Wide-field broadband extreme ultraviolet transmission ptychography using a high-harmonic source

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High-harmonic generation (HHG) provides a laboratory-scale source of coherent radiation ideally suited to lensless coherent diffractive imaging (CDI) in the EUV and x-ray spectral region. Here we demonstrate transmission extreme ultraviolet (EUV) ptychography, a scanning variant of CDI, using radiation at a wavelength around 29 nm from an HHG source. Image resolution is diffraction-limited at 54 nm and fields of view up to \( \sim 100 \) \( \mu \)m are demonstrated. These results demonstrate the potential for wide-field, high-resolution, laboratory-scale EUV imaging using HHG-based sources with potential application in biological imaging or EUV lithography pellicle inspection.

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Coherent diffraction imaging (CDI) is a rapidly evolving technique attractive for imaging at wavelengths where conventional lenses become ineffective [1,2]. CDI methods reconstruct the phase and amplitude of the exit wave field (EWF) from a coherently illuminated sample using the measured scattered radiation. Only the spatial intensity profile of the scattered radiation is collected by the detector; all phase information is lost. However, if the scatter pattern intensity is sufficiently sampled and additional knowledge (i.e., real-space constraints) are provided, the unrecorded phase information can be recovered using computational methods [3,4]. Rapid development of coherent x-ray sources worldwide has led to development of numerous CDI methods applied to both the physical and biological sciences. In the last decade a variation of CDI known as ptychography [5] has advanced the field, allowing for the reconstruction of wide-field images of extended samples. In a ptychography experiment, multiple overlapping regions of the sample are illuminated sequentially and the algorithm uses the overlap between adjacent illuminated regions as the real-space constraint for phase retrieval. The main assumptions of the ptychography method are that the illumination probe and complex transmission of the sample remain unchanged during the scan, and that the probe positions are known precisely. This additional knowledge, combined with sufficient overlap between the scanning positions and appropriately sampled scatter pattern intensities, provides enough constraints for robust reconstruction of the complex-valued probe and object without many of the issues seen in single-diffraction-pattern CDI with support constraint methods. [5].

CDI-based methods have found particular use in the x-ray community because of the difficulty in producing high-quality imaging optics in this spectral region. CDI methods generally require a very high level of coherence that can be provided by high-brilliance light sources such as third- and fourth-generation synchrotron sources or free electron lasers. The main drawback of these large facilities is that they are sparse and the beam time is limited. As a laboratory-scale source of coherent EUV radiation, high-harmonic generation (HHG) can provide the required spatially coherent light. Single-diffraction-pattern CDI with support constraint, in which a single scatter pattern is recorded and the real-space constraint is provided by expectation of an isolated small sample, has been demonstrated with an HHG-based source using binary samples whose transmission is either 1 or 0, giving up to 22 nm resolution [6]. Ptychographic imaging avoids some of the drawbacks of the single-diffraction CDI with support constraint, but it places very high demands on the pointing and intensity stability of the illumination source over the duration of the scan as it requires the acquisition of multiple scatter patterns with the same illuminating probe beam [7,8]. X-ray ptychography at synchrotrons has been very successful and, in a typical x-ray
B4C is spectrally filtered and focused onto a sample by a single 200 nm thick aluminum filter. The generated EUV beam is focused down into a 3 mm long gas cell by a 75 cm focal length lens. The gas cell is filled with argon gas at a pressure of 80 mbar. The EUV and the fundamental IR beam are separated from each other using an HHG source at 29 nm wavelength [9]. The claimed stability of the HHG source was less than 0.6 s per position.

The experimental setup is shown in Fig. 1. The laser generates pulses at 800 nm wavelength with 50 fs pulse length and 1 kHz repetition rate. The generated IR pulses of energy 1.8 mJ are focused into a 3 mm long gas cell by a 75 cm focal length lens. The gas cell is filled with argon gas at a pressure of 80 mbar. The EUV and the fundamental IR beam are separated by a single 200 nm thick aluminum filter. The generated EUV is spectrally filtered and focused onto a sample by a spherical BrC/Si mirror with radius of curvature of 40 cm. The focal length of the mirror is $\sim 7^{\circ}$, resulting in a significant astigmatism of the focused beam. The EUV beam is spatially constrained by a pinhole placed near the beam focus. The structured illumination at the sample plane is therefore defined by near-field propagation of the cropped EUV beam. The sample is moved relative to the pinhole by three-dimensional nanoprecision piezo stages (Smaract 3D SLC-1740). The scattered radiation from the sample is collected in the far-field regime by a 1024 $\times$ 1024 EUV-sensitive CCD camera (Andor DV434) placed 2.55 cm behind the sample. A key feature, essential to the success of the experiments, is the careful attention to all factors that can influence the beam stability. Beam-pointing stability was improved using both passive and active techniques. The fast positional fluctuations caused mainly by air fluctuations and mechanical vibrations were reduced by sealing the beam path between the amplifier and the vacuum system, and mechanical oscillations were minimized. Slow positional variations with frequency below 1 Hz, originating from the laser system, and small air temperature variations in the laboratory during the experiment can adversely affect the stability of the probe illumination. These slower movements were compensated using a custom-built active stabilization system based on a proportional integral derivative controller and two CMOS cameras (DC1545M) (Fig. 1) driving picoactuator actuators that can provide angular precision better than 0.2 $\mu$rad. The measured EUV stability is approximately five times worse than the IR stability due to extra experimental instabilities after the gas cell that cannot be corrected for with active stabilization.

The dynamic range of the collected scatter patterns was increased by exposing the same sample region using different exposure times [5]. To reduce total readout time, only the central regions were read out in the shorter exposures. The image stitching was done by following equation

$$I(k) = \frac{\sum_i W_i(k) I_i(k)}{\sum_i W_i(k) t_i} t_{N},$$

where $I_i(k)$ denotes background subtracted diffraction intensity at the position $k$, $t_i$ is the $i$th exposure time, $t_{N}$ is the maximal exposure time, and $W_i(k)$ are weighting factors. Weighting factor $W_i$ is zero for oversaturated regions and inversely proportional to the expected noise level elsewhere. No mechanical beam block was used in our setup. Exposure times $t_i$ were corrected to avoid systematic errors caused by finite shutter opening and closing times. This method allows the extension of the dynamic range by 3 orders of magnitude without losing the low-spatial-frequency information. The total readout time was increased by a factor of approximately 2 compared to a single exposure. This method was used instead of more common methods such as multi-image accumulation [9], which is slow, or using a static beam stop [10], which results in large missing regions in the collected diffraction patterns. In the presented experiment, the high-dynamic-range method provides a dynamic range of $10^6$ photons/s, equivalent to a flux of $2.7 \text{ mW/cm}^2\text{sr}$ incident on the sample. Exposure times were corrected for the consequences of the curvature of the Ewald sphere and the use of a flat detector [11].

The EUV photon flux incident on the sample is $\sim 6 \times 10^8$ photons/s, equivalent to a flux of $2.7 \text{ mW/cm}^2\text{sr}$ incident on the sample. The use of a single mirror results in polychromatic illumination of the sample [see Fig. 2(b)]. The EUV bandwidth

![Fig. 1. Schematic of the experimental design. After the amplifier, the IR laser is reflected off a first stabilization mirror (M) before passing through a stabilized focusing lens (L). The main beam is then reflected into the gas cell while a small fraction of the beam passes through the back of the dielectric mirror, toward two stabilization CMOS cameras (C1 and C2). The beam path between amplifier and gas cell is approximately 6 m. The main beam passes through the gas cell (G) where the EUV is generated, and is attenuated by an aluminum filter (F), which transmits the EUV beam. A multilayer spherical mirror (XM) is used to focus the EUV onto the aperture (P) and sample (S). The diffraction pattern is measured in the far field by a cooled CCD (XC).](image)

![Fig. 2. Line profile of the far-field diffraction pattern of a double slit placed at the CLC position. The profile is compared to that from a simulated monochromatic spectrum to demonstrate the blurring effect caused by the polychromatic source illumination. The right plot shows the reconstructed EUV spectrum derived from the double-slit diffraction pattern [14].](image)
is limited by the reflectivity spectrum of the B$_4$C/Si mirror, with peak reflectivity around 30 nm and by the absorption edge of argon gas, which strongly attenuates wavelengths above ~30 nm. The experimental geometry (Fig. 1) introduces a significant astigmatism into the beam, and therefore the aperture required for ptychography was placed at the “circle of least confusion” (CLC) between the two foci to maximize EUV flux through the aperture. The sample was positioned approximately 0.1 mm behind the pinhole. It is advantageous to keep this distance in the near-field regime (i.e., Fresnel number $\gg 1$) in order to produce sharp features in the illumination probe. Furthermore, any increase in beam size arising from propagation would result in lower photon flux density on the sample and lower oversampling of the scatter pattern, which would in turn increase demands on beam stability and coherence.

For the data sets presented in this Letter, aperture diameters of 7 and 10 $\mu$m were used. The step size of the scan was chosen to produce 70 ~80% linear overlap to ensure sufficient ptychographic sampling [12], resulting in either 1.5 or 3 $\mu$m steps depending on the pinhole size.

We present results from two samples. Both samples were prepared on a 50 nm SiN membrane. The first sample (Fig. 3) is an aperiodic high-contrast grid pattern produced by electron beam lithography and coated with 100 nm of gold. Figure 3(b) shows an SEM image of the aperiodic grid sample. The second sample consists of 400 nm diameter polymethyl methacrylate (PMMA) spheres deposited onto a 50 nm silicon nitride membrane to produce an extended sample with a random distribution and feature size similar to those found in biological cells [13].

The EUV spectrum incident on the sample was measured by a Young’s slits–based spectrometer [14] in order to obtain the EUV spectrum under the same experimental conditions that were used in the imaging experiments. A double slit with 4 $\mu$m separation and 1 $\mu$m width was milled by a focused ion beam into 200 nm Au-coated silicon nitride and placed in the CLC position. A line profile of the collected high dynamic range far-field diffraction pattern is shown in Fig. 2. The spectral resolution (Fig. 2) was diffraction-limited to approximately 0.5 nm FWHM. However, thanks to a relatively long driving pulse length (50 fs), the produced HHG spectral peaks are very narrow. Despite the use of polychromatic illumination, sharp speckles are visible even when measured using NA > 0.4 in a previous experiment. The main advantage of using the polychromatic spectrum is the increased coherent photon flux density incident on the sample [15] due to the use of a single mirror, which is necessary because of the low reflectivity (peak reflectivity < 30%) of multilayer mirrors in this spectral region.

Reconstructions were performed using the extended ptychographic iterative engine algorithm [5] in combination with the application of relaxation of several constraints. We have used the PolyCDI method [16] to relax for broadband illumination using the reconstructed EUV spectrum. PolyCDI is a valid approximation if the sample can be assumed to be sufficiently achromatic; otherwise, a full multicolor ptychographic method must be implemented [17]. Reduced visibility due to finite coherence length and fast beam fluctuations was relaxed using a convolution-based correction method [18]. Finally, intensity and readout noise relaxations were implemented as in Ref. [19]. The pixel size was estimated from the experimental geometry and more precisely refined during the reconstruction process by a cross-correlation method [8].

The complex reconstruction (i.e., amplitude and phase) of the grid sample is presented in Fig. 3(a). The reconstruction of the complex illumination probe field is shown in Fig. 3(c). Figure 3(d) shows back-propagation of the illumination probe onto the aperture plane where a saddle-shaped wave front cropped by a circular aperture is clearly visible, as expected. The pinhole to sample distance was estimated to be 55 $\mu$m using an angular spectrum model (ASM)-based propagation code [20]. The highest reconstructed spatial frequency was limited by the experimental geometry to 54 nm. There is large attenuation of 29 nm light by the 100 nm thick Au layer, so no phase variation is observed. Due to insufficient flatness of the sample, the majority of the observed phase variation, i.e., fringes around some of the sharp edges, are diffraction effects. The depth of focus (DOF), given as

\[ \text{DOF} = \frac{\lambda}{2(NA)^2}, \]

is approximately 200 nm in this experimental geometry. Thus, the sample-to-pinhole distance must vary by less than 200 nm over the whole scanning region in order to have the whole reconstruction in focus. This is a serious limitation, particularly for higher-NA, short-wavelength imaging. This effect originates from the nature of ptychography, which searches for a single optimal illumination probe for the whole dataset. However, because features in our aperiodic grid sample are sparse and flatness is close to the limit given by Eq. (2), it is possible to numerically “refocus” the reconstruction by the ASM method at different regions of the sample. An example of the numerical refocusing effect is demonstrated in Fig. 4(a). Figure 4(a) shows an example of a cross section of the reconstructed EWF taken at a region of the sample containing a sharp edge. The reconstructed complex data points (stars) were numerically refocused (circles) using the ASM to show a very flat profile on either side of the rising edge,

Fig. 3. (a) Amplitude and phase image of the grid sample. Here, the image hue represents relative phase and image intensity shows the amplitude of the reconstructed complex transmission function. The inset of (a) shows the complex reconstructed probe field on the same scale as the sample (b), which shows an SEM image of the sample. (c) and (d) show the complex electric field distribution of the probe incident on the sample and after back-propagation to the plane of the pinhole aperture. (e) is an example of a single collected scatter pattern.
as expected from the nature of the sample. This knife-edge analysis was applied to multiple sample regions to estimate the resolution. The example of the refocused data in Fig. 4(a) agrees well with the error function complement fit. The knife-edge analysis shows that the 10–90% distance, 58 ± 5 nm, is approximately equal to the pixel size of 54 nm, indicating that the resolution is limited mainly by the experimental geometry and not by the reconstruction process or insufficient EUV signal.

The repeatability of our measurement was evaluated from two independent data sets using Fourier ring correlation (FRC), as shown in Fig. 4. We have used the 1-bit threshold criterion [21] to estimate the resolution. Our aperiodic grid sample possesses a high degree of asymmetry and the distribution of scattered intensity is significantly anisotropic [see Fig. 3(e)]. Thus, the FRC in the vertical direction is reduced at higher spatial frequencies compared to the horizontal direction. The resolution in the vertical direction was estimated from the FRC to be 100 nm and in the horizontal direction it is limited by the experimental geometry to 54 nm, in good agreement with the knife-edge estimates.

A reconstruction of the second extended sample is presented in Fig. 5. The field of view here is ~105 μm, achieved by collecting 976 scatter patterns with a probe aperture diameter of 10 μm moved in steps of 3 μm. The pixel size is again limited by the geometry to 54 nm. The total dose deposited into the sample was approximately 10⁵ Gy.

We have presented diffraction-limited, wide-field transmission ptychography using an HHG-based tabletop source. We have shown that broadband radiation can be acceptable and even beneficial if it provides significantly larger flux for illumination. High resolution and large field-of-view reconstructions of extended samples are now possible, despite the lower stability and flux of tabletop laboratory EUV sources, by application of a variety of appropriate CDI relaxation techniques. As HHG research progresses toward generation of sufficient coherent flux for imaging in the water window, the HHG-based ptychographic microscope should become a highly valuable tool for high-contrast, high-resolution quantitative microscopy.

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**REFERENCES**

1. J. Miao, T. Ishikawa, I. K. Robinson, and M. M. Murnane, Science 348, 530 (2015).
2. P. Thibault and V. Elser, *Condensed Matter Physics* (Elsevier, 2010), Vol. 1.
3. J. Miao, P. Charalambous, J. Kirz, and D. Sayre, Nature 400, 342 (1999).
4. J. R. Fienup, Appl. Opt. 21, 2758 (1982).
5. A. M. Maiden and J. M. Rodenburg, Ultramicroscopy 109, 1256 (2009).
6. M. D. Seaberg, D. E. Adams, E. L. Townsend, D. A. Raymondson, W. F. Schlott, Y. Liu, C. S. Menoni, L. Rong, C.-C. Chen, J. Miao, H. C. Kapteyn, and M. M. Murnane, Opt. Express 19, 22470 (2011), http://www.opticsexpress.org/abstract.cfm?URI=oe-19-23-22470.
7. M. Beckers, T. Senkbeil, T. Giewekemeyer, T. Salditt, and A. Rosenhahn, Ultramicroscopy 126, 44 (2013).
8. F. Zhang, I. Peterson, J. Vila-Comamala, A. Diaz, F. Berenguer, R. Bean, B. Chen, A. Menzel, I. K. Robinson, and J. M. Rodenburg, Opt. Express 21, 13592 (2013).
9. B. Zhang, D. F. Gardner, M. D. Seaberg, E. R. Shanblatt, H. C. Kapteyn, M. M. Murnane, and D. E. Adams, Ultramicroscopy 158, 98 (2015).
10. G. Williams, H. Quiney, B. Dhal, C. Tran, K. Nugent, A. Peele, D. Paterson, and M. de Jonge, Phys. Rev. Lett. 97, 025506 (2006).
11. J. Rodenburg, Adv. Imaging Electron Phys. 150, 87 (2008).
12. T. Edo, D. Batey, A. Maiden, C. Rau, U. Wagner, Z. Pešić, T. Waigh, and J. Rodenburg, Phys. Rev. A 87, 053850 (2013).
13. D. Shapiro, P. Thibault, T. Beetz, V. Elser, M. Howells, C. Jacobsen, J. Kirz, E. Lima, H. Miao, A. M. Neiman, and D. Sayre, Proc. Natl. Acad. Sci. USA 102, 15343 (2005).
14. R. A. Dilanian, B. Chen, S. Teichmann, L. V. Dao, H. M. Quiney, and K. A. Nugent, Opt. Lett. 33, 2341 (2008).
15. M. Ostrov, J. Bussmann, D. Rudolf, R. Bresenitz, J. Miao, W. Brocklesby, and L. Juschkin, Opt. Lett. 40, 5574 (2015).