Topical Review

Table-top nanoscale coherent imaging with XUV light

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Received 16 February 2017, revised 21 August 2018
Accepted for publication 20 September 2018
Published 19 October 2018

Abstract

Modern laser-based XUV light sources provide very high photon fluxes which have previously only been available at large scale facilities. This allows high-performance XUV nanoscale imaging to be implemented in a table-top manner, and thus qualifies XUV imaging as a novel imaging technique complementing electron and visible-light microscopy. This article presents the current state-of-the-art in table-top XUV light sources and matched coherent imaging schemes. Selected experiments demonstrate the unique capabilities of XUV imaging—namely, nanoscale (sub-20 nm) resolution, single shot imaging, imaging of extended samples and 3D imaging of \(\mu\)m-sized objects. In addition, future prospects will be discussed, including scaling to few-nm resolution, extension to the soft x-ray spectral region, chemically-specific imaging at absorption edges and time-resolved imaging on femtosecond time-scales.

Keywords: XUV light, nanoscale imaging, coherent diffractive imaging, high harmonic generation, x-ray lasers, ultrafast imaging

(Some figures may appear in colour only in the online journal)

1. Motivation for nanoscale imaging with XUV light

Imaging of the smallest structures has enabled numerous scientific and technological breakthroughs in this century and the last. While visible-light microscopy provides a large penetration depth and moderate resolution (>200 nm), electron microscopy can provide ultrahigh resolution (<0.1 nm) but is limited to surfaces due to the small penetration depth of the electrons (<100 nm) \cite{1}. Short wavelength light in the XUV and x-ray spectral regions provides complementary opportunities. Its attenuation lengths in matter typically lies between a few and a few hundred micrometers, depending on the material composition and the particular photon energy. Moreover, the short wavelengths enable resolutions down to a few nanometers. While classical x-ray microscopes are limited to \(\sim 12\) nm due to the employed Zone plate optics \cite{2,3}, coherent imaging techniques surpassed this limit and nowadays provide resolutions down to a few nanometers \cite{4,5}. This unique combination of high-resolution and deep penetration allows the acquisition of non-destructive 3D images of \(\mu\)m-sized samples with nanoscale resolution—e.g. uncovering the internal structure of complex integrated circuits or biological cells as displayed in figure 1.

In addition, the various absorption edges in the XUV and soft x-ray spectral region provide manifold opportunities for high-contrast and element-specific imaging.
Unfortunately, light sources—and particularly coherent light sources—are rarely available in the XUV and x-ray spectra region. Third generation synchrotron facilities provide the desired radiation with sufficient photon flux, and have thus been the workhorse for nanoscale imaging with short wavelength light in recent decades. However, high cost and limited beam time have so far hindered the broad use of XUV and x-ray imaging techniques in science and technology.

Laser driven XUV sources are attractive alternatives in a table-top format. Today, high harmonic generation with high power femtosecond lasers can provide the necessary photon flux for imaging with laser-like beam quality and coherence. Consequently, a number of groups have already implemented and successfully applied coherent imaging techniques using high harmonic sources. These table-top experiments demonstrate the feasibility of nanoscale XUV imaging, and represent important first steps towards real-world application in many fields of science and technology.

This review article will provide an introduction and overview of the light sources and XUV optics employed, in chapters 2 and 3 respectively. In addition, the various coherent imaging techniques and selected highlight table-top experiments will be presented in chapter 4. Finally, future opportunities and perspectives will be discussed in chapter 5, followed by a summary (chapter 6).

2. High photon flux XUV sources based on high-harmonic generation

2.1. General characteristics

Lasers, due to their high power, good beam quality and excellent coherence have enabled numerous exciting applications in recent decades. Direct laser emission in the XUV and x-ray range is however challenging, due to the strong increase of spontaneous transition probability towards shorter wavelengths. Thus, the realization of XUV or x-ray lasers requires ultra-short pulses with very high pulse energies (joules) to pump and invert the laser medium efficiently. Consequently, only a few x-ray lasers are operated, at large high-energy laser facilities [8–10].

The process of high harmonic generation (HHG) provides an alternative route via frequency conversion of visible and infrared laser light into the XUV and soft x-ray domain. The process, first reported in 1987 [11, 12], usually relies on noble gasses as nonlinear medium, exposing them to an intense laser field. On a microscopic scale, the basic process can be described in terms of tunnel ionization followed by propagation of a free electron in the laser field and radiative recombination with the mother ion [13].

A typical high harmonic spectrum is displayed in figure 2. It features a plateau of multiple odd harmonics of the fundamental laser light until a distinct cutoff energy. With
long (few-hundred fs) driving pulses, the harmonic lines can be spectrally narrow (dE/E < 10^{-2}), while short few-cycle driving pulses generate broad overlapping harmonic lines or broad spectral continua. Due to the coupling of driving laser and generated harmonic, the HHG beam propagates collinearly with the driving laser and a good laser-like beam quality can be achieved as displayed at the right side of figure 2.

Macroscopically, phase-matching of all involved emitters in the interaction volume and re-absorption of the generated XUV radiation need to be considered to optimize the conversion efficiency of HHG. Moreover, the nonlinear medium—defined by a gas jet, a gas cell or a gas-filled waveguide—needs to be long enough to reach the so-called absorption limit [15]. Unfortunately, even in optimized cases the conversion efficiency into a single harmonic line does not exceed 10^{-5} [15–17]. Hence, the typical average power of HHG sources driven by conventional watt-class driving lasers is in the μW-range, corresponding to ~10^{12} photons s\(^{-1}\).

During the last two decades, modern solid-state lasers have changed this situation dramatically: by incorporating advanced concepts for high average power operation, fiber [18], slab [19] and thin-disk lasers [20] now provide ultrashort pulses at average powers exceeding 1 kW. High harmonic sources based on such laser systems nowadays achieve average power in the mW-range in a single-pass configuration. More detailed information on these sources can be found e.g. in the review article by Hidrich et al. [21].

Recycling of non-converted fundamental laser light is another promising strategy to boost the generated XUV flux [22]. Based on this principle, HHG in enhancement cavities has demonstrated mW average powers recently [23]. Unfortunately, the conversion efficiency of the HHG process dramatically decreases towards higher photon energies. As can be seen in figure 3, the available average power per harmonic line decreases from ~1mW at ~22 eV to ~1 μW at ~70 eV, and reaches <10 nW at ~300 eV. Thus, coherent imaging with HHG sources requires a trade-off between shortness of wavelength and available photon flux. Consequently, all coherent imaging experiments with HHG sources have so far employed <150 eV photon energy (>8 nm wavelength).

### 2.2. Coherence

Besides a sufficiently high photon flux and a good beam quality, a high degree of coherence is important to generate a sufficiently sharp and high-contrast diffraction pattern. Since the high order harmonic is phase-coupled to the driving laser, the degree of spatial coherence of HHG sources is expected to be high. Measurements of the spatial coherence factor have been performed by analyzing the visibility of diffraction patterns behind a double-pinhole or double-slit sample of various separations. Typical examples are displayed in figure 4. For optimized generation conditions and small double-slit separations, fringe visibilities larger than 90% have been achieved (see figure 4(a)). However, it needs to be noted that intensity-dependent phase distortions can alter the spatial coherence, particularly at larger separations, as can be seen in figures 4(b) to (d). Hence, high harmonic sources generally have to be considered as partially spatially coherent [24, 25]. Note that limited spatial coherence degrades the visibility of interference patterns and thus hinders coherent imaging, particularly if sensitive phase retrieval algorithms are incorporated. For coherent diffractive imaging a visibility larger than 75% is required for reliable convergence [26]. A detailed study on the optimization of the wavefront and the coherence of an HHG source for coherent imaging can be found in [27].

Similarly, the temporal coherence properties of high harmonics are important to generate a high-contrast diffraction pattern even at large diffraction angles. These have been studied experimentally using temporal interferometry.

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**Figure 3.** Overview of state-of-the-art HHG sources. The average power per single harmonic is plotted versus photon energy for various reported sources based on different technologies. The black triangles represent results that have been achieved by HHG in an enhancement cavity. The red and blue dots represent results achieved with Ti:Sa solid-state lasers and reported sources based on different technologies. The white triangles represent results that have been achieved by HHG in an enhancement cavity.
Since the high-order harmonics propagate collinearly with the 3.1. Separation of the generated XUV from the driving laser bandwidth of $d\lambda/\lambda = 1/100$ reaches coherence lengths of 100$\lambda$, which corresponds to 3 $\mu$m at 30 nm wavelength.

Thus, a single harmonic line with a typical relative energy coherence time is of the order of the pulse duration of the high harmonic pulse [29] and the corresponding coherence length can be calculated as

$$l_{coh} \approx \frac{\lambda^2}{\Delta\lambda}. \quad (1)$$

3. Efficient XUV separation, delivery, spectral filtering and focusing

3.1. Separation of the generated XUV from the driving laser

Since the high-order harmonics propagate collinearly with the driving laser, efficient separation of the beams is required in application. Thin metal filters can be employed to efficiently reflect and absorb the driving laser while transmitting the generated XUV. Unfortunately, their small thickness and limited thermal conductivity make such filters very sensitive to heating by the driving laser beam. Thus, high average power driving lasers need to be separated beforehand by other means. To date, grazing incidence plates [30], diffraction gratings [22], micro-channel plates [31], and non-collinear generation schemes [32, 33] have been incorporated into HHG setups.

A grazing incidence plate, as shown in figure 5(a), consists of a multilayer coating on a plane substrate. The coating is designed to be anti-reflective for the driving laser wavelength in order to transmit it. In addition, the top layer is chosen to have a high refractive index, and thus a high reflectivity, for the XUV. Unfortunately, the unwanted reflection of the driving laser is increased for smaller angles of incidence, which are required to efficiently reflect XUV wavelengths shorter than $\sim 5$ nm. Grating structures that diffract the XUV light while efficiently reflecting the driving laser allow for efficient separation of the beams, and have been successfully used in enhancement cavity setups. Unfortunately, the diffraction angle is wavelength dependent, resulting in an angular chirp of the XUV beam, as displayed in figure 5(b). Micro-channel plates have been successfully used to diffract the driving laser and to separate it angularly from the XUV laser beam (see figure 5(c)). Although its wavelength is much smaller, the XUV beam is diffracted as well—of course, to correspondingly smaller diffraction angles. This may not be important in experiments with collimated beams. However, if the beam is focused, as is the case for coherent imaging experiments, angles translate into positions, and consequently the intensity profile at the focus is spatially modulated. Non-collinear schemes for HHG completely avoid optics in the XUV beam. Instead, different beams interact in a symmetric non-collinear geometry and generate the harmonics on an axis between them, as illustrated in figure 5(d). A simple pinhole can be used to block the fundamental beams and fully transmit the XUV. Of course, the different generating beams need to be phase stable to avoid amplitude and pointing fluctuations of the generated harmonics [34]. An elegant and intrinsically stable solution is to generate an annular beam with features a zero intensity part in the center [35]. This beam can be focused to a Gaussian-like spot for HHG. Since the divergence of the generated XUV is small, it can be fully transmitted through a pinhole, as illustrated in figure 5(e). This pinhole will, on the other hand, fully block the annular beam if correctly placed. The annular beam method has recently been successfully implemented in a high-photon flux HHG source and demonstrated only 27% reduction in conversion efficiency [33]. The simplicity and near-zero XUV loss favor this method for future experiments with high photon flux HHG sources, particularly at higher photon energies.

3.2. Beam steering, focusing, and spectral filtering

For imaging applications, the generated XUV radiation needs to be steered and focused onto the sample. Moreover, a particular spectral region—usually one harmonic line—needs to be selected from the broad harmonic spectrum. Multilayer-coated XUV mirrors combine both capabilities and are normally employed for this purpose. They provide a sufficiently high reflectivity at normal incidence and the multilayer design allows tailoring of the reflected spectral bandwidth to a certain degree. A typical experimental setup is displayed in figure 6. After high harmonic generation in a gas jet, the driving laser is separated from the generated XUV by two grazing incidence plates and an aluminum filter. Subsequently, two spherical multilayer mirrors (radii of curvature of 2.4 m and 1 m respectively) are used to collimate and re-focus the beam onto the sample. In this particular case a peak reflectivity of 50% is achieved at 68.6 eV photon energy (18 nm
wavelength. These mirrors have an energy bandwidth of 2.2 eV FWHM, which is small enough to suppress the neighboring harmonic lines by at least one order of magnitude. In order to reduce astigmatism, the angle of incidence of the mirrors is set as small as possible (2.5° to the normal). In the end, a 10 μm diameter focal spot is achieved on the sample. Note that spherical mirror telescopes, which are widely used to re-image the source onto the sample, suffer from astigmatism and limit minimum spot size to a few μm [36, 37]. Elliptical, toroidal, or parabolic mirrors can be used if smaller spot-sizes are required [27, 38].

4. Coherent diffractive imaging and related techniques

4.1. Introduction to coherent diffractive imaging

As discussed in section 2, table-top HHG-based XUV sources can now generate high photon flux and coherent radiation up to the water window. The high temporal and spatial coherence of these sources is ideal for CDI, and sub-wavelength resolutions have been demonstrated [37, 38]. In this section, the various modalities of CDI are discussed and results from selected table-top experiments are presented. Figure 7 illustrates the most common coherent imaging techniques—namely, (a) conventional coherent diffractive imaging, commonly referred to as CDI, (b) Fourier-transform holography, and (c) ptychography.

In contrast to conventional imaging methods, CDI techniques avoid the use of image-forming optics after the sample—recording the scattered intensity directly on a camera. In this way, losses and aberrations due to imaging optics are completely avoided. Consequently, these techniques are referred to as lensless imaging techniques.

Since detectors can only measure the intensity of the diffracted light, the phase information usually has to be numerically retrieved with phase retrieval algorithms—except in the case of holography, where interference with a reference wave provides the missing phase information.

In coherent diffractive imaging the highest achievable resolution is given by the Abbe-limit [39], which corresponds to the highest spatial frequency and thus the highest diffraction angle that can be detected with sufficient signal-to-noise ratio (SNR). It is either geometrically limited by the angular acceptance of the detector or noise-limited due to a limited photon flux on the detector.

Besides this, a number of other requirements limit the resolution in coherent imaging techniques. In order to form a diffraction pattern with sufficient fringe contrast, a high degree of spatial and temporal coherence is required. As discussed in section 2, the spatial coherence of high harmonic...
sources is usually high. However, due to the shortness of the pulses involved in high harmonic generation, the spectral bandwidth limits the temporal coherence length. Hence, it is important to keep all path differences between interfering waves in a coherent imaging setup smaller than the coherence length—otherwise, the interference contrast covering the phase information is lost. The resulting minimum spatial resolution \( \Delta r \) can be calculated as

\[
\Delta r \geq \frac{O_a \Delta \lambda}{\lambda},
\]

(2)

with the sample size \( a \), the oversampling ratio \( O \) and the relative spectral bandwidth \( \Delta \lambda / \lambda \) [40]. It is thus important to keep the spectral bandwidth or the size of the sample sufficiently small to achieve high resolutions. Note that in the case of ptychography the sample size \( a \) corresponds to the size of the illuminating beam.

### 4.2. Conventional CDI

In the basic implementation of CDI shown in figure 7(a), an isolated sample is illuminated by an XUV beam and the diffraction from the sample is recorded by an XUV camera. Only the intensity of the diffraction pattern is measured, its phase being retrieved numerically using iterative algorithms [41–43]. A Fourier space constraint (the measured diffracted intensity) and real space constraints (a priori information, e.g., isolated sample or non-negative imaginary part) are applied iteratively until the algorithm converges to a final reconstructed image. To be able to use the Fourier transform as the propagator between the two spaces in these iterative algorithms, the camera is placed in the far field of the wave exiting the isolated sample. In addition, the diffraction intensity has to be sufficiently oversampled on the camera—with an oversampling factor greater than 2, which translates into recorded information on an area at least twice as large as the object [41]. Table-top CDI setups based on HHG sources have demonstrated high spatial resolution in both transmission and reflection geometries [36, 37, 44–48]. The highest resolution achieved to date using a table-top CDI setup is 13 nm. The results of this experiment are shown in figures 8 (a)–(c). Diffraction was measured in transmission geometry at the high numerical aperture of \( NA = 0.7 \) (see figure 8(a)) using an 18.1 nm HHG source. This high NA and high dynamic range diffraction pattern enables high-resolution reconstruction of the sample exit wave with sub-wavelength resolution of 0.72 \( \lambda \) (13 nm), as shown in figures 8(b) and (c) [37]. Note that the phase information on the sample exit wave is available from CDI with the same high-resolution. In reflection mode, a resolution of a few \( \mu \)m was obtained with CDI [46] and a higher resolution of \( \sim 100 \) nm was achieved by introducing an aperture illumination setup [47]. In these experiments, multiple shots of XUV pulses were accumulated in recording the diffraction pattern. Single-shot CDI experiments, which are interesting for time-resolved imaging of non-repeatable processes, have also been realized with HHG-based systems and achieved a resolution of 119 nm (see figures 8(d) and (e)) [45].

### 4.3. Holographic CDI

The phase retrieval algorithms employed in conventional CDI are time-consuming and require lots of computational resources. Convergence speed is also dependent on the SNR and oversampling ratio of the measured diffraction pattern, in addition to issues of twin-image ambiguities and stagnation [41, 49]. These issues can be overcome if the phase of the diffracted field is encoded on the camera during measurement. These techniques are collectively referred to as holographic CDI techniques; the phase of the diffracted wave is encoded
on the camera through interference with a reference wave [50]. The implementation of Fourier-transform holography (FTH) is shown in figure 7(b), where the reference wave originates from a reference structure in close proximity to the sample and interferes with the diffracted wave at the camera. In FTH, a simple Fourier transform performs a direct and unambiguous reconstruction of the object. The achievable resolution is limited to \( \sim 70\% \) of the reference structure size besides the wavelength and NA of the measurement. Since the transmitted intensity of the reference wave scales quadratically with the hole diameter, a smaller reference hole also reduces the SNR of the image, and a compromise often needs to be found. Multiple reference structures can be used instead of one, to improve the SNR of the composite image [51].

The first demonstration of FTH in a table-top setup was performed by Sandberg et al., achieving 89 nm resolution [52] in a multiple reference scheme as depicted in figure 9(a). Iterative phase retrieval algorithms can still be used to refine the resolution by seeding the image from FTH as a starting point. The highest resolution achieved using FTH in a table-top setup is 34 nm (figure 9(b)), and reference holes of just a few wavelengths in diameter were used in this experiment [53]. By refining the resolution using phase retrieval algorithms, the authors demonstrated the smallest features ever

**Figure 8.** Sub-wavelength resolution CDI in a transmission geometry. (a) High-NA diffraction pattern recorded for a transmission sample shown in the inset, (b) reconstructed amplitude using phase retrieval algorithms, and (c) cross-section along the white line in (b) showing a half-pitch resolution of 13 nm. (a)–(c) Reprinted with permission from [37], (OSA). (d) Shows a diffraction pattern measured in a single-shot experiment and (e) the reconstructed amplitude using the diffraction pattern in (d), (e) Reprinted figure with permission from [45], Copyright (2009) by the American Physical Society.

**Figure 9.** (a) Schematic and reconstructed object in multiple reference holography. Reprinted with permission from [52], (OSA). (b) Results from the highest resolution holography experiment, showing autocorrelation of the measured hologram for the sample shown in the inset. Reproduced from [53]. CC BY 4.0.
resolved in a table-top XUV/soft x-ray setup, with a half-distance of only 23 nm. This high-resolution revealed wave-guiding effects in wavelength-sized features [53]. Williams et al presented methods for using FTH in cases where a discrete set of wavelengths is necessary for the application at hand (e.g. attosecond pulses) [54].

Reference structures extended along one or two dimensions have been used in holographic CDI to increase the intensity of the reference wave. This modality is known as holography with extended reference by autocorrelation linear differential operation (HERALDO) and sharp edges along extended references ensure high-resolution reconstructions through a differential operation [55]. The resolution in HERALDO is limited by the sharpness of the edges of the reference structure and not its size. In an experiment shown in figure 10, Gauthier et al demonstrated HERALDO with spatial resolution of 110 nm using a single ~20 fs pulse as illumination [56]. Unfortunately, HERALDO has the disadvantage of direction-dependent resolution and is more sensitive to noise, especially for photon flux-limited scenarios [57, 58].

4.4. Ptychography—scanning CDI on extended samples

The drawback of CDI and FTH is that they usually require isolated samples and are therefore restricted to a small class of specimens. A method that overcomes this limitation is ptychography, which is a scanning type of CDI. Ptychography can be understood as an extension of scanning transmission x-ray microscopy (STXM) in which a focused beam is scanned across the sample and the transmission of the sample is measured using an integrating detector. In the case of ptychography, a spatially resolving detector replaces the integrating detector in measuring the far field diffraction pattern of the sample, thereby enabling resolutions far smaller than the focal spot size but requiring coherent radiation [59, 60]. In contrast to CDI, where the diffraction pattern is oversampled, in ptychography the sample is scanned through the XUV beam and diffraction patterns are recorded for multiple positions, while between adjacent positions the illuminated area partially overlaps (as illustrated in figure 7(c)). By using the redundant information gained from the overlap, the phase problem can be solved by iterative algorithms whereby the illumination of the sample is recovered as well [61, 62]. Since the complex illuminating beam is recovered, algorithms that incorporate partial spatial and temporal coherence of the beam and hence relax the requirements on the illuminating source have been found [63, 64]. Following demonstration that ptychography could be reliably solved for the diffracting object [65], a variety of applications have been demonstrated at large scale facilities. For a recent review on x-ray ptychography see [66].

A variety of ptychography experiments have been performed in a table-top setting using high harmonic radiation; these will be reviewed in the following. Ptychography was first demonstrated in 2014 by Seaberg et al in a reflection geometry using a ~30 nm high-harmonic source [67]. The lateral resolution was later improved to 40 nm × 80 nm [69]. Next, to achieve a near diffraction-limited lateral resolution, the reconstructed phase information was used to recover the height profile of the sample with sub-nanometer precision using a priori information about the material composition of the sample and a rough height range (see figure 11(a)). In a similar experiment published by Shanblatt et al in 2016, structures buried under 100 nm of aluminum were reconstructed and it was possible to model the chemical composition of the layers from the retrieved complex object [70]. In the first experiment in transmission a resolution of 58 nm was achieved [68]. In addition, a sample with a large field of view of ~100 μm × 100 μm (see figure 11(b)) was reconstructed from more than 900 diffraction patterns, proving the achievable stability of such an imaging system. The same group was also able to image biological samples consisting of neural cell structures [71, 72]. Since polycarbonates are highly absorbing at the wavelength of 30 nm used, it was hard to extract phase information from the reconstructions and thereby gain quantitative information about the composition.

In particular, table-top imaging of extended samples at 92 eV (13.5 nm) promises a wide range of application in e.g. inspection of lithography masks in reflection. Gardner et al reported the first table-top ptychography experiment at 13.5 nm in 2017 [38]. They imaged the periodic structures of a zone plate, and a sub-wavelength resolution below 13.5 nm was claimed. Later, the first reflection-type ptychography experiment at 13.5 nm with an angle of incidence of 6°, similar to the situation in EUV lithography, was performed. The experiment was limited to a low NA of 0.13 but allowed for reconstructions of lines with a width of 88 nm, as shown in figure 12(b) [73]. In a different experiment a sample, which consisted of 29 nm tall nickel structures deposited on a Si wafer, was placed under a grazing incidence angle of 80.5° with respect to the normal of the sample plane [74]. In contrast to
the experiment by Mamezaki et al, which was specifically designed for the inspection of lithography masks, such a geometry allows for imaging of a wider range of samples, since the reflectivity under normal incidence is small compared to the reflectivity at grazing incidence. The resolution achieved was rather small, because of a low NA, but the projection of the beam onto the sample yielded a large horizontal extent (see figures 12(d) and (e)) and a large field of view was able to be scanned in a short time.

In a very recent experiment, features of only 43 nm in size have been resolved by combining ptychography with a high photon flux 18 nm source [14, 75]. By the use of Siemens star test samples, features down to 20 nm in size were made available for examination, allowing for a direct reliable real-space resolution test as suggested recently [76]. In contrast to previous experiments, the resolution was estimated by the size of the resolved features for the first time. A detailed comparison with the previously-used knife-edge test showed that a knife-edge can overestimate the achieved resolution and thus Rayleigh-like resolution tests, e.g. on a Siemens-star, should be regarded as the state of the art in future characterization of XUV imaging systems.

5. Future directions

5.1. Coherent diffractive imaging with soft x-rays

The soft x-ray spectral region (<10 nm wavelength) is highly attractive for imaging, since many host materials are highly
opaque and multiple absorption edges can be exploited for high-contrast, high-resolution imaging. Unfortunately, tabletop coherent imaging experiments have so far employed wavelengths larger than 10 nm. The decreasing photon flux available from high harmonic sources is the main reason for this situation. However, a first attempt in 2015 demonstrated that CDI is possible with an 8 nm wavelength high harmonic source \[77, 78\]. Due to the low average power available from the source (~7 nW) and the high losses on the way to the sample (overall transmission 5%), the diffraction pattern suffered from a poor SNR (see figure 13(a)). Nevertheless, a reconstruction of the cross-like samples employed was possible. As can be seen in figure 13(b) the overall structure is successfully reassembled, albeit with low quality and limited resolution. In summary, tabletop coherent imaging has been demonstrated to be already feasible with nW-level sources. Thus, tabletop imaging in the soft x-ray region, including the water window, appears feasible with state-of-the-art sources \[78–81\]. Advances in mid-IR ultrafast laser development \[82, 83\] appear very auspicious, promising to boost the photon flux of HHG sources below 10 nm wavelength and significantly increase the performance of tabletop soft x-ray coherent imaging systems in future.

5.2. Broadband coherent diffractive imaging schemes

High harmonic sources are broadband by nature, include a large number of harmonic lines, and can even form a broadband continuum. In contrast, coherent diffractive imaging requires quasi-monochromatic illumination. Thus, the spectral filtering usually employed selects only a small fraction of the available high harmonic photons to form the diffraction pattern.

On the other hand, a broadband spectrum can provide additional information on materials’ constitutions and, via near- and extended-edge x-ray-absorption fine-structure spectroscopy (NEXAFS and EXAFS) even uncover their chemical states and bonds \[21, 79, 81, 84\]. An example measurement on a 200 nm thick polyimide foil is displayed in figure 14. Contributions from several different carbon orbitals are clearly identified.

Hence, soft x-ray spectro-microscopy techniques, providing a transmitted (or reflected) spectrum at each pixel of a high-resolution image, are highly desirable. Since XUV and soft x-ray spectrometers suffer from a low throughput and limited resolving power, Fourier-transform spectroscopy (FTS) appears an attractive alternative. FTS is based on the coherence of the source, using time-domain measurements of the interference signal of two broadband pulses with variable delay. The spectrum is gained from a single Fourier transform of a delay-dependent signal. A combination of CDI techniques with FTS is feasible if the sample is illuminated by two pulses and diffraction patterns are recorded for different delays, as illustrated in figure 15. The striking advantage of FTS, an improvement of the SNR on the detector, is obvious since the whole spectrum—instead of only a small fraction as in monochromatic illumination—is radiated onto the sample.

The generation of the two pulse replicas and the control of their delay with sufficient (attosecond) precision represent the biggest challenge. So far, XUV interferometers based on thin-film beam splitters have been demonstrated \[86\]. Integrating the interferometer in the driving laser beam before high harmonic generation is an alternative solution currently

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**Figure 13.** (a) Diffraction pattern recorded at 8 nm wavelength and (b) corresponding reconstruction of the exit wave.

**Figure 14.** NEXAFS measurements of a 200 nm thick polyimide foil uncovering contributions from several different orbitals. Reprinted with permission from \[79\], OSA.
under intensive research [87, 88]. Ultimately, nanoscale mapping of chemical composition will become feasible with table-top devices.

5.3. XUV coherence tomography (XCT)

While CDI techniques have demonstrated very high lateral resolution, the depth information is not usually available, except if the material is known and the phase plus amplitude of the transmitted light is carefully analyzed. An alternative method is coherence tomography, which is a non-invasive technique in the VIS and infrared wavelengths commonly used to obtain depth information. Scaling the μm-range axial resolution of optical coherence tomography (OCT) can be achieved by using shorter wavelength XUV light with a broad spectral bandwidth [89]. As discussed in section 2, HHG sources can generate a high photon flux in the spectral range between 30 eV and 100 eV, where Si has high transmission. These sources are thus suitable for coherence tomography in the XUV, known as XUV coherence tomography (XCT), which has a number of potential applications in technology and solid-state physics. In the first demonstration of XCT using an HHG source, Fuchs et al demonstrated an axial resolution of 24 nm using a broadband XUV source generating a relatively flat spectrum from 30 eV–70 eV [90]. The results are shown in figure 16. A scanning stage was incorporated in the setup for imaging in the lateral direction, and a transverse resolution of 23 μm—limited by the XUV spot size—has been achieved. XCT usually requires a reflective layer on top of the sample to be used as a reference. Ghost signals can efficiently be avoided by employing one dimensional phase retrieval algorithms [90].

5.4. 3D tomography

The penetrating power of x-rays enables the combination of CDI with computer tomography, thereby revealing the internal structure of e.g. biological samples. So far tomography in combination with FTH [91], single shot CDI [92, 93], and ptychography [94, 95] were demonstrated at synchrotrons. By combining a variation of FTH [55] with CDI, it was possible to measure the three-dimensional structure of a diatom (see figure 17(a)). With CDI on isolated samples several experiments in combination with tomography were performed. It was, for example, possible to resolve the internal structure of unstained yeast cells (see figure 1(b)) and the three-dimensional structure of a human chromosome (see figure 17(b)). To resolve three-dimensional structures of extended samples, tomography must be combined with ptychography. Several pioneer experiments have already proven this concept. An isotropic resolution of
15 nm was achieved, allowing resolution of the internal structure of integrated circuits (figure 1(b)) and bone material (see figure 17(c)).

So far, CDI experiments combined with computer tomography have been conducted at large scale facilities only. To our knowledge there has until now been no CDI-tomography experiment performed using a table-top XUV source. The main challenge will be to achieve high stability over a long measurement time, since many projections must be acquired. A high photon flux from the source and efficient low-loss XUV optics will be needed to perform such measurements in a reasonable amount of time. Furthermore, the rotating setup necessary to acquire multiple projections must be small, to enable a sufficiently high numerical aperture to achieve high resolutions in the XUV range. A compact apparatus, which enables three-dimensional imaging of extended samples using radiation in the XUV and soft x-ray region, promises to find broad application in technology and particularly in life sciences.

5.5. Time-resolved experiments (pump-probe)

The most striking advantage of high-harmonic sources compared to others is their ultra-short pulse duration, ranging from a few hundred femtoseconds down to 43 attoseconds [96, 97]. Moreover, the generated harmonics are phase locked to the driving laser and to the other harmonics. This enables the observation of ultrafast dynamics with femto- to attosecond precision via pump—probe experiments [98–100]. A combination of the nanoscale resolution of CDI techniques with this high temporal resolution would provide novel insights into ultrafast processes at the nanoscale. Examples include ultrafast electron dynamics, ultrafast magnetization dynamics, and ultrafast heat transport. Repeatable processes might be imaged with multi-pulse accumulation, while non-repeatable processes require single-shot images. Ultimately, the combination of short pulses (with large bandwidth) and high spatial resolution (usually requiring a narrow bandwidth) remains challenging, and advanced approaches relaxing the bandwidth requirements of coherent imaging techniques will be required [101, 102].

6. Summary

In this article, we have presented and discussed the state of the art in table-top nanoscale coherent imaging with coherent XUV light sources based on high harmonic generation. The availability of high harmonic sources with sufficiently high photon flux and excellent coherence properties has enabled nanoscale imaging on the table top with impressive performance. Within a few years, the field has advanced from the first table-top demonstrations on isolated test samples to reliable, routine imaging of extended and complex samples via ptychography with the highest resolutions. Such imaging devices are now ready to address real-world imaging. They will soon complement existing imaging methods in many fields of technology and science, ranging from solid-state physics via nanotechnology up to life sciences. In contrast to synchrotron-based systems, the table-top devices are compact, easy to use, and can be integrated into standard laboratories. These huge advantages will foster their dissemination into research and even industrial applications like actinic semiconductor mask inspection. In future, the output power of high harmonic sources will steadily increase with advances in laser technology. Thus, table-top coherent imaging is expected to be available in the soft x-ray region—including the biologically relevant water window—soon. Increased photon flux of the sources will also enable recording full 3D ptycho-tomography datasets in reasonable measurement times and thus enable unique insight into matter at the smallest scales. Complementary, spectro-microscopy approaches are currently in development, which will allow mapping of chemical elements at the nanoscale.

In addition, the ultra-short pulse durations of high harmonic sources will open unique possibilities in the study of the ultrafast dynamics of e.g. electrons, spins, energy levels, and band-structure at the nanoscale. The development of
adequate techniques enabling both high spatial and temporal resolution is under way.

Acknowledgments

This work was supported by the Fraunhofer Cluster of Excellence Advanced Photon Sources, the Federal State of Thuringia (2015 FGR 0094) and the European Social Fund (ESF).

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