Development of novel methods for broadband lensless imaging

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Abstract

We developed two-pulse imaging, which enables robust and accurate lensless imaging with broadband spectra, without a priori knowledge of the spectrum for image reconstruction. We have shown a method to use supercontinuum coherent light sources together with HERALDO, a Fourier transform holographic imaging technique that uses extended reference structures. We have demonstrated ultra-broadband imaging using HERALDO with 1-D and 2-D reference structures at visible wavelengths. Finally, we have demonstrated our two-pulse method at extreme-ultraviolet wavelengths generated by a broadband HHG source. At these wavelengths no suitable optics are available for high quality image reconstruction. Nevertheless, we have successfully retrieved high-quality images using lensless imaging in the Fresnel regime, with non-collinear beams.
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CHAPTER 1

INTRODUCTION

Ever since the development of the first microscopes in the 16th century, there has been a continuing drive towards higher resolution. Presently, the quality of imaging optics has improved to the point where state-of-the-art microscopes achieve a resolution of several hundred nanometers, which is mainly limited by the wavelength of light. One possible way to reach even better resolution is to develop microscopes that operate with shorter wavelength radiation, such as extreme-ultraviolet or even X-ray radiation. In the X-ray regime, element specific images can be acquired by tuning the frequency of the light to atomic resonances. One of the applications is imaging of biological samples, for which the water window is particularly interesting. The water window is the spectral range located between 2 and 4 nm, where oxygen (thus also water) is transparent, but nitrogen and carbon are still absorbing. This gives rise to good opportunities to image biological samples.

For imaging with visible wavelengths excellent diffractive optics, such as lenses are available. However, imaging optics for X-ray wavelength ranges are much more problematic to manufacture, due to the lack of suitable materials for lenses, as well as the extreme tolerances needed for the surface quality at these short wavelengths.

Lensless imaging methods provide a promising alternative in this case\(^1\). In lensless imaging, the imaging optics are essentially replaced by computer algorithms, and the ‘microscope’ consists of only a light source and a camera. The final step in these algorithms is that the recorded diffracted light field is numerically propagated back to the object position. Therefore, the main requirement for image reconstruction is that the phase of the recorded diffraction pattern can either be measured directly, or retrieved from the measured diffraction intensity through numerical means.

Numerical phase retrieval algorithms have been developed and used successfully in coherent diffractive imaging (CDI)\(^2\)-\(^5\). One important ingredient is that the imaging conditions are set up such that they fulfill some support requirement, which usually means that the sample is an isolated sample in an empty background that fills more than half the field-of-view.

Direct phase measurements can be performed in a lensless geometry by adding a reference scatterer close to the sample. This reference scatterer provides a known reference wave which interferes with the light diffracted from the sample. The resulting far-field interference pattern is an off-axis hologram, and encodes the phase difference between the reference and object fields. A 2D Fourier transform of this hologram directly provides both amplitude and phase of the diffracted object field, and this method is known as Fourier transform holography (FTH)\(^6\)-\(^9\).

A major requirement of any lensless imaging method is that the light source is fully coherent (both spatially and temporally), as any form of decoherence directly results in a decreased visibility of the diffraction pattern. This has limited the application of lensless X-ray imaging to large facilities such as synchrotrons and X-ray free electron lasers (XFELs), but recent breakthroughs in high-harmonic generation (HHG) have led to the development of fully coherent table-top soft-X-ray sources. While HHG sources exhibit high spatial coherence, their temporal coherence is very short. This is intrinsic to the HHG process, which occurs on attosecond timescales and generates broadband light. In practice this means that lensless imaging with such a broadband source will result in a superposition of diffraction patterns at different wavelengths. While spectral filtering before imaging is possible, it results in a highly inefficient use of the available photon flux.
Chapter 1 Introduction

We developed a two-pulse lensless imaging method, which enables spectrally resolved lensless imaging with ultra-broadband light sources [9]. This approach combines lensless imaging with methods from Fourier-transform spectroscopy, and allows efficient use of the full spectrum of ultra-broadband sources for lensless imaging. We demonstrated this method in the far-field (Fraunhofer diffraction) using an octave-spanning supercontinuum light source. Secondly, we extended the method to Fourier-transform holography with extended reference structures (HERALDO). Furthermore, we discuss the concept of two-pulse lensless imaging, as well as our experimental results and the technical requirements for performing HERALDO with broadband sources using both one-dimensional (1D) and two-dimensional (2D) reference structures, and show several methods to increase the signal-noise ratio in the reconstructions. Finally we demonstrated two-pulse Fresnel diffractive imaging with a XUV source down to a wavelength of 47 nm.

1.1 Phase problem

When an object is illuminated it can reflect, diffract or absorb the light. The shape and structure of the object are imprinted on the phase front of the light. The light propagates perpendicular to the phase front, so the intensity distribution and phase front transforms while propagating through space. In classical imaging setups a lens is used to diffract the light is such a way that an image of the object is formed after passing through the lens.

In lensless imaging, rather than using a lens to create an image on a CCD, the intensity of the light diffracted on the sample is captured directly in the far field. An image can then be obtained through numerical back-propagation of the recorded field, provided that both its intensity and phase are known. The phase of the light field needs to be retrieved as well, as this encodes the propagation of the light. To retrieve the phase two approaches are currently available: iterative phase retrieval and holography.

![Figure 1.1: Artist impression of lensless imaging. A sample (left) is illuminated with a coherent light source. The scattered light is directly captured with a detector (right).](image)

In this section I introduce a mathematical framework describing light propagation and image formation. Throughout the whole section a typical lensless imaging setup as depicted in figure 1.1 was considered. A 2D object \( o(x, y) \) was illuminated. The transferred light, an electric field, propagated to the detector at distance \( z \) [8, Chapter 4]:

\[
E(x', y') = \frac{z}{j \lambda} \int \int o(x, y) \frac{\exp(j kr_0)}{r_{01}} \, dx \, dy
\]

where \( j = \sqrt{-1}, k \) the wavenumber of the light and distance \( r_{01} = \sqrt{x'^2 + (x' - x)^2 + (y' - y)^2} \).
1.2 Iterative phase retrieval techniques

The detector measured the intensity of the electric field:

\[ I(x', y') = |E(x', y')|^2 \]  \hspace{1cm} (1.2)

To reconstruct the object the direction in which of the light was propagating, the phase, should be known. The phase was lost when capturing the light with the detector. To recover this phase different methods where known: iterative phase retrieval algorithms and holography techniques.

**1.2 Iterative phase retrieval techniques**

![Diagram showing iterative phase retrieval algorithm]

Figure 1.2: Gerchberg Saxton type iterative phase retrieval algorithm. The amplitude is replaced by the measured data in the detector plane. In the image plane a support constraint is applied.

Many iterative phase retrieval techniques are based on the Gerchberg-Saxton[10] algorithm. The original light field is recovered in successive iterations. In every iteration the reconstruction is projected back and forth, while applying constraints in the object and detector plane. In the detector plane the constrain is given by the measured data, while the constraint mostly consists of a priori known information about the object, for example that the space surrounding the object is black. An iteration of the original GS algorithm consists of different steps, visualized in Fig. 1.2:

1. Apply (measured) information about recording plane
2. Propagate back to object plane
3. Apply support constraint in image plane
4. Propagate to recording plane

After many iterations this algorithm might converge [11] and reconstruct the image.

Often, these kind of algorithms need some a priori knowledge about the object, this information is called the support. An often used constrained is that the space around the object is black, or the object is real valued, i.e. it has a constant phase. The requirement of such support constraints is one of the drawbacks of iterative phase retrieval algorithms.
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To capture the light from the sample the detector can be placed in the near field (Fresnel diffraction) or far field (Fraunhofer diffraction). In the Fresnel zone, the intensity and the phase of the light field are strongly coupled. Small changes in detector distance and in wavelength will then result in different intensity patterns. Iterative phase retrieval algorithms can be used for both near and far field intensity patterns.

There are different approaches to overcome the limitation of an a priori known support constraint by combining several measurements. One of them is ptychography [5, 12], where the detector is placed in the far field of the sample. Multiple diffraction patterns are obtained for different lateral positions of the sample. The overlapping parts are used as support for the other measurements.

Two other approaches are available for near field imaging. In the first approach the intensity of the light field is measured at multiple distances from the object [13, 14]. In this approach the measurement at one distance acts as a support for other distances. The intensity data from the first distance is propagated to the next distance by the Fresnel propagation integral in Eq. 1.1 depicted in figure 1.3. Here intensity is replaced by the measured intensity data, while the propagated phase is retained. For this approach either the detector or the sample has to be moved during the experiment.

Another approach to overcome the support limitations is multi-wavelength Fresnel imaging [9]. In this scheme the intensity of the light is measured at different wavelengths, and therefore no detector or sample movement is necessary. When the sample imprints the same phase on the light for the used wavelengths, the measurement at one wavelength can act as a support for the other wavelengths. In this scheme the measured intensity data for one wavelength is propagated back from the measurement plane to the object plane. Now the wavelength in the Fresnel propagation integral in Eq. 1.1 is changed and the field is propagated forward to the detector plane, see figure 1.4. Successively the measurement at this wavelength can be used as support: the intensity is replaced by the measured intensity while the phase information is retained. By propagating between several wavelengths, typically 2-5, the object is reconstructed in a fairly low number of iterations, typically

Figure 1.3: Simulation of iterative phase retrieval using measurements at different distances from the object. In every iteration the image is propagated between the measured distances. Every measured data at a certain distance acts as support for the data measured at other distances.
1.3 Holography

Direct phase measurements can be performed by lensless Fourier transform holography, in which the diffraction pattern is recorded in the far field of the sample and where a known reference wave is used to encode the phase, shown in figure 1.1. In many realizations this reference wave is generated by a reference scatter close to the sample. The simplest realization of such reference is a pinhole in transmission, generating a spherical wave. Because no iterative steps are necessary this is a direct and deterministic technique.

Interference between the object wave \( o(x, y) \) and the known reference wave \( r(x, y) \) provides a phase measurement. The superposition \( e(x, y) \) of both light fields at the sample plane propagates to the recording plane as \( E(k_x, k_y) \). When the light is recorded in the far field of the sample, the Fraunhofer approximation applies. Therefore the propagation from the object plane to the recording

Figure 1.4: Iterative phase retrieval using measurements at different wavelengths. In every iteration the image is propagated from one wavelength to the other measured wavelengths via the image plane. Here the measured data is used as support.

This approach only works if the sample imprints the same phase on the light for the used wavelengths.

All these iterative phase retrieval algorithms depend on some support, which should be known in advance, or constructed by movement of the detector or sample, or retrieved by measuring at different wavelengths. Moreover all implementations of such algorithms, for example error reduction (ER) or hybrid input-output (HIO), have problems with converge and stagnation if no tight support is used [11].
plane, described by Eq. 1.1, reduces to a Fourier transformation:

\[
E(k_x, k_y) = \frac{e^{j\pi}}{\lambda} \int \int (o(x, y) + r(x, y)) \frac{\exp(jkr_0)}{r_0^2} \, dx \, dy
= \exp(jkr) \exp(j\frac{k^2r_0^2}{\lambda^2} + k_0^2) \int \int (o(x, y) + r(x, y)) \exp \left(-\frac{2\pi}{\lambda^2} (k_x x + k_y y)\right) \, dx \, dy
= A(k_x, k_y) F\{o(x, y) + r(x, y)\} = A(k_x, k_y) (O(k_x, k_y) + R(k_x, k_y))
\]

where \( A(k_x, k_y) \) contains a phase factor and a constant scaling depending only on \( z \).

The detector measures the intensity, the diffraction pattern, an example shown in figure 1.1 on the detector:

\[
I(k_x, k_y) = |E(k_x, k_y)|^2 = |O(k_x, k_y) + R(k_x, k_y)|^2
= O(k_x, k_y) O^*(k_x, k_y) + R(k_x, k_y) R^*(k_x, k_y) + O(k_x, k_y) R^*(k_x, k_y) + R(k_x, k_y) O^*(k_x, k_y)
\]

To obtain a reconstruction, the measured intensity is propagated back to the sample plane by an inverse Fourier transformation \( F^{-1} \). However, because only the intensity \( I \) of the light field can be measured directly, the light field at the sample plane is not reconstructed directly, but:

\[
F^{-1}(I(k_x, k_y)) = o \circ o + r \circ r + o \circ r + r \circ o
\]

where the cross-correlation function between \( o(x, y) \) and \( r(x, y) \) is defined as:

\[
o \circ r \equiv \int \int o(\xi, \zeta) r^*(\xi - x, \zeta - y) \, d\xi \, d\zeta
\]

where \( * \) denotes the complex conjugate. Notice that if \( r \) is chosen as a delta function, \( o \circ r \) and their complex conjugate reduce to \( o \) and \( o^* \) and therefore the object can be retrieved directly.

### 1.4 HERALDO

Pinhole Fourier-transform holography provides a direct reconstruction using conceptually simple mathematics. However, manufacturing a delta function or a pinhole next to the object is challenging and a finite point source will blur the image and might reduce the resolution. A larger source will decrease the resolution, but might increase the flux of the reference beam and so increase signal of the reconstructed object. Creating an extended reference structure like a line segment or a trapezoid structure provides much more flexibility [16].

A robust method to reconstruct images from holograms created with extended references is HERALDO, a differential operator based technique, which was developed by Podorov et al. [16] for square references and generalized to arbitrary references by Guizar-Sicairos et al. [15]. The main advantage is that many extended reference sources are easier to manufacture than pinhole reference sources. When an extended object is used as reference in classical holography, the resulting reconstruction is smeared out by the reference. With HERALDO a differential operator is applied to use a sharp feature of the extended reference to reconstruct the object.

The key point of this method is the fact that a sharp feature in the reference object can be reduced to a delta function by applying differential operators. A 1D example is a line segment, which can be described as a rect function (\( \text{rect}(x) = 1 \) for \( |x| < 1 \) else \( 0 \)), shown in figure 1.5a. When applying a differential operator this reduces to two delta functions, shown in figure 1.5b:

\[
\frac{\text{drect}(x)}{dx} = \delta(x + \frac{1}{2}) - \delta(x - \frac{1}{2})
\]

For higher dimensional reference structures, such as 2D squares, multiple differential operators should be applied to reduce a corner to a delta function. An example with a parallelogram as
Figure 1.5: Example of derivatives used by HERALDO. (a) A derivative of a block function reduces to (b) a positive and negative delta function. (c) A directional derivative reduces a parallelogram to (d) a negative and a positive line. A second derivative in the other direction reduces the two lines to (e) two positive and two negative delta functions.

reference is shown in figure 1.5-е. A wide class of objects are suitable as reference, for example a nano tube or a AFM tip. The sharpness of the edge of sharp feature used to reconstruct the image is a fundamental limit of the resolution of the reconstruction. An unsharp edge results in a wider delta function, illustrated in Fig. 1.6, which blurs the reconstruct.

A full mathematical treatment was given in [15], a summary follows. For a given reference \( r(x, y) \) a linear differential operator \( \mathcal{L}\{\cdot\} \) is defined for which the reference reduces to a sum of delta functions and some other function \( g(x, y) \):

\[
\mathcal{L}\{r(x, y)\} = A\delta(x - x_0)\delta(y - y_0) + g(x, y)
\]  
(1.8)

where \( A \) is a constant and

\[
\mathcal{L}\{r(x, y)\} = \sum_{k=0}^{n} a_k \frac{\partial^n}{\partial x^n \partial y^k}
\]  
(1.9)

is a linear differential operator and \( a_k \) are constant coefficients. When this operator is applied to the inverse Fourier transform in equation 1.5 this results in a sharp reconstruction:

\[
\mathcal{L}\{o \odot o + r \odot r + o \odot r + r \odot o\} = \mathcal{L}\{o \odot o\} + \mathcal{L}\{r \odot r\} + o \odot g + g \odot o + A^*o(x + x_0, y + y_0) + Ao^*(x_0 - x, y_0 - y)
\]  
(1.10)

the last two terms in equation 1.10 are the reconstruction and the complex-conjugated reconstruction of the object.

HERALDO provides much flexibility in the choice reference and setup layout, and both inline and off-axis holograms and both dark and bright field reconstructions can be obtained. In the first HERALDO paper by Podorov et al [16] a simulation of a bright field inline hologram was shown. An absorbing object was placed inside an transmitting square reference structure. This was also experimentally demonstrated [17][31]. In many other HERALDO experiments an off axis dark field setup is used [17][23], where both the reference and sample are transmitting light, while the separating space is dark. Guizar-Sicairos and Fienup [15] propose a more flexible bright field setup,
where both the object and the reference are absorbing and the reference is formed by an obscuring a part of the beam by an edge, for example a sharply pointed atom force microscope tip.

The main advantage of HERALDO over iterative phase retrieval approaches is that it provides a direct phase measurement, so it does not suffer from problems related to iterative phase retrieval algorithms, such as convergence and stagnation problems. Furthermore HERALDO references have less manufacturing limitations than conventional pinhole holography and HERALDO needs less time consuming calculations than extended reference holography by means of deconvolution [24-26].

HERALDO has been experimentally demonstrated with visible light [17], but also with X-rays from a synchrotron source [21, 31] and with single pulses from a soft X-ray HHG setup [18]. It can also be combined with polarization dependent contrast techniques, such as x-ray magnetic circular dichroism [22, 23].

1.4.1 Separation conditions

A full reconstruction of the object is retrieved, if we are able to distinguish the last two terms of equation 1.10 from the other auto and cross correlation terms. This is possible when the separation conditions [15] are fulfilled:

1. The reconstructed object should not overlap with its own autocorrelation. Consider an object that can be contained in a circle with radius \( \rho \), then the distance between the center of the object and reconstructing delta function should be more than \( 3\rho \).

2. The reconstructed object should not overlap with the autocorrelation of the reference structure \( r \otimes r \). If this cannot be avoided, then the autocorrelation of the reference structure might be subtracted if prior knowledge of the reference structure is available. This information can be recorded by illuminating the reference structure without the object in place.

3. The reconstructed object should not overlap with other reconstructions \( \mathcal{L} \{r \otimes \alpha\} \)

4. The reconstructed object should not overlap with other terms \( g(x, y) \) of the reference.
1.5 Two-pulse imaging

In a setup with an obscuring tip and bright background, the reconstruction always overlap with the autocorrelation of the illuminating beam. This autocorrelation function can be obtained separately by autocorrelating a measured beam profile.

1.5 Two-pulse imaging

Figure 1.7: (a) Sample containing star and triangular HERALDO reference. (b) Simulation of a far-field monochromatic diffraction pattern at a wavelength of 800 nm. (c) Simulation of far-field broadband (400 nm wide spectrum, centered at a wavelength of 600 nm) diffraction pattern of a.

Figure 1.8: Basic principle of Fourier transform spectroscopy. (a) Two pulses with a time delay $T$. (b) The spectrum of such a pair consists of the spectrum of a single pulse, plus a modulation with a frequency proportional on the inverse time delay.

Lensless imaging techniques usually depend on the diffraction of light on a sample. As the diffraction angle is wavelength dependent, this leads to problems when the source spectrum is not monochromatic. In Fig. 1.7 a numerical simulation is presented that shows the influence of the source spectrum on a resulting diffractoin pattern. A sample structure is depicted in Fig. 1.7, and a monochromatic diffraction pattern of this sample is displayed in Fig. 1.7. Since broadband diffraction patterns consist of a superposition of monochromatic diffraction patterns they will exhibit a structure that is smeared out in the radial direction. An example of such a broadband diffraction pattern is shown in Fig. 1.7, in which this effect is clearly visible. Such smearing hides all sharp features, and hinders or even inhibits image reconstruction. This effect obviously limits high-quality HERALDO image reconstruction.

From an application perspective, many coherent X-ray sources, including HHG, produce only broadband light. Current X-ray imaging approaches using synchrotron or XFEL sources therefore include a monochromator for this reason. But this approach is highly inefficient, as most of the source spectrum is filtered away. This is especially problematic when the starting flux is already low, which is typically the case for HHG sources.
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Our approach to acquire monochromatic diffraction patterns is two-pulse imaging [9]. This method depends on the interference between two coherent pulses with a time delay $T$, of which the principle is shown in Fig. 1.8. Depending on the time delay $T$ different wavelengths will interfere constructively and others destructively, resulting in spectral interference as shown in Fig. 1.8b. By measuring diffraction patterns over a range of time delays, the relative intensity of all spectral components scattered onto a pixel can be revealed by Fourier-transforming the time-domain signal. This results in a collection of quasi-monochromatic diffraction patterns that spans the full source spectrum, with a spectral resolution limited only by the scanned time delay. Each of these quasi-monochromatic diffraction patterns enables image reconstruction by HERALDO as well as for other lensless imaging techniques.

An advantage of this approach is that the full source spectrum is used throughout the entire measurement, which is an advantage that is common to many Fourier-transform-based techniques [27, 28]. This results in an efficient use of the available photon flux, and makes the method suitable for implementation with low photon flux sources as HHG sources [9]. Furthermore, the spectrum of the source does not have to be known in advance, and this method is remarkably robust against jitter and noise in the spectrum [9]. Moreover this method has many other features in common with Fourier-transform-based methods [27, 28], including insensitivity for constant background stray-light.

The highest frequency (shortest wavelength) that can be resolved using this Fourier transform method is given by the smallest possible time delay $T_{\text{min}}$:

$$\nu_{\text{max}} = \frac{1}{2T_{\text{min}}} \quad (1.11)$$

The factor two follows from the Shannon-Nyquist sampling limit. All lower frequencies are resolved.

The resolution, the smallest frequency step, that is obtained depends on the longest time step $T_{\text{max}}$:

$$\Delta \nu = \frac{1}{2T_{\text{max}}} \quad (1.12)$$

1.6 Maximum resolution

In order to retrieve an image of an object, the object is illuminated with a light source (unless the object itself is a light source). By reflection, diffraction of absorption in the object, the intensity and direction of the light are altered. The scattered light leaves the object at certain angles, depending on the spatial frequencies contained in the object. If the angle of the scattered light is too large, it will not get captured by the imaging system, because of a limited size of the lens or the detector. The maximal angle at which is captured is given by the numerical aperture (NA) of a system.

The resolution of an imaging system is closely related to the NA of the system, and is often defined by the distance where two points are still distinguishable. For visible light telescopes this is a useful criterion, because the observed stars are point sources. An often used definition of resolution is the Rayleigh criterion, but it is inaccurate for systems with coherent light sources, such as most lensless imaging systems.

To get a better understanding of the limiting factors for resolution of imaging systems, I first explain image formation of classical microscopes and telescopes in order to understand the Rayleigh criterion. In the second part of this section I investigate the consequences of the use of coherent light for lensless imaging on the Rayleigh criterion.

When a point is imaged, not all light will get captured. The light is clipped by the lens or detector in the setup, which can be described by a window function. The most common window function is a block function, created by the circular aperture of a lens. Because of this window function the imaged point will expand in size to a point spread function (PSF). In most systems with a circular aperture the PSF will be an Airy pattern [29, Chapter 4] [8, Chapter 1, 4], and in one dimensional systems the PSF will be a sinc function. The Rayleigh criterion states that two points are indistinguishable when the distance between the centers of their PSF is shorter than the distance between the center and the first zero of the PSF, as shown in figure 1.9d. With other words, the intensity between the two maxima should decrease to less than 74% of the maxima.
1.6 Maximum resolution

Figure 1.9: Example of influence of finite hologram size for a one dimensional hologram. (a) Two point sources and (b) the absolute value of their Fourier transform. (c) Window function created by finite hologram. (d) A graphical representation of the one dimensional Rayleigh criterion. Two points are imaged using a diffraction limited system and broadened to a sinc function due to this diffraction limit. The normalized electric field in the image plane of both point sources is shown, note that the first zero of the field overlaps with the maximum of the field from the other point. The normalized intensity of the sum of the fields of both points is shown, together with the intensity interpretation of the Rayleigh criterion. A multiplication with the window function in the Fourier plane results in a convolution and thus broadening in the image plane.

For a traditional microscope the Rayleigh criterion is expressed as:

$$r = \frac{1.22 \lambda}{2 \pi \sin(\theta)} = \frac{0.61 \lambda}{NA}$$  

(1.13)

where $r$ the resolution, $n$ the refractive index, $\theta$ the maximum angle of incidence and NA the numerical aperture. The maximum resolution increases when scattered light at higher angles is captured, so the NA is larger.

In a lensless Fourier transform holography setup the resolution is also limited by the NA of the system, which is in most cases limited by the size of the captured hologram. In contrast to classical, incoherent light, imaging, every criterion for the resolution depends on the phase distribution associated with the object and illumination. The Rayleigh criterion is only valid for very specific phase distributions. In the following paragraph I showed a one dimensional example of a limitation of the resolution.

The result of the limited size of a one dimensional hologram on the resolution can be derived by considering two coherent point sources with a certain phase difference $\phi$ in the object plane and their Fourier transform in the far field, see figure 1.9a and b. Further consider a finite sized detector, which can be seen as a multiplication with a window (block) function in the far field, shown in
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Figure 1.10: Two points with several phase differences $\phi$ are imaged with a finite size hologram. The points with $\phi = \pi/2$ satisfies the classical Rayleigh criterion, shown in dashes. For other $\phi$ the Rayleigh criterion is no applicable.

The initial phase distribution strongly influences the imaged intensity distribution, illustrated in figure 1.9c. The result in the reconstructed image is the convolution of the original image with a sinc function, shown in figure 1.9d.

The above paragraphs demonstrated that the Rayleigh criterion is not useful for coherent holographic systems. Another resolution definition can be used: the maximal spatial frequency, or minimum spatial wavelength $\lambda$ in the sample that a system can detect, which is limited by the largest detected scattering angle. The minimum spatial wavelength $\lambda$ can be expressed as:

$$\lambda = \frac{\lambda}{2NA} = \frac{z\lambda}{2\xi}$$

(1.14)

where $\xi$ is the radius of the hologram and $z$ the distance between the object and the hologram. When a lens is used to place the hologram in the far field, the focal length $f$ is used for the distance $z$.

1.7 High harmonic generation

In order to develop microscopes operating at shorter wavelengths suitable light sources at these wavelengths are needed. To generate X-ray radiation several methods are available. In all these methods accelerating electrons are responsible for the X-ray generation. The most common source is the X-ray tube. The coherent properties of such tube are comparable to a light bulb. However, for lensless imaging coherent light is needed, comparable with laser light. The coherence of a light source can be increased by spatially and spectrally filtering of the light. However, this highly reduces the residual photon flux. Synchrotron sources provides a much higher photon flux at X-ray wavelengths. In a synchrotron electrons are stored in ring by a magnetic field. By bending the electron beam in this ring, X-ray photons are radiated by bremsstrahlung. The intensity and coherence of the X-ray beam is enhanced by sending the electron beam through an undulator.
1.7 High harmonic generation

Figure 1.11: Artist impression of the HHG process. (1) A bound electron in potential of an atom. (2) The extreme high electric field of light pulse tilts the potential and accelerates the electron. (3) The electric field of light pulse flips and accelerates the electron back to the atom. (4) The high energy electron recollides with the atom and emits X-ray radiation.

X-ray beams with an even higher degree of coherence are generated in free electron lasers (FELs) or high harmonic setups. In a FEL, bunches of electrons are accelerated, mostly in linear accelerators. These high energetic electrons are sent through an undulator, containing a periodically spatially poled magnetic field and will start to radiate. This radiation can stimulate the other electrons in the bunch to radiate. This self self-amplified stimulated emission (SASE) is comparable to laser action and provides the coherent properties of the X-ray radiation. Currently build sources, FLASH and XFEL in Hamburg, LCLS in Stanford, SCSS and SACLA in Japan and SwissFEL in Switzerland, provide very intense (peak spectral brightness of $10^{33}$ photons/s/mm$^2$/mrad$^2$/0.1%-BW) X-ray beams with a high temporal coherence. The drawbacks of these sources are the high building (> $10^9$ euro) and operational costs.

Relatively inexpensive and accessible alternatives are table-top high harmonic generation (HHG) sources. In these sources extremely high intensity laser light is focused in a gas cell, gas capillary or gas jet. The laser light accelerates electrons bound to an atomic in the gas, shown in Fig. 1.11. The electric field of the laser light is comparable to the coulomb potential, which binds the electron. In the first half cycle of the passing light wave the electron is accelerated from the atom. In the second half cycle the electric field tilts the other way around and the electron is accelerated towards the atom. When the electron recollides with the electron, a short burst of X-rays is radiated. If the propagation velocity of X-rays and laser light is matched in the medium (phase matching), these short X-ray bursts constructively interfere to broadband X-ray pulses. The shortest generated wavelength depends on the wavelength and intensity of the incident light, the ionization potential of the atom and the phase matching conditions.

In order to generate X-ray radiation starting with visible light, harmonics above the 100th harmonic should be generated. All lower harmonics are also generated and the incident energy is split over all generated harmonics, resulting in extreme broadband light with low spectral density. Therefore, only a low flux of X-ray photons is generated.

For lensless imaging monochromatic light is needed. To obtain such small bandwidth a monochromator is used often. Monochromators are discarding the light outside the selected bandwidth and the current monochromators are photon inefficient, with typical efficiencies of even 3dB loss for the selected wavelength. This, in combination of the low spectral density of HHG sources results...
in a low photon flux. Our two-pulse imaging enables spectrally resolved lensless imaging with broadband light sources, such as HHG sources. It is photon efficient and so it has a great potential to use with HHG sources [9]. However, it has some additional requirements on the source: it needs two identical, delayed, X-ray pulses. Because good optical elements like beam splitters for X-ray wavelength are problematic to manufacture, it is not practical to split one X-ray pulse into two, delay one and recombine them. In contrary, in the visible regime this task is almost trivial and two time delayed visible pulses could be used to generate two X-ray pulses. However, in a geometry where the focal spots overlap, the ionization induced by the first pulse would distort the phase-matching conditions for the second pulse, resulting in time-dependent intensity modulations and nonlinear phase shifts in this second pulse. This would severely distort a Fourier-transform scan.

An elegant way to overcome this limitation is a scheme with two mutually coherent X-ray beams [30]. In this scheme a visible pulse is split and delayed with an interferometer, whereby the pulses travel different paths and are focused in two closely separated spots in the gas, which results in the generation of two non-collinear X-ray beams. In this scheme the time delay can be altered without influencing the intensity of the two X-ray pulses.
CHAPTER 2

HERALDO WITH AN ULTRA-BROADBAND LIGHT SOURCE

Our development of broadband lensless imaging methods started in the near infrared regime, where coherent sources and high quality optical elements were available. In this chapter the first broadband HERALDO with 1D and 2D reference results are shown, together with signal to noise ratio (SNR) improvement by averaging over several wavelengths. Furthermore limitations of the high dynamic range of captured diffraction patterns are treaded. Finally, a simulation of the influence of reference shape and size on reconstruction quality.

2.1 Setup

We set up an experiment aimed at demonstrating lensless imaging with ultra-broadband light using HERALDO. To investigate the influence of the shape of the reference object, we performed measurements using both a 1D and a 2D reference structure. The sample with a 1D reference structure is shown in Fig. 2.1a, while a sample combined with a 2D reference structure is shown in Fig. 2.1b. The setup used in the experiment is schematically depicted in Fig. 2.1c. A 15 fs pulsed titanium-sapphire laser running at 80 MHz was used as light source. The spectrum of the laser was 80 nm wide with the central wavelength \( \lambda = 800 \text{ nm} \). To generate an even broader spectrum the light passed through a nonlinear photonic fiber and broadened to more than one octave. The spectrum after the PCF is shown in Fig. 2.1d. To produce a pair of coherent pulses with a controllable time delay, the output of the PCF is sent into a Michelson interferometer. By adjusting the length of one of the arms in the interferometer, the time delay between the two pulses can be scanned, which was achieved by mounting one of the interferometer end mirrors on a closed-loop piezo stage. After the interferometer, the pulse pair is used to illuminate the sample and the resulting diffraction pattern was captured with a 1936 \( \times \) 1465 pixels, 14 bit, 4.54 \( \mu \text{m} \) pitch, CCD camera in the far field.

The achievable resolution of an imaging system is typically determined by the numerical aperture. In a lensless imaging system the numerical aperture is mainly limited by the physical dimensions of the CCD and the resolution \( \Lambda \) is given by \( \Lambda > \frac{\pi \alpha^2}{\lambda z/(2 \xi)} \), where \( z \) is the distance between the sample and the CCD, \( \lambda \) the wavelength of the light and \( \xi \) is the height of the CCD.

In order to use HERALDO, the CCD must be placed in the far field, meaning that the Fraunhofer approximation must be fulfilled, which can be expressed as: \( \pi \alpha^2 \ll \lambda z \) [Chapter 4], where \( \alpha \) is the radius of sample, \( \lambda \) the wavelength of the light and \( z \) the distance between sample and CCD. This expression gives a lower limit for the distance between sample and CCD, which for our experimental parameters results in a distance \( z \gg 0.15 \text{ m} \). However, such a large distance would severely limit the numerical aperture of the system, thus resulting in a low resolution. To overcome this numerical aperture limitation the CCD was placed in the focal plane of a lens with a focal length \( f = 5 \text{ cm} \). This geometry resulted in a resolution \( \Lambda = 4.5 \text{ \( \mu \text{m} \) for } \lambda = 600 \text{ nm light.} \)

In the two-pulse scan, the delay was scanned over a distance of 100 \( \mu \text{m} \) in 500 equidistant steps during the experiment, which resulted in a minimal resolvable wavelength of 400 nm (0.74 PHz)
Figure 2.1: (a) Scanning electron microscope picture of the 1D HERALDO holography sample, created by focus ion beam etching. Besides the transmitting star pattern, horizontal and vertical line shaped reference structures were present. (b) Visible light microscope picture of the 2D HERALDO holography sample, produced by laser micro machining. The corners of the transmitting square reference structure had different curvature. (c) Schematic image of two pulse Fourier transform imaging setup. (d) Spectrum of the incident light, reconstructed from our two pulse Fourier transform scan. The wavelengths of different reconstructions showed in Fig. 2.2 are marked with boxes. The grey area indicates the spectral information used in Fig. 2.3b.
2.2 Broadband HERALDO results

and a frequency resolution of 3 THz. After every step, an image of the diffraction pattern was taken with different exposure times between 0.01 and 3 ms.

2.2 Broadband HERALDO results

Figure 2.2: (a) Ultra-broadband diffraction pattern from the sample shown in Fig. 2.1a. (b) HERALDO reconstruction from this ultra-broadband diffraction pattern. The vertical line in Fig. 2.1a is used as reference. (c) Quasi-monochromatic (λ = 559 nm, Δλ = 5 nm) diffraction pattern obtained by two pulse Fourier transform imaging. (d) HERALDO intensity reconstruction from (c) with a resolution of 6 µm. (e) Three reconstruction for different wavelengths (λ = 559, 695 and 885 nm, marked with boxes in Fig. 2.1a) shown together. For shorter wavelengths a higher resolution is achieved. (f) Enlarged view of (d). The sharp features in the center of the star are used to determine the resolution (c,d,f) Shown with an unbiased linear gray scale.

The 1D HERALDO imaged sample consists of a star pattern with a diameter of 200 µm as object and two line segments as HERALDO reference structures of which an electron microscope image is shown in Fig. 2.1a. This sample was etched in a 100 nm gold layer deposited on a fused silica substrate.

We performed two-pulse lensless imaging using HERALDO, of which results are shown in Fig. 2.2. The obtained broad band diffraction pattern is shown in Fig. 2.2a. A strong radial smearing is present and direct reconstruction from this ultra-broadband data did not result in a proper image, as shown in Fig. 2.2b. A quasi-monochromatic (Δλ = 5 nm) diffraction pattern retrieved by the two pulse Fourier transform imaging method is shown in Fig. 2.2c: note that the strong radial smearing has disappeared, and much more detailed features are visible in this diffraction pattern.

To obtain a good HERALDO reconstruction first a raised cosine filter is applied on this quasi-monochromatic data, sacrificing the outermost 100 pixels to reduce edge effects. Subsequently a HERALDO reconstruction is performed by multiplying the data with a differential phase mask and
Fourier transforming back to the image plane. In this reconstruction, shown in Fig. 2.2d and enlarged in Fig. 2.2f, a sharp image of the star object is observed, with a resolution of 6 \( \mu \)m, which is close to the diffraction limit of 4.5 \( \mu \)m for our system. Reconstructions can be performed throughout the whole spectrum and reconstructions at three different wavelengths are shown together in Fig. 2.2e. Although this particular sample does not have a spectrally dependent response, it does demonstrate the ability of our method to obtain images at multiple wavelengths simultaneously. If a sample would display spectrally dependent scattering, its spectroscopic response can be retrieved from these reconstructions at different wavelengths.

2.3 Multi-spectral signal to noise ratio improvement

Figure 2.3: Information from different wavelengths can be used to enhance SNR. In (a) the intensity of a monochromatic reconstruction is shown. In figure (b) six reconstructions at different wavelengths were scaled and averaged which results in less noise, clearly visible as a smoother background and image. Intensities showed with an unbiased linear gray scale.

Since diffraction patterns are obtained for all spectral components in the source spectrum, this information about different wavelengths can be used to improve the reconstruction. Specifically, if the sample does not have wavelength dependence, information about different wavelengths can be used to enhance the signal to noise ratio. Reconstructions are performed for different spectral components, scaled according their wavelength, and subsequently averaged. Multiple frames in a small wavelength range, shown in grey in Fig. 2.3b, are combined, of which a single component is shown in Fig. 2.3a. The captured diffraction patterns for these spectral components show similar structure and have similar SNR, but different numerical aperture and field of view. Fig. 2.3b shows the result of combining six frames after appropriate transverse scaling. Clearly visible are the smoother background and constant signal level in the combined reconstruction compared to the frame in Fig. 2.3a.

2.4 High dynamic range and 2D HERALDO

As the two-pulse imaging shows encouraging results in combination with 1D HERALDO, a logical next step is to extend the approach to higher dimensional reference structures. For this purpose,
Figure 2.4: Results from windmill sample. (a) Diffraction pattern captured with exposure time of 0.021 ms. The center of the diffraction pattern is not saturated, but a low SNR is obtained for high spatial frequencies. (b,c) Diffraction pattern captured with exposure time of 0.300 and 3.000 ms. The center of the diffraction pattern is over-exposed, but the high spatial frequencies have better SNR than in a. (d) High dynamic range combination of a and b. (e) High dynamic range combination of a, b and c. (f,g,h) Intensity of 2D HERALDO reconstruction of a, d and e, shown with n unbiased linear gray scale. Including longer exposure times enhances resolution and smoothness of the image. (i) Horizontal cut through the center of the diffraction patterns a-c. In thick lines is shown which data is used in e.
Chapter 2 HERALDO with an ultra-broadband light source

we used the windmill target combined with a square reference structure, shown in Fig. 2.1b. A two-pulse imaging procedure was used with nearly identical parameters as used in the 1D-HERALDO experiment, resulting in a similar collection of spectrally resolved diffraction patterns. By applying two derivatives the windmill was reconstructed on the corners of the square reference, as shown in Fig. 2.4. Not all corners of the reference structure had the same sharpness, which resulted in different qualities of reconstructions.

One of the challenges to overcome when using Fourier transform holographic imaging techniques is the limited dynamic range detectors. In practice, capturing all the light with sufficient SNR is a challenging task due to the large dynamic range of the diffraction pattern. The light that is not scattered by the object is focused at the center of the hologram, resulting in a high signal at the central pixels. Information about low spatial frequencies, which carries information about the global outline of the object, is also focused near the center of the diffraction pattern, while significantly less light is scattered on the higher spatial frequencies, containing information about edges and the smaller features in the object. A simulation of the lack of low spatial frequencies is shown in Fig. 2.5, where the real part of the reconstruction is shown. Due to the direction of the derivative, the reconstructions at both ends of the line shaped reference have a different sign. The central pixels of the data shown in Fig. 2.2 were removed digitally. Due to the missing semi DC-terms an offset was introduced. The sharp edges of the beam block caused the ringing effects. In this simulation the reconstruction disappeared for beam blocks with a radius larger than 180 px. A 180 px beam block corresponds to filtering all spatial wavelengths larger than 37 µm. This illustrated that in order to obtain a high resolution reconstruction both high spatial frequencies, information about edges, and low spatial frequencies should therefore be captured with sufficient SNR.

To achieve a sufficient high dynamic range with a conventional CCD detector, multiple exposures with different exposure times are taken, shown in Fig. 2.4a-c, and stitched together using the following procedure. Starting with the shortest exposure time, overexposed areas and their direct surrounding (about 10 pixels) are identified and masked out manually, an example of an overexposed area is shown in Fig. 2.6. Furthermore areas with decent signal, SNR larger than approximately 5, are selected manually in the diffraction patterns and are used to normalize the different exposure times. Then the different exposures are combined, shown in Fig. 2.4d,e, and the result of combining three exposures of 0.021, 0.3 and 3 ms is shown in Fig. 2.4f–g. Notice the increase in resolution and contrast after combining images from all three exposure times, shown in Fig. 2.4g compared to shorter exposure times, shown in Fig. 2.4e,f. To give an impression of the gain in dynamic range, horizontal cuts through the center of the diffraction patterns are shown in Fig. 2.4, also showing the used parts of different exposures.

An alternative method to overcome this high dynamic range limitation is covered in section 2.6: reconstruct the low spatial frequencies by iterative phase retrieval algorithms.

2.4.1 Negative sample

In first HERALDO paper by Podorov et al. [16] an absorbing sample in a square aperture was simulated. In such a negative sample (bright field setup), the background is transmitting light, while the object and reference are absorbing. This configuration offers much more flexibility in choosing samples and reference structures. Guizar-Sicairos and Fienup [15] propose several other references, including an obscuring tip. However, no experimental proof has been demonstrated yet of HERALDO with an obscuring tip as reference. The only bright field experiments that have been presented used parallelogram-shaped apertures [17,31].

Several attempts to reconstruct different HERALDO samples were unsuccessful. We tried to reconstruct the gold layer star-line sample in reflection geometry. However, the contrast between reflections from the gold and the bare fused silica substrate was too low for reconstruction. In a setup with a negative USAF target in transmission with a stripped fiber tip as HERALDO reference, the scattering from the fiber was insufficient for reconstruction. Also limiting was the size of the light beam, which meant that the samples had to be sufficient small. Finally, samples printed with a laser printer on overhead sheet lack optical density and resolution.

In a configuration with a negative sample, only a small fraction of the light interacts with the
Figure 2.5: Result of beam block. (a) The centre of the 1402x1402px diffraction pattern is blocked digitally for different radii. (b) Real part of resulting reconstruction. (c) Detailed view of left of reconstructed image
sample, resulting in a very large intensity at low spatial frequencies and a low intensity at high spatial frequencies in the far field. The extremely high dynamic range needed to capture this complete diffraction pattern is experimentally challenging and is a likely explanation for the lack of published experimental results with negative samples. Due to the steep slopes of the central peak in the diffraction pattern, combining different exposure times is demanding and often impossible. In order to capture the high intensity at low spatial frequencies a fairly low flux is needed, even when using the shortest available exposure time of most CCD cameras, resulting in very long exposure times (seconds) in order to capture the weak high spatial frequencies.

If the light source is almost saturating a CCD detector at very low exposure times and no shutter is used, the intense beam is illuminating the detector during readout, which causes a readout line at short exposure times and bleeding effects at long exposure times. In such situation it is beneficial to use a beam block to remove the intense center of the beam and protect the detector. A higher flux, and so shorter exposure times, can then be used to capture the high spatial frequencies.

In order to block the light, the beam block should be optically dense. Furthermore, it should only block the high intensity center. The beam block should be placed very close near the detector to reduce diffraction from the edge of the beam block itself. For the visible regime I tried several beam blocks. The beam block was attached to a coverslip, which was glued on a C1 ring, which placed 1 mm from the CCD detector. We found that both permanent marker on glass and laser printer toner on overhead sheet are insufficiently optically dense to act as beam block. Sand grains have the right size and are optically dense, but hard to attach to a glass surface.

2.5 Image quality depends on reference shape and size

The quality of a holographic reconstruction is strongly dependent on the reference source. In classical Fourier transform holography with a 0-D reference source, a pinhole, both the size of the pinhole
and the flux through the pinhole are relevant. The size determines the maximum resolution, and the flux determines the visibility of the fringes in the hologram, which determines the SNR in the reconstruction [24][22]. The visibility of the fringes depends on the relative amplitude between object field and reference field and varies for different spatial frequencies. The optimal flux from the reference will therefore be a function of reference shape and desired resolution. The maximal visibility of the fringes is obtained by a reference similar to the object. However, HERALDO requires a simple reference, such as a line or a triangle.

In many setups the object and reference structure are illuminated by the same source, and changing the flux from the reference is only possible by changing the area of the reference. This strongly suppresses the phase space for optimisation. In practice including this spatial frequency depending part is not feasible and often ignored.

For pinhole holography there is a maximum in overall visibility of the fringes if the flux through the object is equal to the flux of the reference. Gauthier et al. [18] propose the same reasoning for extended HERALDO references. However, in HERALDO only the boundary waves from the reference contribute to image reconstruction. The sharpness of the edge of the reference determines the relative amplitude of the boundary wave, and should be manufactured as sharp as possible in order to obtain the highest resolution in combination of a relatively low dynamic range in the diffraction pattern.

2.6 Iterative phase retrieval algorithms in combination with holographic techniques

Fourier-transform holographic techniques provide a direct and deterministic reconstruction in the form of a cross-correlation between the object and the reference structure. Besides being a direct reconstruction method, it gives an additional advantage when imaging weak scattering samples. By using a strong reference, the object signal is amplified. However, the cross-correlation also imposes some disadvantages. The resolution and contrast of the reconstruction are always limited by the quality and intensity of the reference beam. Iterative phase retrieval algorithms are not limited by such a reference beam, and in some cases such algorithms can therefore increase the resolution and contrast of the reconstruction.

2.6.1 Contrast improvement in HERALDO measurements

Compared to Fourier-transform holography, HERALDO offers much more flexibility in terms of reference structure fabrication, as well as a potential signal-to-noise advantage because of the stronger reference wave. But it should be noted that this signal-to-noise advantage is not always present, and depends on the exact reconstruction procedure and final image resolution. In HERALDO, the observed far-field holographic cross-correlation consists of a convolution between the object and the reference structure. The strength of this cross-correlation is therefore proportional to the area of the reference structure. When taking the derivative, however, the resulting field only contains a contribution from the edge of the reference structure (assuming that the reference structure has a constant intensity). The size of that contribution depends on the sharpness and area of the edge of the reference, which limits the maximal resolution.

In this sense, HERALDO itself does not make effective use of the available reference light in the way that a deconvolution would. The advantage of HERALDO is in its simplicity: a deconvolution procedure is highly sensitive to errors, and requires accurate knowledge of the shape and intensity distribution of the reference structure (rather than just the knowledge that the structure contains a sharp edge). However, this advantage in signal-to-noise can be regained by using a Gerchberg-Saxton-type iterative phase retrieval algorithm on the original measured far-field hologram, using the result of the HERALDO reconstruction to provide a high-quality object support. Although this approach comes at the cost of increasing the complexity of the reconstruction procedure, it does provide both the signal-to-noise improvement and the high resolution offered by HERALDO.
Here we demonstrate that the contrast of a reconstruction can be improvement by combining an iterative phase retrieval algorithm and HERALDO. For Gerchberg-Saxton type iterative phase retrieval algorithms a support constrained is essential. Using only the knowledge about our simple HERALDO reference as support was insufficient for high resolution reconstruction with an error-reduction algorithm. However, the high resolution HERALDO reconstruction, as shown in Fig. 2.2f, was sufficient as support in order to obtain high resolution reconstructions. The results of 150 runs of 200 iteration steps with random initial start phases were averaged, and reconstructions were obtained with resolution of 8 µm. Both the resolution and contrast can be increased by using both the HERALDO reconstruction and the knowledge about the reference structures as support. This resulted in a reconstruction, shown in Fig. 2.7, with in a two orders of magnitude increase of contrast compared to the HERALDO reconstruction. The resolution, in contrast, slightly deteriorated to 7 µm. Iterative phase retrieval algorithms are highly sensitive to noise, therefore the noise in the measured diffraction pattern might reduce the resolution of this reconstruction.

2.6.2 Beam block removal and resolution improvement

In some cases, such as X-ray diffraction experiments, a beam block is used to protect the CCD from intense unscattered light and so the center of the diffraction pattern cannot be measured. The missing parts can be reconstructed using iterative phase retrieval algorithms. The missing center of the diffraction pattern results mainly in a DC offset. Holographic and HERALDO reconstructions can be used as initial guess and as a solid basis for the support constraints of these algorithms. These algorithms can not only recover the low spatial frequencies, but in some cases also increase the resolution. In experiments with low SNR and a high quality HERALDO reference, iterative phase retrieval algorithms were capable of reconstructing the low spatial frequencies, but a higher resolution was obtained by HERALDO alone. However, when only a weak HERALDO reference is available, such as the edge of a silicon nitride window, iterative phase retrieval algorithms reconstructed a higher resolution.
For extreme ultraviolet (XUV) and X-ray wavelengths, optical components are either not practically realizable or especially complex to manufacture at sufficient quality for high resolution imaging. In this regime we can take full advantage of lensless imaging methods. This chapter we demonstrate two-pulse imaging with broadband XUV light. Furthermore, we extend our two-pulse method to non-colinear beams and show lensless imaging using a multi-wavelength phase retrieval algorithm [9].

3.1 Setup

A schematic of the setup used for two-pulse lensless imaging with HHG radiation is given in Fig. 3.1. We start with intense near infrared ~40 fs pulses emitted by a Ti:Sapphire amplifier system (Spectra Physics Spitfire Ace). From this beam, a pulse pair with variable time delay is produced by passing half the incident beam through a pair of quartz wedges. The time delay $T$ can be scanned by moving one of the wedges further into the beam. This particular interferometer implementation is highly stable: it is nearly common path, and there are no mirrors in the setup that only reflect one of the pulses. These properties eliminate most of the noise caused by vibrations, air flow and acoustics present in conventional Michelson interferometers. We used a $1^\circ$ wedge for scanning the delay, mounted on a closed-loop piezo-stage with 5 nm resolution and 500 $\mu$m scan range. A 100 nm movement of the stage resulted in a 1.32 nm optical path length difference between the pulses, enabling scans with sub-nm step size.
A single wedge in the other interferometer arm introduces an angle between the beams. The lens (focal length $F$), that focuses the pulses into the HHG gas cell, is placed at a distance $F$ behind the point where the beams cross. This geometry ensures that the two beams run parallel to each other behind the lens. Both beams focus near the end of the gas cell with a small transverse displacement between the focal spots. In a geometry where the focal spots overlap, the ionization induced by the first pulse would distort the phase-matching conditions for the second pulse, resulting in time-dependent intensity modulations and nonlinear phase shifts in this second pulse. This would severely distort a Fourier-transform scan. Because of the transverse separation that we introduce in our setup, both pulses will produce HHG independently, allowing clean Fourier-transform images to be recorded \[30\]. The separation between the spots is determined by the angle of the wedge and the focal length $F$, and is set at 0.2 mm. The focal length $F$ is 350 mm, and the focal spot diameter is 40 $\mu$m FWHM. Harmonics are produced in Xenon gas at ~50 mbar pressure in the gas cell, with an energy of 0.2 mJ in each pulse. Behind the cell, a differential pumping stage is used to prevent re-absorption of the produced HHG.

We use the full HHG flux directly to image a Nickel grid attached to a 300 nm thick Aluminium foil. The Al foil reflects the fundamental beam, while transmitting radiation below 70 nm wavelength (harmonics 13 and higher). In this experiment, the Nickel grid is placed at 40 cm behind the focus. The diffracted light is detected in a transmission geometry 40 cm behind the sample using an XUV-sensitive CCD camera (Andor Technology) with 1024x1024 pixels, a pixel size of 13 $\mu$m, and a bit depth of 12 bits. In a typical Fourier-transform scan, 512 diffraction images are recorded as a function of time delay. Between consecutive images, the time interval is increased in steps of 44 attoseconds (step size 13.2 nm). At each time step, 5 frames are recorded with 0.3 second exposure time and averaged to improve the signal-to-noise ratio.

### 3.2 Phase retrieval results

We use the full HHG flux directly to image a Nickel grid, with 20 $\mu$m wide bars, attached to a 300 nm thick Aluminium foil. Fig 3.2a displays a broadband diffraction pattern of the Nickel grid recorded with the transmitted HHG spectrum. Fig 3.2b shows the HHG spectrum used in these imaging experiments, as derived from the two-pulse scan itself. Three harmonics are present, spanning a wavelength range between 47 and 63 nm. Lower harmonics are not transmitted by the Al foil and the Xenon gas, while the HHG phase-matching cutoff is near harmonic 17. Spectrally resolved diffraction patterns at harmonics 13, 15 and 17 are used as input for the multi-wavelength phase retrieval algorithm. Due to the transverse displacement between the two HHG pulses, the recorded data contain two displaced copies of the object's diffraction pattern. This complication can be accounted for by incorporating an additional deconvolution step in the image reconstruction procedure. Small angular deviations between the two beams can also be handled, enabling two-pulse imaging even with non-collinear beams from a split-wavefront interferometer \[33\][34].
Figure 3.2: Broadband lensless imaging with extreme-ultraviolet radiation. (a) Broadband diffraction pattern of a Nickel grid on a 300 nm thick Al foil, recorded using harmonics 13, 15 and 17. (b) Spectrum extracted from the two-pulse Fourier-transform scan. (c) Retrieved image of the grid after 10 iterations of the multi-wavelength phase retrieval algorithm, using diffraction patterns at 3 different harmonics as input. The regular features of the grid are clearly visible, along with several damage spots induced by intense laser irradiation in earlier experiments. (d) Enlarged view of part of the image in c, showing the 20 µm wide grid lines.
The aim of the project was to develop imaging techniques that can be used with broadband XUV and X-ray sources, such as the high harmonic generation source that has been built in the Laser-lab. For classical imaging, diffractive optics, such as lenses and mirrors, are needed to refocus the light. For XUV and X-ray wavelengths, such optical components are either not practically realizable or especially complex to manufacture at sufficient quality for high resolution imaging. Lensless imaging methods, such as holography and iterative phase retrieval algorithms, provide an alternative route towards imaging at these wavelengths. However, these methods have always required monochromatic light in order to provide high quality reconstructions.

We developed two-pulse imaging, which enables robust and accurate lensless imaging with broadband spectra, without a priori knowledge of the spectrum for image reconstruction. This is a situation often encountered with HHG sources, where efficient use of the available photon flux is essential for practical imaging applications.

We have shown a method to use supercontinuum coherent light sources together with HERALDO, a Fourier transform holographic imaging technique that uses extended reference structures. Our two-pulse imaging method enables holographic imaging at all spectral components of the incident light. We have demonstrated ultra-broadband imaging using HERALDO with 1-D and 2-D reference structures at visible wavelengths. Moreover different spectral components can be used to enhance the contrast in our reconstructions. Furthermore we demonstrated the effect of limit dynamic range of CCD detectors on the quality of the image reconstruction and provide a method to overcome this limitation. Also we have demonstrated that iterative phase retrieval algorithms can increase the contrast of the reconstruction by two orders of magnitude when the HERALDO reconstruction is used as support.

Finally, we have demonstrated our two-pulse method at extreme-ultraviolet wavelengths generated by a broadband HHG source. At these wavelengths we have shown multi-wavelength Fresnel-imaging with non-collinear beams.

Several interesting additional features of our two-pulse imaging technique, HERALDO and multi-wavelength phase retrieval can be envisioned. One of them is the imaging of a sample with a wavelength-dependent response to the incident light. The spectroscopic characteristics of such a sample can be probed with two-pulse imaging. Another feature of two-pulse imaging that remains to be investigated further is that it is almost insensitive for constant background illumination. In addition, bright field HERALDO has yet to be demonstrated, which probably requires a beam block to reduce the dynamic range of the diffraction pattern, in combination with an iterative phase retrieval algorithm to recover the spatial frequencies that are eliminated by the beam block. Iterative phase retrieval algorithms are a powerful tool to increase contrast and resolution of HERALDO algorithms. However, more advanced algorithms have to be developed in order to enhance HERALDO reconstructions of objects that contain multiple intensities, such as gray scale images, and to enhance bright field HERALDO reconstructions.
Sinds de ontwikkeling van de eerste microscoop in de 16e eeuw is er een continue ontwikkeling gaande om kleinere dingen te kunnen zien. Tegenwoordig is de kwaliteit van lenzen zo goed dat de resolutie van een goede microscoop voornamelijk wordt begrenst door de golflengte van het licht. Een methode om de resolutie te verhogen is om kortere golflengtes te gebruiken, bijvoorbeeld extreem ultraviolet licht of Röntgenstraling. Röntgenstraling heeft een golflengte die meer dan 100 keer korter is dan zichtbaar licht, dus de hoogst mogelijke resolutie is in principe dan ook 100 keer beter. Het is echter erg moeilijk om goede optische elementen voor deze golflengtes te maken, omdat de brekingsindex van bijna alle materialen bij deze golflengtes hetzelfde zijn en omdat de oppervlakte kwaliteit extreem goed moet zijn.

Lensless imaging methodes zijn wel toepasbaar voor deze golflengtes. De lenzen uit de microscoop worden in dit geval namelijk vervangen door computer algoritmes. De intensiteit van het licht dat van een sample afkomstig wordt direct opgevangen met een detector. Als we ook de richting van het licht weten, kunnen we vanaf de detector terug rekenen hoe het object eruit zag (sectie 1.1). De lokale voorplantingsrichting van het licht wordt bepaald door de fase. Deze fase kunnen we op twee manieren achterhalen. We kunnen het direct meten door er een bekende lichtbundel bij te mixen, deze methode is bekend als holografie (sectie 1.3). Een andere methode is toepasbaar als we al wat weten over het sample. We gebruiken deze informatie dan om de fase van het licht iteratief terug te rekenen (sectie 1.2).

Voor beide methodes geldt dat het enkel werkt met monochromatisch laserlicht. Voor coherente bronnen in extreem ultraviolet- of Röntengolflengtes is dit een grote beperking. Wij hebben nu een techniek ontwikkeld op basis van twee laserpulsen, die deze beperking opheft (sectie 1.5). Hiermee kunnen we in een meting alle golflengtes tegelijkertijd meten, en deze later met een computer weer uit elkaar filteren. Dit geeft ons de mogelijkheid om ook bij breedband lichtbronnen lensless imaging technieken toepassen.

We hebben een opstelling gebouwd en technieken ontwikkeld die holografie zonder lenzen mogelijk maken. Eerste hebben we holografie met breedband zichtbaar licht gedemonstreerd met behulp van onze twee pulsen methode (hoofdstuk 2). Daarna hebben we een hoge harmonische generatie opstelling gebouwd om onze twee pulsen methode te demonstreren met breedband extreem ultraviolet licht (hoofdstuk 3). We hebben een iteratieve terugrekenmethode gebruikt om plaatjes te reconstrueren van een aluminium filter met een ruitjespatroon van nikkel.
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