Spectral characterization of fully phase-matched high harmonics generated in a hollow waveguide for free-electron laser seeding

F Ardana-Lamas\textsuperscript{1,2,4}, G Lambert\textsuperscript{3}, A Trisorio\textsuperscript{1}, B Vodungbo\textsuperscript{3}, V Malka\textsuperscript{3}, P Zeitoun\textsuperscript{3} and C P Hauri\textsuperscript{1,2,4}

\textsuperscript{1} Paul Scherrer Institute, SwissFEL, CH-5232 Villigen PSI, Switzerland
\textsuperscript{2} École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland
\textsuperscript{3} Laboratoire d’Optique Appliquée, ENSTA-CNRS-Polytechnique, F-91761 Palaiseau, France
E-mail: fernando.ardana@psi.ch and christoph.hauri@psi.ch

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\textbf{Abstract.} We present a bright and coherent soft x-ray source based on high harmonic generation delivering up to $10^{10}$ photons per second centered at 120 eV within an 80 eV bandwidth. The source profits from fully phase-matched harmonic generation in an unmodulated hollow waveguide. Under these conditions, the resulting high harmonic spectrum is shown to be flat-top up to the cutoff photon energy and in line with the theoretical single-atom response. The source is characterized in view of seeding a free-electron laser and is shown to overcome the free-electron laser noise floor for wavelengths as short as 8.9 nm. This opens the perspective toward direct high harmonic seeding of a free-electron laser at soft x-ray wavelengths.
1. Introduction

High-order harmonic generation (HHG) produced by laser-field-induced ionization in a gas offers fully coherent radiation in the XUV and soft x-ray spectral region. An HHG table-top source finds use in numerous applications such as molecular orbital imaging, ultrafast electron dynamics in atoms, shell-selective magnetic probing and others [1–3]. Moreover, it delivers pulse durations down to a few tens of attoseconds. Those pulses are excellently suited for time-resolved measurements, but the low photon flux restricts their use for photon yield demanding applications such as seeding of a free-electron laser (FEL).

FELs are the new generation of large-scale facilities that deliver ultrashort and bright pulses down to angstrom wavelength, at a peak brilliance of up to $\approx 10^{30}$ photons s$^{-1}$ mm$^{-2}$ mrad$^{-2}$ within 0.1% bandwidth. In contrast to HHG sources, FELs provide orders of magnitude higher flux at femtosecond pulse durations but suffer from a partial incoherent beam. The lasing process based on self-amplified spontaneous emission (SASE) offers excellent transverse coherence. However, the longitudinal coherence property is affected by the incoherent merge of numerous longitudinal modes, each of them coherent on its own.

The spectral and temporal profiles of SASE FELs present several spikes that differ from shot to shot. To improve the performances of FELs, different seeding approaches have been proposed, such as high-gain harmonic generation [4], echo-enabled harmonic generation [5], self-seeding [6] and also HHG seeding [7].

HHG seeding is one of the most straightforward ways of transferring the coherence properties of an external source to FEL radiation by mode locking the individual longitudinal FEL modes. Indeed, seeded FELs with HHG have been recently demonstrated at XUV wavelengths [7–9]. Scaling the seeding approach toward significantly shorter wavelengths is a crucial user request since seeding offers, in addition to the full coherent FEL pulse, an inherent synchronization between the FEL and the external laser used for experiments.

This is favorable for pump–probe investigations since the typical time jitter issues associated with SASE FELs (typically a few hundreds of femtoseconds) are avoided. However, injection at shorter wavelengths (i.e. < 50 nm) poses significantly higher demands on the HHG peak power since the FEL SASE shot noise scales with $\lambda^{-1}$. This requirement is in conflict with HHG conversion efficiencies that decrease for higher photon energy.

One principal limitation to upscaling HHG toward higher peak power at shorter wavelengths originates from the phase mismatch between the high harmonics (HH) and the fundamental wavelength. Different approaches have been explored in the past to overcome
velocity mismatch and to achieve higher conversion efficiencies at shorter wavelengths. One possible approach is quasi-phase matching where harmonic generation is suppressed in the region where harmonics interfere destructively [10]. Counter-propagating beams or modulated waveguides have also been used to extend harmonic generation toward shorter wavelengths by modulating, respectively, laser intensity and gas density [11, 12]. This approach complicates the generation process and does not provide phase matching across a large spectral bandwidth. Directly phase matching the harmonic emission by compensating the different dispersion contributions (plasma, waveguide, etc) is considered to be the most efficient approach for broadband harmonic generation.

The choice of the driving wavelength defines up to which photon energy full phase matching can be achieved. The use of longer driving wavelength entails not only higher cutoff energy but also lower efficiencies. For example, using a 2 µm driving laser, a cutoff energy of 450 eV has been achieved but with a photon flux of 10^6 photons per shot in a bandwidth of ~250 eV [13]. In this paper, we present significantly higher photon flux per shot up to 160 eV by using a conventional Ti:sapphire laser (0.8 µm) at a repetition rate compatible with FELs and at narrower bandwidth. The latter is favorable for seeding the FEL, which has typically a rather narrowband emission (≤0.5%). In the past, efficient HHG driven by Ti:sapphire technology has been reported by exploring loose focusing conditions [7] with low-repetition-rate, high-energy lasers (up to 100 mJ, ≤10 Hz), HHG with few-cycle pulses [14] as well as using a two-color driving laser field [15]. For operating a seeded FEL, the most robust laser technology at hand is required which, we believe, is presently the Ti:sapphire laser technology commercially available at a repetition rate compatible to FELs (0.1–10 kHz). In view of FEL seeding, we explore in this paper fully phase-matched HH generation in an unmodulated capillary hollow waveguide driven by a technologically mature Ti:sapphire laser system.

We demonstrate efficient harmonic emission with up to 10^{10} photons s^{-1} and present a detailed study of phase-matching conditions for different laser and gas parameters. At fully phase-matched conditions, a flat-top spectrum is reported with a spectral bandwidth of 80 eV at a central photon energy of 120 eV. The high peak power of this source opens up new opportunities for seeding an FEL at photon energies in the soft x-ray region down to 10 nm (124 eV), thus significantly shorter than state of the art. Realistic parameters for HHG-driven lasers for future seeding experiments at such short wavelengths are estimated by interrogating FEL simulations.

2. Experimental setup

In our experiment, performed at the Laboratoire d’Optique Appliquée, the 1 kHz Ti:sapphire laser delivers 40 fs full-width at half-maximum, 6 mJ pulses centered at 805 nm. The laser beam is focused with a lens (f = 1.5 m) into a capillary that is continuously flooded with gas. The capillary is made up of two identical sapphire blocks. Each of the glass blocks carries a 33 mm long laser-drilled semi-cylindrical channel with 100 µm radius, which forms a waveguide after assembling the two parts. The gas inlet is provided with two additional channels that are placed close to both ends of the capillary and perpendicular to the laser guiding channel. The sketch of the experimental setup is shown in figure 1(a).

Different gases (He, Ne and Ar) are injected into the laser waveguide with pressures up to 200 mbar. Behind the capillary a set of metallic filters is used to separate the HH from the infrared radiation. Additionally, a pair of flat and concave mirrors coated with ZrO₂ are used to filter out the remaining infrared radiation and to focus the harmonic beam on the CCD.
camera after passing through a 1000 lines mm\(^{-1}\) transmission gold grating. Alternatively, the spectrometer (dichroic mirror, focusing mirror and grating) could be removed to record the HH footprint. The spectrometer has been calibrated in wavelength using the absorption edge of aluminum and the zero order of the grating; see figure 1(b).

For FEL seeding, bandpass filters are not needed since the undulator itself acts as a wavelength-selective filter that is resonant only at the FEL emitting wavelength. Taking this into account, it is important to retrieve the real HHG spectrum and power distribution at the target position. As a consequence, the measurements are corrected with the transmission curves of the filters [16] and the quantum efficiency (QE) of the CCD camera.

In figure 2, corrected and uncorrected (raw) spectra are compared. The reflectivity of the ZrO\(_2\) mirror for an incident angle of 10\(^\circ\) is also plotted, which indicates that the HH spectra are significantly affected by the drop in mirror reflectivity for photon energies in the cutoff region.

### 3. Phase-matched harmonic generation

To efficiently generate high-order harmonics, phase matching between the fundamental laser and the generated radiation must be achieved, preferably along an elongated distance to enhance the HH yield. In principle, capillaries offer potentially more efficient HHG than a gas jet due to the high laser intensity confined in the waveguide and longer interaction with the gas. In capillaries, there are three main contributions of phase mismatch between fundamental wavelength and harmonic radiation [17]: waveguide dispersion, dispersion from neutral atoms and free-electron dispersion. The total phase mismatch is given by

\[
\Delta k = \frac{q u_{11}^2 \lambda_L}{2 \pi a^2} - q P \left[ \frac{2 \pi}{\lambda_L} (\Delta \delta) - \eta \left\{ \frac{2 \pi}{\lambda_L} (\Delta \delta) + N_{\text{atm}} r_e \lambda_L \right\} \right], \tag{1}
\]

where \(q\) is the harmonic number, \(P\) the gas pressure, \(a\) the radius of the capillary, \(u_{11}\) the mode factor, \(\Delta \delta\) the difference in refractive index between the driving laser and the harmonics, \(\lambda_L\) the wavelength of the laser, \(\eta\) the ionization fraction of the generating media, \(r_e\) the classical electron radius and \(N_{\text{atm}}\) the gas density.
The critical ionization fraction, $\eta_{cr}$, 

$$\eta_{cr} = \frac{2\pi \Delta \delta}{2\pi \Delta \delta + r_e N_{atm} \lambda_L^2},$$

is defined as the ionization fraction for which phase mismatch caused by the neutral gas is compensated by the corresponding contribution from the electron gas. In our capillary, it is the laser intensity that defines the fraction of ionization. If $\eta \leq \eta_{cr}$, full phase matching can be obtained according to equation (1) by adjusting the gas pressure. In this way, the (positive) mismatch of the capillary and free electrons can be compensated by the (negative) mismatch of the neutral atoms. For ionization fractions above $\eta_{cr}$, phase matching cannot be compensated anymore by adjusting the gas pressure and the HH yield significantly drops.

Figure 3(a) illustrates the maximum photon energy that can be achieved within full phase-matching conditions as a function of fundamental wavelength using He and Ne as generation media. For Ti:sapphire lasers emitting at 0.8 µm, the maximum photon energy is 185 eV (6.7 nm) in He and 115 eV (10.8 nm) in Ne. Harmonics above the maximum cutoff energies (indicated by the red and blue lines for He and Ne, respectively) are not fully phase matched and increasing the pressure will further decrease the harmonic yield. In the red region phase matching of HH can only be achieved in He, while in the blue region full phase matching can be achieved for He and Ne. For a given laser intensity defining the fraction of ionization and thus the maximum reachable HH cutoff energy, the gas pressure needs to be adapted adequately in order to achieve full phase matching.

Under our experimental conditions, a maximum laser intensity of $7 \times 10^{14}$ W cm$^{-2}$ is achieved. From figure 3(b) an ionization ratio of the order of 1% can be estimated for this laser intensity. The achievement of full phase matching by adapting the pressure is observed in our experiment. In figure 4, the HH spectrum generated in He is shifted toward shorter wavelengths for higher pressures. Furthermore, the spectral shape becomes flat-top for a pressure of 200 mbar. This is a signature that full phase matching is achieved for all harmonic
Figure 3. (a) Maximum HH cutoff energy in He and Ne for fully phase-matched HHG in a capillary, as a function of laser wavelength. (b) Ionization ratio versus laser intensity for He and Ne.

Figure 4. Experimental HH spectra recorded in He (a) and Ne (b) at different pressures. For Ne the scaling factor is given in the legend, and the highest photon yield is achieved at 50 mbar pressure. One pixel corresponds to 1% bandwidth at 160 eV and 0.5% at 80 eV.

order \( q \). In the fully phase-matched case the plateau starts to cut off at 140 eV, which represents the limit of the spectrometer optics. The absolute photon number per second is also shown in figure 4, under consideration of the transfer function of the system (figure 2). We would like to draw attention to the linear scale in figure 4. In our experiment, further increase of the pressure was not feasible due to limited capability of our vacuum pumping system.

For Ne, at the same laser intensity, opposite behavior is observed. By increasing the pressure, phase matching shifted the highest HH from 160 eV toward lower energies (100 eV). This observation is again in agreement with equation (1), since to produce HH photon energies \( \geq 115 \text{ eV} \) the ionization of Ne needs to be above the critical ionization fraction, according to figure 3(a), while He is still below the critical ionization fraction.

In the investigated spectral range, the transmission in He is higher than in Ne, which makes He more suitable for HHG; see figure 2.
Fully phase-matched ($\Delta k = 0$) HHG over such a broad spectral range and independent of the harmonic order is possible for a capillary, since this configuration allows for compensating the different phase terms with the pressure as a free parameter. In principle, the capillary could be even more elongated since the coherence length in this configuration could be significantly longer than the capillary length. For HHG in a gas jet or a gas cell, the positive dispersion term (from the waveguide) is lacking and furthermore the laser–gas interaction is significantly shorter. In addition, the Gouy phase shift across the focus limits the phase-matched generation of harmonics. In principle, phase matching could be redeemed to a certain extent by means of loose focusing geometries or filamentation.

An additional confirmation for full phase-matching HHG is shown in figure 5. When full phase matching is achieved, the experimental HH spectrum becomes similar to the spectrum calculated from the single atomic response corrected for the transmission properties of the experimental setup (Ti filter, QE correction). The theoretical spectrum is calculated according to the Lewenstein model [18] mimicking HHG in He at a laser intensity of $7 \times 10^{14} \text{ W cm}^{-2}$.

Those findings suggest the use of longer capillaries to further enhance the HH photon flux.

4. Free-electron laser seeding perspectives

Seeding an FEL with an HHG source provides peculiar advantages, such as improvement of spectral shape, coherence properties, time synchronization and reduction of the FEL gain length, which helps in achieving the saturation regime with less undulators. For successful seeding, the HH peak power needs to be above the FEL shot noise by typically two orders of magnitude [19].

Unfortunately, the shot noise of FEL increases with the resonant photon energy of the FEL, while HHG conversion efficiency dramatically decreases at shorter wavelength. This fact makes it challenging to seed FELs in the soft and hard x-ray region and makes the field of HHG

**Figure 5.** Comparison of the theoretical single-atom response (red) and the experimental spectrum (blue) in the fully phase-matched condition. Experimental conditions (Ti filter, QE of the camera) have been taken into account for both spectra.
seeding a very active research field [15, 20]. Nevertheless, the presented improvements in HH phase matching open up opportunities for direct seeding in the soft x-ray region.

Based on figure 4(a), the peak power spectrum of the HH was computed for different central photon energies assuming a 1% bandwidth and an HH pulse duration half the driving laser pulse. The calculations are shown in figure 6. This will establish a lower limit for the harmonic intensity. Losses produced by the two ZrO$_2$ mirrors (25% each) used for beam transport are included in the calculation.

To see whether the HH peak power is sufficient to efficiently seed an FEL, the FEL shot noise has been estimated according to [21]

$$P_{sn} = 6\sqrt{\frac{\pi}{\rho_3}} \frac{P_b}{N} \sqrt{\log \left( \frac{N}{\rho_{3D}} \right)}$$

with $P_b$ the electron beam peak current, $N_e = \frac{P_b}{\lambda r}$ the number of electrons per radiation wavelength and $\rho$ the FEL Pierce parameter. To estimate the shot noise, an electron beam with a normalized emittance of $\epsilon_n = 0.43$ mm mrad, average beta function $\beta = 10$ m, peak current $P_b = 2.7$ kA, a kinetic energy of $E_k = 2.1$ GeV and energy spread $\Delta E = 250$ keV is assumed. These values correspond to the SwissFEL soft x-ray beam line covering 1–7 nm. The undulator period is $\lambda_u = 40$ mm and the undulator parameter $K_0 = 1.2$–3.2.

In figure 7, FEL shot noise is compared with HH peak power. With the presented results, the HH power in the FEL bandwidth is twice above the shot noise across the fully phase-matched spectral region up to 140 eV (8.8 nm).

A similar seed-to-SASE power ratio has recently been shown to be sufficient for observing seeding effects at longer wavelengths (61 nm; 20 eV) [7]. The capillary approach presented here provides similar HH-to-FEL power ratio up to 140 eV.

In our experiments, laser energies up to 3 mJ are coupled into the capillary. Further upscaling of the HH peak power seems feasible by combining the capillary approach with cutting-edge Ti:sapphire laser technology. Modern lasers are capable of delivering up to 100 mJ per pulse at a repetition rate compatible with FELs (typically 100 Hz). Assuming linear scaling, the resulting HH peak power would be sufficient to seed FELs down to 10 nm.
For seeding at shorter wavelengths (\(<7.5 \text{ nm}; >165 \text{ eV}\)), the driving laser wavelength needs to be chosen in the mid infrared [22, 23] in order to shift the HH cutoff toward higher photon energies. For seeding the soft x-ray beam line at SwissFEL, for example, an estimated laser pulse energy of up to 100 mJ in the infrared is required to provide adequate seeding conditions. The development of such a laser is currently under investigation at the Paul Scherrer Institute in Switzerland.

5. Conclusion

In this paper, fully phase-matched HHG in a sapphire hollow waveguide has been presented using a conventional Ti:sapphire system emitting at 800 nm. The HH source delivers up to \(10^{10}\) photons s\(^{-1}\) in a bandwidth of 80 eV around a central photon energy of 120 eV.

Phase matching conditions of HH generation have been studied as a function of the pressure, and different behaviors of the phase matching for harmonic generation below and above the critical ionization ratio are observed, in excellent agreement with theoretical expectations.

The fully phase-matched HHG source delivers a flattop spectrum and close to kW peak power within a 1% bandwidth up to photon energies of 140 eV. In our experiments, the HH peak power surpasses the FEL shot noise by a factor of 2. At increased laser power, the methodology presented here offers the potential for seeding FELs at wavelengths significantly shorter than state of the art.

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