Femtosecond pulse x-ray imaging with a large field of view

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\textbf{Abstract.} Femtosecond pulse x-ray imaging is demonstrated in a sample-multiplexed Fourier transform holography scheme. Parallel imaging of multiple samples over an extended field of view is achieved by exploiting the coherence properties of the free-electron laser (FEL) source and the large profile of the unfocused x-ray pulse. The resulting photon flux density per pulse allows for damage-free single-pulse imaging with moderate image resolution. We envision the application of the method for femtosecond time-resolved pump–probe experiments with the feasibility of recording multiple steps in time with a single pulse. Furthermore, the scheme presented allows for a characterization of FEL radiation pulse parameters.

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1. Introduction

The emerging free-electron laser (FEL) sources delivering coherent (soft) x-ray radiation are currently opening up the field of ultra-fast single-pulse imaging with high spatial resolution. Due to the available high flux density and the small photon wavelengths of a few nanometers or less, an image resolution of some tens of nanometers has been demonstrated and significantly higher spatial resolution is envisioned with FELs operating in the Ångstrom wavelength range coming online \[1\]–\[3\]. Images can be recorded with a single x-ray pulse representing a snapshot of the sample system on a femtosecond (fs) time scale. These experiments aim for structure analysis of small, single (i.e. non-periodic) objects, especially in biology \[4\] and for monitoring a sample’s evolution on very short time scales \[5, 6\]. Moreover, pump–probe experiments with fs time delay allow dynamic studies of objects away from equilibrium \[7, 8\].

At FELs, the imaging methods of choice are coherent diffractive imaging (CDI) and x-ray holography. Both methods take advantage of the high degree of coherence of the delivered radiation. In CDI, the real-space image is retrieved from the coherent diffraction pattern of the sample alone. The phase problem is solved by applying several constraints and using iterative algorithms (phase retrieval) \[9, 10\]. In contrast, the phase information in holography is encoded in the diffraction pattern itself by interfering the scattered radiation with a reference beam. Two different geometries are common for x-ray holography and both were already demonstrated at an FEL: in-line holography \[11\] and Fourier transform holography (FTH) \[2\]. Both methods introduce a small pinhole to generate the reference beam. In in-line geometry, this pinhole is located on the optical axis upstream of the sample, whereas in FTH the pinhole is placed next to the sample in the object plane \[12, 13\]. The latter arrangement can be extended to multiple references and objects, giving the FTH method a large flexibility for sample geometries and interesting opportunities for time-resolved measurements \[14\]–\[16\].

The highest resolution in single-pulse imaging achieved so far is of the order of 40–75 nm. \[1, 2\]. In order to be able to detect the necessary signal under large scattering angles in these experiments, the x-ray pulse needs to be tightly focused. Due to the high-energy fluence per pulse (>1 J cm\(^{-2}\)) at the sample, single-pulse imaging experiments were carried out at the expense of destroying the sample. If the incident photon density is significantly reduced below the typical damage thresholds for condensed matter, the image reconstruction for a desired resolution becomes more challenging. So far, all experiments where the sample survived required multiple FEL pulses \[11, 17\].

In this paper, we show single-pulse imaging results achieved via FTH with a very ‘diluted’, i.e. unfocused, FEL beam. We successfully reconstruct a single-pulse image without detecting any damage on our sample. Furthermore, by employing the large dimensions of the beam and
the coherence properties of the FEL radiation, we are able to simultaneously image several samples over a large field of view (FOV). In contrast to experiments aiming for imaging of very small samples with very high resolution, our study focuses on the parallel imaging of many samples achieving moderate resolution. We envision that this type of experiment can play an important role for systematic time-resolved studies of sample series with time resolution below the temporal jitter of FEL sources. In addition, we believe that the proposed method is also very suitable for time-resolved experiments at high-harmonic generation (HHG) sources as our experimental conditions fit the parameters of these sources. Femtosecond single-pulse FTH imaging was recently achieved at HHG sources [18, 19].

The paper is structured as follows. Firstly, we will give an introduction to the FTH method with focus on the object and reference multiplexing techniques that are heavily employed in the present work. Secondly, we will introduce the experiment including the sample design and experimental setup. In the main part, the imaging results of a selected single pulse will be presented and discussed in detail. We will explicitly demonstrate how the properties of the FEL radiation influence the image quality. The last section will summarize our results and give an outlook for the application of the presented techniques for time-resolved measurements.

2. Theory: Fourier transform holography (FTH) with x-rays

The development of FTH in the x-ray wavelength range was largely promoted by the increasing availability of highly coherent intense x-ray beams at modern third-generation synchrotrons and by the increasing capabilities in nano-fabrication. In particular, the method was successfully applied for imaging of magnetic nano-structures [16], [20]–[22] and biological samples [2, 23].

The principle of the method is simple: the incident coherent x-ray beam illuminates the sample and a separated small pinhole which is transversely offset from the sample in the sample plane. The scattered wave field from the sample and the approximately spherical wave emerging from the pinhole can then interfere and form a hologram on a two-dimensional (2D) detector placed in the far-field. The real-space image is digitally retrieved by a discrete 2D Fourier transform of the hologram [13, 20].

In practice, the sample–reference arrangement has to be adapted to the coherence properties of the x-ray beam. The distance between the sample and the reference hole has to be smaller than the transverse coherence length of the beam, in order to allow interference between the scattered waves. The small x-ray coherence lengths of a few \( \mu m \) up to some tens of \( \mu m \) that are typically realized at synchrotron beamlines require elaborated sample designs integrating the actual sample and the reference. When using soft x-rays, this problem is commonly solved by masking the sample with an opaque metal film (e.g. a 1 \( \mu m \) thick gold layer). An object hole defining the FOV and a reference pinhole are then cut into the film using, e.g., focused ion beam (FIB) lithography. By using this mask technique, the necessary separation between object and reference is also achieved for extended samples such as continuous magnetic films.

Mathematically, FTH can be treated within the Fourier optics formalism [24]. If \( a(\mathbf{r}) \) is the electromagnetic wave at the exit plane of the sample (exit wave), its diffraction pattern recorded with an area detector in the far-field will be the squared magnitude of the Fourier transform

\[
|\mathcal{F}[a(\mathbf{r})]|^2 = \mathcal{F}^*[a(\mathbf{r})] \mathcal{F}[a(\mathbf{r})].
\]
The Fourier transform of the diffraction pattern is the so-called 2D Patterson map \( P \), which is the autocorrelation of \( a(r) \), i.e. \( a(r) \) convolved with its inverse conjugate \( a^*(r) \):

\[
P = \mathcal{F}^{-1}[\mathcal{F}[a(r)] \mathcal{F}[a(r)]] = a(r) \star a(r) = a^*(r) \star a(r).
\]

(2)

Here, \( \star \) denotes the cross-correlation and \( \ast \) the convolution of two functions. For convenience, we choose \( r = 0 \) in the center of the object. If a reference pinhole \( b(r - r_b) \) located at position \( r_b \) in the object plane is added to the object, the Patterson map becomes \[25]:

\[
P = (a^*(-r) + b^*(-r - r_b)) \ast (a(r) + b(r - r_b))
\]

\[
= a^*(-r) \ast a(r) + b^*(-r - r_b) \ast b(r - r_b) + a(r) \ast b^*(-r - r_b) + a^*(-r) \ast b(r - r_b)
\]

\[
= a^*(-r) \ast a(r) + b^*(-r) \ast b(r) + a(r) \ast b^*(-r) + a^*(-r) \ast b(r).
\]

(3)

The first two terms again describe the autocorrelation of both objects. The cross-correlations between object and reference are relevant for the image formation. If \( b(r) \) is a delta-like reference object, the terms reduce to \( a(r + r_b) \) and \( a^*(-r + r_b) \), respectively. In other words, the sample exit wave field is reconstructed at position \(-r_b\) and its complex conjugate, the so-called twin image, at position \( r_b \). This reconstruction represents an unambiguous, direct solution without the need for iterative phase-retrieval algorithms.

For a successful reconstruction, two implications for the experiment can already be derived from equation (3). (i) The separation vector \( r_b \) has to be large enough to ensure that the autocorrelations and the reconstructions do not overlap in the Patterson matrix. Since the extent of autocorrelation is twice as large as the object itself, this criterion is fulfilled if the magnitude of \( r_b \) is larger than 1.5 times the sample diameter. On the other hand, the distance between sample and reference must not exceed the x-ray coherence length in order to allow interference between the exit waves. (ii) The resolution of the image is limited by the radius of the reference hole, because the reconstruction is obtained by the convolution of the object with the reference.

The features of the Fourier transform used in the calculation above can be employed to a larger extent if more than one sample and more than one reference is introduced to the problem. It can be shown that the so-called sample and reference multiplexing are possible with FTH [14, 15, 26]. Let us assume an arrangement with two objects \( a_1(r - r_{a1}) \) and \( a_2(r - r_{a2}) \) and two references \( b_1(r - r_{b1}) \) and \( b_2(r - r_{b2}) \) located at positions \( r_{a1}, r_{a2}, r_{b1} \) and \( r_{b2} \) in the same plane. The Patterson map will then contain all the autocorrelations (\( P_k \)), the cross-correlations between different samples (\( P_k \)) and the desired cross-correlations between samples and references forming the images (\( P_k \)) and twin images (\( P_k \)):

\[
P = P_1 + P_2 + P_3 + P_4
\]

\[
= [a_1^*(-r - r_{a1}) + a_2^*(-r - r_{a2}) + b_1^*(-r - r_{b1}) + b_2^*(-r - r_{b2})] \\
\ast [a_1(r - r_{a1}) + a_2(r - r_{a2}) + b_1(r - r_{b1}) + b_2(r - r_{b2})],
\]

(5)

\[
P_1 = a_1^*(-r) \ast a_1(r) + a_2^*(-r) \ast a_2(r) + b_1^*(-r) \ast b_1(r) + b_2^*(-r) \ast b_2(r),
\]

(6)

\[
P_2 = a_1^*(-r) \ast a_2(r - r_{a2} + r_{a1}) + a_2^*(-r) \ast a_1(r - r_{a1} + r_{a2}) \\
+ b_1^*(-r) \ast b_2(r - r_{b2} + r_{b1}) + b_2^*(-r) \ast b_1(r - r_{b1} + r_{b2}),
\]

(7)

\[
P_3 = b_1^*(-r) \ast a_1(r - r_{a1} + r_{b1}) + b_1^*(-r) \ast a_2(r - r_{a2} + r_{b1}) \\
+ b_2^*(-r) \ast a_1(r - r_{a1} + r_{b2}) + b_2^*(-r) \ast a_2(r - r_{a2} + r_{b2}),
\]

(8)
\[
P_4 = a_1^* (-r - r_{a1} + r_{b1}) * b_1(r) + a_2^* (-r - r_{a2} + r_{b2}) * b_2(r) \\
+ a_1^* (-r - r_{a1} + r_{b2}) * b_2(r) + a_2^* (-r - r_{a2} + r_{b1}) * b_1(r).
\] (9)

For each sample, reconstructions using both references and their associated twin images are obtained, yielding eight images altogether. Obviously, this calculation can be extended to more objects and/or references. The separation vectors \(r_{a1}, r_{a2}, r_{b1}\) and \(r_{b2}\) have to be carefully chosen, in order not to let images overlap with cross-correlations or other images. Since all objects and references usually have a small extent compared to the distances between them, the positions of the cross-correlations and images in the reconstruction can be read directly from the equations above, e.g., object \(a_1(r)\) is reconstructed at \(r_{a1} - r_{b1}\) and \(r_{a1} - r_{b2}\). If the distance between the samples is increased and becomes much longer than the x-ray coherence length, the diffraction patterns of the samples will add up incoherently on the detector and the cross-correlation terms will vanish in the reconstruction \([15]\). Of course, a reference pinhole should be associated with each sample within the coherence length, in order to achieve an image reconstruction with high contrast.

Another limitation related to the object–reference distance is given by the number of detector pixels used in the experiment. This limitation is explained by the Nyquist theorem. If a detector with \(n \times n\) pixels of edge size \(s\) is placed in a distance \(D\) from the sample, an object–reference pair of a distance \(d_{\text{max}}\) is expressed by a modulation with a period of \(\lambda D/(sd_{\text{max}})\) pixels in the hologram on the detector with \(\lambda\) being the photon wavelength. This modulation is only sampled for periods equal or larger than two pixels. On the other hand, the image resolution \(d_{\text{min}}\) is dependent on the maximum scattering angle recorded with the detector. If the hologram is centered on the detector, the resolution is calculated with \(d_{\text{min}} = 2\lambda D/(ns)\). Combining both conditions, \(d_{\text{max}}\) and \(d_{\text{min}}\) are limited by \(d_{\text{max}}/d_{\text{min}} = n/4\).

In summary, FTH in the x-ray regime is a lensless imaging method, which is dependent on coherent illumination. The hologram reconstruction yields unambiguous images with separated twin-images. The resolution is limited by the reference source size and the maximum scattering angle encoded. In order to overcome this limitation, reconstructions from FTH can be used as an input for phase-retrieval algorithms \([2, 18, 27]\). The possibility of simultaneously imaging multiple samples is a feature of the FTH method. This multiplexing technique can be used for extending the FOV and via a more sophisticated layout for imaging a full series of samples under identical conditions or alternatively identical samples under spatially varying conditions. Reference multiplexing is mainly employed for enhancing the contrast and signal-to-noise ratio of the images. By increasing the number of references or by introducing more elaborated reference concepts, the scattering signal of the sample is largely enhanced, helping to reconstruct an image of the sample \([2, 3, 15, 28]\). Especially with respect to femtosecond imaging applications at FELs, holography concepts can be employed to realize time-resolved experiments \([8, 15]\).

3. Experimental details

We designed a sample containing in total 16 separated FTH test specimens. The specimens were realized by small holes in a thin metal foil and each specimen consisted of a dot matrix number representing the object and an associated single reference aperture. The foil was fabricated using the direct ultra-violet (UV) Lithographie, Galvanoformung, Abformung (LIGA) process,
Figure 1. (a) Complete sample design using object multiplexing: 16 dot matrix numbers (marked by blue circles) are arranged in a $4 \times 4$ matrix with a distance of 880 $\mu$m to each other. In this figure, the spacing was reduced for better visibility. (b) SEM image of an object number. The numbers are formed by circular apertures in a $3 \times 5$ matrix. (c) SEM image of a reference aperture. Every number is imaged by a 2.5 $\mu$m-diameter circular reference aperture located in a distance of 120–340 $\mu$m from its object (yellow circles). (d) The center region of the simulated autocorrelation map of (a). Relative distances up to 1300 $\mu$m are shown, including all cross-correlations between nearest and second nearest neighbors. As an orientation selected cross-correlations between objects (purple) and between one object and different references (red) are marked in (a) and (d).

which allowed us to produce micro-structures with high aspect ratio. In brief, an UV-positive resist (AZ 9260) was deposited on a silicon substrate and then exposed to UV radiation passing through a mask. The mask was produced via electron-beam lithography. After exposure, the photo-resist was developed and the resulting voids were plated with a Ni–Fe alloy to a thickness of 5 $\mu$m. Then the foil was removed from the substrate.

The layout of the sample foil is presented in figure 1. The specimens form a matrix of $4 \times 4$ objects with a separation of 880 $\mu$m resulting in a complete sample size of $2.6 \times 2.6$ mm$^2$. A separate reference for each object is placed in a 120–340 $\mu$m distance. The vectors connecting the objects with their references are systematically shifted, enabling a sample-multiplexed FTH experiment. The objects are formed by one- or two-digit $3 \times 5$ dot matrix numbers with 5 $\mu$m dot diameter and 10 $\mu$m pitch. The references have a diameter of 2.5 $\mu$m. For all holes, scanning electron microscope (SEM) images reveal only very few, small deviations from the circular shape. The total open area of the sample is 4300 $\mu$m$^2$.
Based on the sample design, we simulate the FTH experiment. Assuming fully coherent illumination, the hologram is calculated by taking the magnitude squared of the sample layout’s Fourier transform. A further inverse Fourier transform of the hologram generates the expected reconstruction. As already pointed out, an image of each object is reconstructed for each reference. Consequentially, each object is imaged 16 times for our sample. In practice, the number of successful reconstructions is limited by the spatial coherence length of the incident light and by the number of detector pixels, as discussed above. The simulated reconstruction presented in figure 1 shows up to nine images of every object, i.e. the images that are generated by the nearest reference pinhole (distance 120–340 µm), the nearest-neighbor references (561–986 µm), and the second nearest-neighbor references (956–1431 µm). Due to the strategic reference arrangement, the object numbers form a dense square without any overlap in the reconstruction. On the actual sample, the objects are located far away (880 µm) from each other. The number arrangement in the dense square represents Dürer’s magic square on his Melencolia I copperplate print from 1514.

The proposed multi-object layout and the simulated reconstruction already demonstrate how the number of objects is maximized in such an experiment. If the objects are assumed to have a rectangular shape of equal size \((a_1 \times a_2)\) (red boxes in figure 1(d)) and have minimum spacing, their positions in the reconstruction—given by the object–reference vectors—fall on a grid with spacings \(a_1\) and \(a_2\). The area in the reconstruction which can be filled up with these single-object images is confined by reconstructions created by references attributed to neighboring objects (yellow box in figure 1(d)). If, on the real sample, the objects are again located on a rectangular lattice with spacings \(A_1\) and \(A_2\), the available area is \(A_1A_2/2\) as one half is taken by the twin images. The area is slightly reduced by the autocorrelation, which has an extent of \(2a_1 \times 2a_2\). The maximum number of objects is then \(A_1A_2/(2a_1a_2) - 2\).

For the present case \((A_1 = A_2 = 880 \mu\text{m}, a_1 = 90 \mu\text{m}, a_2 = 60 \mu\text{m})\) the theoretical maximum number of objects would have been 70. But of course for an increasing number of objects, the object array on the sample expands and a larger beam profile is needed. In addition, the object–reference distances have to be further increased and limitations due to the spatial coherence length and the number of available detector pixels come into play.

The sample is used in a holography scattering experiment in transmission geometry. The sample foil is held in a frame on a manipulation stage in a high-vacuum chamber. A charge-coupled device (CCD) camera (Princeton Instruments, \(n^2 = 2048 \times 2048\) pixels, \(s^2 = (13.5 \times 13.5) \mu\text{m}^2\) pixel size) records the hologram in a distance of \(D = 1.02\,\text{m}\) from the sample (figure 2).

The experiments were carried out at beamline BL3 of the Free-electron Laser in Hamburg, Germany (FLASH) [29]. A beam-splitting autocorrelator device [30, 31] was installed upstream of the experimental chamber. The FEL delivered single 30 fs long pulses with a repetition rate of 5 Hz, and a photon wavelength of \(\lambda = 23.5\,\text{nm}\). The mean single-pulse energy was \(14\,\mu\text{J}\), which corresponds to \(5.6 \times 10^{11}\) photons per pulse at the experiment when considering a beamline transmission of 34% at this wavelength [31]. The statistical nature of the self-amplified spontaneous emission (SASE) process used in FEL sources heavily influences the source parameters. The time length of a pulse can vary by \(\pm 10\,\text{fs}\) and the time structure is mostly dominated by one mode, but can contain up to four temporal modes. [29, 30, 32, 33]. These modes also appear in the energy domain leading to a broadening of the average spectrum of about 1%. The width of a single-pulse spectrum is often smaller. Nevertheless, the number of modes, the peak photon energy, the width of the energy spectrum, and the total pulse energy are
Figure 2. (a) Sketch of the experimental setup. During the experiment, a beamsplitter shearing off half of the incident femtosecond pulse was installed at the beamline. For the actual experiment, only one half of the FLASH beam was used to illuminate the right half of the sample matrix. The hologram was recorded with a CCD camera. The hologram was reconstructed by a digital 2D fast Fourier transform. (b) Image of the incident beam on a fluorescence screen. The faint vertical stripes are markers with 1 mm separation on the screen.

fluctuating for individual pulses. The fluctuations in beam position or direction were translated to intensity variations by a 3 mm pinhole behind the undulator. Single pulses were selected by using a fast-shutter.

The measurements were performed without any focusing optics. The experimental chamber was located approximately 70 m downstream from the undulator exit. Due to the natural beam divergence, the spot size at the sample position was broadened to a diameter of 5 mm corresponding to a photon density on the sample of \( I_0 = 2.9 \times 10^4 \) photons \( \mu \text{m}^{-2} \). Taking the transmission, i.e. the open area, of the sample and the CCD’s quantum efficiency of 41\% into account, we expected \( 5.1 \times 10^7 \) detected scattered photons per pulse. For the time of the experiment, only one half of the FLASH beam was available, while the other half was blocked in the autocorrelator. Consequently, only half of the sample was illuminated (figure 2), and the number of photons that we expected to detect reduces to \( 2.5 \times 10^7 \) photons.

4. Results and discussion

The single-pulse diffraction pattern of the right half of our sample is presented in figure 3 after subtraction of the CCD dark current. Because of the weak illumination and the mask nature of the sample, a central beamstop was not necessary. We recorded \( 2.2 \times 10^7 \) scattered photons on the whole detector area. Taking the intensity fluctuation of the FEL and the finite detector size into account, this value is in good agreement with our previous estimate and corresponds to a mean density of detected photons on the sample of \( 1.1 \times 10^4 \) photons \( \mu \text{m}^{-2} \). The sample
survived the pulse without any damage, as examined by SEM afterward. Although only one single-pulse image is presented in the paper, the sample was exposed to several hundreds of pulses during the whole experiment. We are confident that even more sensitive samples would withstand such a broadened FEL beam, since the applied energy fluence of approximately $7 \times 10^{-5}$ J cm$^{-2}$ per pulse was more than three orders of magnitude smaller than the typical ablation threshold for most materials [34].

Figure 3. Single-pulse hologram ($2048 \times 2048$ pixels) of the right half of the sample on a logarithmic intensity scale. The diffraction of the circular apertures in the sample dominates the pattern. The modulations are caused by the large number of object and reference apertures. Because of the diluted incident beam a central beamstop was omitted. The zoom-in shows a $30 \times 30$ pixels wide area of the hologram center with linear intensity scale from 35 photons (black) to 180 photons (white) per pixel. Due to the multiplexed sample layout, spatial intensity modulations with high frequencies arise.

The magnitude and the phase of the exit wave are reconstructed by a 2D fast Fourier transform of the hologram. In figure 4, we show the magnitude of the complete reconstruction. For the Fourier transform, the hologram was zero padded to a matrix of $4096 \times 4096$ pixels and centered in the matrix. The resulting pixel resolution of the reconstruction is 433 nm. The autocorrelation of every object is generated in the center of the matrix. At the edges and corners, the cross-correlation between neighboring objects are formed. The existence of these cross-correlations directly proves the high spatial coherence of the FEL radiation. The cross-correlation in the corners is the evidence of interference of waves emerging from objects that are 1.3 mm apart, which equals $5.5 \times 10^4$ times the wavelength.

Next to the autocorrelation, eight objects, i.e. the numbers, and their twin images are reconstructed, representing the right half of the sample. Each image is formed by the cross-correlation of the dot matrix number with its associated reference aperture. Reconstructions of numbers generated by references with larger distances also become visible at the edges and in
Figure 4. Digital reconstruction (magnitude) of the hologram shown in figure 3. In the center, the sample images created by the closest, associated references become visible. Moreover, the large transverse coherence of the FEL radiation allows for recording the nearest and second nearest neighbor cross-correlation at the edges and in the corners. The red boxes mark the zoom-in regions for figure 6.

The corners, although the contrast is drastically reduced. The reasons for this are the decreasing coherence on these lateral length scales, and additionally the influence of the pixel structure of the detector, which is described by the modulation transfer function (MTF) [24]. For a pixel detector, the ability to sample intensity modulations with small periods deteriorates with decreasing period, with the Nyquist cut-off at a two-pixel period. Because narrow intensity modulations translate to large real-space distances, the image contrast for far distant object–reference pairs is further reduced. We calculate the MTF in order to correct for these effects by the squared Fourier transform of the point spread function (PSF), which is the response of a detector to a delta-like signal. For low-energy x-rays, a good approximation for the PSF of a CCD is the square pixel shape. The result is shown in figure 5, which we used to correct the reconstruction with regard to the influence of the MTF. A zoom-in on our diffraction pattern (zoom-in in figure 3) illustrates the pronounced modulations with only a two-pixel period that appear as a result of the object multiplexing. These high-frequency modulations are strongly effected by the CCD’s MTF.

In order to allow a more detailed look into the image reconstruction, figure 6 collects four zoom-in regions of figure 4 with applied MTF correction and with individually adjusted brightness scale. As figure 6(a) reveals, we are able to clearly image all illuminated numbers with high contrast, resolving the smallest features in our sample. The photon flux of a single pulse is sufficient to simultaneously image multiple objects. Since our sample consists of open apertures, the values for the reconstructed magnitudes (figure 6(a)) and phases (figure 6(b)) represent the incident wave at the respective position of the aperture on the sample. Evidently, holographic imaging of a hole array can be used for detailed wavefront diagnostics. Note that every object is individually imaged by its associated reference and the position in the
Figure 5. The MTF of a CCD detector calculated by Fourier transforming the pixel shape. This function was used to correct the reconstruction shown in figure 6. Note that the unit ‘pixels’ is used as dimension in Fourier (hologram) space and real space.

Figure 6. Zoom-ins on the reconstruction presented in figure 4 with (180 × 260) μm² FOV. The intensity was corrected for the influence of the MTF and individually scaled for every image from zero to maximum intensity. (a) Magnitude of the central reconstructions. (b) Combined magnitude and phase of (a). The phase is encoded as hue, the magnitude as saturation. The intensity scale is reduced in order to improve the visibility of the phase variations (0–45,000 photons). (c) Magnitude of the reconstructions at the upper edge, (d) Magnitude of the reconstructions at the left edge. (e) Magnitude of the reconstructions in the upper left corner.

reconstruction is determined by the vector linking object and reference. The numbers are separated much more on the real sample. In addition, the phase is reconstructed relatively to the phase of the particular reference beam. Since every number is imaged by another reference aperture, the phase relation between different numbers remains unknown for our sample layout.
Figure 7. Zoom-in on the top row of the image of the number ‘7’, as shown in figure 6(a), and an intensity slice through the center of the reconstructed apertures. The gray boxes illustrate the width and the position of the apertures in the real sample.

Obviously the intensity varies transversely over the sample, but it is locally flat over the size of an individual number. In contrast, the reconstructed phase modulates on a much shorter length scale of 20–40 $\mu$m. The variation in intensity and phase can be explained by initial variations in the FEL pulse in conjunction with the diffraction of the light at the beamline pinhole. These diffraction rings are also visible on the fluorescence screen behind the beam splitter, as presented in figure 2(b).

An even more detailed view on the smallest features of our samples is presented in figure 7, in combination with an intensity slice through the reconstruction. The circular apertures with a diameter of 5 $\mu$m and a spacing of 10 $\mu$m are well reproduced and separated. The contrast, i.e. signal-to-noise ratio, reaches a value of approximately 21. The apparent diameter (full-width at half-maximum (FWHM)) of the reconstructed apertures is slightly reduced to 4 $\mu$m compared to the real structure. At the softened edges of the reconstructed apertures the resolution achieved in our experiment can be quantified by the distance between the 10% and 90% points of the rising edge. We achieve 1.8 $\mu$m resolution based on this criterion.

The reconstructed intensity values allow us to estimate the number of coherent detected photons that form the reconstruction image, i.e. that are able to interfere. We approximate a coherent photon density ($I_c = U^*U$) that is constant over the area taken by an object ($A_{\text{obj}}$) with its associated reference ($A_{\text{ref}}$). The exit waves of the object $a(r)$ and the reference $b(r)$ can then be written as

$$a(r) = \begin{cases} U & : r \in A_{\text{obj}}, \\ 0 & : \text{ else,} \end{cases} \quad b(r) = \begin{cases} U & : r \in A_{\text{ref}}, \\ 0 & : \text{ else.} \end{cases}$$

For our estimate, we integrate the reconstructed intensities $J(r)$ of the object reconstruction ($A'_{\text{obj}}$), i.e. in practice we sum up all the intensities $J_r$ in our resolution elements (pixels) that have the area $A_{\text{res}} = (0.433 \mu m)^2$ given by the size of our detector:

$$\int_{A'_{\text{obj}}} J(r) \, dr = \sum_{A_{\text{obj}}} J_r A_{\text{res}} \equiv J(A'_{\text{obj}}) A_{\text{res}}.$$
On the other hand, when following equation (3), the total integrated intensity of the object reconstruction is determined by

\[
\int_{A_{\text{obj}}} J(r) \, dr = \int_{A_{\text{obj}}} (a(r) * b^*(\xi)) \, dr = \int_{A_{\text{obj}}} \int_{A_{\text{ref}}} a(\xi) \cdot b^*(r + \xi) \, d\xi \, dr \\
= U^* U \int_{A_{\text{obj}}} \int_{A_{\text{ref}}} d\xi \, dr = I_{c} A_{\text{obj}} A_{\text{ref}}.
\]

(12)

Combining equation (11) with (12), we obtain

\[
I_{c} = \frac{J(A'_{\text{obj}}) A_{\text{res}}}{A_{\text{obj}} A_{\text{ref}}}. 
\]

(13)

As an example, we calculate the total number of photons that built up one of the dot reconstructions shown in figure 7 by using equation (13), and with the knowledge of the actual sizes of our object aperture (5 µm diameter) and reference (2.5 µm diameter). The integrated reconstruction intensity (∫) of the dot is \(6.0 \times 10^6\) photons, which corresponds to \(2.9 \times 10^4\) photons passing through both apertures and forming the image. This value is, of course, dependent on the position of the object on the sample, since the FEL beam intensity is not uniform and dependent on the distance of the object to its reference because of the decreasing coherence with increasing distance.

By applying this procedure to all reconstructions, we end up with a mean density of coherent detected photons of \(I_{c} = 7.3 \times 10^3\) photons µm\(^{-2}\). The ratio of this value with the total density of detected photons can serve as a measure for the degree of coherence. We obtain a ratio of 69% for our experiments using object–reference distances smaller than 340 µm and using small scattering angles.

Beyond the reconstruction of objects with their associated reference, we obtain images of objects generated by references of neighboring objects. For these object–reference pairs the distance increases to ca. 600 µm (figure 6(c)), 750 µm (figure 6(d)) and 950 µm (figure 6(e)). The contrast of these reconstructions drops down to 48%, 43% and 17% compared to the images presented in figure 6(a), respectively. With these reconstructions, we prove that the large transverse coherence length of the FLASH pulses can be exploited to image extended objects on length scales up to a few hundred micrometers. Based on these results, we estimate a transverse coherence length of 600 µm at the position of our experiment, which is in agreement with double slit measurements in [35] and with interferometry experiments using the autocorrelator device in [30].

Our sample and setup layout was designed with the goal of performing single-pulse imaging with a—compared to previous studies—substantially extended FOV. This extension was realized by using far distant object–reference pairs and by imaging multiple samples. The measurements benefited from the high spatial coherence of the FEL source and from the large beam profile. Although we did not focus on high-resolution imaging in this borderline-type of experiment, it is worth discussing the resolution reached in our measurement and the resolution limiting factors as the results are also of value for FEL beam diagnostics. Usually, in synchrotron-based FTH, the resolution is only limited by (i) the size of the reference pinhole and (ii) the maximum detected scattering angle. In our present FEL study, we additionally encountered effects stemming from (iii) the spectral distribution of the radiation. In the
Figure 8. Simulation of the cross-correlation of a 5 µm sample aperture (red) with a 2.5 µm reference aperture (green). Due to the convolution, the reconstruction is washed out and the resolution is reduced as seen in the line scan of the normalized intensities.

following, we discuss these limitations in detail and show their influence on the resolution of our image.

Ad (i) The FTH image reconstruction is determined by the convolution of the object with the reference (equation (3)). Consequently, the finite size of the reference in the experiment will always limit the resolution of the reconstruction unless the reference exit wave is exactly known so that it can be deconvolved. If the reference is treated as a circular aperture, the image resolution cannot be enhanced to more than 1.4 times the radius of the reference [26]. The situation for the present experiment is demonstrated in figure 8. The convolution with the circular aperture of a reference (2.5 µm diameter) was calculated for the circular aperture function of the object dot matrix (5 µm diameter). Because of the rather large size of the references, the sharp edges of the dots are strongly washed out. Due to this effect, the resolution of our reconstruction cannot exceed 1.7 µm. On the other hand, larger references transmit more light and the visibility of the modulations in the hologram—and thus the contrast of the reconstructions—is substantially enhanced [14].

Ad (ii) The achievable resolution in every optical imaging experiment is limited by the maximum detected scattering angle, usually referred to as the numerical aperture of the optical system. In particular for single-pulse x-ray imaging, the highest scattering angle which is detected with statistical significance is dependent on the incident photon density and scattering cross-section of the sample. The reason is that for non-periodic objects the number of scattered photons drastically reduces for higher scattering angles [36]. Analyzing the scattering pattern presented in our work, the resolution is not yet limited by the scattering intensity because we detected photons up to the rim of the CCD. But as photons would be hardly detected for much higher angles, a considerably higher resolution than achieved in our experiment is not possible when using unfocused single FLASH pulses.

In the FTH setup, the angular acceptance of the 2D detector defines the maximum numerical aperture of the system. The resolution of our detection system is limited to 1.8 µm. This value was also experimentally reached in our image reconstructions (figure 7) and therefore our resolution is diffraction-limited.
Figure 9. In order to simulate the influence of the detector, the 5 µm sample aperture (red) was convolved with the PSF, which is the Fourier transform of the detector shape. The normalized intensity line scan reveals that as a result the edges of the aperture soften and the diameter (FWHM) reduces.

The estimate above does not account for the detector shape influences. Because of the square shape of the detector, the maximum detected scattering angle is different for different scattering directions, which leads to slight artifacts in the reconstruction. Quantitatively, this effect is again treated in terms of the transfer function of the optical system, which is defined by the detector shape [24]. The influence on the image formation is then calculated by determining the PSF in the image domain, which is the Fourier transform of the transfer function, and convolving the object with the PSF (figure 9). From the simulation of our experimental conditions, we expect small shape deviations, a softening of the sharp object edges and a slight reduction of the reconstructed aperture size when measured at FWHM. These effects agree with our experimental findings (figure 7). In order to reduce effects stemming from the detector shape, a proper radial symmetric windowing can be applied to the hologram, with the drawback of losing information and further reducing resolution.

Ad (iii) The monochromaticity (\(\lambda/\Delta\lambda\)) at synchrotron beamlines is usually higher than 1000. There, influences from the wavelength distribution can be neglected. In contrast, the spectral distribution of the unmonochromatized FEL radiation at FLASH is of the order of \(\lambda/\Delta\lambda \approx 100\). Moreover, this distribution fluctuates from pulse to pulse, dependent on the pulse width and mode structure. As a consequence, the quality of an image taken with a single pulse is dependent on the respective pulse quality. Images in which several pulses are accumulated depend on the properties of all pulses. Expressed more emphatically, every single pulse image is slightly different, making it complicated to analyze image series, especially because the spectral distribution of the pulses cannot yet be measured simultaneously.

In order to get a more quantitative idea of how the FEL pulse quality influences the image, the effect of a single-mode spectrum with a Gaussian distribution (width \(\Delta\lambda\)) on the reconstruction is calculated. We assume a delta-like object and reference connected by the vector \(r\) (\(r = |r|\)). For small scattering angles, this arrangement is expressed in the hologram by a modulation with pixel period \(\lambda D/(rs)\), with \(D\) being the sample–CCD distance and \(s\) the pixel size. In the digital reconstruction of the \((n \times n)\)-sized hologram the object will appear at the position \(x(\lambda) = n sr/(\lambda D) - r/r\), measured in pixels. Thus, the distance \(x = |x(\lambda)|\) of the reconstruction from the origin of reconstruction matrix is dependent on the wavelength. For a distribution
Figure 10. The wavelength distribution of the FEL pulse reduces the image resolution. The influence on the reconstruction is simulated by a 1D convolution of the object with the wavelength distribution. For the presented example, a Gaussian distribution with $\lambda/\Delta\lambda = 100$ and an aperture–reference pair with 216 $\mu$m distance (corresponding to the number ‘13’ in our sample) was assumed. The influence on the image presented throughout the paper is not as strong as predicted by this values, indicating an FEL pulse with a single narrow spectral mode. But we also detected pulses with much lower quality (figure 11).

of wavelengths, the reconstruction will smear out along the direction of $r$ with a width of $\Delta x = n sr/(\lambda^2 D) \cdot \Delta \lambda$ or translated to real-space distances with a width of $\Delta r = r \cdot \Delta \lambda/\lambda$. In other words, the spatial resolution due to $\Delta \lambda$ is linearly deteriorated with the distance between object and reference. The generalization of this calculation from delta-like to extended objects leads to a 1D convolution of the object, with the wavelength distribution scaled with the distance $r$ along the direction of $r$.

The result of an example calculation for an aperture dot in the number ‘13’, which has its reference in a distance of $r = 216 \mu$m, is shown in figure 10. The circular aperture function is convolved along one dimension with a Gaussian distribution. Due to the assumed broad spectral width of the FEL radiation ($\lambda/\Delta\lambda = 100$), the sharp edges of the dot are smeared out over 2.2 $\mu$m in the simulated reconstruction. This effect is stronger than our observation in the single-pulse measurement. But the broadening is indeed detected in our experimental reconstructions. In figure 11, we show two reconstructions of the number ‘13’ stemming from holograms taken with different single FEL pulses. The first image is taken from the reconstruction presented throughout the paper. Here, the smearing is hardly visible, indicating a narrow one-mode spectrum of the FEL pulse used. In contrast, the second reconstruction taken with another pulse is strongly washed out along the object–reference direction. The intensity slice through a reconstructed object dot suggests the presence of at least two predominant spectral modes. When analyzing reconstructions of the same object from the same FEL pulse, but created by different reference holes, the influence of the wavelength distribution also becomes apparent (figure 12). As expected, the strength and the direction of the smearing clearly change for different object–reference pairs. For small distances, the influence is very weak and the resolution is dominated by the effects discussed above, but it plays a critical role for large distances.

Obviously, these types of experiment provide a single pulse characterization of the FEL beam that can even be performed on a shot-to-shot basis together with a coherent scattering experiment.

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Figure 11. The number ‘13’ created by the same reference, but imaged with different FEL pulses. While the spectral distribution was small in the first case and an influence on the image reconstruction is not visible, the second image is clearly deteriorated. In this case, the spectrum of the FEL pulse cannot be described by a single Gaussian peak, but contains a dominating spectral mode that is strongly broadened only at the lower-energy side, which is a typical behavior of the FLASH source [32]. The red arrows indicate the direction of the vector, linking the object with its reference. The smearing of the reconstructed apertures clearly follows this vector in strength and direction. Line cuts are presented in the right panel.

Figure 12. Zoom-ins on the reconstruction shown in figure 4 displaying the number ‘7’ imaged by four different references. The red arrows point in the direction of the respective reference position and their lengths are proportional to the distance between the object and the reference. The strength and the direction of the smearing caused by wavelength distribution of the FEL pulse clearly follow this vector.

5. Summary

In this paper, we demonstrate the possibility of non-destructive FTH imaging using single FEL pulses with soft-x-ray wavelength. Our images represent a femtosecond snapshot of the sample. Moreover, we prove the feasibility of object and reference multiplexing over large transverse distances by exploiting the enhanced spatial coherence at this x-ray source. We are able to simultaneously image eight objects made up of 104 dots in total, but based on our sample design it is straightforward to increase the number of objects considerably. Some of our objects are multiply imaged by different references with object–reference distances up to 950 µm. The resolution of 1.8 µm in our experiments is diffraction-limited for the given size of the detector.
This value can be easily decreased by shrinking the FOV, increasing the number of detector pixels and focusing the x-rays. Additionally, by using a well-known test sample containing only open apertures, we are able to perform basic FEL beam characterizations such as coherence properties, wavefront characteristics and spectral properties. We show explicitly how these parameters influence the image reconstruction.

Apart from diagnostic purposes, we anticipate the application of the imaging concept presented here to femtosecond serial imaging experiments at FEL or HHG sources. The goal of these experiments will be to follow the sample evolution on a femtosecond time scale, making it necessary to image the sample several times at different points in time. Holographic imaging methods can solve this problem by translating well defined spatial distances into time delays. In the longitudinal direction, this concept was demonstrated in [8]. This work illustrates how the concept can be utilized in the transverse direction, following the concept proposed by some of the authors of this paper in [15]. Again, the technique will be dependent on identical samples in order to realize an image series, but by applying object multiplexing, the series will be taken with the same single FEL pulse and fluctuations in the pulse properties will thus not play a role.

The idea is to slightly tilt the sample plane with respect to either an incoming laser pump pulse or the FEL probe pulse and thereby delay the arrival of the pulse in dependence on the object position. Already a tilt of 2° would introduce a time delay of 100 fs between neighboring objects for the sample geometry used in the present study. Each object row would then represent a time step allowing for systematically studying the time-resolved evolution of identical samples. Another way of employing the object multiplexing technique is to vary the object properties spatially over the sample, but keep external conditions identical for all objects. This concept was developed for magnetic imaging experiments with strong external magnetic fields at synchrotron sources [16], but is ideally suited for single-pulse experiments at fluctuating FEL sources. By probing multiple objects with the same FEL pulse, statistical variations are reduced, enabling quantitative analysis of the experiment.

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