features in figure 5.5 a). Ergo, no overlap has occurred between the recovered images and their complex conjugate counterparts, and the on-axis autocorrelation. It is

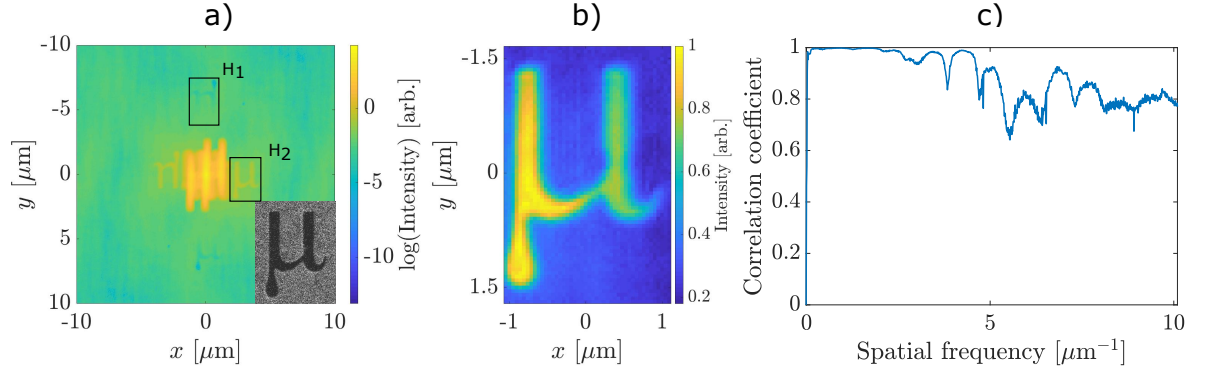


Figure 5.7: a) A zoomed view of the object plane found by inverse Fourier transforming the data shown in figure 5.6 b). The inset is a SEM image of the actual aperture for comparison. The location of two unique images are labelled $H_{1,2}$. b) A zoomed view of H_2 . c) The FRC curve used to calculate the resolution achieved in subfigure b).

worth noting that if an overlap had occurred, there are numerical methods available to subtract the autocorrelation and reveal the obscured information [178].

5.3 Summary

To conclude, the first demonstrations of lensless imaging using an XUV lensless microscope within the author's research group has been presented. These images obtained through CDI and FTH show resolutions down to under 100 nm, or approximately 3 times the imaging wavelength of 32 nm. Although the absolute value of the resolutions claimed in this thesis may vary depending on the technique used to determine it, it is clear that ~ 100 nm scale features within the apertures have been successfully resolved.

Chapter 6

Brightness Improvement



THE poor conversion efficiency of HHG from the IR into the XUV typically restricts harmonic sources to average powers of a few microwatts or lower [87, 88]. The low average power limits their use in applications such as seeding XFELs [179], production of isolated attosecond pulses [180] and lensless imaging [39]. The high flux and low divergence radiation demanded by these applications motivates the search for increasingly bright harmonic sources [89].

The brightness [89, 181] of a beam of radiation can be defined as

$$B = \frac{N}{\Delta t \cdot d\Omega \cdot A_s} \approx \frac{N z^2}{\Delta t \cdot \pi w(z)^2 \cdot A_s}, \quad (6.1)$$

where N is the number of photons delivered by the beam in a time interval Δt , A_s is the source size, and $w(z)$ and $d\Omega$ are respectively the beam radius and solid angle subtended by the source in a plane a distance z from the source. The approximation $d\Omega \approx \frac{\pi w(z)^2}{z^2}$ is valid when the distance z is much larger than the source size A_s . The product $A_s \cdot d\Omega$ is commonly called the **beam parameter product (BPP)** [182] and is a metric of beam quality that is conserved in an ideal lossless optical system [183]. The brightness is therefore increased as the flux is increased and or the BPP is minimised

toward the limit of a Gaussian beam. This corresponds to a higher density of photons in the focal plane that enables imaging to be performed with a commensurately reduced exposure time. Equivalently, for a fixed exposure time, data with a larger **S:N** will be obtained. As a knock on effect, a reduced exposure time enables experiments to be completed faster and thus with a lower susceptibility to mechanical drifts or vibrations. This is particularly desirable for **HHG** driven imaging experiments because the low conversion efficiency of the IR into the **XUV** is typically countered by using longer exposures.

This chapter explores a method that increases by a factor of 5 whilst maintaining a fixed experimental footprint the brightness of high harmonic radiation produced by a driver beam of constant peak focal intensity. This was achieved by using a **SLM** to shape the transverse intensity profile of the focussed driver pulse from Gaussian to approximately supergaussian of order $n = 2 - 3.5$ with a **SLM**. The **SLM** imprinted a circular binary phase mask of magnitude π and radius ρ onto the collimated driver beam upstream of the **HHG** interaction. As ρ was reduced, the driver focal spot radius, w_0 , and supergaussian order, n , both increased whilst the longitudinal position of the focal plane remained fixed, preserving the f-number (f/N) and spatial footprint of the experiment.

At a fixed peak focal intensity, the enlarged spot size increased the number of harmonic emitters, increasing the harmonic flux. However since n increased in tandem with w_0 , the harmonic source size w_q increased by a larger amount than w_0 , further increasing the harmonic flux. These changes occurred without changing the lens used to focus the driver beam, making this approach highly experimentally convenient.

Previous work to enhance the brightness of high-harmonic beams can be loosely categorised into three approaches. In approach 1) the flux $\frac{N}{\Delta t}$ is increased by increasing the repetition rate [184] or pulse energy of the driving laser in combination with a loose

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focussing geometry [96]. In approach 2) the BPP is reduced toward the theoretical minimum via wavefront control of the driving laser in a feedback loop [94, 95]. In approach 3) phase matching [90] and quasi-phase matching [91, 92] are exploited to increase the harmonic yield.

In approach 1), a high pulse energy [185] driver is focussed by a lens of focal length often greater than 3 m, which may be inconvenient experimentally and requires more expensive laser systems. This route toward brighter sources is therefore inaccessible to small research facility scale laboratories which only have access to driver lasers of modest pulse energy.

In approach 2), the link between driver wavefront quality and harmonic brightness is exploited [186]. However, the highly non-linear nature of HHG means that small variations in the parameters of the driving laser have a dramatic influence on the spatial properties and conversion efficiency of the harmonic beam. This includes the amplification of small driver phase variations that are imprinted into the harmonic beam, increasing the harmonic BPP.

6.1 Beam Path

The beam path of the harmonic beam in this experiment is identical to that shown in figure 4.2 from section 4.2. For convenience, that figure is replicated here as figure 6.1.

In the experiment described in this chapter, the 300 μm thick nickel gas cell was backed by 50 mbar of argon, and the pulse energy of the driver beam was varied in the range 0.3 – 1.1 mJ. Due to the non-linear self focussing [187] experienced by the driver beam during propagation through L_1 and a vacuum chamber window, the longitudinal position of the focal plane of the driver was dependent on its energy. At higher pulse

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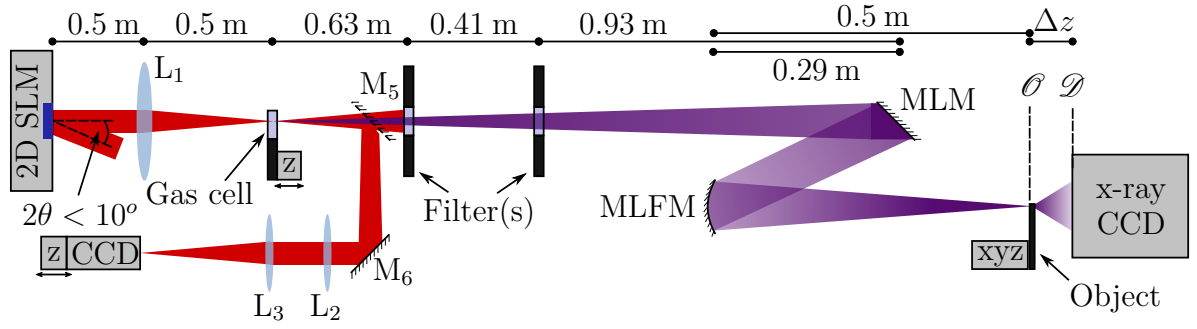


Figure 6.1: Schematic of the harmonic beamline. L_1 is the driver focussing lens, MLM is a multilayer XUV mirror, MLFM is a multilayer XUV focussing mirror. $\mathcal{O}$ is the object domain and $\mathcal{D}$ is the detector domain. Z and XYZ are linear and three axis stages, M_5 and M_6 are beamsplitters that each reflected 10% of the incident energy, the former of which is removable. L_2 is a $f = 50$ cm collimating lens and L_3 is a $f = 50$ cm refocussing lens.

energies, the driver focal plane retreated toward L_1 by several gas cell widths. Calibration of this movement was achieved by inspecting the longitudinal shift of the focal plane using the unitary magnification reimaging line comprising $M_{5,6}$, $L_{2,3}$ and a CCD mounted upon a translation stage.

The object in the $\mathcal{O}$ domain comprised a silicon wafer with a sharp edge that was used to characterise the reimaged harmonic source through the knife edge technique described in section 6.2.3.

Establishing the position of the gas cell relative to the focal plane was a nontrivial task. Multiple methods were tested ranging from the use of spiral phase masks [188], to the reimaging of a knife edge located coincident with the downstream face of the gas cell itself. These methods proved unsatisfactory and so the harmonics themselves were used as an indicator of gas cell position instead. At very low backing pressures of approximately 30 mbar and driver pulse energies of approximately $100 \mu\text{J}$, the gas cell was scanned through the focal volume. At increments smaller than the $300 \mu\text{m}$ gas cell width, the spatially integrated harmonic fluence was recorded as a function of the

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longitudinal position of the gas cell, and exhibited a single peak. The aforementioned backing pressure and pulse energy ensured that no phase matching was present and the HHG could be considered in the single atom regime. Ergo, the single peak in integrated fluence was attributed to the highest intensity of the driver laser which is necessarily coincident with the focal plane for a Gaussian beam with negligible aberrations.

Variation in the longitudinal position of the cell would also have caused the longitudinal position of the focussed harmonic beam to have changed. Using the thin lens imaging formula, a 1 mm longitudinal shift of the gas cell induced an approximately 50 μm shift in the $\mathcal{O}$ plane location. The distance between the $\mathcal{O}$ and $\mathcal{D}$ domains, Δz , was found by illuminating two ≈ 150 nm diameter pinholes located in the $\mathcal{O}$ plane with the harmonic beam of known wavelength. By observing the fringe pattern detected in the $\mathcal{D}$ domain, Δz was found to be (3.9 ± 0.2) cm.

The number of photons arriving at the detector within the exposure time was taken to be proportional to the number of CCD counts. For the measurements presented in this chapter, the CCD preamplifier gain was set to unity, the readout rate fixed at 2 MHz in ‘high sensitivity’ mode, and the exposure time was set to 1 second to avoid saturation. For these conditions the nonlinearity of the CCD is expected to be less than 2% according to a manufacturer supplied specifications sheet. Since the peak driver intensity was fixed, we assume that the spectral content of the harmonics produced is independent of the spatial shaping.

6.2 Definitions

It is important to note that there are several different definitions used to determine the parameters of a beam [146] which can make it difficult to compare different measurements of beam properties such as brightness. In this thesis, unless otherwise stated, the

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definitions of beam diameter calculations described by ISO11145 [189] are used. The remainder of this section sets down the nomenclature and equations used for this chapter.

6.2.1 Supergaussian

A supergaussian intensity distribution of order n and e^{-2} radius w_0 can be written as

$$I_{\text{sg}}(x, y, n) = I_0 \exp \left[-2 \left(\frac{\sqrt{(x - \bar{x})^2 + (y - \bar{y})^2}}{w_0} \right)^n \right], \quad (6.2)$$

where, I_0 is the peak intensity, (x, y) are the transverse focal coordinates and the transverse position of the centre of mass of the profile is denoted $(\bar{x}, \bar{y})$. When $n = 2$, equation 6.2 recovers a Gaussian distribution.

For a beam with a supergaussian transverse intensity profile, pulse energy E_0 and full width at half maximum (FWHM) pulse duration τ , the peak on axis intensity is given by

$$I_0 = \frac{4^{(1/n)} E_0}{\pi w_0^2 \Gamma \left[\frac{2+n}{n} \right] \tau}, \quad (6.3)$$

where $\Gamma[\dots]$ is the Gamma function [190]. For $n = 2$, equation 6.3 reduces to $I_0 = (2E_0)/(\pi w_0^2 \tau)$, the standard normalisation for a Gaussian beam, $I_{\text{sg}}(x, y, 2)$.

6.2.2 Second Moment Width

The second moment transverse width along the x axis for an arbitrary beam is [191]

$$\sigma_x^2 = \frac{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} (x - \bar{x})^2 I_{\text{sg}}(x, y, n) dx dy}{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} I_{\text{sg}}(x, y, n) dx dy} \quad (6.4a)$$

$$= 2^{(-2/n)} w_{0,x}^2 \frac{\Gamma[3/n]}{\Gamma[1/n]}. \quad (6.4b)$$

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By rearranging equation 6.4b, w_0 can be expressed in terms of σ and n :

$$w_0 = \sqrt{\frac{2^{(2/n)} \Gamma[1/n]}{\Gamma[3/n]}} \sigma. \quad (6.5)$$

This multiplicative factor reduces to 4 for $n = 2$ revealing the relation $w_0 = 2\sigma$ for a Gaussian beam.

In order to retrieve a beam width comparable to the commonly employed Gaussian e^{-2} width from a highly asymmetric or badly defined beam profile, it is assumed that once a value of σ has been calculated, the value of n is set to $n = 2$ in equation 6.5 giving a generalised beam width, $W = 2\sigma$ [146]. Given that the detector in figure 6.1 is not located in the $\mathcal{O}$ domain, it is not anticipated that the detected beam profile will necessarily resemble a supergaussian beam. Instead, a badly defined profile that peaks on axis is anticipated that requires the aforementioned generalised treatment where n is assumed to equal 2.

6.2.3 Knife Edge

In practice it is often experimentally difficult to directly image or characterise the primary harmonic source due to the presence of the high intensity driver focus: In past studies, the harmonic source size has been inferred from measurements of the hole drilled through a harmonic generation gas cell by the driver beam [89], or from the driver spot size when using a gas jet. To avoid the uncertainties implicit in this approach, the author reimaged the harmonic source as shown in figure 6.1 to a location upstream of a CCD detector. It is inconvenient to locate the detector exactly in the harmonic source plane ($\mathcal{O}$ domain) because the reimaged source size is smaller than the detector pixel size. This forces the detector to be placed some arbitrary distance Δz downstream of the reimaged source in the $\mathcal{D}$ domain to provide enough space for a knife

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edge measurement [192] to be performed, characterising the transverse intensity profile in the $\mathcal{O}$ domain. A knife edge measurement involves recording the energy transmitted past a beam block with a sharp edge (a ‘knife edge’) as a function of the transverse position of the edge. From equation 6.2, the energy transmitted past the knife edge is

$$\begin{aligned} F_{\text{sg}}(x, n) &= \int_{-\infty}^{\infty} \int_{-\infty}^x I_{\text{sg}}(x, y, n) dx dy \\ &\propto \text{sgn}\{x - \bar{x}\} \frac{\gamma\left[\frac{1}{n}, 2\left(\frac{x-\bar{x}}{w_0}\right)^n\right]}{2\Gamma\left[\frac{1}{n}\right]}, \end{aligned} \quad (6.6)$$

where we have assumed $I_{\text{sg}}(x, y, n)$ to be separable in x and y , and $\text{sgn}\{\dots\}$ and $\gamma[\dots]$ are the sign and lower incomplete gamma functions respectively [190]. When $n = 2$, equation 6.6 reduces to the error function [190] commonly used to fit Gaussian knife edge data.

6.2.4 Beam Parameter Product

The BPP of a beam is defined as the product of its source size and divergence in the far field. It is minimised in the case of a Gaussian beam with a constant phase. In one dimension, using the standard Gaussian optics expression for the beam size $w(z)$,

$$\text{BPP} = w_0 \cdot \frac{w(z)}{z} \quad (6.7a)$$

$$= \frac{w_0}{z} \cdot w_0 \sqrt{1 + \left(\frac{\lambda z}{\pi w_0^2}\right)^2} \quad (6.7b)$$

$$= \frac{\lambda}{\pi} \quad \text{for } \lambda z \gg \pi w_0^2, \quad (6.7c)$$

where $w(z)/z$ is taken to be the divergence of the beam in the far field. This result reveals that for a perfect Gaussian beam, the BPP is invariant to the spot size w_0 . Any deviation of the profile from Gaussian will increase the BPP, lending a larger contribution to beam divergence and decreasing the beam brightness for a given photon

flux.

6.3 Harmonic Field

For the q^{th} harmonic order, the complex field E_q is given by [193]

$$E_q \propto |E_{\text{IR}}^\perp|^{q_{\text{eff}}} \exp[-i(q\Phi_{\text{IR}} + \Phi_{\text{dipole}})], \quad (6.8)$$

where $E_{\text{IR}}^\perp$ is the transverse profile of the amplitude of the driver beam in the generation plane, q_{eff} is the effective non-linearity of the HHG interaction, Φ_{IR} is the phase of the driving radiation also in the generation plane, and Φ_{dipole} is the dipole phase of harmonic q .

In figure 6.1, the gas cell as shown in figure 6.2, is located at the focal plane of the $f = 50$ cm focussing lens, coincident with the supergaussian shaped spatial profile. Since the 4 mm Rayleigh range of the driver beam was much larger than the $300 \mu\text{m}$ thickness of the gas cell, Φ_{IR} is assumed to be constant.

The transverse intensity profile of the driving radiation could be measured as a function of longitudinal position in the focal region by inserting beamsplitter ‘M₅’ and using the 4-f unitary magnification imaging system shown in figure 6.1. These measurements showed that the transverse intensity profile of the driver did not vary over the $300 \mu\text{m}$ thickness of the gas cell, an observation supported by ASM simulations.

For pressures in the range 50 – 80 mbar, the spatially integrated harmonic signal increased linearly with pressure, with no evidence of roll-off, implying that the harmonic emission was not phase matched and hence that any improvement in the brightness can be attributed to the spatial shaping alone. The changes induced in the geometric

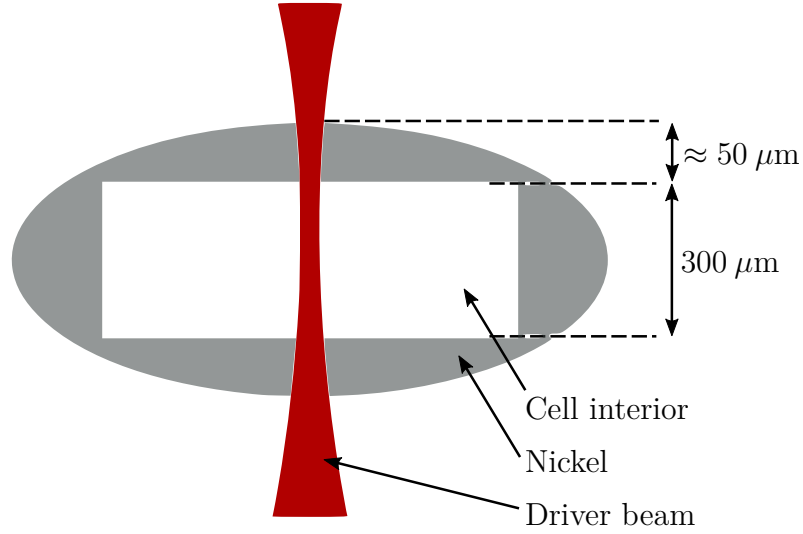


Figure 6.2: Schematic cross section through the gas cell. The hollow nickel tube (light grey) is clamped around a $300\,\mu\text{m}$ spacer (not shown) during construction, which distorts the manufacturer specified $50\,\mu\text{m}$ tube wall thickness. The argon gas is pumped into the cell interior (white) where the driver (red) drives HHG.

phase by spatially shaping the driver focus (described in section 3.3.2) could be used in principle to optimise phase matching conditions. However, to characterise the change in brightness of a harmonic beam due only to shaping the driver focus, the regime of phase matching was avoided in this experiment.

The dipole phase Φ_{dipole} varies approximately linearly with the local driver intensity with a proportionality constant α_q^j depending on the j^{th} electron trajectory in the continuum [115]. In the treatment below, only the short ($j = s(\text{hort})$) trajectories are considered since the more divergent long trajectories were blocked by the small aperture of the filter mounts during propagation from plane P to plane S in figure 6.1. Ignoring the constant Φ_{IR} term, this allows the harmonic field to be written as

$$E_q \propto |E_{\text{IR}}^\perp|^{q_{\text{eff}}} \cdot \exp[-i(\alpha_q^s |E_{\text{IR}}^\perp|^2)]. \quad (6.9)$$

The parameter α_q^s can be calculated using the methods described in reference [115].

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Note that for a reimaged harmonic source with magnification factor M , the knife edge fit equation shown in equation 6.6 is modified to

$$F_q(x, n) \propto \text{sgn}\{x - \bar{x}\} \frac{\gamma\left[\frac{1}{n}, 2\left(\frac{M w_0(x - \bar{x})}{w_q^2}\right)^n\right]}{2\Gamma\left[\frac{1}{n}\right]}. \quad (6.10)$$

Here $M = 0.28$ is the magnification of harmonic source S from harmonic source P and n remains the supergaussian order of the driver focus. Equation 6.11 has been substituted for the driver spot size w_0 in order to eliminate q_{eff} .

6.3.1 Harmonic Source Size

From equations 6.2 and 6.9, w_q^P , the e^{-2} intensity radius of the primary harmonic source [6, 116] generated by a supergaussian driver, can be written as

$$w_q^P = \frac{w_0}{\sqrt[n]{q_{\text{eff}}}}. \quad (6.11)$$

If the thickness of the gas cell is much less than that of the Rayleigh range of the driver then for a fixed supergaussian order, the number of harmonic emitters, and hence the harmonic flux, is proportional to $(w_q^P)^2$. However, in this work, the supergaussian order n increased as w_0 was increased, and hence we expect the harmonic flux to increase faster than w_0^2 .

Ordinarily, manipulating w_0 would require both the IR focussing lens to be changed, and therefore the location of the gas cell relative to the SLM or equivalent mirror. However, through the method explored in this chapter it is shown that w_0 can be manipulated through the phase of the collimated driver beam prior to the HHG interaction. This enables control over the brightness of the harmonic beam without changing the experimental setup.

In this chapter, we characterise the reimaged harmonic source in the $\mathcal{O}$ plane which involves no loss of generality since the BPP is conserved in a lossless imaging system [183]. In the schematic depiction of the experiment in figure 6.1, the aluminium filters and MLO reduce the flux and increase the astigmatism of the beam respectively, but this is of no consequence since only relative changes in the source brightness are important in this work.

6.4 Beam Shaping

The optimal profile for the driver field for a given peak intensity is that of a tophat [194] transverse amplitude profile with constant phase: In this case, according to equation 6.11, when $n \rightarrow \infty$, $w_q = w_0$. This would maximise the number of harmonic emitters for a given w_0 , maximising flux whilst providing a larger source thus a smaller contribution to divergence. The tophat amplitude profile of diameter A , described by

$$\Pi_{-\frac{A}{2}, \frac{A}{2}}(x) = \begin{cases} \sqrt{I_0} & \text{for } |x| < \frac{A}{2} \\ 0 & \text{for } |x| > \frac{A}{2} \end{cases} \quad (6.12)$$

is shown in figure 6.3 (a).

We employ a 2-f geometry between the SLM, lens and gas cell in order to eliminate any residual field dependent phase in the plane P, as explained in section 2.2.3. Accordingly, to generate the field described by equation 6.12 in the plane P, the field displayed on the SLM should be

$$\text{FT}^{-1} \left[\Pi_{-\frac{A}{2}, \frac{A}{2}}(x) \right] = A\sqrt{I_0} \text{sinc} \left(\frac{A\pi X}{\lambda f} \right), \quad (6.13)$$

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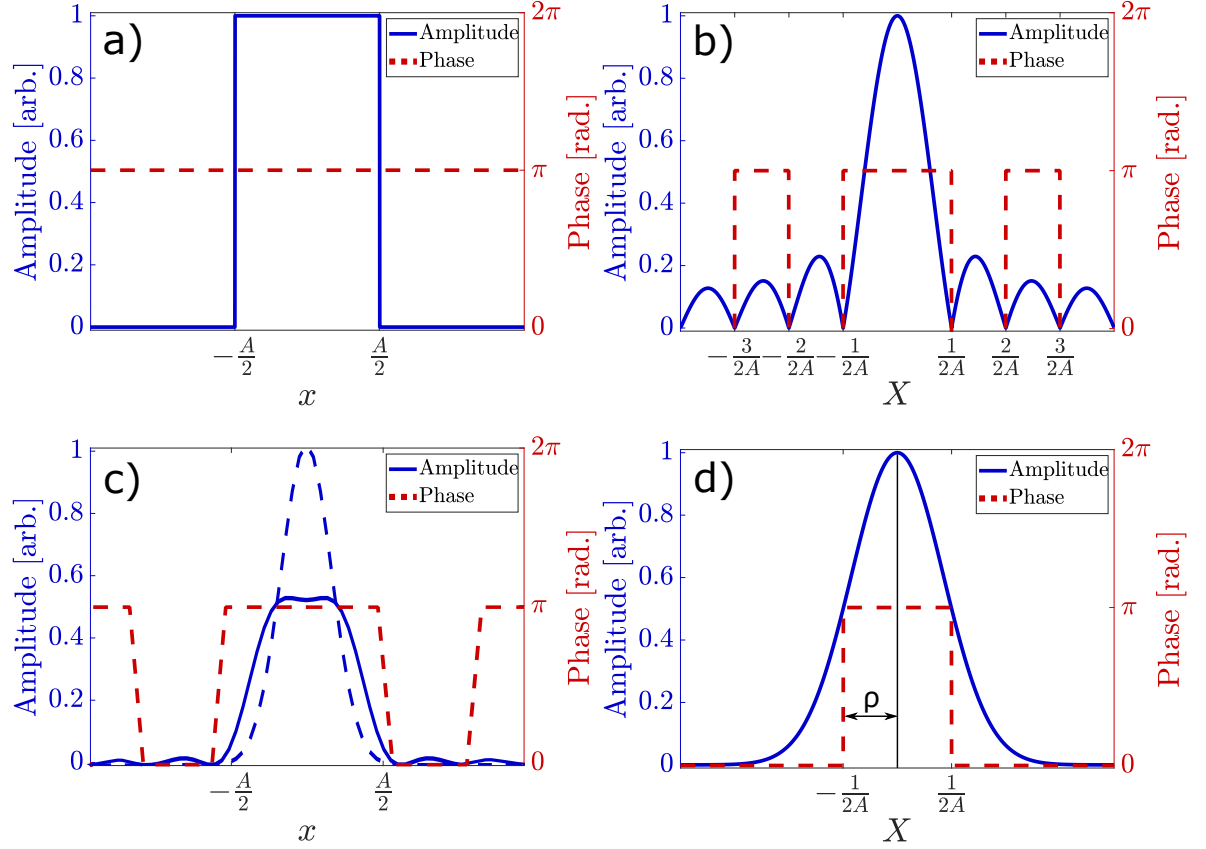


Figure 6.3: The amplitude (blue curves) and phase (red curves) of the driver field, as a function of the transverse focal coordinate x (a, c), and as a function of the transverse SLM coordinate X (b, d). (a) shows the optimal focal profile comprising a tophat amplitude and constant phase. (b) shows the field that must leave the SLM in order to produce the optimal focal field shown in (a). (d) shows a schematic representation of the field leaving the phase-only SLM in the experiment comprising a collimated Gaussian amplitude and modulated phase. (c) shows the simulated focal field produced by focussing the field shown in d), when the value of ρ is $\rho = 4.6$ mm. Also shown by the dashed blue line, is the amplitude of the focal field produced by focussing the field shown in d) without any phase step.

where f is the focal length. This field is shown in figure 6.3 (b). Note that in two dimensions using the radial coordinates r and R in the focal and SLM planes respectively, the Fourier transform of the circularly symmetric tophat function is

$$\text{FT}^{-1} \left[\Pi_{-\frac{A}{2}, \frac{A}{2}}^{2D}(r) \right] = A \sqrt{I_0} \frac{J_1(4\pi AR/\lambda z)}{4\pi AR/\lambda z}, \quad (6.14)$$

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where J_1 is the Bessel function of the first kind and first order. The zeros in the sinc function occur when $X = \pm m \frac{\lambda f}{A}$. For the example of an 800 nm driver creating a 50 μm diameter tophat at the focus of a 50 cm lens, the first four zeros are (8, 16, 24, 32) mm. The corresponding zeros in J_1 can be found at $R = 0.61(\lambda z)/(2A)$. For the remainder of this chapter only the 1D case is considered for the sake of brevity. Although the above equations allow the calculation of the radial positions of the phase steps in figure 6.3 b), the active area of the SLM was too small to display these additional steps: Throughout this experiment, only one phase step was ever generated. In addition, the phase-only nature of the SLM used in this experiment prevented the amplitude from being shaped to that shown in figure 6.3 b). This is why the experimentally achieved field leaving the SLM is that shown in figure 6.3 d).

A number of methods exist to produce phase masks that transform a collimated Gaussian beam into a focal spot of uniform intensity [195–197]. The binary phase mask [198] of magnitude π and radius ρ described above was used in the work presented in this chapter. A single phase step of these parameters was imprinted into the collimated Gaussian driver beam, creating the field shown in figure 6.3 (d) leaving the SLM.

Figure 6.3 (c) shows the calculated transverse intensity profile of the beam produced by focussing the beam shown in fig 6.3 (d), which in turn is generated by reflecting an ideal collimated Gaussian beam from a SLM displaying the binary phase profile. It can be seen that when $\rho = 4.7$ mm, the transverse intensity profile of the focussed beam is approximately described by a supergaussian with a piecewise linear transverse variation in phase.

Simulation were undertaken using the ASM method in order to assess the longitudinal evolution of the amplitude and phase in the focal region of beams shaped by binary masks. These showed that within the gas cell, and for regions in which the intensity exceeded $1 \times 10^{14} \text{ W cm}^{-2}$, the transverse phase variation in any plane was less than

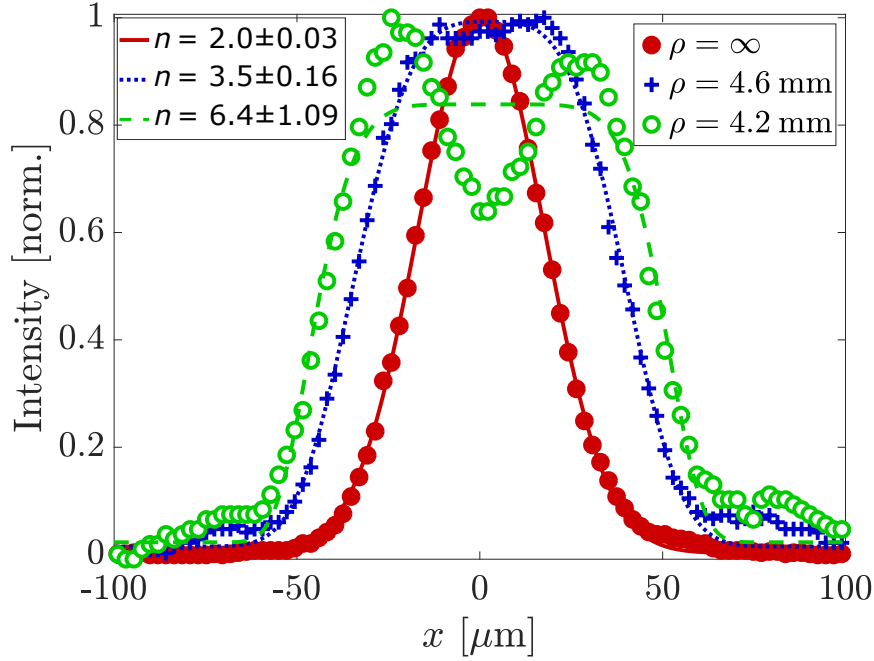


Figure 6.4: Measured transverse intensity profiles of the focussed driver pulses for a range of values of ρ (points), together with fits to equation 6.2 (solid, dotted and dashed lines). The n values relate to the two dimensional fits as described in the text.

0.09 radians. Ergo, the assumption that Φ_{IR} is constant within regions within which harmonic were generated is valid.

The transverse intensity profiles of the focussed driver beams were recorded for a range of values of ρ . Three example lineouts along the x axis with fits to equation 6.2 in one dimension are shown in figure 6.4. Additional one dimensional fits to lineouts along the y axis yielded fit parameters within the uncertainties of their x axis counterparts. When $\rho \rightarrow \infty$ the SLM displays a uniform phase across the chip and is therefore equivalent to a planar mirror. As seen in figure 6.4, as ρ is decreased interference between light focussed from the inner and outer regions of the beam leaving the SLM causes the transverse intensity profile of the focal spot to become flatter, corresponding to a supergaussian profile of larger n . For our geometry, when ρ decreases below approximately 4.6 mm an on-axis minimum develops and the desired higher order

supergaussian profile deteriorates. Additionally, an excessive pulse energy is required in order to maintain a fixed peak intensity for such profiles. Consequently, we take the minimum usable value of ρ in this work to be $\rho \approx 4.6\text{mm}$, corresponding to $n = 3.4 \pm 0.16$.

For all data presented in this chapter, the energy of the driving pulses was adjusted using equation 6.3 to give a peak on-axis focal intensity of $I_0 = 4.7 \times 10^{14} \text{ W cm}^{-2}$, corresponding to the expected peak focal intensity of an unshaped ($n = 2$) Gaussian beam with a spot size $w_0 = 34 \mu\text{m}$, pulse energy $E_0 = 300 \mu\text{J}$ and FWHM pulse duration $\tau = 35 \text{ fs}$. Equation 6.3 assumes a perfect supergaussian profile free of aberrations and hence any deviation from this would reduce the achieved experimental peak intensity.

6.5 Results

Five different phase masks were investigated, corresponding to $\rho = \infty, 5.7 \text{ mm}, 5.2 \text{ mm}, 4.8 \text{ mm}$ and 4.6 mm . As w_0 and n both increase, the pulse energy was also increased in order to maintain the same fixed peak intensity as described above. Figure 6.5 shows the pulse energy and spot size as ρ is decreased from unshaped.

Figure 6.6 a) shows, as a function of ρ , the change in both the driver spot size and the supergaussian order of the driver profile. It can be seen that n increases monotonically as ρ is decreased to the experimental limit of $\rho = 4.6 \text{ mm}$.

The data in figure 6.6 b) shows that as ρ is decreased and w_0 increases by a factor of 1.98 ± 0.02 , the radius of the reimaged harmonic source w_q^S increases by a factor of 2.36 ± 0.26 . This corroborates the anticipated faster than quadratic increase in harmonic source size with the driver spot size. The corresponding effective nonlinearity calculated by equation 6.11 gives $q_{\text{eff}} = 2.6 \pm 0.6$.

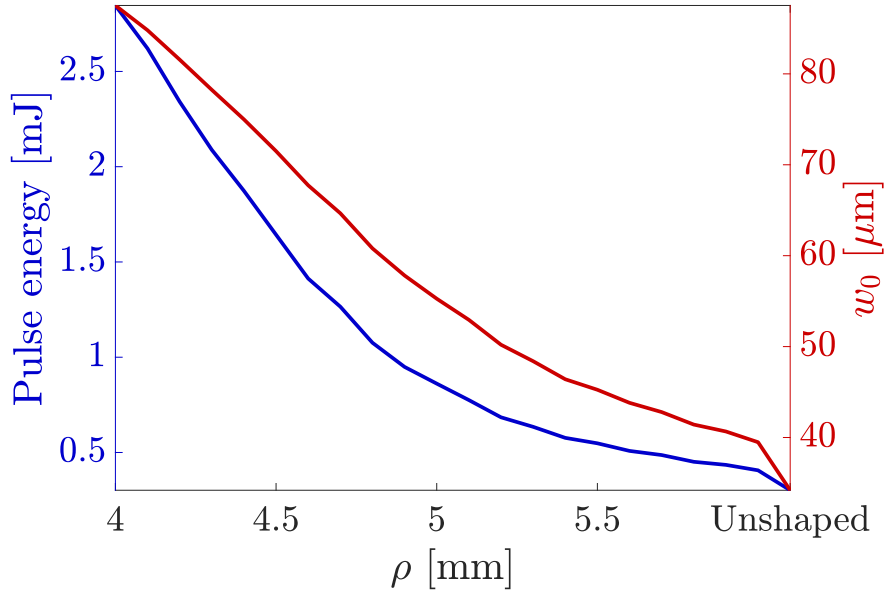


Figure 6.5: The change in pulse energy (blue) specified by equation 6.3 as the driver spot size (red) is increased as a function of ρ . The unshaped case corresponds to a $34\mu\text{m}$ radius Gaussian spot with a pulse energy of $300\mu\text{J}$.

The monotonic increase in w_q^S as w_0 increases correlates with the increase in the detected flux shown in figure 6.7 a) as a larger number of harmonic emitters is accessed by the enlarged source. As w_0 increases, the harmonic BPP also increases due to the effect of the increasing value of n , without which, the BPP would remain constant. Although increasing n gives an increasing contribution to the beam divergence, in the next section it will be shown that the decrease in the beam divergence caused by the increasing source size dominates over the range of parameters considered here.

Figure 6.7 a) shows a linear decrease in the detected beam size $W(z)$ by a factor of 2 as w_0 is increased from $34\mu\text{m}$ to $68\mu\text{m}$ ($\rho \rightarrow \infty$ to 4.7mm). This trend is interrupted at larger w_0 values due to the emergence of the on-axis minima in the driver intensity profiles shown in figure 6.4 when ρ is decreased below $\rho \approx 4.7\text{mm}$. The doubling of w_0 would imply a factor of 4 increase in the detected flux. However, figure 6.7 a) shows that the increase in flux peaks at a factor of 5 ± 0.65 . This is due to the larger increase in w_q^S compared to w_0 caused by the increasing value of n .

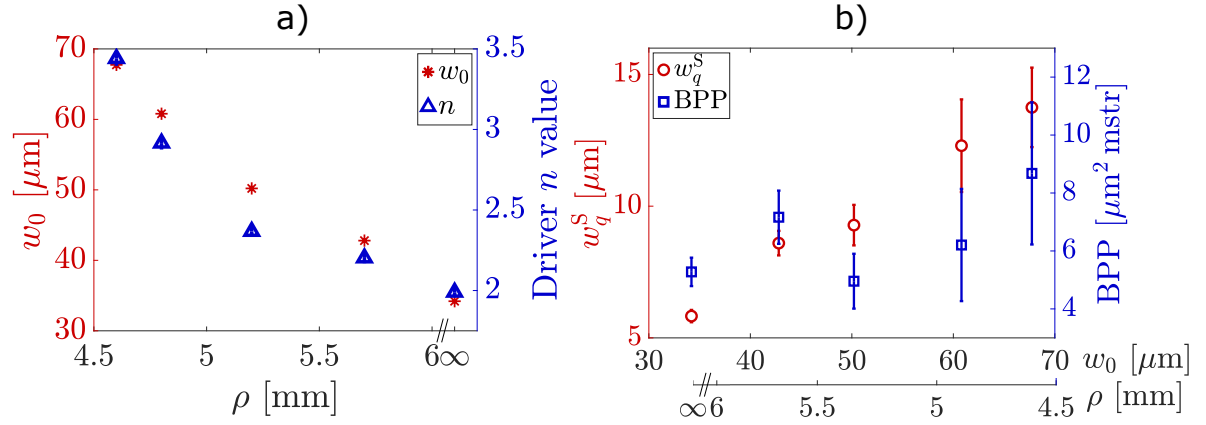


Figure 6.6: a) The measured variation in the driver spot radius (red asterisks) and the driver supergaussian order n (blue open triangles) as a function of ρ . The uncertainties in n and w_0 are smaller than the data points. b) The variation in the source radius of the reimaged harmonic beam (red open circles) and harmonic BPP (blue open squares) as a function of w_0 . An axis of ρ is also shown illustrating its inverse proportionality with w_0 .

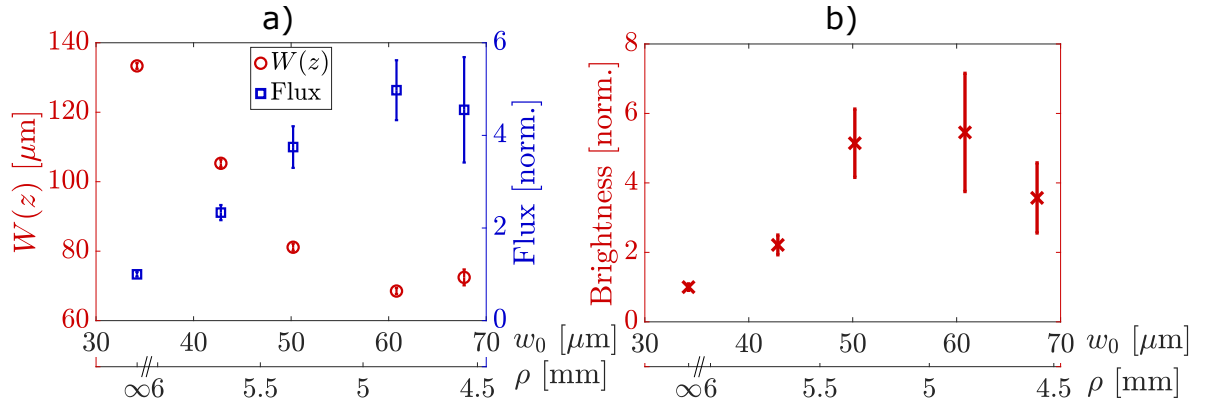


Figure 6.7: a) The variation in the detected harmonic beam size $W(z)$ (red open circles) and integrated harmonic fluence (blue open squares) as a function of w_0 . b) Variation of the brightness of the harmonic beam calculated according to equation 6.1 as a function of w_0 . An axis of ρ is also shown in both subplots to reference the independent variable manipulated during the experiment.

6.6 Simulations

Simulations were performed in order to gain insight into the effects of increasing the spot size and supergaussian order of the driver beam. In order to fully capture all the possible effects, the ranges of driver spot size and corresponding supergaussian order

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were extended over wider ranges than could be investigated experimentally (as described in section 6.6.1). Due to the larger maximum spot size produced by this extension, an arbitrary propagation distance of $z = 15$ m was used in the simulations to ensure that even the largest simulated sources were propagated into the far field using a Fourier transform. Given that this chapter is only concerned with relative changes in the source brightness, this disparity between the simulated and experimental propagation distance is considered unimportant as it manifests only as a scaling in the detected profile size $W(z)$.

The remaining simulation parameters used were; a wavelength of $\lambda = 32$ nm, driver spot sizes ranging from $w_0 = 34$ to 102 μm with supergaussian orders $n = 2$ to 13 , an effective nonlinearity of $q_{\text{eff}} = 2.6$, a peak focal intensity of 4.7×10^{14} W cm^{-2} and corresponding trajectory coefficient $\alpha_{25} = 1.04 \times 10^{-14}$ $\text{cm}^2 \text{W}^{-1}$.

6.6.1 Extending the Simulation Range

In order to circumnavigate the undesired complications introduced by the emerging on-axis minimum in the shaped transverse intensity profiles, the function $n(w_0) = a w_0^b + 2$ was fit to the measured variation of the supergaussian order with the driver spot size w_0 . The fit could then be extrapolated beyond the range of ρ (and hence w_0) for which viable supergaussian beams were generated in the experiment. Figure 6.8 shows the data, fit and extrapolated curve over a range of values that encompasses $w_0 = 100$ μm . The fit revealed that the power relating the driver spot size and supergaussian order in the data shown in figure 6.8 was $b = 4.77 \pm 0.01$.

Using the extended range of n and w_0 , harmonic sources were simulated in accordance with equation 6.9 and propagated $z = 15$ m into the far field. The sizes of the harmonic beams were calculated in both the source and $\mathcal{D}$ planes using equation 6.4b. In the source plane (P), the supergaussian order in equation 6.4b was set to the value

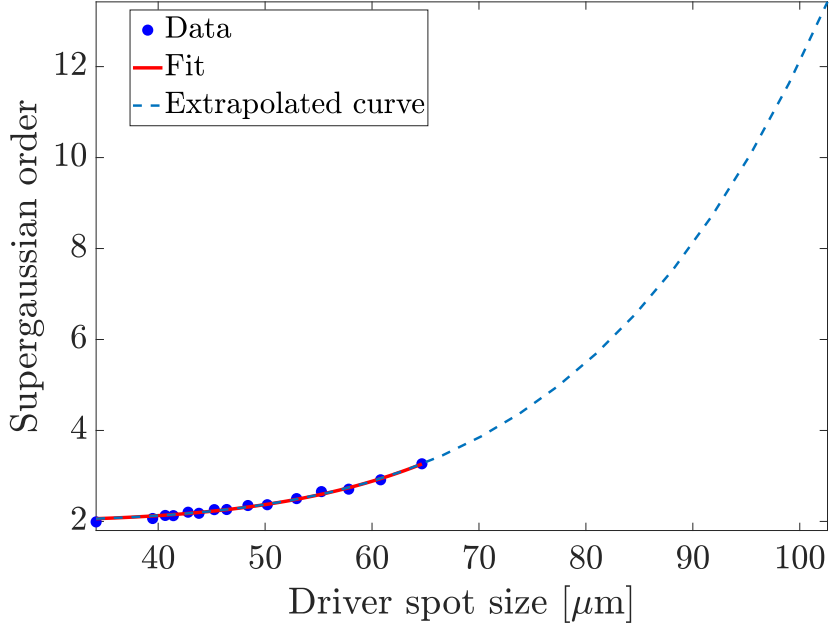


Figure 6.8: Measured (blue dots) and extrapolated (dashed line) supergaussian order as a function of the driver spot size w_0 . The fit to the power law described in the text is shown in red. The fit curve is extrapolated (blue dashed line) to include large n values.

used to create the source, whilst in the $\mathcal{D}$ plane, it was set to $n = 2$, in accordance with the convention outlined in section 6.2.2.

Given the detected beam sizes in both planes, the BPP was then calculated according to equation 6.7a. The simulations were repeated with the dipole phase term in equation 6.9 switched on and off to assess the effect of this term on the beam properties.

6.6.2 Gaussian Harmonic Sources

Figure 6.9 shows simulations where the supergaussian order has been fixed to $n = 2$ for all sources considered. In figure 6.9 a) the dipole phase is inactive. The source size w_q increases linearly with driver spot size w_0 , behaviour implied by equation 6.11 when both n and q_{eff} are held constant. As expected, the detected beam size $W(z)$ decreases by the same factor, resulting in a BPP that remains constant at its theo-

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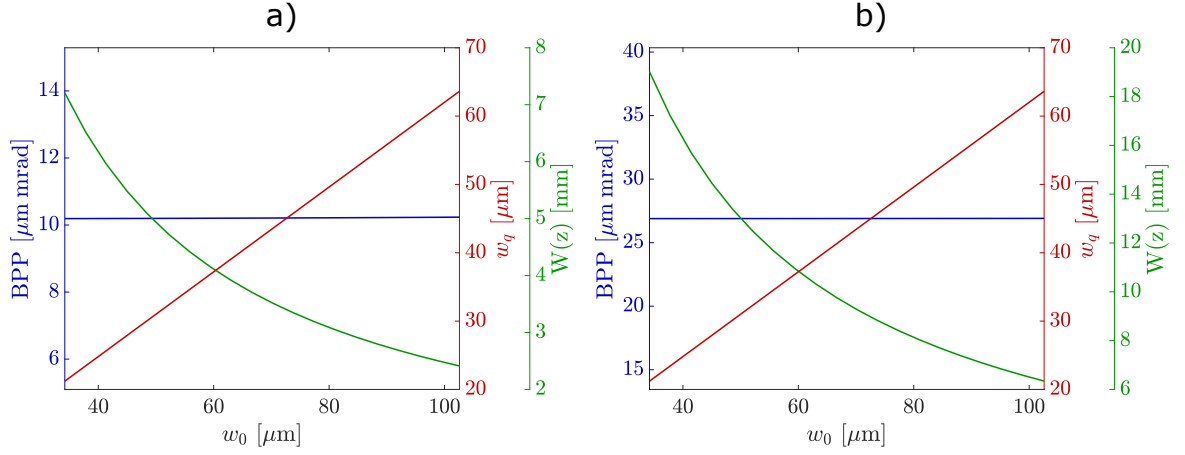


Figure 6.9: Calculated harmonic source size w_q (red curve), simulated far field size $W(z)$ (green curve) and calculated BPP (blue curve) as a function of the driver spot size w_0 for the case where the dipole phase is inactive (a) and active (b). All sources have supergaussian orders fixed at $n = 2$.

retically minimum value of $10.2 \mu\text{m mrad}$ which can also be calculated from equation 6.7c.

In figure 6.9 b) the dipole phase is active whilst the supergaussian orders of every source remained fixed at $n = 2$. All the trends and fractional changes found in figure 6.9 a) are preserved, however, the detected profile sizes $W(z)$ are increased by a factor denoted by γ that varies between 2.64 and 2.63. This value can be found by expanding the form of the dipole phase:

$$e^{(i\phi_{\text{dipole}})} = \exp[i\alpha_q I(r)] \approx \exp\left\{i\alpha_q \left[I_0 - \frac{1}{2}I_0(r/w_0)^2\right]\right\}. \quad (6.15)$$

Neglecting the constant phase contribution, the expanded dipole phase can be modelled as the phase of a diverging lens described by $\phi_{\text{lens}} = (\pi r^2)/(\lambda f)$ with focal length f :

$$f = \frac{\pi}{2\lambda\alpha I_0} w_0^2. \quad (6.16)$$

To find the beam size subsequent to propagating a distance z with the dipole phase, ray transfer matrices [199] can be used. These relate the incident and outgoing beam

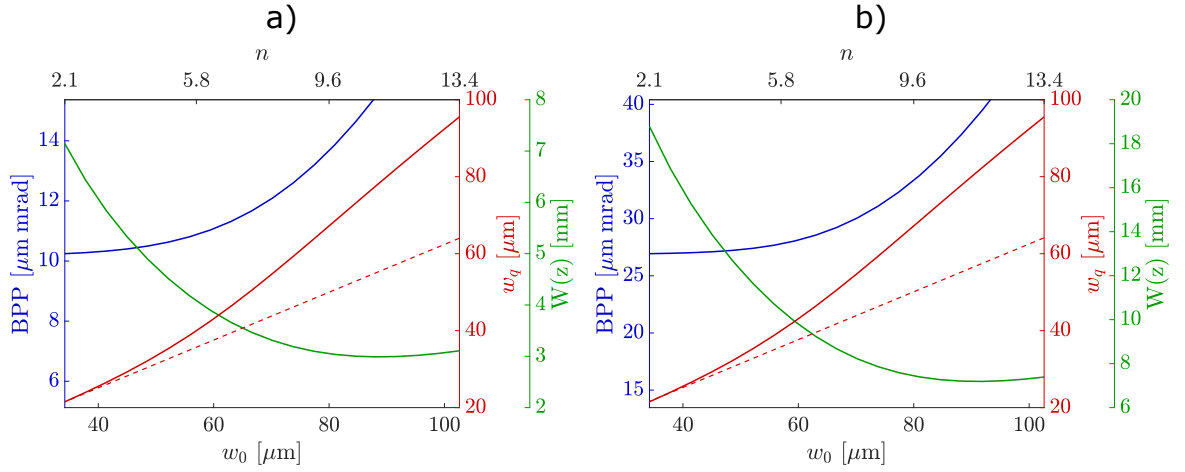


Figure 6.10: Calculated harmonic source size w_q (solid red curve), simulated far field size $W(z)$ (green curve) and calculated BPP (blue curve) as a function of the driver spot size w_0 for the case where the dipole phase is inactive (a) and active (b). The dashed red curves show the $n = 2$ variation for reference. All sources have supergaussian orders that vary with driver spot size according to the extrapolated curve shown in figure 6.8.

parameters y and θ denoting beam height from the optical axis and divergence angle respectively:

$$\begin{bmatrix} y_{\text{in}} \\ \theta_{\text{in}} \end{bmatrix} = \begin{bmatrix} 1 & z \\ 0 & 1 \end{bmatrix} \cdot \begin{bmatrix} 1 & 0 \\ -\frac{1}{f} & 1 \end{bmatrix} \cdot \begin{bmatrix} y_{\text{out}} \\ \theta_{\text{out}} \end{bmatrix} \quad (6.17)$$

For the example of the first Gaussian beam of source size $w_q = 21.2 \mu\text{m}$ (with corresponding $W(z) = 19 \text{ mm}$ found in figure 6.9 b)), equation 6.17 gives a value of $W(z) = 20.0 \text{ mm}$, corresponding to $\gamma = 2.73$. The difference between this figure and the one found numerically from the simulation of 2.63 is attributed to the truncation of the Taylor series in equation 6.15. The relative change in γ will be explored toward the end of this section. The calculated BPP in figure 6.9 b) is also larger than in 6.9 a) by the same γ factor as the source sizes remain the same.

6.6.3 Supergaussian Harmonic Sources

Figure 6.10 shows simulations where the supergaussian orders of the sources vary with w_0 according to the extrapolated curve in figure 6.8. In figure 6.10 a) the dipole phase

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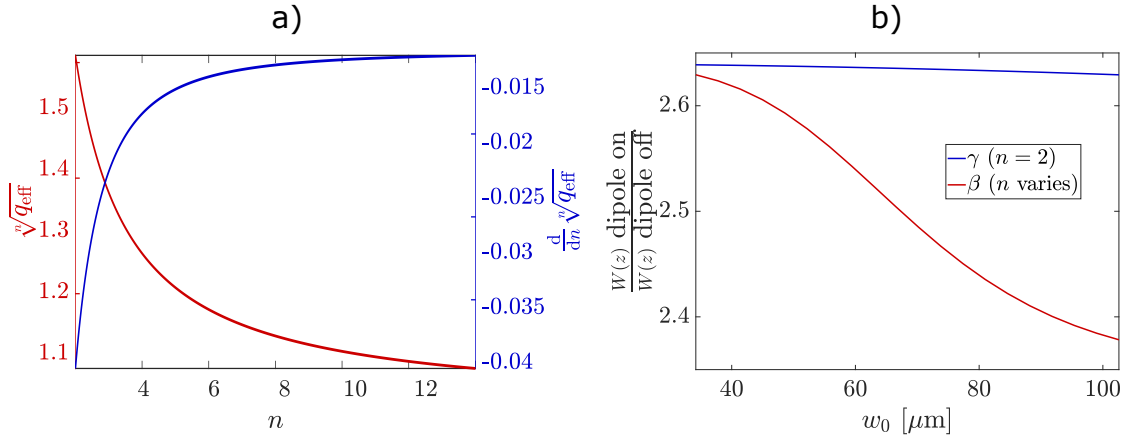


Figure 6.11: a) The variation in the value of the denominator in equation 6.11 as a function of n (red line). The derivative of this is shown by the blue line, revealing that the maximum rate is constrained to low values of n . b) The ratio of the detected beam widths when the dipole phase is active and inactive, as a function of w_0 , for the cases of $n = 2$ (blue curve, γ) and n varies (red curve, β).

is inactive. As the driver spot size increases by a factor of 3, the harmonic source size increases by a factor of 4.44. The increase in supergaussian order further increases the source size, in addition to the increase due to the increase in driver spot size. In equation 6.11, if the denominator is held constant, $w_q \propto w_0$, but when n is allowed to vary, this linear proportionality is no longer present. The red curve in figure 6.11 a) shows the variation in the denominator of equation 6.11 as a function of n . Its value tends asymptotically to unity in the limit of $n \rightarrow \infty$ where the harmonic source must necessarily match the driver spot size exactly. This implies that the faster rate of growth of w_q compared to w_0 must be constrained to lower values of n : The blue curve in figure 6.11 a) shows the derivative of $\sqrt[n]{q_{\text{eff}}}$ with respect to n , revealing that the maximum rate of increase in harmonic source size occurs when n is small. It is beyond this regime that both red curves in figure 6.10 depart from the linear relationships shown by the dashed lines.

Over the range of values considered within the simulations in figure 6.10 a), the detected beam size $W(z)$ decreases by a factor of 2.30. The counterpart decrease in the

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simulations of Gaussian sources in figure 6.9 a) was a factor of 3. Given that the only difference between these two simulations is the inclusion of varying n in the former, it can be deduced that the disparity between these two factors must be attributable to increasing n . As the supergaussian order of a harmonic source increases, increasingly high spatial frequencies are required to construct the transverse profile of the beam, and hence its divergence increases.

In figure 6.10 b) the dipole phase is active whilst the sources again have supergaussian orders that vary with driver spot size according to the extrapolated curve shown in figure 6.8. As the driver spot size increases by a factor of 3, the harmonic source size again increases by a factor of only 4.44, now however, the detected beam size decreases by a factor of 2.54. In figure 6.11 b), the red curve shows the ratio of the detected beam sizes for the cases where dipole phase is active and inactive, denoted by β . Similarly to the Gaussian simulations, the dipole phase introduces a defocus that enlarges the values of $W(z)$ by a factor β which varies from 2.63 to 2.38. The monotonic decrease of β with w_0 when n varies implies that the inclusion of the dipole phase drives the divergence of the beam down with increasing supergaussian order. This is due to the decreasing contribution to beam divergence as the intensity dependent dipole phase becomes smoother. However, over the range of n considered in these simulations, this only reduces the detected beam profile size by $\approx 10\%$.

6.6.4 Summary of Simulations

A brief summary of the four simulations presented above will now be provided. In all cases, w_q , n and the dipole phase affect the detected profile size and therefore the BPP. Figure 6.12 gives a schematic representation of the changing beam sizes in each situation.

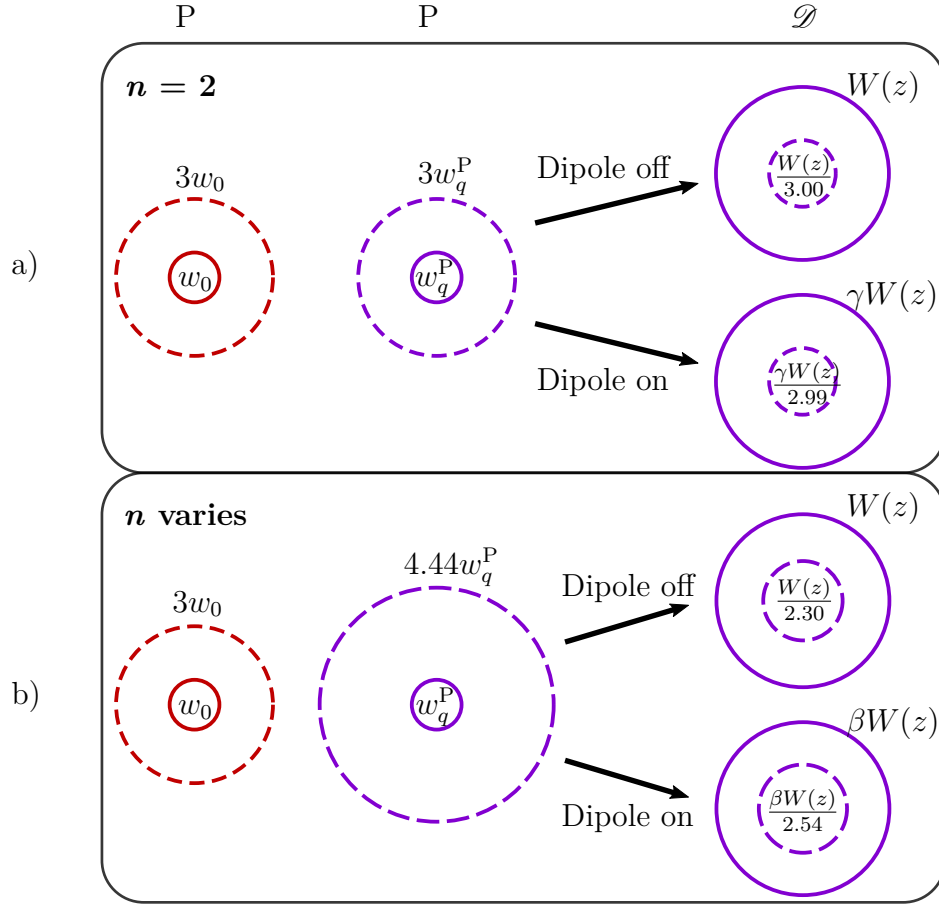


Figure 6.12: A schematic depiction of the changing relative sizes of the simulated driver spot (red, plane P), primary harmonic source (violet, plane P) and detected harmonic profile (violet, plane $\mathcal{D}$) as the simulated w_0 value increases by a factor of 3 when a) $n = 2$ and b) n varies according to figure 6.8. The solid circles correspond to the sizes of the simulated unshaped beam (when $\rho \rightarrow \infty$), whilst the dashed circles correspond to the sizes of beams when w_0 has been increased by a factor of 3. The factors of γ and β are those referred to in the text and shown in figure 6.11 b).

Gaussian sources with no dipole phase (figure 6.9 a))

When $n = 2$ and the dipole phase is inactive, $W(z)$ is inversely proportional to w_q , resulting in a BPP independent of w_q . This is shown in the upper branch of figure 6.12 a).

Gaussian sources with the dipole phase (figure 6.9 b))

When $n = 2$ and the dipole phase is active, the value of $W(z)$ is a factor of $\gamma = 2.635$ larger for all values of w_0 than when the dipole phase was inactive. There is an $\approx 2\%$ decrease in γ as shown by the blue curve in figure 6.11 b) meaning the effect of the dipole can be considered predominantly as a defocussing term for Gaussian sources. This results in a BPP that is larger than the case with no dipole phase by the same factor γ with an $\approx 2\%$ variation. This is shown in the lower branch of figure 6.12 a).

Supergaussian sources with no dipole phase (figure 6.10 a))

When n varies and the dipole phase is inactive, the higher divergence compared to Gaussian sources of the same size is attributed to the increasing supergaussian order. The balance between the decrease in $W(z)$ caused by increasing w_q (dominating at smaller w_q values) and the increase in $W(z)$ caused by increasing n (dominating at larger n values) can be seen in the turning point of the green curves in figure 6.10 at $w_0 \approx 90 \mu\text{m}$. This can be considered the junction between regimes where the major contribution to beam divergence changes from increasing w_q to increasing n . The departure from the inverse proportionality of $W(z)$ and w_q drives the BPP up with n . This is shown in the upper branch of figure 6.12 b).

Supergaussian sources with the dipole phase (figure 6.10 b))

When n varies and the dipole phase is active, the absolute values of $W(z)$ are larger by the factor β than that compared to the supergaussian case with an inactive dipole phase. However the decrease in β as w_0 and n are increased over their full range is only $\approx 10\%$. This can be seen in the red curve in figure 6.11 b). This additional reduction in $W(z)$ will eventually plateau as $n \rightarrow \infty$. Ergo, the inclusion of the dipole phase again defocusses the beam by a factor β that changes with increasing n . The change in β is $\approx 10\%$ resulting in a BPP qualitatively similar to that expected with no dipole phase

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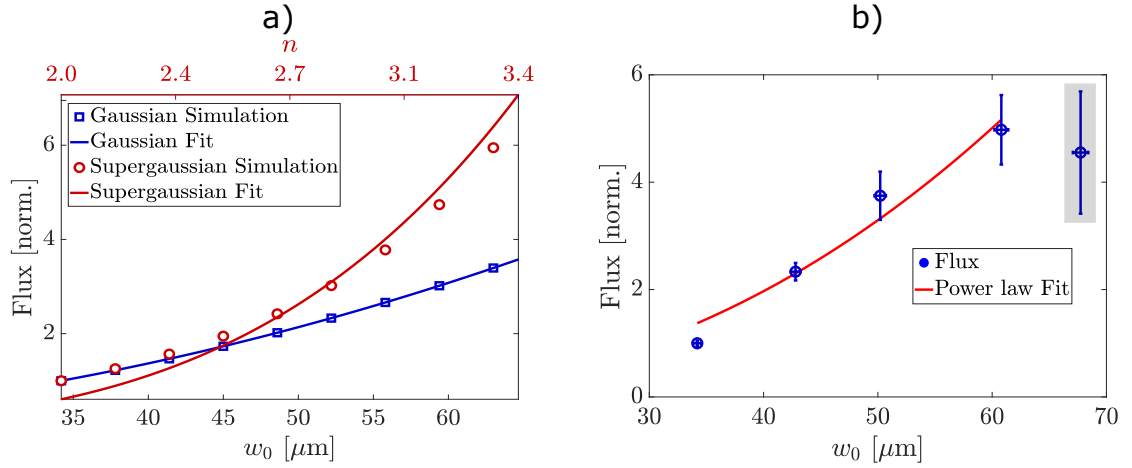


Figure 6.13: a) The calculated photon flux for the cases of Gaussian (blue) sources and supergaussian (red) sources. The fit was performed over the extended range of w_0 , only a subset of which is shown in subfigure a). Note the upper horizontal axis of increasing supergaussian order that applies only to the red supergaussian curve. b) Fit of a power law to the measured harmonic flux data shown in figure 6.7 a). The final data point contained within the grey shaded box has been excluded from the fit as the driver spatial shaping had begun to break down.

albeit approximately twice as large. The change in the dipole phase associated with increasing n lends a comparatively small contribution to beam divergence than that compared to w_q and n . This is shown in the lower branch of figure 6.12 b).

6.6.5 Detected Harmonic Signal Scaling

The photon flux calculated as the spatially integrated value of the harmonic intensity profiles, as a function of driver spot size w_0 is shown in figure 6.13 a). The blue data points represent the values corresponding to Gaussian sources with $n = 2$ of varying widths whilst the red data points represent the values corresponding to the supergaussian sources of varying widths and varying supergaussian orders. Fits were made to these simulated data according to a power law of the form $\text{Flux}(w_0) = a w_0^b$ with parameters a and b . When $n = 2$, the expected $b = 2$ scaling was found from the blue curve, whilst $b = 3.86 \pm 0.09$ from the red curve in the supergaussian case.

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The same power law fit was made to the experimentally detected harmonic flux data and is shown in figure 6.13 b). The final data point in the grey shaded box was excluded from the fit as it corresponds to the case of degraded shaping in the driver intensity profile. The fit to the experimental data reveals a power scaling of 2.3 ± 1.72 , the large uncertainties arising from the small number of data points and their large error bars in the dependent variable. Although a quadratic scaling relationship is encompassed in the uncertainty, there is a reasonable justification in both simulation and theory for this scaling to increase beyond quadratic. It is explained by the larger increase in w_q compared to w_0 over the range of n values used experimentally. With the improved scaling, a larger amount of flux can be generated from a harmonic source of increasing size. These factors combine to increase the brightness of the detected beam by a factor of 5 ± 0.65 when n is varied in tandem with w_0 .

6.7 Conclusions and Outlook

To conclude, a method for increasing the brightness of a harmonic source at the focus of a fixed lens has been presented. This was achieved through the use of a SLM to generate a supergaussian transverse driver focal intensity profile of increasing order and size. As the supergaussian order increased from $n = 2$ to $n \approx 3.4$, the produced harmonic source grew in size at a faster rate than the driver profile, accessing a greater number of harmonic emitters without changing the experimental footprint of the system. These factors combined to increase the harmonic flux as $w_0^{2.3 \pm 1.72}$ rather than the expected w_0^2 for a Gaussian beam. The BPP increased as the supergaussian order increases, but the harmonic divergence overall decreased as a result of the dominant effect of a spatially larger source. As a result, the brightness increases by approximately a factor of 5 without the need to change the focussing optic in the experiment.

The poor flatness of modern XUV optics will have increased the M^2 of the harmonic

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beam upon reflection. This effect would be deleterious for the absolute brightness, however, in this chapter only the relative change in brightness with driver shaping has been considered. Consequently the conclusions drawn remain valid. In the absence of shaping, a similar increase in brightness could be achieved with the use of a longer focal length lens. In order to achieve the factor of 5 increase in brightness demonstrated in this chapter a lens with a focal length 2.24 times longer would be required. However, this method sacrifices the more efficient distribution of IR photons in the harmonic source plane, an advantage that becomes more attractive when one considers the low conversion efficiency of HHG.

The harmonic source was reimaged, allowing the properties of the source to be measured directly, rather than inferred from those of the driver. The size of the harmonic source was determined from knife edge scans of the reimaged source fitted to equation 6.10, which assumes a supergaussian source, rather than the traditional Gaussian profile. The uncertainties in the deduced harmonic brightness are quite large; the largest contribution to these arises from the pointing instability of the harmonic beam which means the knife edge scans characterise the combination of multiple spatially displaced harmonic foci. This could be minimised with the appropriate stabilisation system.

It was found that the largest gains in harmonic source size were constrained to low driver supergaussian orders between 2 and 4. Beyond this range, the rate of harmonic source size increase slows asymptotically with increasing n . This implies that the advantages of shaping the driver transverse focal intensity profile from $I_{\text{sg}}(x, y, 2)$ to $I_{\text{sg}}(x, y, 4)$ may provide a larger and more easily accessible enhancement than shaping the profile to much higher values of n . This removes the need to solve an engineering challenge of achieving high order shaping, the complexity of which likely scales non-linearly with the desired supergaussian orders involved. As a result, with further refinement of the driver shaping to more accurately, repeatably and stably generate $n = 2 - 4$ supergaussians,

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the experimentally achieved brightness increase may be maximised toward its theoretical limit.

The fixed spatial footprint of this experiment lends a degree of flexibility to HHG setups. In this chapter, we have increased the driver spot size by a factor of 2 using only a phase-only SLM and a fixed lens. With the inclusion of the additional rings in the binary phase mask discussed in section 6.4, we anticipate that the degradation in the driver shaping may be mitigated, and larger w_0 values may be achieved, allowing access to a loose focussing geometry without sacrificing the compact nature of a HHG experiment.

Chapter 7

Engineered Illumination



THIS chapter is based upon the author's work detailed in reference [7]. It describes a method of manipulating multiple XUV beams without physical optics, thereby mitigating any additional reduction in efficiency of a HHG experiment, outside that implicit in the generation process itself.

As an example use of the increased degree of flexibility afforded by this approach, the spatial distribution of the Gaussian illumination impinging upon a sample in a FTH experiment is optimised to the sample shape. This enables the most efficient use of available photon flux, providing images with a superior S:N for a given exposure time.

7.1 Motivation

The low conversion efficiency in HHG from infrared to harmonic photons typically restricts harmonic sources to μW or nW of average power [87, 88]. This is in part due to the diffusion of the electronic wavepacket during its time in the continuum as described in section 3.2.2. Several approaches have been developed to increase the efficiency of HHG, including the use of phase-matching [90], quasi-phase matching [91, 92] and high-repetition rate driver lasers [93]. However, little attention has been given to

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controlling the spatial properties of harmonic beams, despite the fact that many applications, such as imaging, would benefit greatly from this degree of control: Being able to specify the spatial distribution of the illumination impinging upon an object would allow the optimum use of the limited available flux, mitigating the low conversion efficiency.

The manipulation of XUV radiation is hindered by the high degree of absorption in the majority of optical materials in this spectral range, where the real part of the complex refractive index approaches unity [200]. The resultant complexity and inefficiency of optics in this spectral range [201, 202] compounds the inefficiency of the HHG process itself, motivating the use of hybrid optics that combine diffraction and refraction [203, 204]. However, their construction remains complex and expensive [205].

Spatial shaping of XUV radiation has been demonstrated by splitting the fundamental beam in an interferometer to create multiple IR driver beams [206, 207], each of which produces a separate harmonic source. However, this implementation affords only limited control over the positions and relative powers of the XUV beams, and the number of independent beams cannot be changed easily. Alternatively, the spatial manipulation of a harmonic beam can be achieved with a split XUV mirror [208]. However, this optomechanical approach limits the number of independent beams to the number of mirror facets, which cannot be changed.

By spatially shaping the phase of the collimated driver beam with a SLM, the transverse intensity distribution in the harmonic generation plane is shaped. This in turn specifies the transverse intensity distribution of the harmonic source according to equation 6.8. When reimaged, a scaled version of the primary source is produced in the secondary source plane.

For the particular example of FTH with multiple reference holes as described in section

2.3.2, the traditional approach to uniformly illuminate both the object and all surrounding references would be to relocate the object a longitudinal distance Δz away from the secondary source plane. This ensures that the width of the incident beam $w(\Delta z)$ is large enough to illuminate all the necessary features with an appropriate amount of energy. To accommodate the spatially isolated object constraint, a binary transmissive mask is often applied to the object. To further satisfy the holographic separation condition, each reference must be separated from the centre of the object by a minimum of 3 object radii. This results in a significant amount of illumination impinging upon regions between the object and references that must necessarily be opaque. Ergo, to avoid this wastage of valuable flux, the ideal spatial shape of the illumination would consist of N independent but mutually coherent harmonic beams: One beam coincident with the object and $N - 1$ beams located at the reference holes.

The contrast in the recovered FTH image is proportional to the visibility of the interference fringes which is maximised when the amplitudes of the object and reference waves leaving the back of the sample are comparable [26]. In the case of an object with a complex index of refraction therefore, the image quality may require incident object and reference wave amplitudes that are unobtainable with a single Gaussian illuminating beam.

7.2 Beam Path

The beam path of the harmonic beam(s) in this experiment is identical to that shown in figure 4.2 from section 4.2. For convenience, that figure is replicated here as figure 7.1, which shows a schematic diagram of the beam path when $N = 2$ driver foci are created, both of which generate harmonic beams.

The detector used in this experiment was a back illuminated x-ray CCD (iKon-L,

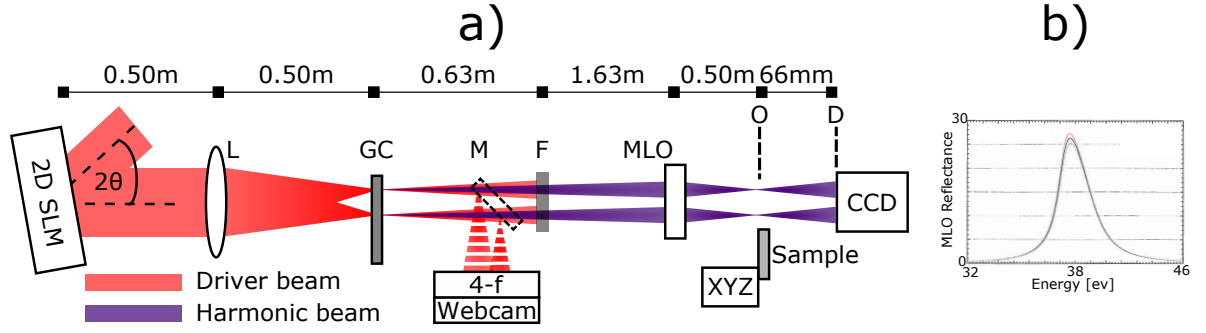


Figure 7.1: a) Schematic diagram of the experimental beamline downstream of the SLM. The labels are as follows: SLM is the spatial light modulator, θ is the angle of incidence to the SLM which was $< 5^\circ$, L is the IR focusing lens, GC is the nickel gas cell and M is a beamsplitter that when in place, deflects the driver beam onto a CCD through a 4-f unitary magnification reimaging line that allows inspection of the focal plane. F represents Aluminium filters, MLO represents a pair of multilayered XUV optics and XYZ represents a 3 axis optically encoded translation stage. The object and detection planes are denoted ‘O’ and ‘D’ respectively. b) Reflectance curve of both MLO.

ANDOR), comprising 2048×2048 square pixels of side $13.5 \mu\text{m}$ with a fill factor of 100%. During experimentation, the chip was cooled to -45°C .

7.3 Driver Phase Mask

To generate N driver foci, the SLM displayed a phase mask comprising N contiguous annuli. Each annulus encoded a different phase which dictated the transverse displacement of both the foci that were formed. This phase can be determined by observing the shift invariance of the Fourier transform. From equation 4.11 relating the field on the SLM to the field at the focus of a lens with focal length f , we can write

$$T_{\text{slm}}(x, y) \propto \iint_{-\infty}^{\infty} U(X, Y) \exp \left[i \frac{2\pi}{\lambda f} (Xx + Yy) \right] dXdY \quad (7.1)$$

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where (x, y) and (X, Y) are coordinates in the SLM and focal planes respectively. With the substitution $X' = X + \Delta X$, where ΔX is a constant, gives

$$T_{\text{slm}}(x, y) \propto \exp \left[i \frac{2\pi}{\lambda f} \Delta X x \right] \iint_{-\infty}^{\infty} U(X' - \Delta X, Y) \exp \left[i \frac{2\pi}{\lambda f} (X' x + Y y) \right] dX' dY. \quad (7.2)$$

Dropping the dummy variable X' , equation 7.2 reveals that a focal displacement of ΔX can be achieved by imprinting a phase gradient denoted by $\dot{\phi}_{\text{trans}}$ onto the beam in the conjugate domain [209]. Generalising to two dimensions, this phase gradient is expressed as

$$\dot{\phi}_{\text{trans}} = \frac{2\pi}{\lambda f} (\Delta X x + \Delta Y y). \quad (7.3)$$

The radii of the N annuli are initially calculated to ensure that each contains $1/N$ of the driver laser power. Assuming an incident collimated Gaussian beam, this requirement yields

$$\int_0^{2\pi} \int_{r_n}^{r_{n+1}} I_0 e^{-2\left(\frac{r}{w}\right)^2} r dr d\theta = \frac{1}{N} \int_0^{2\pi} \int_0^{\infty} I_0 e^{-2\left(\frac{r}{w}\right)^2} r dr d\theta, \quad (7.4)$$

where w is the e^{-2} radius of the beam, I_0 is the peak intensity, and r and θ are the radial and azimuthal coordinates in the SLM plane. This implies that

$$e^{-2\left(\frac{r_n}{w}\right)^2} - e^{-2\left(\frac{r_{n+1}}{w}\right)^2} = \frac{1}{N}. \quad (7.5)$$

Solving for r_{n+1} results in the recursion relation:

$$r_{n+1} = w \cdot \sqrt{\frac{1}{2} \log_e \left[\frac{1}{\exp\{-2\left(\frac{r_n}{w}\right)^2\} - \frac{1}{N}} \right]}, \quad (7.6)$$

where $r_1 = 0$ mm.

Equation 7.6 enables the remaining zone radii to be calculated ensuring a theoretically equal distribution of driver power exists within each zone. In practise however,

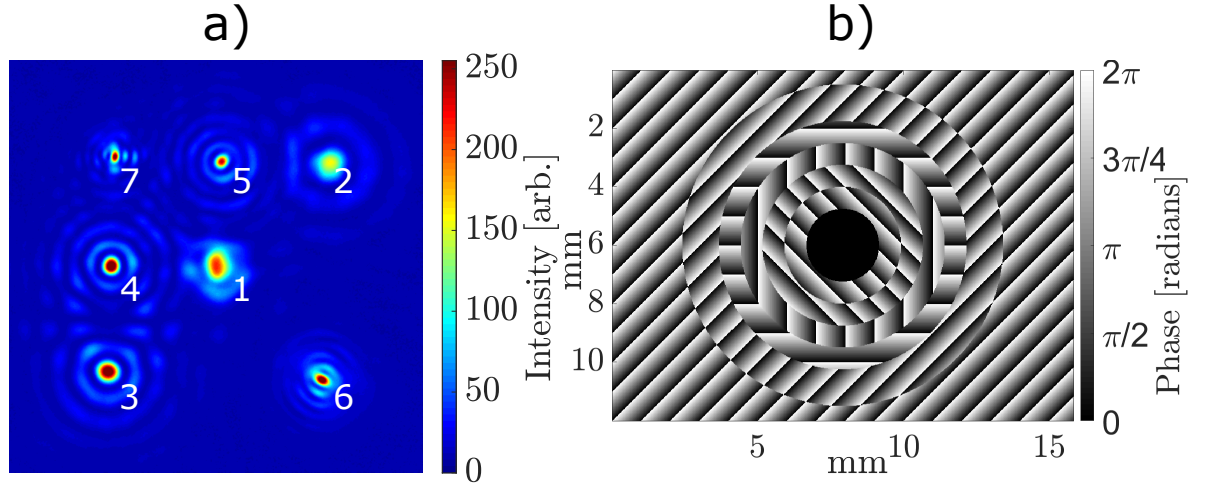


Figure 7.2: a) Measured transverse intensity profile of a $N = 7$ shaped driver beam at the focus of a $f = 50$ cm lens. b) The $N = 7$ phase mask imprinted onto the collimated Gaussian beam prior to focussing. The numbers shown next to the foci in a) indicate the corresponding annulus responsible for their creation in b), numbered in order of increasing annulus radius.

the zone radii were adjusted experimentally from initial values calculated from equation 7.6 to account for inhomogeneities of the incident beam. With this approach the spot size of the focus generated by each annulus will tend to decrease with n since $r_n > r_{n-1}$. To obtain foci of equal intensity the radii r_n can be iteratively optimised.

Figure 7.2 shows an example phase mask and the experimentally measured focus the shaped beam produces, showing 7 spatially separated foci. The positions of these foci can be controlled by adjusting the phase gradients in accordance with equation 7.3.

7.4 Driver and Harmonic Beams

For the FTH demonstration, the driver focus was separated into $N = 2$ foci. The phase mask therefore comprised two zones: an inner zone of radius 3.3 mm and no phase shift; and an outer zone, comprising the remainder of the mask which displayed a linear phase gradient of $\dot{\phi}_{\text{trans}} = 2356.2 \text{ rad m}^{-1}$ in $\hat{x}$ such that the first focus was on

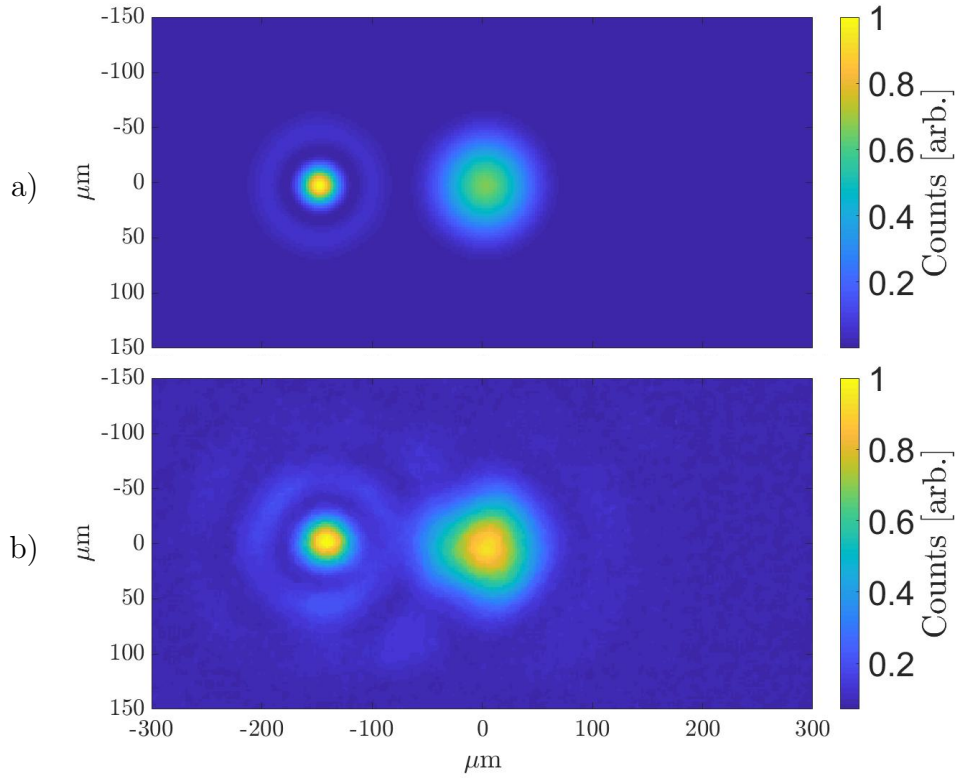


Figure 7.3: a) Simulation of the $N = 2$ driver focal intensity produced downstream of the phase mask described in the text. The design separation was $-150 \mu\text{m}$. b) Measured intensity profile of a $N = 2$ shaped driver beam.

axis and the second was designed to be $-150 \mu\text{m}$ off axis in $\hat{x}$. This is shown in figure 7.3.

Figure 7.4 a) shows the measured transverse line out of the driver focal distribution shown in figure 7.3 b). Overlaid are the corresponding simulated transverse line outs of the intensity and phase calculated by [ASM](#) propagation of a Gaussian amplitude with a $N = 2$ phase mask through a $f = 50 \text{ cm}$ lens. These data show the separation of the two foci to be $(146 \pm 1) \mu\text{m}$, which compares very well with the design separation of $150 \mu\text{m}$.

The phase steps in figure 7.4 a) occur at points where the simulated intensity drops to zero. Across the two intensity peaks the simulated phase remains constant as desired. [ASM](#) simulations of the longitudinal evolution of the phase indicate that it remains

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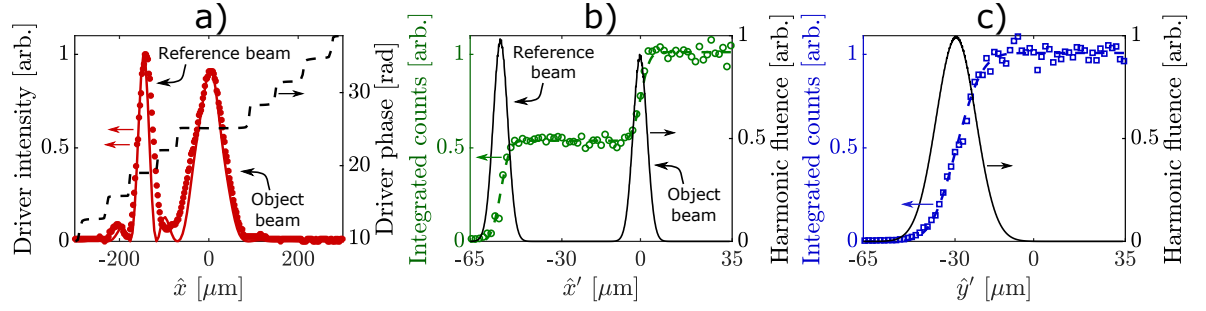


Figure 7.4: a) Measured transverse intensity profile (red dots) in the driver focal plane ($\hat{x}$) of the IR focus produced by a $N = 2$ SLM mask with parameters given in the text. The simulated intensity (red, solid) and simulated phase (black, dashed) profiles are overlaid. b) Measured variation of the harmonic signal as a function of the position of a knife edge inserted in the harmonic beam with its edge parallel to $\hat{y}'$ coordinates in the harmonic focal plane shown by green points, along with the fit (dashed green line). Only every third data point is shown, however the fit was made to the complete dataset. The transverse fluence profile of the harmonic beam in the $\hat{x}'$ dimension (solid black line) was calculated by differentiating [192] the corresponding fit. c) Shows the counterpart data to (b) but in the perpendicular transverse dimension.

circularly symmetric across the intensity peaks over a distance larger than the gas cell thickness.

Using 1 mJ of pulse energy, the driver focal intensity distribution shown in figure 7.4 a) was used to generate harmonics in 70 mbar of argon. In order to establish the spatial separation of the reimaged harmonic sources in $\hat{x}'$, the CCD signal was recorded as a function of the transverse position of a knife edge located 50 cm downstream of the MLFM. Figure 7.4 b) shows data together with a fit to the sum of two error functions [190]. Fitting to an error function assumes that the reimaged harmonic foci have Gaussian intensity profiles.

The green dashed curve in figure 7.4 b) reveals that $(53 \pm 0.5) \%$ of the harmonic energy is located in the reference beam. The spot sizes in $\hat{x}$ of the reference and object beams are $(5.4 \pm 0.7) \mu\text{m}$ and $(4.9 \pm 0.5) \mu\text{m}$ respectively. They are separated by $(52.4 \pm 0.5) \mu\text{m}$, which is consistent with the propagation of the harmonic beams over

the 2.27 m between the gas cell and the MLFM, and the demagnification of the MLFM. The scan along the $\hat{y}$ dimension shown in figure 7.4 c) reveals only one step, with a deduced spot size of $(7.6 \pm 1.3) \mu\text{m}$. Taken together, the results show the formation of two harmonic beams with beam centres separated along the $\hat{x}'$ -axis.

7.5 Sample Design

To provide a proof-of-principle demonstration of optimized **XUV** holographic imaging, **FTH** was undertaken with two **XUV** beams generated by the method described above. The sample comprised a $200 \times 200 \times 0.05 \mu\text{m}$ silicon nitride (Si_3N_4) membrane supported within a $5 \times 5 \times 0.525 \text{ mm}$ silicon wafer (Silson Ltd). This was sputter coated with gold of 150 nm thickness. A pattern was then milled through both the gold and Si_3N_4 substrate with a gallium-based focussed ion beam (FEI company) to act as an object.

The object was accompanied by near and far reference holes. Reference holes were milled, laterally displaced from the object. The near reference was located at $8 \mu\text{m}$ in $\hat{x}$ from the object to satisfy the holographic separation condition. The far reference was located at $52.4 \mu\text{m}$ in $\hat{x}$ from the object matching the harmonic foci separation for convenience: The sample was constructed after the separate harmonic beams had been demonstrated. An electron beam image of the sample is shown in figure 7.5.

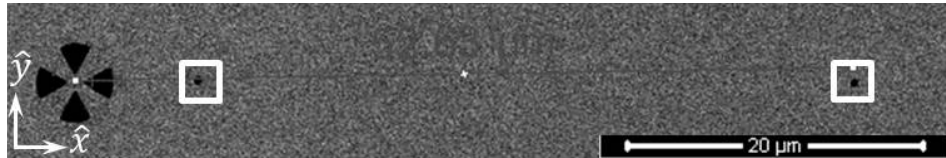


Figure 7.5: Scanning electron microscope image of the FTH sample that comprises the object to the left and two reference holes outlined by white squares. The faint horizontal grey line is a ruler drawn on the SEM image in the focussed ion beam software.

7.6 Comparison

The HHG beam shaping method presented here allows the object and reference holes to be illuminated efficiently by a pair of beams with user defined relative intensities. This is not possible with unshaped illumination without moving the sample away from the focus, which wastes XUV flux. Ergo, an experiment with shaped illumination should provide images with a significantly higher S:N than obtained from their unshaped counterpart.

For a fair comparison, both the relative and absolute intensities illuminating the object and reference pinhole should be comparable for the $N = 1$ (Unshaped) and $N = 2$ cases. Figure 7.6 illustrates the two cases schematically. In figure 7.6 b), the $N = 2$ case comprises spatially separate but mutually coherent object and reference beams. Their respective intensities at the object and far reference (R_2) are I_{obj} and I_{R_2} , with a ratio $\alpha_2 = \frac{I_{obj}}{I_{R_2}}$.

Figure 7.6 a) shows two cases corresponding to $N = 1$. For a single beam centred on the object (dashed curve) it is impossible to achieve $\alpha_1 = \alpha_2$, where $\alpha_1 = \frac{I_{obj}}{I_{R_1}}$. However, by moving the object longitudinally a distance Δz away from the focus, it is possible to achieve $\alpha_1 \approx \alpha_2$.

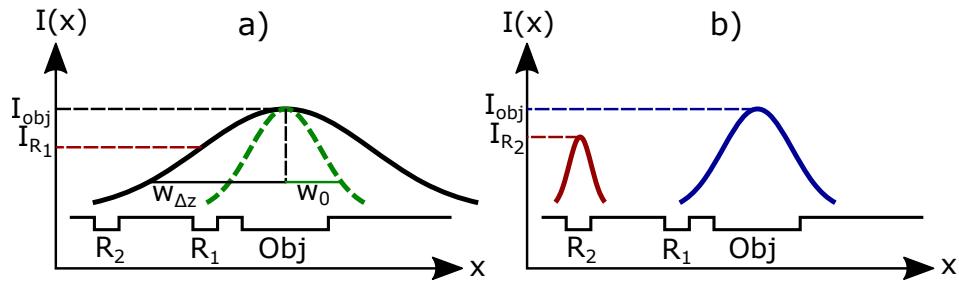


Figure 7.6: Schematic illustrations of: a) Illumination of the object and reference hole R_1 by a single beam; and b) illumination of the object and reference hole R_2 by a pair of harmonic beams. The labels ‘Obj’, R_1 and R_2 refer to the object, near reference and far reference respectively. More detail is in the text.

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The spot size of the unshaped **XUV** beam required to give $\alpha_1 \approx \alpha_2$ is

$$w_{\Delta z} = \frac{d}{\sqrt{\frac{1}{2} \log_e \left(\frac{1}{\alpha} \right)}}, \quad (7.7)$$

where d is the fixed distance of $8 \mu\text{m}$ between the centres of the object and R_1 . From figure 7.4 b), we have achieved $\alpha_2 = 0.88 \pm 0.02$ and hence $w_{\Delta z} = (31.6 \pm 0.2) \mu\text{m}$ is required for α_1 to match α_2 .

The longitudinal movement of the sample required to fulfil this criteria was found experimentally to be $\Delta z = 2.5 \text{ mm}$ downstream of the unshaped harmonic focus. This shift reduced the minimum theoretically resolvable feature size from 76 nm to 73 nm [20]. With this shift the object is illuminated by a spherical wavefront when it is placed out of the harmonic focus; simulations suggest the effect this has on the recovered image is negligible.

The procedure above allowed the ratio of the intensities of the beams illuminating the object and reference pinhole to be the same for the two configurations. However, for a fair comparison it is also necessary to use the same absolute intensities for the two geometries. With $N = 1$, the detected harmonic fluence was measured to be factor of 3.125 larger than in the $N = 2$ beam case, in part due to the spatial chirp imparted onto the beam by the shaping process. This was accounted for by increasing the pulse energy and adjusting the CCD exposure time for the $N = 1$ case to be $3.125^{-1} = 0.32$ times that required for the $N = 2$ case.

Subsequent results are therefore plotted against normalised exposure times where the normalisation factors for the $N = 1$ and $N = 2$ beam experiments are 0.32 and 1 respectively. This allows the comparison of results where the total harmonic flux incident upon on the sample is equivalent.

7.7 Results

Figure 7.7(a) and 7.7(b) show the detected holograms recovered for the cases of $N = 1$ and $N = 2$ beams for a normalized exposure times of 120, corresponding to 38.4 and 120 seconds respectively, in accordance with the constraints of section 7.6. When plotted on a log scale, the hologram shown in fig. 7.7(b) has visible interference fringes up to the outer limits of the CCD chip, corresponding to locations containing high spatial frequencies. Figures 7.7(c) and 7.7(d) show images of the object recovered for the $N = 1$ and $N = 2$ cases respectively. In the $N = 2$ case the recovered image is less noisy and exhibits a superior S:N, which has been defined as

$$\text{S:N} = \frac{\mu_{\text{sig}}}{\sigma_{\text{bg}}}. \quad (7.8)$$

in section 2.4 where μ_{sig} is the mean of the image signal and σ_{bg} is the standard deviation of the background. The $N = 1$ image has a more uniform intensity in the transparent regions of the object which results from the more uniform illumination provided by the larger beam used in this case.

To calculate the S:N of the images with equation 2.58, the image signal is taken to be the pixel values in figs. 7.7(c) and 7.7(d) within the area of the object found by performing a similarity transformation of the SEM image onto the retrieved images. The background is taken as the remaining pixels values not included in the image signal. Figure 7.7(e) shows the calculated S:N as a function of normalised exposure time for the cases of $N = 1$ and $N = 2$, within the constraints described in section 7.6. It can be seen that the S:N of the recovered image is larger, and increases with normalized exposure time more quickly, when the sample is illuminated by $N = 2$ beams.

Fourier ring correlation [74] curves were calculated for the data presented in figs. 7.7(a)

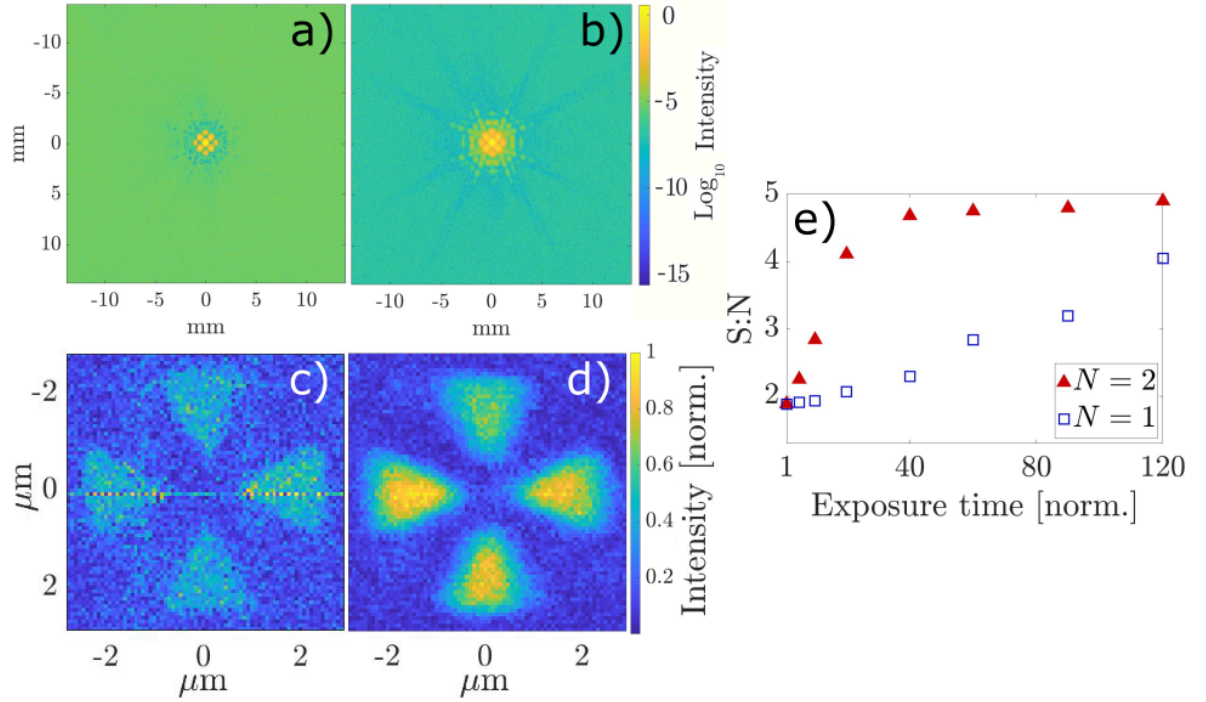


Figure 7.7: Detected holograms on a log scale for normalised exposures of 120 for $N = 1$ (a) and $N = 2$ (b) beams. Recovered images for normalised exposures of 120 for $N = 1$ (c) and $N = 2$ (d) beams. e) Variation of the S:N ratio as a function of normalized exposure time for the case of $N = 1$ (open blue squares) and $N = 2$ (full red triangles) beams.

and 7.7(b). Resolutions of 580 nm and 420 nm were found for ring correlation thresholds of 0.5 and 0.143 [210]. These values are consistent with the expected resolution limit in an FTH image of approximately 70% of the reference diameter [26]. The diameters of the reference pinholes used in this experiment were roughly 490 nm.

7.8 Conclusions and outlook

To conclude this section, a method for the programmable and independent manipulation of the relative positions and intensities of multiple harmonic beams has been presented. The potential advantages of this method were demonstrated by FTH imaging of an object with two, independently-controllable HHG beams. It was found that FTH imaging with two HHG beams yielded reconstructed images with superior S:N than equivalent

imaging with a single beam under comparable circumstances.

Pending the next generation of higher damage threshold LCoS adaptive optics [167], the number of HHG beams that can be generated with this method may be restricted. The throughput of the beamline in figure 7.1 is limited by the damage threshold of the LCoS SLM to approximately 1 W for 1 kHz repetition rate 30 fs duration laser pulses. For harmonic generation to occur, the peak intensity of each beamlet must exceed a threshold of approximately $1 \times 10^{14} \text{ W cm}^{-2}$ and with the power and focussing geometry available to the author, only two harmonic beams could be generated. Tighter focusing would allow the generation of more harmonic beams, the maximum number being limited by a requirement that the Rayleigh range of the driver foci must be larger than the thickness of the gas cell. Alternatively, static bespoke phase plates with higher damage thresholds could be used to generate an increased number of harmonic beams albeit at the expense of flexibility.

The work described here paves the way for new classes of experiments enabled by the ability to generate multiple, independently controlled harmonic beams. In combination with other advances such as shaping the driver foci in both space and time [211–213], additional information may be revealed in HHG experiments ranging from interferometry [206] to high harmonic metrology [207].

Within the field of imaging, multi-beam ptychography [214, 215] and blind digital holography [216] in the XUV would benefit greatly from the use of multiple harmonic beams. High resolution images could be obtained at a fraction of typical exposure times, mitigating limitations of stability in long exposure experiments. In addition, the maturity of high harmonic sources has led to an abundance of experiments that would benefit from multiple, controlled XUV beams [83, 217, 218].

Chapter 8

Conclusion



HIS thesis has motivated and described the construction of an XUV lensless microscope capable of imaging with a resolution of approximately 3 times the imaging wavelength of 32 nm. Experimental results comprising the first ever demonstration of lensless imaging in the XUV within the author's research group have been provided. Further, two novel advances in the manipulation, flexibility and enhancement of XUV radiation using a SLM have been presented. The highlights of these results will be summarised here. The final section of this chapter will outline how this work could be extended.

8.1 Summary of Results

Chapter 5 presented the first results of CDI and FTH imaging performed by the author. The binary transmissive apertures employed by the author acted as a testbed to establish the capabilities of the lensless imaging system. Resolutions of the order of 3 wavelengths were reported, as evidenced by the FRC curve in figure 5.7 c) and the reconstruction of certain features of known size present in the scanning electron microscope images.

Chapter 6 described a novel method for increasing the brightness of a harmonic beam

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8.1. SUMMARY OF RESULTS

by a factor 5 ± 0.65 at the focus of a fixed lens. This was achieved by imprinting a single radially symmetric binary phase step of magnitude π into the collimated driver beam before the HHG interaction. This enlarged the spot size and changed the transverse spatial profile from a Gaussian to a supergaussian. The harmonic source size grew at a faster rate than the driver spot size when the order of the supergaussian profile was increased from $n = 2$ to $n \approx 3.4$. This accessed a greater number of harmonic emitters at a faster rate, changing the scaling of detected flux with driver spot size from a power of 2 to a power of 2.3 ± 1.72 without the need to change the focussing optic.

Chapter 7 described a novel method for manipulating a broadband beam of harmonic radiation by spatially shaping the source distribution. This technique enables all constituent frequencies of the harmonic beam to be manipulated simultaneously without the introduction of angular dispersion commonly associated with post generation manipulation by refractive optics. This was achieved by imprinting a phase mask comprising N annular zones onto the driver beam prior to the HHG interaction. Each zone contained a phase gradient that transversely displaced a user specified amount of driver energy in the harmonic generation plane, directly moulding the harmonic source to the desired design. Further, using $N = 2$ independent harmonic beams, an improved FTH experiment was demonstrated. For a fixed total harmonic fluence, images with a far superior S:N were recovered using illumination redistributed with this method than were without. A corollary of this result is that it allows a given S:N to be achieved with a shorter exposure time, advantageous for experiments sensitive to drifts in the beam pointing and sample stability.

The advances detailed in chapters 6 and 7 should provide groups around the world with a new level of flexibility in HHG experiments that facilitates a greater degree of control over harmonic radiation.

8.2 Future Prospects

With the aforementioned advances, avenues for new experiments involving multiple and or brighter harmonic beams have been opened. In a non exhaustive list, the author provides a few examples of experiments that may prove useful and or yield new, interesting results:

1. For any work involving a [SLM](#), the author would advise the installation of a feedback loop to automatically correct for aberrations in the driver beam prior to the [HHG](#) interaction, saving the user a great deal of time and effort. To accomplish this, a feedback loop would inspect the collimated beam profile at multiple longitudinal points downstream of the [HHG](#) interaction and adjust the aberration correction displayed on the device to optimise the cylindrical symmetry of the beam prior to focussing.
2. In section [4.4.4](#) a method of achieving both amplitude and phase control over the driver beam was outlined. This did not lead to the desired level of control at the focus however due to the inefficiency of the operating principal. However, more efficient alternate methods of achieving complex control such as that detailed in section [4.4.4](#) may yet enable such control at the focus of a focussing lens. This would enable the phase and amplitude of the focal intensity profile to be decoupled and manipulated independently during [HHG](#). This may enable the user to properly investigate the behaviour of the intensity dependant dipole phase.
3. As an extension of point 2, the control over the complex field may enable the user to modulate the phase of the harmonic beam illuminating a lensless imaging target in a known and predictable manner. This a priori information could then be fed into a [IPRA](#) and the rate of convergence recorded. It is reasonable to assume that the convergence would be more reliable and or faster with this additional information.

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4. The field of information transport using orbital angular momentum (OAM) is an emerging method in information transport using radiation [219]. It has been found that during HHG, OAM is multiplied by the harmonic order when it is imprinted into the harmonic beam [220]. As a result, investigations of how to best imprint and decode OAM from the XUV could reveal interesting results.
5. As an extension of the work presented in chapters 6 and 7, an investigation into the behaviour of phase matching with shaped driver profiles may be performed. If some form of phase matching could be achieved, the restriction of using a gas cell with a thickness much less than the driver Rayleigh range may be lifted. This would enable more harmonic flux to be generated from shaped harmonic beams, further increasing the generated harmonic flux along with the advantages already outlined.

8.3 Quotes

This section contains some notable quotes and excerpts from conversations involving Dr David Lloyd (DL), Prof. Simon Hooker (SH), Dr Kevin O’Keeffe (KO) and Mr Daniel Treacher (DT).

DL: *"The laser’s broken."*

DT: *"What’s wrong with it?"*

DL: *"There’s no light coming out of it."*

DL: *"You just want to swim in a lake infested with dinosaurs and you’re not happy unless you’re stabbing someone."*

DL: *"So what’s ‘Barry’ short for?"*

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8.3. QUOTES

DT: "*Definitely 'Barrold'. It has to be that.*"

DL: "*You know that guy from Whiplash? J. K. Simmons? Am I that guy?*"

DT: "*I didn't want to say it, but yes. Yes you are.*"

DL: "*That's not my tempo.*"

DL: "*So, what's the plan?*"

DT: "*Erm..."*

SH: "*I'd be surprised if that took more than a few weeks at most.*"

DT: "*Well, I'm full of surprises.*"

DL: "*So either this'll work, or my corpse will fly across the lab and you'll have to drag my body out to the rubbish.*"

DT: "*Sounds good. Go for it.*"

KO: "*Ooooooooo.*"

DT: "*Ooooooooo.*"

DT: "*Here's an idea, what if we did the whole thing in reflection geometry?*"

DL: "*That is, without doubt, the worst idea you've ever had.*"

DL: "*Good work.*"

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