XUV digital in-line holography using high-order harmonics

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Abstract

A step towards a successful implementation of time-resolved digital in-line holography with extreme ultraviolet radiation is presented. Ultrashort XUV pulses are produced as high-order harmonics of a femtosecond laser and a Schwarzschild objective is used to focus harmonic radiation at 38 nm and to produce a strongly divergent reference beam for holographic recording. Experimental holograms of thin wires are recorded and the objects reconstructed. Descriptions of the simulation and reconstruction theory and algorithms are also given. Spatial resolution of few hundreds of nm is potentially achievable, and micrometer resolution range is demonstrated.

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1 Introduction

Advanced coherent and short-pulse photon sources in the XUV and X-ray range are being extensively developed with the goal to provide both temporal and spatial resolution, thus allowing scientists to explore at the same time structure and dynamics of matter. Examples of this development are the effort to build X-ray free electron lasers at various places in the world [1] and to develop seeded soft-x-ray lasers [2]. One of the first experiments performed at FLASH, XUV-free electron laser facility in Hamburg is time-resolved X-ray holography using a clever single-shot design [3]. Many smaller sources are those provided by high-order harmonic generation in gases [4]. These sources have the advantage of being table-top and affordable in university laboratories. The temporal resolution reaches the attosecond time scale, while the wavelength range varies from the UV to the soft X-ray spectral regions. The highest conversion efficiencies, of the order of a few times $10^{-5}$, and energy per pulse (fractions of $\mu$J) are obtained when Xe or Ar are used for generating the harmonics, with wavelengths typically between 30 and 50 nm. The spatial coherence has been measured to be excellent, comparable to that of the generating laser beam [5,6].

These properties make this radiation an interesting candidate for time-resolved microscopy [7], in particular using holographic approaches [8], at least in the long-wavelength part of the XUV range (around 30 nm) where the number of photons per pulse and per harmonic can be as high as $10^9 - 10^{10}$. A few promising experiments have been performed, using, in particular, digital holographic techniques in the XUV [5,9] and X-ray range [10–13]. Bartels et al. [5] used a very simple setup, where an object was placed directly in a harmonic beam, containing harmonics of orders 17 through 23. The resulting interference patterns, produced by the diverging harmonic beam, acting as a reference wave, and the waves diffracted from the object, were recorded by an X-ray CCD camera. The resolution achieved was not very high due to a low numerical aperture, and the use of several harmonics blurred the image. However, images of the tip of a scanning microscope could be reconstructed. Morlens et al. [9] used two multilayer spherical mirrors to focus the beam on the sample in order to achieve a larger numerical aperture and to select only one harmonic, the 25th. Holograms of spider threads were recorded and reconstructions with resolution below one micrometer could be successfully performed. In both cases, however, acquisitions of over 1000 shots were needed to achieve high signal/noise ratio. Recently Tobey et al. [13] performed time-resolved holography using high-order harmoni-
ics. The experiment was done with a simple single-reflection geometry and without any focusing optics. In the present work, we use high-order harmonics produced by a powerful 10 Hz, 35 fs, titanium-sapphire laser system to perform digital in-line holography of micrometer wires at 38 nm wavelength, using focusing optics with higher spatial resolution than in the previous experiments. High quality holograms are obtained by adding about 100 images. Single shot images are also obtained, though with low signal to noise ratio. Images are successfully reconstructed by using a reconstruction algorithm. In Section 2 we describe the experimental method and results, while in Section 3, we give details of our reconstruction algorithms and apply them to the analysis of the experimental holograms. In Section 4, we discuss how to improve the experiments to realise high-quality single shot digital in-line holography, leading the way to time-resolved holographic microscopy in the femtosecond regime.

2 Experiment

Fig. 1 describes our experimental setup. The laser beam is a low-energy arm of the 40 TW laser system at the Lund Laser Center, a titanium-sapphire laser based on the chirped-pulse amplification technique. Pulses are delivered at a 10 Hz repetition rate, 35 fs duration, 800 nm central wavelength and up to 100 mJ energy. 10 mJ are actually used to generate harmonics by focusing loosely (1 m focal length) the laser beam in an argon gas jet. After that the fundamental beam is blocked by a 200 nm-thick aluminium filter. Fig. 2 shows a typical harmonic spectrum recorded with a flat-field XUV spectrometer. A multi-layer coated Schwarzschild objective is used to focus tightly the harmonic beam and at the same time to select a narrow spectral region. An object (wire) is located a few mm after the image point of the harmonic source and holograms are recorded by using imaging multichannel plates (MCP) coupled to a phosphor screen. A CCD camera is used to record the image on the phosphor screen.

A Schwarzschild objective is an optical system consisting of two spherical mirrors, one convex and one concave of which the centers of curvature coincide. A tight focusing can be obtained. In our design the two radii of curvature are equal to 3.39 and 9.17 cm respectively, with a resulting focal length of 2.69 cm. The Schwarzschild objective is placed 200 cm from the gas jet producing the harmonics and the detector 30 cm further away. The object is located about 3 mm after the focus of the Schwarzschild objective. To avoid losing significant XUV energy, we use it in a slightly off-axis geometry as shown in Fig. 1 without introducing significant aberration. Due to this off-axis configuration, the beam going out of the objective is forming an angle of 10° with the ingoing beam. It is coated with a multilayer structure giving a transmission window centered at $\lambda = 37 \pm 3$ nm. The maximum transmission is approximately 40%.

The wavelength transmission of the objective allows us to select the 21st harmonic. The inset at the top of Fig. 2 represents the far-field profile of that harmonic, with a 1 mrad divergence. The energy available in the 21st harmonic is measured to be 5 nJ which corresponds to $10^9$ photons per pulse, reduced by a factor of 5 after transmission through the Al filter.

The theoretical resolution of the experiment is given by:

$$R = \frac{\lambda}{2\tan(\theta)} = \frac{\lambda L}{a},$$

where $\theta$ is the half angle of aperture, $\lambda$ the wave-
length of the radiation used, \( L \) the distance between the object and the detector and \( d \) the diameter of the detector area illuminated by the beam. In our setup, the half angle of aperture after the Schwarzschild objective is 4.3°, leading to a resolution of 250 nm. Recorded holograms of wires of diameters varying from a few tens to a few micrometers are shown in Fig. 3. These results are obtained after integration of 200 images.

3 Reconstruction

In this Section the algorithm used for simulation of holograms and reconstruction of experimental images is presented. Many innovative techniques have been put together to allow reconstruction of experimental datas. Due to the tight focusing of the beam on the object, the beam has gaussian properties, that have been taken into account. Aliasing is also avoided in this process. Furthermore the problem of the twin images is solved by introducing an iteration process.

Our reconstruction algorithm is based on the Fresnel-Kirchoff integral, illustrated in Fig. 4 [14]. Every point in the object plane \( O (x_o, y_o) \) is the source of a spherical wavelet. Their addition in the hologram plane \( H (x_h, y_h) \) is the electromagnetic field:

\[
E_H(x_h, y_h, Z + L) = \frac{i}{\lambda} \int_S E_O(x_o, y_o, Z) \frac{e^{-ikr}}{r} \times \cos \theta dx_o dy_o, \tag{2}
\]

where \( r \) is the distance from the point \( P_1 \) to the point \( P_2 \), \( \theta \) is the angle between \( P_1 P_2 \) and the normal to the planes, \( S \) is the area over which the integral is carried out. \( Z \) and \( L \) define the position of the object and hologram planes as illustrated in Fig. 4.

In the paraxial approximation, \(|x_h - x_o|, |y_h - y_o| \ll L, \cos \theta \simeq 1\), \( r \) can be expressed to first order as:

\[
r \approx L + \frac{(x_o^2 + y_o^2)}{2L} + \frac{(x_h^2 + y_h^2)}{2L} - \frac{(x_o x_h + y_o y_h)}{L}. \tag{3}
\]

Using \( F^H_O(x_o, y_o, Z) = E_O(x_o, y_o, Z) \exp(-ikx^2_{ho}y^2_{ho}) \) and its Fourier transform \( F^H_O \), the resulting field can
be written as

$$E_H(x_h, y_h, Z + L) = \frac{ie^{-ikL}}{\lambda L} e^{-ik\frac{x_h^2 + y_h^2}{2L}} F_O \left( \frac{kx_f}{2\pi L}, \frac{ky_f}{2\pi L}, Z \right). \quad (4)$$

We now consider a third plane, \((F)\), called far-field plane, far from the object and hologram planes, as shown in Fig. 5. The electric field in the far-field plane can be determined from the electric field in any of the two other planes [15], as

$$E_F(x_f, y_f, Z + L) = \frac{ie^{-ikL_1}}{\lambda L_1} e^{-ik\frac{x_f^2 + y_f^2}{2L_1}} F_O \left( \frac{kx_f}{2\pi L_1}, \frac{ky_f}{2\pi L_1}, Z \right) = \frac{ie^{-ikL_2}}{\lambda L_2} e^{-ik\frac{x_f^2 + y_f^2}{2L_2}} F_H \left( \frac{kx_f}{2\pi L_2}, \frac{ky_f}{2\pi L_2}, Z + L \right). \quad (5)$$

\(L_1\) and \(L_2\) are the distances between \(F\) and \(O\), and between \(F\) and \(H\), respectively (see Fig. [5]). By dividing the two equations an expression for \(F_H\) as a function of \(F_O\) is obtained, and vice versa.

To simulate a hologram we can then calculate

$$E_H(x_h, y_h, Z + L) = e^{ik\frac{x_h^2 + y_h^2}{2L}} F_H \left( x_h, y_h, Z + L \right) = e^{ik\frac{x_f^2 + y_f^2}{2L}} FT^{-1} \left[ G F_O \left( \frac{kx_f}{2\pi L_1}, \frac{ky_f}{2\pi L_1}, Z \right) \right], \quad (6)$$

where

$$G = \frac{L^2}{L_1} e^{-ik(L_1 - L_2)} e^{-ik\frac{x_f^2 + y_f^2}{2L}} \left( \frac{L}{L_1} - \frac{1}{L_2} \right). \quad (7)$$

Similarly, to reconstruct the object, we use

$$E_O(x_o, y_o, Z) = e^{ik\frac{x_o^2 + y_o^2}{2L}} \times \text{FT}^{-1} \left[ G' F_H \left( \frac{kx_f}{2\pi L_2}, \frac{ky_f}{2\pi L_2}, Z + L \right) \right], \quad (8)$$

where

$$G' = \frac{L_1}{L_2} e^{-ik(L_2 - L_1)} e^{-ik\frac{x_f^2 + y_f^2}{2L}} \left( \frac{L}{L_2} - \frac{1}{L_1} \right). \quad (9)$$

This reconstruction method works quite well for plane or collimated waves. However in our case, tight focusing optics is placed before the object, leading to a strong beam divergence. The beam cannot be approximated as a plane wave anymore. Moreover, the typical size of the object is orders of magnitude smaller than the size of the detector and the pixel sizes in the hologram and far-field planes are large. The phase factors of different terms of \(E_H\) and \(E_O\) therefore vary very rapidly, leading to aliasing and picture replicas [16]. To overcome this problem, we introduce a new hologram plane, hologram plane \((H')\) (see Fig. 5). This plane is situated close to the object plane and there is almost no magnification between the two planes so that the algorithm described above can be applied. A simple transformation is then made to obtain the hologram in \(H\). More formally, the field in the object plane can be written as

$$E_O(x_o, y_o, Z) = \mathcal{E}_O(x_o, y_o, Z) \exp(-ik\frac{x_o^2 + y_o^2}{2Z}), \quad (10)$$

where \(\mathcal{E}_O\) is a slowly varying term (product of the object transmission field and the slowly varying part of the reference field, taken to be Gaussian) while the phase term is a rapidly varying spherical term. We apply the following coordinate reduction:

$$(x_h, y_h) \mapsto (x'_h, y'_h) = (\frac{L'}{L} x_h, \frac{L'}{L} y_h), \quad (11)$$

with \(L' = LZ/(L + Z)\). After some elementary mathematical manipulation, the field in the hologram plane can be expressed as:

$$E_H(x_h, y_h, Z + L) = \frac{Z - L'}{Z} e^{-ik(L - L')} \times \text{FT}^{-1} \left[ G H' \left( x'_h, y'_h, Z + L' \right) \right]. \quad (12)$$
where $\overline{E}_{H'}(x_{h'}, y_{h'}, Z + L')$ is the field in $H'$ corresponding to the slowly-varying field $\overline{E}_O$ in the object plane. The terms in front $\overline{E}_{H'}$ in Eq. (12) are, apart from a constant factor, pure phase terms that vanish when calculating the hologram intensity. This equation can be interpreted as follows: the diffraction calculation with a very divergent reference wave is equivalent to the diffraction calculation with a plane wave, on a plane $H'$ separated from the object by $L'$, followed by a magnification by $(L + Z)/Z$. The simulation algorithms described before can now be applied on $\overline{E}_O$, using $H'$. To get the real hologram in $H$ we just make a scale change using Eq. (11). The reconstruction algorithm is based on the same idea.

In the reconstruction process, the phase information is lost because only the intensity of the hologram is recorded. To retrieve this information, an iteration process inspired from the Gerchberg-Saxton algorithm, similar to the one described in [17], has been implemented. The aim of this process is to eliminate the twin images following reconstruction in in-line holography. Here are the different steps:

1. Reconstruction of the object, using the intensity information of the hologram (no phase information).

2. Simulation of a new hologram using the real part of the reconstructed object as object field.

3. Replacement of the intensity of this new hologram by the intensity of the original hologram. The phase of the simulated hologram is now used as a first guess of the phase of the field on the hologram plane.

4. Reconstruction of the object using this last field guess.

5. Repeat steps 2 through 4 until convergence.

Fig. 6 illustrates the performance of this algorithm on simulated data. In (a) the object is represented and in (b) the simulated hologram. The object is reconstructed in (c) without iteration and in (d) with 100 iterations. The iteration process clearly improves the quality of the reconstruction. We also studied the simulation and reconstruction of an hologram generated by many harmonics. In (e) the hologram is simulated with three harmonics (the 19th, the 21st and the 23rd). Despite the relative blurring of the fringes in the hologram, the object can still be reconstructed in (f) also with 100 iterations.

In the experiment, the images are recorded by a digital camera and processed by a computer program. Thus the influence of the hologram pixel size has to be taken into account to estimate the experimental resolution, which will depend on the pixel size of the reconstructed object given by the algorithm. As seen in Eq. (6) and (8) we need to calculate numerically several 2D Fourier transforms and their inverses to...
simulate holograms or reconstruct object from holograms. These calculations involve a discretization of \( H \) and \( O \) in arrays of pixels. Understanding the relations between these pixel sizes is crucial. The size of the reconstructed pixel of \( O \) has indeed to be of the order of the theoretical optical resolution in order to reach the full power of the method. Numerically, a reconstructed object with pixel size smaller than the optical resolution will exhibit diffraction blurring of the order of the optical resolution. For simplicity we consider the shape of the pixels of the object and of the hologram as squares.

In the case of Fresnel diffraction calculated with a 2D Fast Fourier Transform, the pixel sizes in \( F \) and in \( O \) are related by the simple expression:

\[
\text{pix}_{\text{farfield}} = \frac{\lambda L_1}{N \text{pix}_{\text{object}}},
\]

where \( L_1 \) is the distance between the far field plane and the object, \( N \) is the number of pixels in one row of the object plane, \( \text{pix}_{\text{farfield}} \) is the length of the pixel side in \( F \) and \( \text{pix}_{\text{object}} \) is the length of the pixel side in \( O \). A similar expression relates the pixel size in \( H \) and in \( F \), so that \( \text{pix}_{\text{hologram}} = \text{pix}_{\text{object}} L_2 / L_1 \).

Taking into account the reduction factor introduced before, we have

\[
\text{pix}_{\text{object}} = \frac{L_1 Z}{L_1(L + Z) - LZ} \text{pix}_{\text{hologram}} \quad \text{as} \quad L_1 \rightarrow \infty.
\]

The pixel size of the reconstructed object depends on \( L_1, L \) and \( Z \) and on the pixel size of the hologram. The minimum value is reached when \( L_1 \rightarrow \infty \). To obtain an even smaller pixel size, we should thus reduce the pixel size of the hologram by interpolating the experimental hologram or recording with a device with smaller pixels. Note however that, the resolution of the experiment is not limited by the reconstruction algorithm but only by the optical resolution given by Eq. [1].

The recorded holograms are reconstructed using the algorithm described above and are presented in Fig. 7 We observe that the reconstruction is possible for all the objects and that the phase retrieval process is efficient. It is possible to record and reconstruct holograms of wires of 6 \( \mu \)m diameter, which is still far above the theoretical resolution of our experimental setup (250 nm). By comparison a single shot image of a 18 \( \mu \)m wide wire is shown in Fig. 8. The reconstruction is still possible but with reduced quality, due to the low signal-to-noise ratio.

4 Conclusion

Holograms of thin wires are recorded using high order harmonics at 38 nm. A good spatial resolution is achieved with the help of a Schwarzschild objective.
Since single shot holograms are recorded and reconstructed, we are able to reach the femtosecond temporal resolution range. A reconstruction program for digital in-line holography has been developed, tested and implemented. An iteration process was also introduced to improve the phase retrieval process of the reconstruction. With the help of this program we are able to reconstruct different objects.

This work proves that digital in-line holography using XUV radiation is a possible alternative to microscopy and a step in its optimization has been taken. Although there are many improvement that can - and should - be done, this is a step towards successfully performing time-resolved holography with extreme ultraviolet radiation. The main improvements that will be made are mostly experimental. It seems important to select only one harmonic order to have a hologram which can be entirely reconstructed. This could be possible by adding other multilayer optics efficiently selecting one harmonic order. Simultaneously, the harmonic photon flux should be improved by at least a factor of 10 to increase the signal-to-noise ratio. Promising techniques using, for example, improved phase matching [18, 19] or multicolor fields [20, 21] make this within reach. In the future the possibility of single-shot measurements gives this technique a great potential for imaging femtosecond processes.

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