Topical Review

Single-pass high harmonic generation at high repetition rate and photon flux

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Abstract

Sources of short wavelength radiation with femtosecond to attosecond pulse durations, such as synchrotrons or free electron lasers, have already made possible numerous, and will facilitate more, seminal studies aimed at understanding atomic and molecular processes on fundamental length and time scales. Table-top sources of coherent extreme ultraviolet to soft x-ray radiation enabled by high harmonic generation (HHG) of ultrashort pulse lasers have also gained significant attention in the last few years due to their enormous potential for addressing a plethora of applications, therefore constituting a complementary source to large-scale facilities (synchrotrons and free electron lasers). Ti:sapphire based laser systems have been the workhorses for HHG for decades, but are limited in repetition rate and average power. On the other hand, it has been widely recognized that fostering applications in fields such as photoelectron spectroscopy and microscopy, coincidence detection, coherent diffractive imaging and frequency metrology requires a high repetition rate and high photon flux HHG sources. In this article we will review recent developments in realizing the demanding requirement of producing a high photon flux and repetition rate at the same time. Particular emphasis will be put on suitable ultrashort pulse and high average power lasers, which directly drive harmonic generation without the need for external enhancement cavities. To this end we describe two complementary schemes that have been successfully employed for high power fiber lasers, i.e. optical parametric chirped pulse amplifiers and nonlinear pulse compression. Moreover, the issue of phase-matching in tight focusing geometries will be discussed and connected to recent experiments. We will highlight the latest results in fiber laser driven high harmonic generation that currently produce the highest photon flux of all existing sources. In addition, we demonstrate the first promising applications and discuss the future direction and challenges of this new type of HHG source.

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

1. Introduction

Initially dubbed ‘a solution looking for a problem’ the laser has beyond any doubt revolutionized many scientific and industrial fields [1, 2]. The ability to produce short pulses of coherent light has fascinated researchers ever since the first demonstration of the laser in 1960 [3] and has enabled the achievement high field strengths. This led to the discovery of nonlinear phenomena [4] and their explanation by the quantum-mechanical perturbation theory in the early 1960s [5]. In the following years efforts were made to further reduce the pulse durations and increase the intensities, i.e. the electrical field strengths, available from laser sources to enable the tracking of ultrafast processes and the study of light–matter interactions. In that regard the realization of mode-locked laser oscillators with femtosecond pulse duration [6] and the chirped pulse amplification (CPA) scheme [7] allowed the achievement of field strengths of the order of inner atomic fields. Using these high intensity lasers has led to several experiments that showed significant deviations from the established perturbative description of nonlinear optics, i.e. above-threshold ionization [8], non-sequential double ionization [9] and high harmonic generation (HHG) [10, 11]. The latter experiments were an extension of harmonic generation [4] to address the ultraviolet spectral region, at which point the occurrence of a plateau region, i.e. a large number of harmonics with approximately the same intensity, and a sudden cutoff frequency [12], could not be explained by existing theory [5]. It was found early on that the maximum harmonic order or the photon energy $E_{\text{max}}$ that can be obtained in the process is proportional to $E_{\text{max}} = I_p + 3U_p$, where $I_p$ is the ionization potential and $U_p$ the ponderomotive potential, i.e. the mean kinetic energy an oscillating electron acquires in the laser field [12]. This finding has motivated many of the early works that were aimed at extending the cutoff to shorter wavelengths [13–18]. Furthermore, this universal cutoff law led to extensive studies and was finally explained by a semi-classical explanation based on a so-called three step model [19, 20]. Due to the high field strengths of the laser, the atom potential is bent such that a potential barrier exits, where an electron can tunnel through, which leads to ionization and constitutes the first step. This free electron is accelerated in the second step by the laser field and can recombine, upon field-reversal, with the parent ion in the third step. During recombination a highly energetic photon is radiated that has a photon energy equal to the kinetic energy of the returning electron and the binding energy. This simple semi-classical model explains most of the observed experimental phenomena very well. For example it supports the occurrence of a cutoff frequency

$$\omega_c = \frac{(I_p + 3.17U_p)}{c},$$

where is $I_p$ is the ionization potential and $U_p = 9.33I^2/\lambda^2$ ($I$—intensity in $10^{14}$ W cm$^{-2}$, $\lambda$—wavelength of the driving laser in $\mu$m) the ponderomotive potential [21]. This cutoff law matches the original experimental findings very well. The three step model was validated by a fully quantum-mechanical model, known as the Lewenstein model [22]. All of the aforementioned models describe single atom effects, but it was also recognized that phase-matching effects play a pivotal role in not only maximizing the yield of HHG [23, 24], but also obtaining some of the attractive properties of the harmonics, e.g. coherence and ultrashort pulse duration. It was known from ‘conventional’ harmonic generation that spatial and temporal coherence are transferred to the newly generated fields, which is a natural consequence of the perturbation theory [5]. However, it was not convincingly clear if this held for the non-perturbative process of HHG. Theoretical and experimental studies were then used to show that the generated harmonics can indeed be generated with good coherence properties if proper phase-matching conditions are achieved [25]. Another interesting feature that attracted significant attention is the time structure of the emitted harmonics. The comb-like structure of the HHG spectrum in fact looks very similar to the longitudinal modes of a mode-locked ultrashort pulse oscillator that produce a train of sub-femtosecond pulses if they are phase-locked [26, 27]. It was theoretically shown that propagation, i.e. phase-matching, effects are necessary to appropriately phase the harmonics and generate attosecond pulses [28, 29]. New measurement techniques finally allowed the validation of these attosecond pulse trains (APTs) [30, 31]. The occurrence of APTs is a consequence of the simple three step model that predicts one emission event every half-cycle of the driving laser field. Moreover, the latter hinted at the possibility of creating single isolated attosecond pulses (IAPs) by using a temporal polarization variation of the driving laser [32] or few-cycle pulses with less than two cycles in duration [33]. This, essentially, would restrict the emission of the highest harmonic orders to one event per laser pulse and simple frequency filtering would create the IAP [32, 33]. Impressive advances in laser technology allowed the production of energetic 5 fs pulses from post-compressed Ti:sapphire lasers [34, 35] that led to the first experimental observation of IAPs in 2001 [36]. Later on this was further complemented by the ability to control the carrier envelope phase [37] and, therefore, fully control the waveform of the electrical field of the laser pulse constituting the technological foundation for the field of attosecond physics [38, 39], which has emerged as a completely new research direction [40–42]. Starting from simple systems, attosecond physics is currently aimed at investigating complex atomic and molecular systems on their natural (electronic) time scales and holds the promise of the investigation of many fundamental processes in physics, chemistry and biology [41, 43, 44].
One of the most charming advantages of HHG sources is their realization on a table-top, making them particularly attractive as an alternative to large-scale facilities, such as synchrotrons or free electron lasers, which only offer restricted user access. Due to their outstanding properties, i.e. coherence, short wavelength and femto- to attosecond pulse durations, high harmonics have found numerous applications over the years. The application fields are as diverse as atomic and molecular physics and spectroscopy [45–56], surface science [57–61], material science [62–66], microscopy [67–73] and other emerging fields [74, 75]. However, one of the bottlenecks is the conversion efficiency that ‘typically’ is of the order of $10^{-6}$ for plateau harmonics, and even lower for cutoff harmonics, leading to a limited photon flux, which hinders or prevents many potential applications [42, 76].

In recent years, three important requirements for further advancement of the field were identified [77–80]. The first obvious requirement is the possibility of extending phase-matching concepts to much shorter wavelengths so as to address the absorption edges of many materials, improve resolution in microscopy and eventually enable zeptosecond pulses [81, 82]. In that regard impressive progress has been made by using long wavelength ultrashort pulse drivers to reach into keV photon energy regions [81, 83–86]. The other two challenges are to increase the energy of extreme ultraviolet (XUV) pulses to address nonlinear effects in this spectral region [78, 87, 88] or to increase the number of XUV pulses per second, i.e. their repetition rate, and the overall photon flux at the same time. The latter is particularly important for exploiting novel applications of coherent short wavelength sources [77, 79]. A high repetition rate source is, for example, beneficial for avoiding space-charge effects in photoemission spectroscopy and microscopy [58, 59, 77, 89, 90], addressing novel experiments that rely on coincidence detection of ionization fragments [56, 91] or enabling frequency metrology in the XUV [92]. Experiments building on coincidence detection have to be operated such that less than one fragmentation event per laser shot is induced on average to unambiguously correlate fragments to each other. Consequently, a sufficient statistic requires the acquisition of many laser shots and can only be achieved with multi-kHz sources, which is beyond the operating parameters of current free electron lasers [91, 93].

However, achieving HHG at high repetition rates puts stringent requirements on laser technology or the generation conditions of HHG. When simply keeping the average power constant, the required focusing will be so tight that efficient generation might be prevented due to various limitations as recently discussed in [94]. On the other hand, a simple comparison to 1 mJ, 1 kHz (1 W of average power) Ti: sapphire laser systems, which are the workhorses for HHG and attosecond science, reveals that kilowatt level femtosecond lasers are required for MHz repetition rates.

Over the years different techniques have emerged to tackle this problem, and two of these approaches circumvent the aforementioned severe requirements on laser technology. The first one of these methods utilizes field enhancement on certain nanostructures to create the necessary field strengths directly with multi-10 MHz oscillators, and was first demonstrated in 2008 [95]. Extensive research has been devoted to promote this field, but it is still heavily debated whether the nanostructures remain intact and whether the emitted radiation is coherent HHG or incoherent atomic line emission [96–100].

A second approach uses passive high finesse external resonators to enhance incoming low energy, but high repetition rate ultrashort laser pulses [101]. Consequently, this method is referred to as femtosecond enhancement cavities (fsEC), which are operated between 10 MHz and 250 MHz [92, 102–106]. The enhancement factors that can be achieved are of the order of 1000, leading to several kilowatts of average power inside the fsEC. Harmonic generation is achieved in a focus inside of the evacuated resonator, where a gas jet supplies the necessary nonlinear medium. One of the most challenging parts is to couple-out the so generated radiation, which co-propagates with the circulating laser power. Different methods, e.g. Brewster plates [101], XUV grating on top of a highly reflecting mirror for the fundamental [107], anti-reflection coated grazing incidence plates [108, 109], drilled holes in cavity mirrors [110, 111] or the use of non-collinear geometries and higher order modes have been suggested to resolve this issue [112, 113]. The technology of fsEC has matured since the first successful demonstration in 2005 [101]. For example, a high XUV power of up to 200 μW at 12.7 eV (intra-cavity) was generated [92]. Moreover, the latter experiment succeeded in realizing a stable XUV frequency comb and used this for precise transition measurements, which opened up fascinating prospects in frequency metrology [92, 114]. Recent years have seen steady efforts to increase the intra-cavity average power, achieve higher photon energies or even to realize isolated attosecond pulses at multi-MHz repetition rates [113, 115, 116].

The third technique, which will be the subject of this review, directly uses high average power lasers to drive the HHG process. In principle, this is motivated by rapid progress in ytterbium-doped femtosecond laser technology that has seen power levels in the kilowatt range for Innolaser [117], thin-disk [118] and fiber lasers [119]. In this paper we attempt to review recent advances in HHG with high repetition rate and average power lasers. We solely focus on the use of single-pass harmonic generation and refer to existing literature on the related subjects of using enhancement cavities or plasmonic field enhancement [95, 99, 100, 104, 105, 120, 121]. General aspects of HHG, such as the dependency of cutoff and efficiency on the pulse duration, and the possibility of efficiently generating harmonics in tight focusing geometries are presented in section 2 and underlined with experimental results. In sections 3 and 4 we will describe advances in fiber laser technology and subsequent nonlinear compression and optical parametric amplification, which are the fundamental building blocks for achieving efficient HHG with high repetition rate lasers. Recent experimental advances in high repetition rate HHG and attosecond pulse generation will be reviewed in section 5, concluding with the current state-of-the-art in this field. One of the major motivations for the realization of such new XUV sources is their expected broad application range, which will be underlined by first
experiments described in section 6. We will conclude with a summary and outlook on further potential in this exciting field, but we will also address challenges that are expected along the way.

2. Efficient HHG with low pulse energies: phase-matching in the tight focusing regime

On the single atom level the process of HHG is well described by the three step model explained above. The emitted radiation of such a single atom is described by the dipole moment, which depends on many factors, e.g. the intensity or wavelength of the driving laser as well as the gas medium used for generation. However, a macroscopic buildup of all the emitters located inside the generation medium and therefore the highest conversion efficiency can only be achieved when the emission of the single emitters is phase-matched. This can be investigated by considering the wave-vector mismatch \( \Delta k = qk_0 - k_q \) between the fundamental \((k_0)\) and the generated \(q\)th harmonic \((k_q)\). Only if the phase velocities of both fields are equal, i.e. \( \Delta k = 0 \), will efficient buildup of the signal occur. For the process of HHG the wave-vector mismatch term is time- and density-dependent and contains terms due to the dispersion of atoms and free electrons, focusing and the phase of the atomic polarization. It can be written as [24, 42, 122–125]

\[
\Delta k(t, \rho) = qk_0 - k_q = \left[ \frac{2\pi \rho}{\lambda_0 \rho_0} \Delta \delta \right] \left( 1 - \frac{\eta(t)}{\eta_{C,q}} \right) - \frac{1}{\varepsilon_R + \frac{z_0^2}{z_R}} \left( z_R + \frac{z_0}{\varepsilon_R} \right)
\]

where \( \lambda_0 \) is the laser wavelength, \( \rho_0 \) the number density at standard condition (1013 mbar), \( \rho \) the number density in the interaction region, \( \Delta \delta = \delta_0(\lambda_0) - \delta_0(q) \) the refractive index difference between the fundamental \((\lambda_0)\) and \(q\)th harmonic \((\lambda_q)\) [123], \( z_0 \) and \( z_R \) the Rayleigh range and position relative to the focus, and \( \alpha \) the phase-coefficient for the dipole phase [25, 126, 127]. Equation (2) shows that pressure-induced phase-matching is only possible if the fraction of the ionized medium \( \eta \) is smaller than the so-called critical ionization \( \eta_c \), which is typically of the order of a few percent depending on the gas species and harmonic order [42, 123]. If this condition is met, choosing the appropriate density, i.e. pressure, in the interaction region leads to \( \Delta k = 0 \) and, therefore, phase-matching. However, this constrains the usable intensity to avoid excessive ionization and imprints a strong dependence of the efficiency and phase-matching properties on the pulse duration.

This fact is illustrated in figure 1(a), which shows the fractional ionization \( \eta \) in xenon calculated for laser pulses with a central wavelength of 1030 nm using the established ADK model [128]. The intensity of the 500 fs (blue), 100 fs (green) and 30 fs (red) pulses has been chosen such that 95% of the critical ionization, which is indicated by the dotted black line, is reached at the pulse peak \((t = 0)\). (b) Based on (a) the signal of the 25th harmonic in xenon has been calculated, using a one-dimensional model [24], for pulse durations between 20 fs and 500 fs, showing a clear increase of conversion efficiency (see text for more details).

![Figure 1](image-url)
model introduced by Constant et al [24, 124], which allows the study of the buildup of the HHG signal along a certain laser pulse. This model includes an empiric scaling law for plateau harmonics, modeling the HHG signal dependence to be proportional to $\rho^{8.2}$ [24, 124]. Therefore, a higher intensity will significantly increase the single atom dipole moment. In addition, phase-matching will only be transiently achieved during the laser pulse due to the temporally varying wave-vector mismatch (equation (2)). As a consequence, the HHG conversion efficiency increases rapidly with decreasing pulse duration, as illustrated in figure 1(b), which shows the temporally integrated (over the laser pulse) signal of the 25th harmonic in xenon for a laser pulse of varying duration and a central wavelength of 1030 nm. For each pulse duration, similar to the calculations in figure 1(a), the intensity has been chosen such that $\eta(t = 0)/\eta_c = 0.95$ and phase-matching can be achieved. Furthermore, absorption limited conditions have been chosen [24]. Figure 1(b) shows that this simple model gives an increase in the HHG signal by more than one order of magnitude when going from 500 fs to 30 fs. As already mentioned the achievable signal by more than one order of magnitude when going to 30 fs.

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mentioned above. The other quantities (see table 1) can be scaled accordingly and it turns out that the harmonic conversion efficiency $\eta' = \eta$ is independent of the focusing conditions [135, 136].

As a side note it has to be mentioned that the scaling laws deduced here seem to be even more universal, and can also be applied to other nonlinear processes such as filamentation [94]. Interestingly, other limiting effects such as absorption, defocusing or defocusing also scale accordingly, leaving the conversion efficiency unchanged [136]. However, there might be other limitations, such as the difficulty of confining the high density gas medium to very small spatial scales [135]. Furthermore, at high densities the presence of neighboring atoms or avalanche ionization effects might prevent downscaling even before reaching the limits, e.g. of the paraxial approximation [94].

The discussion above is general and can be applied to every HHG experiment. To test the validity of the findings regarding tight focusing we performed an HHG experiment with an optical parametric chirped pulse amplifier system delivering $\sim 8$ fs pulses at 820 nm central wavelength (sections 4.1, 5.4, 5.5). The focus in that experiment was as small as $w_0 = 15 \mu m$ and different experimental parameters were tested and optimized [136]. Figure 3 shows the number of photons s$^{-1}$ for the harmonic orders 17–27 with respect to the xenon backing pressure applied to a 150 $\mu m$ gas nozzle. Interestingly, more than $10^{12}$ photons s$^{-1}$ can be obtained in the strongest harmonic (H17, 25.7 eV) leading to a conversion efficiency as high as $8 \cdot 10^{-6}$, which is very close to benchmark experiments performed under loose focusing conditions [24]. Furthermore, the minima, which appear in the harmonic signal with respect to pressure (figure 3(a)) can be used to measure the phase-matching conditions and show that the efficiency is only limited by reabsorption in the generating medium [136].

In summary, it has been shown that efficient, i.e. absorption limited, HHG is possible even in tight focusing geometries if appropriately dense gas targets can be supplied. In this case there is, theoretically, no difference between using a high or low energy laser system, which has been experimentally verified by achieving $8 \cdot 10^{-6}$ conversion efficiency with a focal spot size of $w_0 = 15 \mu m$. It is rather important to use ultrashort laser pulses of 10 cycles or less in duration to avoid excessive ionization and achieve sufficiently high efficiencies and photon energies. Ideally, these parameters can be obtained at the highest possible average power to achieve unprecedented photon flux levels in HHG.

3. High average power fiber lasers

The requirements for achieving a high photon flux coherent short wavelength source were described in section 2. A high average power ultrashort pulse laser system is necessary, which can be realized by InnoSlab [117], thin-disk [118] or fiber lasers [119]. Here, we focus on fiber lasers and the power scaling concept of coherent combination, since the other technologies have been little used for HHG so far [137, 138]. Regardless of the laser architecture used, techniques to obtain shorter pulses have to be applied to the high average power lasers. Due to the limited gain bandwidth of the ytterbium ions, which are used as the laser medium, these systems emit rather long pulse durations of a few hundreds of femtoseconds. Techniques that can be applied to each of the aforementioned laser architectures will be described in section 4.

3.1. Femtosecond fiber CPA

The most severe limitations in the amplification of ultrashort laser pulses with fibers are damage and nonlinearity, which are generally avoided by reducing the intensity in the amplifiers as much as possible [139]. Therefore, a first concept that can be applied is CPA, i.e. stretching the pulses in...
time prior to amplification and compressing them back afterwards [7]. Another important aspect is the diameter of the signal beam propagating through the fiber. Obviously, a larger beam diameter allows for higher pulse energies and, therefore, for higher peak powers, while avoiding detrimental effects on the temporal pulse quality via nonlinearity. On the other hand, standard step-index fibers are only single-mode for signal core diameters of less than 15 μm [140]. Increasing the diameter further leads to multi-mode output, which decreases the spatial beam quality, leading the fiber to lose one of its trademark features. Therefore, new micro-structured fiber designs have been developed such as the photonic-crystal fiber (PCF) [141] and large-pitch fiber [142] that allow effective single-mode operation at signal core diameters of up to 100 μm.

Over the last few decades tremendous improvements have been made in increasing the average power, pulse energy and peak power of femtosecond fiber laser systems. The increase on the average power side has been mostly driven by the availability of high power semiconductor based pump diodes with a high enough beam quality to couple the light into the fiber. As a consequence, femtosecond fiber lasers running at very high repetition rates have been demonstrated at average powers close to 1 kW [119] and pulse energies of up to 2.2 mJ [143]. However, with regard to the average power the observation of a threshold-like degradation of the beam quality above a certain average power value constrains further power scaling. This effect, which is called mode-instabilities, is currently a major research topic aimed at understanding and mitigating this effect [144–146].

3.2. Coherent combination as a power scaling concept

In order to scale the performance of femtosecond fiber laser systems even further, the use of multiple parallel main amplifiers, together with coherent combination of the amplified pulses, can be employed. Assuming identical behavior of the amplifiers and perfect efficiency of the combination process, this concept enables increasing the average power, peak power and pulse energy by a factor of \( N \), with \( N \) being the number of amplifier channels [147].

A generic setup of such a system is shown in figure 4. The pulses coming from a front-end system are split up into the \( N \) parallel amplification channels. This ensures mutual coherence of the input pulses for each amplifier. After amplification, the pulses are recombined again using a suitable combination element. In order to realize the coherent combination at the output, the path lengths in the channels need to be matched with sub-wavelength precision. This is implemented by making the path lengths adjustable with elements such as piezo-mounted mirrors or phase modulators. By employing an active phase stabilization system, these adjustments can be continuously made during operation to compensate for path-length fluctuations. In addition, differences in the temporal and spectral properties of the pulses after amplification, caused by differences between dispersion and nonlinear effects in the various channels, have to be kept low to avoid a drop in combination efficiency [148]. If these conditions are fulfilled, then the coherent combination concept can be successfully implemented in experimental setups. It has to be noted that fiber laser systems are particularly appropriate for this concept, due to the excellent parallelization abilities, but it can also be applied to other amplifier architectures [149].

So far, systems comprising four parallel main amplifiers delivering gigawatt peak powers and average powers of more than 0.5 kW have been demonstrated [150], as well as peak powers of up to 22 GW [151]. These performance values clearly surpass currently available systems using a single fiber amplifier. It is expected that the performance will increase further in the future by increasing the number of channels [147], which can be supported by further compact integration of the channels, e.g. by using multicore fibers [152]. However, regardless of using fiber CPA systems with or without coherent combination or other approaches (Innoslab, thin-disk), the achieved pulse durations are between 200 fs and 500 fs, which is too long for efficient HHG.

4. Generation of ultrashort to few-cycle pulses with fiber lasers

High average power femtosecond laser systems can be routinely realized by applying the coherent combination concept as a power scaling approach to fiber lasers (section 3). However, it has been discussed in detail in section 2 that approximately 10 cycle pulses, i.e. 34 fs pulses at 1030 nm central wavelength, have to be used for efficient HHG. The requirements on pulse duration become even more demanding when IAPs have to be generated, which require carrier envelope phase stable sub-2 cycle pulses when no other gating methods are applied [78].

In the following, the concepts of optical parametric chirped pulse amplification (OPCPA) and nonlinear compression (NC), along with their respective advantages and
4.1. Optical parametric chirped pulse amplification

Optical parametric amplification (OPA) is a second order nonlinear process, which allows the amplification of a weak signal beam of frequency $\omega_s$ with a strong pump beam at frequency $\omega_p$. During this process a new frequency component, the so-called idler ($\omega_i$), is generated due to energy conservation ($\omega_p = \omega_s + \omega_i$) [153]. OPA has attracted a lot of interest because of e.g. its huge single-pass gain, the ability to address signal wavelength ranges that are not covered by conventional laser materials and the potential for enormous phase-matching bandwidths allowing the amplification of few-cycle pulses [154, 155]. The combination of the concepts of OPA and CPA eventually led to the technology of OPCPA, which was first realized by Dubietis et al in 1992 [156]. Another important aspect is that the OPA process preserves the carrier-envelope phase (CEP) stability of the seed signal, regardless of the phase stability of the pump beam, because all phase variations of the pump are transferred to the newly generated idler wave [157]. High repetition rate and high average power fiber ytterbium-doped laser systems, e.g. fiber lasers, have proven to be ideal pump sources for ultrabroadband OPCPA systems. In consequence, the combination of different high average power laser systems with OPCPA has led to an immense increase in the repetition rate of intense few-cycle laser sources by two orders of magnitude with respect to Ti:sapphire driven systems [158–163].

Figure 5 shows the schematic realization of such a high average power fiber chirped pulse amplifier (FCPA) system and a two-stage OPA pumped by the frequency doubled FCPA output. Spectral phase control of the few-cycle pulses is achieved with a combination of a spatial light modulator (SLM) and chirped mirrors. Due to the high average power of the FCPA, the system can deliver up to 22 W of average power at a repetition rate of 1 MHz [158].

However, this system was limited by the onset of thermal effects inside the nonlinear crystals, such as change of phase-matching conditions, thermal lensing and damage due to thermally induced stress [164]. Even though the OPA process does not store energy within the crystal (unlike the conventional laser process), residual linear absorption of all interacting waves has been identified to cause detrimental heat load. In particular, the idler can easily penetrate infrared absorption bands and its restriction to spectral regions with high transmission can significantly reduce thermal effects inside the nonlinear crystal. Nevertheless, the observed effects are challenging further average power scaling of ultrabroadband OPCPA systems. However, several techniques might reduce thermal effects, such as better heat removal from the crystal by bonding it to materials with a high thermal conductivity [165] or the distribution of the heat load into several spatially separated OPA stages, which can be recombined afterwards [166, 167].

4.2. Nonlinear compression in gas-filled hollow waveguides

The concept of nonlinear compression builds on a simple concept that was proposed in 1969 for the first time [168]. It relies on the nonlinear interaction of an intense laser pulse with matter. More precisely, third order nonlinearity leads to frequency modulation (temporal chirp), the so-called nonlinear phase, via the optical Kerr effect. This process, called self-phase modulation, in turn leads to a broadening of the spectrum, which is the key aspect. Obviously, the subsequent removal of the chirp via dispersive delay lines leads to temporal compression of the pulses and a peak power
Figure 6. Generic setup of a nonlinear compression experiment used for high average power fiber laser. The setup can be either operated with one or two compression stages as described in the text.

enhancement if the losses are low enough. In principle, it can be applied to arbitrary laser systems that are bandwidth limited by different factors, which makes it a very attractive and common approach. The spectral broadening is often performed in waveguides to enhance the interaction length and to avoid spatial chirp. The choice of the waveguide is dictated by the performance of the laser system, most importantly by its peak power (pulse energy). For sub-μJ level pulses common solid core (glass) fibers can be used that are limited by the onset of self-focusing at 4 MW (6 MW) of peak power for linear (circular) polarization. For higher pulse energies gas-filled waveguides can be used. In 1996 the use of gas-filled capillaries was successfully applied to mJ-class Ti:sapphire laser systems [34, 169–171]; an approach that has become the standard in this pulse parameter regime. The use of laser pulses with several μJ has been difficult in that regard for years, since capillaries have to be filled with extremely high-pressure gas or small-diameters have to be used, leading to unacceptably high propagation losses, and solid fibers would simply be destroyed. With the availability of a novel PCF, called Kagome, this situation changed dramatically. Kagome PCFs behave like small-core capillaries, but have a significantly lower propagation loss [172]. Therefore, they are excellent candidates for pulse compression of tens of μJ pulses [173–176]. It has to be noted that all three waveguide architectures (solid core, Kagome, capillary) have already shown high average power operation, e.g. 0.9 μJ, 23 fs pulses at 250 W in solid core fibers [177], 7 μJ, 30 fs at 76 W in Kagome fibers [176] and 0.55 mJ, 26 fs pulses at 135 W in capillaries [178].

Despite the potential for achieving phase-matching even in the tight focusing regime, a driving laser system requires a few microjoules of pulse energy. Therefore, the common approach to addressing this parameter regime relies on gas-filled waveguides, where both Kagome fibers or capillaries are employed.

Figure 6 shows a generic setup of a typical nonlinear compression experiment. A fiber laser system delivers 200–300 fs pulses with energies between a few microjoules and a few millijoules, which are coupled to the waveguide of choice (capillary or Kagome) with a length of ~1 m. The gas filling, i.e. the type and pressure of the gas, is adapted to the input parameters of the laser system. In that regard the two major limitations of self-focusing and ionization have to be considered [179]. For a given input peak power the first effect limits the gas pressure (nonlinear refractive index n2) and, therefore, the achievable spectral broadening. Ionization, on the other hand, occurs for too-high intensities at the entrance of the waveguide and can be avoided by either using a gas with higher ionization potential or using a larger core size [179]. The waveguide itself sits in a V-groove that is mounted inside a water-cooled tube to ensure stable operation even with high average power. After spectral broadening the pulses are compressed in time to typical durations of <30 fs after the first compression stage. Then the output can be either used directly (sections 5.1 and 5.3) or further pulse shortening may be achieved in a second compression stage, which is identical in its setup and yields 6–8 fs pulses (section 5.2). Furthermore, it has to be mentioned that in general the process of self-phase modulation maintains the carrier envelope phase stability. Although not yet realized, further advances in fiber laser technology will aim to also implement CEP stability for nonlinearly compressed fiber laser systems.

This very simple approach enables us to address a broad range of output parameters (pulse energy, pulse duration, repetition rate) for various experiments in HHG, as outlined in the following section.

In summary, both methods (OPCPA, NC) can be applied to obtain ultrashort to few-cycle laser pulses at high average power using fiber lasers (or other) as front-end. It has been shown that OPCPA systems are the choice if tunability (section 5.4) and broadband amplification is required. Moreover, it is straightforward to implement CEP stability, since, e.g. established state-of-the-art Ti:sapphire oscillator technology can be used as an OPCPA seed. However, as discussed above, the linear absorption of pump, signal or idler puts severe constraints on the average power scalability of this concept and it remains open, i.e. advanced heat spreading techniques help to overcome this issue. On the other hand, gas-filled waveguides seem to offer superior average power scaling. Recently, kilowatt level cw lasers were successfully coupled and transmitted through both capillary and Kagome, hinting at a possible power increase in the near future [180]. Moreover, the advances in coherent combination of femtosecond fiber lasers should help to reach new power levels. Although not yet demonstrated, and technically challenging, the stabilization of the carrier envelope phase of CC-FCPA systems in combination with nonlinear compression will be a significant step towards >100 W few-cycle laser systems, which will be of extreme importance for many strong field applications.

5. High harmonic and attosecond pulse generation with fiber lasers

Experiments on high repetition rate HHG started in 2003 [133] and soon after many groups followed with different approaches, i.e. plasmonic field enhancement, fsEC and single-pass HHG, as described in the introduction. In recent
years some work has also been devoted to the use of commercially available easy-to-use lasers with high repetition rate for HHG [59, 181–183]. The potential of fiber lasers in that regard was realized in 2009 when the first high harmonic generation experiments were conducted by Boullet et al [184]. In that experiment a 270 fs fiber laser with an average power of up to 28 W and 1 MHz repetition rate was used. As shown in figure 7 it was possible to generate harmonics at various repetition rates up to 1 MHz.

Although not explicitly stated, it can be expected that the conversion efficiencies were low because 270 fs pulses were used. Nevertheless, further work resulted in a significant photon flux of $>10^{12}$ photons s$^{-1}$ per harmonic (H15, 18 eV) despite the use of 500 fs driving pulses [185]. In parallel it was shown that high average power fiber lasers in combination with an optical parametric chirped pulse amplifier and nonlinear compression (sections 3 and 4) offer perfect parameters for increasing the conversion efficiency [186, 187] already resulting in microwatt level harmonics [188]. In recent years the investigation and realization of phase-matching in tight focusing geometries [135, 136] led to significant advances in achieving unprecedented photon flux and repetition rates simultaneously, which will be outlined in this section.

All of the experiments summarized in this section have utilized fiber lasers as front-end for either nonlinear compression or an OPCPA system as shown in the schematic setup in figure 8.

The latter is used to obtain the required ultrashort pulses at high average power (sections 3 and 4). HHG is then achieved by focusing the pulses with a lens or a mirror onto a gas target supplied by a nozzle with a round orifice. Of course, there are alternative target geometries such as gas cells [189], squeezed tubes [135] or waveguides [42], which are not discussed here since they can be challenging to realize for tight focusing geometries. The generated harmonics and the fundamental laser are separated by one or several thin metal filters, unless otherwise stated, and are then analyzed with a flat-field grating spectrometer.

### 5.1. High photon flux XUV sources

As mentioned earlier, fiber laser driven HHG was able to achieve microwatt level harmonics [185, 188], but in the early stages still lagged behind many other experiments in terms of conversion efficiency. On the other hand, the emergence of coherent combination techniques allowed the increase of the average power of fiber lasers and also nonlinear compressed pulses [147, 178]. For the experiments discussed in this section, which are described in more detail in [190], a 4-channel FCPA system was used, which delivered 270 μJ, 340 fs pulses at a repetition rate of 600 kHz (163 W of average power). As discussed in section 2 much shorter pulses are required to efficiently convert the laser radiation into the XUV spectral region. This is achieved by propagating the pulses in a krypton filled capillary for spectral broadening and subsequent pulse compression with chirped mirrors. After this pulse compression step $>130 \ μJ$, 29 fs pulses with $>80$ W of average power are used for HHG. For that purpose the output of the compression stages is focused to a focal spot diameter of 90 μm ($1/e^2$-intensity) inside a xenon or krypton gas
Due to the high average power of the laser the separation of the XUV from the fundamental cannot directly be done with aluminum filters. Therefore, both the infrared laser and the generated harmonics pass a chicane of grazing incidence plates, which are fused silica substrates that possess an anti-reflection coating for the laser radiation at an angle of incidence of 75° and an additional top-SiO2 layer to reflect the harmonics. This method suppresses the infrared laser to roughly 1%, allowing us to use additional aluminum filters to further reduce the stray light before the harmonics are analyzed with a flat-field grating spectrometer or an XUV photodiode. Figure 9 shows the results of HHG at 600 kHz in xenon (a) and krypton (b) gas after all experimental parameters are optimized. Obviously, phase-matching can be achieved, which is indicated by the good conversion efficiency and Gaussian-like beam profiles of the generated harmonics obtained (figure 9). Using xenon gas, it is possible to obtain several plateau harmonics between 25 eV and 33 eV with >100 μW of average power. The strongest harmonic that was generated was harmonic 25 (30 eV) with 143 μW of average power, which corresponds to 3 \cdot 10^{13} \text{ photons s}^{-1} and constitutes the highest photon flux that has been obtained for all harmonic sources in that wavelength region so far (section 5.6). Strong plateau harmonics with more than 30 μW of average power (>5 \cdot 10^{12} \text{ photons s}^{-1}) can be obtained up to \sim 40 eV by using krypton gas. All the mentioned power levels are corrected for filter and spectrometer transmission and the detection efficiency of the used CCD camera as described in [190].

5.2. High photon flux soft x-ray generation up to the water window

As described in section 5.1, the use of \~30 fs pulses from a combination of a fiber laser and nonlinear compression facilitates strong harmonics between 25 eV and 40 eV (figure 9). However, the phase-matching aspects discussed in

**Figure 9.** High harmonics generated in xenon (a) and krypton (b) with a 600 kHz fiber laser. The upper panels show the spatial spectral lineout of the harmonics and the respective harmonic order. The curves on the lower panel were obtained by integrating along the spatial direction of the upper panel data. The numbers on each harmonic order are the average power levels that can be calculated by an additional spectral integration of the respective harmonic orders. The power levels are the ones generated, i.e. directly after the gas jet. (Reproduced from [190].)
section 2 (figure 2) prevent the efficient generation of higher photon energies and shorter pulses are required to extend the harmonic cutoff.

This is addressed by adding a second nonlinear compression stage for further pulse shortening. The experiment was performed with a 1 mJ, 210 fs coherently combined fiber laser system and the first compression stage delivered the same 30 fs pulses as used in section 5.1, but with a higher pulse energy of 550 μJ. These pulses are further compressed in time by a second stage that provides sub-8 fs pulses with up to 350 μJ of pulse energy and an average power of up to 53 W at 150 kHz repetition rate [191]. However, the silver mirrors used for beam steering of the few-cycle laser pulses show thermal effects at this high average power.

As a consequence, the experiments on HHG with this laser could only be conducted at a repetition rate of 100 kHz, but this rather technological challenge appears to be solvable in the near future [180]. The few-cycle pulses are sent to the same high harmonic generation setup as described in section 5.1, but are focused with a spherical mirror to a focal spot size of 45 μm (1/e²-intensity) to an intensity of up to 2 · 10¹⁵ W cm⁻² depending on the actual generation conditions (gas, harmonic order). Figure 10 shows the results for HHG at 100 kHz with neon (a) and helium (b) gas that are both supplied by a 150 μm orifice gas nozzle. The experimental conditions were optimized for a maximum harmonic signal at 120 eV in neon (figure 10(a)) yielding up to 3.1 · 10⁹ photons s⁻¹ in a 1% bandwidth at 120 eV [191]. This value, which was obtained by accounting for the detection efficiency as described in section 5.1, is comparable or even higher than other realizations of high photon energy HHG (section 5.6) [192–194]. A maximum cutoff of ~180 eV of the harmonics can be achieved when lowering the gas pressure, which most likely minimizes phase-mismatch in that spectral region. An even further extension of the cutoff can be achieved when switching to helium gas and using the maximum available laser intensity. This allows us to extend the HHG signal to more than 300 eV (blue curve in figure 10(b)), which is an important spectral region for biological applications. The reason for the ladder is that carbon has an absorption edge at 283 eV, while oxygen is still transparent up to its absorption edge at 530 eV. Therefore, natural contrast between water and biological cells that contain carbon can be achieved. The insertion of a parylene filter clearly shows the carbon K-edge implying that the water window was achieved with this high repetition rate laser. The achieved photon flux above 283 eV is estimated to be 10⁷ photons s⁻¹, which might be improved in future, but already compares well with reported values [83].

5.3. Using μJ pulses for 10 MHz high harmonic sources

The results presented in sections 5.1 and 5.2 were obtained by a combination of fiber lasers and nonlinear compression in gas-filled capillaries, which allows efficient HHG at up to 0.6 MHz repetition rate and an order of magnitude improved photon flux. However, a significant increase in repetition rate is required, e.g. to address applications in photoemission spectroscopy and microscopy or frequency comb metrology in the XUV. This operation regime has been addressed with a
Yb:YAG Innoslab amplifier in combination with nonlinear compression in a standard solid core fiber, allowing us to generate HHG at repetition rates as high as 20 MHz, which is still the highest value achieved for single-pass harmonic generation [137]. Figure 11 shows the harmonic spectrum that was obtained in the experiment of Vernaleken et al [137].

One issue that arises with the use of solid core fibers for NC is the restriction in pulse energy to the sub-μJ level, which, in consequence, requires extremely tight focusing and may approach the limits of the phase-matching, as discussed in section 2. The aforementioned experiment used a 4.6 μm focus to obtain $4 \cdot 10^{13}$ W cm$^{-2}$ and the reported conversion efficiency was 5 $\cdot$ 10$^{-11}$ (1 nW at H15, 18 eV).

In recent years, a novel type of waveguide, a so-called Kagome photonic crystal fiber, has attracted significant interest, since it offers some unique properties. The most important one is that it essentially behaves like a small-core capillary allowing a variety of nonlinear effects to be addressed [195–197]. Moreover, despite its typically small core size of <60 μm the propagation losses are orders of magnitude lower as compared to conventional capillaries [198]. In combination with very broadband transmission bands, which sets them apart from photonic bandgap fibers, these fibers are ideally suited for nonlinear compression of multi-μJ pulses. Additionally, very promising progress has been made to use them for nonlinear compression of 100 W class laser systems [176, 199]. Consequently, the use of Kagome fibers is eminently suitable for fiber lasers at high repetition rate. In this particular experiment a $\sim$9 μJ, 250 fs fiber laser operating at 10.7 MHz (90 W of average power) has been coupled into a Kagome fiber with a core diameter of 57 μm (39 μm mode field diameter). The Kagome PCF is then filled with krypton gas at 7 bar of pressure to achieve the required spectral broadening for subsequent pulse compression with chirped mirrors. Due to the above-mentioned low propagation losses of the Kagome fiber the transmission of the nonlinear compression setup is $\sim$80% resulting in 7 μJ, 31 fs pulses with 76 W of average power, which are available for HHG. The HHG setup (figure 8) is again similar to that described in sections 5.1 and 5.2: a lens focuses the incoming infrared laser beam to a focal spot diameter of 22 μm (1/e$^2$ intensity) to achieve intensities of $\sim$7 $\cdot$ 10$^{13}$ W cm$^{-2}$, the xenon gas target is supplied by 65 μm orifice gas nozzle (continuous gas flow) with $\sim$6 bar of backing pressure, the harmonics are separated from the fundamental by one or several aluminum filters and the generated XUV radiation is analyzed with a flat-field grating spectrometer.

The results of HHG in xenon with this low energy laser are shown in figure 12, and more detail regarding this experiment can be found in [176]. The optimization of the experimental conditions is first done at a lower repetition rate of 234 kHz (figure 12(a)). Gaussian-like spatial profiles for the harmonics are obtained (figure 12(a)) with a higher divergence as compared to figure 9, which is due to the tighter focusing. The average power of the harmonics is obtained by accounting for the efficiencies of the detection system (aluminum filters, spectrometer, CCD) as described in sections 5.1 and 5.2. Even at this low repetition rate of 234 kHz, where the nonlinear compression stage delivered $<2$ W of average power, the 23rd harmonic contains more than $3 \cdot 10^{11}$ photons s$^{-1}$ and 1.5 μW of average power (inset in figure 12). This shows that phase-matching can be achieved.
even in tight focusing geometries as outlined in section 2. Subsequently, the repetition rate is gradually increased from 234 kHz to 4 MHz and then to 10.7 MHz, leading to an overall increase of the detected signal by a factor of 32. Therefore, the obtained average power of H23 at 10.7 MHz is as high as 51 μW (10^{13} photons s^{-1}) while all harmonics between H17 and H25 have at least 25 μW (>5 · 10^{12} photons s^{-1}) of average power. Therefore, the use of fiber lasers and Kagome fibers for nonlinear compression facilitates the unique combination of high photon flux and repetition rate HHG and, at the moment, constitutes the most compact way to realize this. Moreover, gas-filled waveguides for nonlinear compression offers the advantage of simply adapting the nonlinear compression to the pulse parameters of the laser system by changing the gas type or gas pressure, which also enables generating HHG at various parameters (e.g. repetition rate or photon energy). This is exemplarily illustrated in figure 13, where harmonics have been generated with a higher pulse energy of 14 μJ in argon gas (figure 13(a)), or with even lower pulse energy of 2.2 μJ in xenon gas. Although, the generation conditions are not particularly optimized in this case, it shows the great flexibility offered by fiber lasers in combination with nonlinear compression. In particular, the use of only 2.2 μJ could pave the way for a further increase in repetition towards the 100 MHz level, which might also allow the generation of frequency combs in the XUV via single-pass HHG.

5.4. IAPs at MHz repetition rates
A completely different operation regime is required when so-called IAPs have to be generated. In this case the requirements on the driving laser system are even more demanding, i.e. sub-2 cycle pulses with stable carrier envelope phase when using amplitude gating. If this has to be combined with high repetition rates, it calls for unprecedented performance levels of ultrafast laser systems.

OPCPAs, as described in section 4.1, offer the advantage of large enough bandwidth to obtain few-cycle pulses and carrier envelope phase stability. Moreover, the use of fiber lasers as pump lasers allows for high average power few-cycle pulses. In this particular experiment a standard FCPA was frequency doubled to pump a two-stage OPCPA system (figure 5) that delivered 14 μJ, 2.1 cycles pulses with repetition rates between 150 kHz and 600 kHz (2.1 W–8.4 W average power) [158]. These pulses were focused with an off-axis parabolic mirror to a

Figure 13. High harmonic generation with 14 μJ pulses in argon (a) and with 2.2 μJ pulses in xenon (b).

Figure 14. High harmonic generation and IAP generation at 150 kHz with a few-cycle CEP-stable OPCPA system. The bandpass filtered cutoff harmonics are shown with respect to the carrier envelope phase (a), while lineouts show positions of modulated (b) and continuous (c) HHG spectra. (Reproduced from [200].)
focal spot diameter of 32 μm (1/e² intensity) and intensities of up to 4 · 10¹⁴ W cm⁻². The generated harmonics were separated from the driving laser by a 200 nm thick aluminum foil and additionally filtered by another 200 nm thick zirconium filter. This filter combination creates a spectral bandpass filter between 50 eV and 73 eV (figure 14(a)), which was used to select the cutoff harmonics and therefore the IAP [32, 33]. Figure 14 shows the harmonic spectra behind the filters with respect to the carrier envelope phase. The variation of the latter changes the electrical field under the pulse envelope from a sine-shaped field that corresponds to two XUV emission events and a modulated harmonics spectrum (figure 14(b)) to a cosine-shaped field that creates only one XUV emission event and a continuous harmonics spectrum (figure 14(c)). The spectral changes with respect to the carrier envelope phase were simulated under experimental conditions and excellent agreement was found as described in detail in [200]. A numerical simulation of the results predicts that an IAP with a duration of 338 as was generated [200]. Due to the use of fiber lasers this experiment can be performed at repetition rates of up to 600 kHz, which is two orders of magnitude higher than previous IAP realizations [200]. Such a high repetition rate source opens up unique possibilities for applications such as attosecond photoelectron spectroscopy and microscopy (atto-PEEM) and many others [201].

5.5. Tunable harmonics and structural resonances in argon

As mentioned in section 4.1 OPCPA systems offer the possibility of tuning the central wavelength. This can, for example, be achieved by changing the delay between the pump and the signal (figure 5) in the OPCPA system that was used for IAP generation (section 5.4). Due to the temporally stretched (chirped) signal, the change in delay will cause different spectral components to be amplified, which will change the center of mass of the spectrum to approximately between 780 nm and 860 nm. In turn, this will lead to a change of the photon energy of the generated harmonics in argon (a) and krypton (b) gas as shown in figure 15.

In this experiment the delay was changed by applying a sinusoidal driving signal to a piezo-driven mirror with a period of 2/3 Hz. As shown in the lower traces in figure 15, this shifts the harmonic positions accordingly. Interestingly, the achieved shift is large enough to cover the complete spectral range shown here (upper panel in figure 15). Although not depicted here, the same OPCPA system was also used to generate harmonics with photon energies up to 200 eV [193]. Therefore, this system allows arbitrary photon energies up to the latter value to be addressed. The wavelength scans shown in figure 15 show additional interesting features. For example, the scan in argon (figure 15(a)) shows a very pronounced narrowband peak once the harmonic signal is tuned to 26.6 eV. As it turns out, this peak, and the other peaks located between 28 eV and 30 eV, correspond to resonances in the photoabsorption spectrum (window-type Fano resonances) as discussed in [202]. The harmonics are generated in absorption limited conditions and the window-type resonances lead to reduced absorption and, therefore, an enhanced macroscopic harmonic yield within a narrow spectral region [202]. Due to the small bandwidth, the harmonic line at 26.6 eV also has a relative bandwidth of only ΔE/E = 3 × 10⁻³ (figure 15(c)). It is interesting to note that this resonant enhancement is a macroscopic effect, while the observation of enhanced harmonic emission in plasma HHG is attributed to an increased single atom response [203]. Due to the potential of generating narrowband harmonics with increased efficiencies operating near the resonances, a
Figure 16. State-of-the-art in high photon flux HHG. The graph shows the highest reported literature values of single harmonics, and all of these numbers refer to the point of generation. The squares show values obtained by using Ti:sapphire lasers, circles stand for results of fsECs and triangles for fiber lasers. The results have been taken from the following publications: ref.1 [207], ref.2 [24], ref.3 [208], ref.4 [194], ref.5 [92], ref.6 [111], ref.7 [190], ref.8 [191], ref.9 [176], ref.10 [205], ref.11 [206], ref.12 [209].

powerful source for spectroscopic or imaging applications could be achieved [202, 204].

5.6. State-of-the-art high photon flux sources

The progress in fiber laser driven HHG has been outlined in the last few sections. Obviously it is important to put this into perspective with other HHG experiments that have been performed with various approaches and laser systems. Figure 16 shows the current state-of-the-art of average power levels in HHG, whereby it was attempted to pick the record values for each of the most commonly used laser systems, i.e. Ti:sapphire lasers, fsECs or fiber lasers. For a more detailed overview of progress in the respective fields we refer to other publications and references therein [76, 104, 105, 121].

As mentioned earlier Ti:sapphire based laser systems are the workhorses in the fields of high harmonics and attosecond science and, consequently, much work has been devoted to achieve the highest possible conversion efficiencies to increase the photons s⁻¹, i.e. the average power, that can be obtained in a single harmonic. This has resulted in several microwatts per harmonic at repetition rates between 10 Hz and 1 kHz, as shown in figure 16 (ref.1–ref.4). Most of this work was performed around the year 2000 and for a long time little progress was made in terms of increasing the photon flux. However, in recent years great effort has been put into exploiting the stronger single atom response at shorter wavelengths by working with the second or third harmonic of Ti:sapphire lasers, which has already yielded more than 100 µW at 22.3 eV (ref.10 in figure 16) [205] or 1.5 µW at 97 eV (ref.11 in figure 16) [206]. According to the cutoff law (equation (1)) shorter wavelengths are supposed to significantly reduce the achievable photon energy, but it appears that this can be overcome and even water window harmonics can be generated by using appropriate generation conditions [206].

After the first demonstration in 2005 the use of enhancement cavities has rapidly led to the highest photon flux values for wavelengths longer than 50 nm (<25 eV) with up to 200 µW (intra-cavity) in a single harmonic at 12.7 eV (97 nm, ref.5 in figure 16) [92] or 0.5 mW out-coupled power at 149 nm (not shown in figure 16) [102]. Moreover, they have successfully been pushed into the 100 eV regime recently (ref.7 in figure 16) [111].

On the other hand, the photon flux values obtained by fiber laser driven HHG (sections 5.1, 5.2 and 5.3) can be considered the highest of all laser systems for photon energies between 30–150 eV, which makes them highly attractive for a multitude of applications (see section 6). It should also be noted that there is still great potential for further increasing these values by orders of magnitude in the next few years, as outlined in section 7.

6. Potential applications of high repetition rate sources

The availability of the unique combination of high repetition rate and photon flux XUV sources opens up manifold opportunities for applications. In the following some first experimental results of these new sources are exemplarily shown and introduced.

In general, it can be distinguished between photon hungry applications that essentially need as much flux as possible and others that are mostly benefiting from an increase in
repetition rate. The former ones are for example imaging and microscopy techniques while the latter ones are photoelectron spectroscopy and coincidence detection.

Additionally, all presented experiments may be combined with pump-probe techniques which allow to study matter on femto- to attosecond time scales. This is a particular advantage of HHG sources compared to synchrotrons and FELs. While femto-sliced synchrotrons are limited to ~100 fs pulse duration and suffer from limited photon flux [210], pump-probe experiments at FELs require pump pulse from a synchronized excitation laser and probe pulses delivered by the FEL. Due to the long optical path lengths the resulting jitter between both pulses is currently of the order of some-ten femtoseconds [211], which limits time resolution. Table-top HHG offer inherent synchronization of the driving laser and the high order harmonics overcoming above-mentioned issues.

6.1. Coherent diffractive imaging with a fiber laser driven HHG source

Naturally, the process of HHG addresses the XUV to soft x-ray spectral regions, i.e. very short wavelengths can be generated. Microscopy techniques are widespread for investigating small scale structures, but the size of the resolvable features is more or less limited by the wavelength of the illumination. Therefore, shorter wavelengths, in principle, allow us to see smaller features. However, the fabrication precision of imaging optics for short wavelength sources, e.g. zone plates, limits the achievable resolution, giving the need for alternatives. In that regard, the technique of lensless imaging or coherent diffractive imaging (CDI) has been very promising, since it eliminates the need for optics and only relies on capturing a diffraction pattern [73, 212]. Therefore, it has the potential to achieve better resolution than x-ray microscopes based on zone plates. The investigation of small samples is particularly interesting for nanotechnology and nanoelectronics where inspection techniques are required. Furthermore, the large penetration depth of short wavelength radiation, as compared to electrons, for example, enables three-dimensional images to be obtained, as recently demonstrated [213]. CDI is also expected to be of importance for the investigation of biological samples, e.g. by exploiting natural contrast in the water window, and has recently been used to classify cancer cells [214].

High harmonics are particularly suitable, since they are coherent and can be realized with table-top setups. Consequently, CDI has been realized with various harmonic sources pushing the resolution to 22 nm with 13 nm illuminating wavelength [67, 215]. However, the typical integration times are still several tens of seconds to minutes, preventing real-world application to time-resolved measurements. The use of a fiber laser driven HHG source has recently significantly reduced the integration times to 1 s, while still maintaining sub-70 nm resolution due to the high photon flux available [72]. Moreover, in that case very narrowband harmonics were used, which enabled sub-wavelength resolution of 0.8 λ (26 nm) with an illuminating wavelength of 33.2 nm (figure 17). This first demonstration experiment shows great potential, and it can be expected that with further increase of the photon flux and the availability of shorter wavelengths, sub-10 nm resolution with real-time image capturing will be possible. In addition, other imaging and holography techniques could be used to further extend the application range of the high photon flux sources [68, 213].

6.2. Photoemission spectroscopy

Another very interesting application field can be found in solid state physics and material science for investigating the electronic structure of matter, where techniques such as (time-resolved) photoelectron spectroscopy or microscopy are routinely addressed by XUV light sources, e.g. synchrotrons, free electron lasers or HHG sources. The latter are now becoming more widespread in the field, because they allow for excellent timing accuracy, ultrashort pulse durations to investigate ultrafast processes, they are realized on a table-top and, therefore, can be more easily accessed than large-scale facilities [58]. These ultrashort XUV to soft x-ray pulses are required to investigate ‘fundamental interactions between charge, lattice, orbital, and spin dynamics in real time’ [64]. As such they will help the understanding of correlated-electron materials, switching processes in magnetic materials and chemical reactions on surfaces [64].

Another advantage, in particular, of high repetition rate HHG sources is that they reduce or eliminate space-charge effects, as pointed out in various publications [58, 77, 90, 163]. Figure 18 shows such a measurement with a 1 MHz HHG source, which shows that space-charge effects can be effectively eliminated [39]. Therefore, the sources presented here (sections 5.1–5.3) can advance the field and potentially allow multi-dimensional studies that were not feasible up to now.

6.3. Coincidence detection

Experiments that rely on coincidence detection are a prime example of applications that would benefit from high repetition rate HHG sources [79, 91, 93]. A generic setup of such an experiment is shown in figure 19. The probe beam is obtained via HHG of the laser while the pump beam is either a small part of the infrared laser or low-order harmonics of it. These two beams overlap on a gaseous target (atoms or molecules) and two detectors for different ionization fragments and their properties are used in coincidence detection mode. This mode requires experimental conditions to be chosen such that statistically less than one event per laser shot is detected to distinguish, e.g., different ionization channels. The variation of the delay between pump and probe beams allows dynamical processes to be captured. Here, the inherent timing stabilization of the beams due to the origination from the same laser is a key advantage of table-top HHG sources as compared to, e.g., free electron lasers.

Consequently, different experiments have already been performed, e.g. to investigate molecular dynamics [49, 56, 216–218], or to probe double ionization on attosecond time scales.
However, such measurements typically require several million laser shots to gather a significant statistic, which leads to long measurement times, e.g., 100 h in [216]. Here, the fiber laser driven high repetition rate HHG sources might initiate and enable a new class of experiments by significantly shortening the measurement time and increase signal-to-noise ratios, e.g., in the study of molecular dynamics. A recent experiment with a 50–100 kHz source at 68.6 eV has emphasized this unique potential by demonstrating coincidence measurements on inner-shell ionized gas-phase iodomethane (CH$_3$I) molecules [209].

6.4. Near edge x-ray absorption fine structure (NEXAFS) spectroscopy

Quasi-continuous harmonic spectra as generated by few-cycle pulses can be applied to NEXAFS spectroscopy [86] that can potentially be combined with pump-probe techniques to study ultrafast dynamics [220]. For example, the two spectra shown in figure 10(b) allow information on the used parylene filter and its chemical structure at the carbon K-edge to be obtained, as shown in figure 20. Ultimately, further advances might facilitate the mapping of chemical elements and states with nanometer resolution when combining it with, e.g., the CDI techniques outlined above [221].

7. Summary and future direction

In this review the most important developments in single-pass high repetition rate HHG sources over the last few years have been recapitulated. It has been emphasized that one crucial aspect of these novel sources has been the possibility of achieving phase-matching, even in tight focusing geometries. The use of a high density gas target leads to $\Delta k = 0$ and also compensates for the smaller interaction volume. Theoretical considerations have shown that conversion efficiencies are independent of the focusing geometry if the same intensity, pulse duration and central wavelength is used [135, 136]. These findings have been verified experimentally by achieving $8 \cdot 10^{-6}$ conversion efficiency for a 30 $\mu$m focal spot size [136]. In combination with important advances in fiber laser technology, OPCPAs and nonlinear compression, the required high average power ultrashort pulse or even few-cycle lasers have become available. This has finally led to the most

Figure 17. Coherent diffractive imaging with a fiber laser driven HHG source. The object (a) is illuminated with a single harmonic at 33.2 nm delivered by high repetition rate fiber laser. The captured diffraction pattern (b) is used to reconstruct the object as shown in (c). A subwavelength resolution of 0.8 $\lambda$ is achieved, as suggested by the phase retrieval transfer function in (d). The scale bar in (a) and (c) corresponds to 1 $\mu$m. More details can be found in [72]. (Reproduced from [72].)
powerful coherent XUV and soft x-ray sources, which have been realized with table-top setups. For example, it is possible to generate 143 μW in a single harmonic at 30 eV (3 × 10^{13} photons s^{-1}) at 0.6 MHz repetition rate when using a >80 W, sub-30 fs nonlinearly compressed fiber laser [190]. The achievable photon energy range could be extended well into the water window with a sub-8 fs, 35 W laser operated at 100 kHz [191]. The latter spectral region will be of extreme importance for biological and spectroscopic applications. The source delivers 3 × 10^9 photons s^{-1} in a 1% bandwidth at 120 eV, which is among the highest photon fluxes ever achieved in this spectral region. The use of a Kagome PCF as waveguide for nonlinear compression opens a route to address more advanced parameters, i.e. a significantly increased repetition rate. Furthermore, this paves the way for compact and portable XUV sources for many applications. It has been possible to efficiently generate high harmonics with up to 10.7 MHz repetition rate, where the strongest harmonic has 51 μW (27.7 eV) of average power (10^{13} photons s^{-1}) [176]. On the other hand, the fiber laser pumped OPCPA constitutes a unique source of few-cycle pulses with controlled carrier envelope phase at a high repetition rate. Consequently, it has enabled the generation of isolated attosecond pulses at up to 0.6 MHz repetition rate, an increase of two orders of magnitude as compared to previous realizations [200]. In addition, the central wavelength of the OPCPA systems can be easily shifted allowing the generation of harmonics at arbitrary photon energies (up to the cutoff). During these experiments it was also discovered that window resonances in the absorption cross sections of the generation gas have a strong influence on the macroscopic buildup [202], and might be utilized for future narrowband HHG [204]. Therefore, it can be concluded that fiber laser driven high repetition rate coherent XUV sources are now providing an extremely attractive set of parameters for the fields of HHG and attosecond science [176, 185, 190, 191, 200, 209, 222].

As a consequence, they are now finding their way into various application fields. First experiments on coherent diffractive imaging have already yielded a significant advance by demonstrating sub-wavelength and real-time imaging thanks to the high photon flux [72]. Moreover, the sources are now also being used for photoemission spectroscopy [59, 223–225], which constitutes one of the fields where the repetition rate is particularly beneficial for avoiding space-charging. It can be expected that further advances in source parameters will allow multi-dimensional studies in surface science or advances in (attosecond) microscopy techniques. Another application that benefits from the high repetition rate, is a coincidence detection experiment, where important progress has been recently made with fiber laser driven HHG sources [209]. In addition, there are many other potential and impactful fields, e.g. absorption spectroscopy [226–228], where the sources discuss here could be used. More generally, they can foster any existing application in terms of integration (measurement) time and signal-to-noise, or even enable novel experiments.

For example laser spectroscopy of high energy transitions in highly charged ions, which constitute a dominant fraction...
of the visible matter in stars, supernovae, stellar clouds and hot (e.g. fusion) plasmas, puts enormous demands on the laser source [204]. So far investigations on highly-charged ions at storage rings have been mainly limited to hyperfine-transitions due to the lack of suitable XUV laser sources. This source has to be synchronized with the highly charged ion circulation in a storage ring at MHz revolution frequency and needs to provide at least $10^{12}$ photons per second within a narrow relative energy bandwidth of $\Delta E/E < 10^{-3}$ in the XUV [204], a combination of requirements that can most likely be fulfilled by the next generation of fiber laser based XUV sources. Besides fluorescence spectroscopy, XUV-pump–XUV-probe experiments seem feasible to determine the ultrashort lifetimes of the corresponding excited states.

Despite the rapid progress over the last few years there are certainly many exciting developments that can be expected in the next few years, but there will also be challenges along the way. First of all, further advances in coherently combined femtosecond FCPA systems will enable reliable kilowatt (and beyond) lasers in the next few years [147]. Recent investigations on the scalability of the nonlinear compression setup suggest the same favorable scaling behavior for that important building block [180], so that kilowatt drivers, potentially combined with CEP stability, will be made available for HHG. However, one important challenge will be the separation of the high average power lasers from the generated harmonics. Obviously, the commonly used metal filters will simply burn under high average power illumination, calling for new concepts to tackle this challenge. The issue is somewhat comparable to out-coupling in enhancement cavities. For that reason, it is not surprising that some of the concepts, e.g. gracing incidence plates [109], have already been used for HHG experiments with high power lasers. The difficulty with these elements is that they have to be used at gracing incidence to reflect as much of the harmonics as possible, which makes the design of high power capable broadband anti-reflection coatings difficult. In addition, the reflectivity of common transparent materials used as a top-layer drops towards higher photon energies (>150 eV). Other potential ways to solve this issue might include other filter ‘geometries’, such as the recently proposed multi-channel plate [229], or alternative interaction geometries, e.g. non-collinear HHG [112, 230–232] or the use of annular beam profiles [233] or Bessel–Gaussian beams [234, 235].

So far, most of the high average power laser systems are operated at a wavelength of 1 \( \mu \)m, yielding an unprecedented photon flux level in HHG. However, it is a known fact that the single atom response is much stronger at shorter wavelengths [236–238], and that phase-matching becomes more favorable [205, 206]. Therefore, it can be expected that frequency doubling the output of the above-presented nonlinear compression stages will lead to a significant increase in HHG conversion efficiency, holding the promise of milliwatt level harmonics in the near future. On the other hand, cutoff scaling, i.e. the increase of achievable photon energy, has been a fascinating topic ever since the first demonstration of HHG and still attracts a lot of attention. Using longer wavelength driving lasers in combination with improved phase-matching conditions has led to >keV coherent x-rays with impressive bandwidth in the last few years by using a 3.9 \( \mu \)m OCPA system [81, 239]. In that regard, current developments in 2 \( \mu \)m fiber laser technology [240–243], which have already seen power levels of >100 W with femtosecond pulses, could play a pivotal role in significantly increasing the available photon flux in the water window and beyond.

In conclusion, high repetition rate and photon flux HHG sources have come of age, and are now enabling sophisticated applications. Future developments are expected to further boost their average power into the milliwatt regime making them as attractive as synchrotrons or free electron lasers by offering complementary research opportunities with powerful and versatile ultrashort pulse sources that are eminently suitable for investigating ultrafast pump-probe processes due to low timing jitter. In addition, it is expected that their operation range will be extended into the water window and even the keV spectral region with unparalleled flux levels, further bolstering the enormous application potential.

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