

# FULLY PHASE MATCHED HIGH HARMONIC GENERATION IN A HOLLOW WAVEGUIDE FOR FREE ELECTRON LASER SEEDING

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## Abstract

A bright high harmonic source is presented delivering up to  $10^{10}$  photons per second around a central photon energy of 120 eV. Fully phase matched harmonics are generated in an elongated capillary reaching a cut-off energy of 160 eV. The high HHG photon fluence opens new perspectives towards seeding free electron lasers at shorter wavelengths than the state of the art. Characterization of the phase matching conditions in the capillary is presented.

## INTRODUCTION

Bright table-top VUV and soft x-ray sources based on laser-field induced ionization in a rare gas found a prominent application in free electron laser facilities, which is seeding. Overlapping a fully coherent seed pulse generated by high-order harmonic generation (HHG) with the electron bunch in the undulator results in locking the FEL longitudinal modes and consequently in an improved longitudinal coherence of the FEL pulse. Proof of principle experiments have been performed at  $\approx 160$  nm [1] and at wavelengths around 50 nm [2,3].

Seeding an FEL with a HHG source is one of the most straightforward ways to transfer the coherence properties of an external source to the FEL radiation. In addition to an enhanced longitudinal coherence of the FEL pulse, seeding with an external laser offers a laser pulse, which is inherently synchronized to the FEL x-ray emission. This is most favorable for x-ray/nIR pump-probe investigations in the femtosecond range since the typical arrival time jitter associated to SASE FELs is significantly reduced. At modern SASE FELs, a typical timing jitter of about hundred femtoseconds or more is typically present at the experimental stations.

While seeding at VUV wavelength was successful, injection at wavelengths shorter than 50 nm has remained a challenge using state of the art HHG sources. At shorter wavelengths significantly higher HHG peak power is required since the FEL SASE shot noise scales with  $\lambda^{-1}$ . This requirement is in conflict with the HHG conversion efficiency since the latter decreases for higher photon energy.

In this paper we present recent results on enhanced

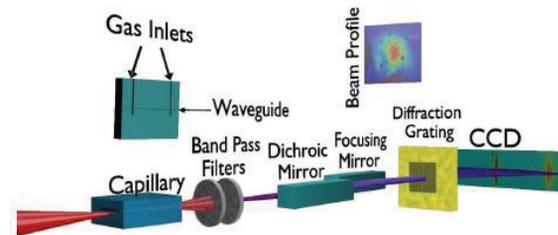


Figure 1: Experimental setup for HHG. (From ref [8])

HHG in an elongated capillary waveguide. One of the limitations to upscale HHG to higher photon fluence and peak power at shorter wavelengths is given by the phase mismatch between the fundamental driving laser wavelength and the harmonics. In the past, different schemes have been studied to overcome this detrimental velocity mismatch in order to enhance the conversion efficiency, such as quasi phase matching, counter propagating beams, modulated waveguides and HHG by a two-color driving laser [4-7]. Since these enhancing techniques complicate the generation process and does not allow phase matching across a large spectral bandwidth we investigated a more simple approach based on an unmodulated, elongated waveguide. We consider direct phase matching by compensating the different HHG dispersion contributions (plasma, neutral gas, waveguide etc.) to be the most efficient and most robust approach for broadband harmonic generation.

## EXPERIMENTAL SETUP

For the presented investigations recently published in [8] a 40 fs, 6 mJ Ti:sapphire laser system is used at 1 kHz repetition rate and at a central wavelength of 805 nm. HHG is launched by focusing the laser beam loosely ( $f=1.5$ m) into a capillary which is continuously flooded with a rare gas (Fig. 1). The waveguide is made of sapphire glass and carries a 33 mm long laser-drilled channel with an inner diameter of 200  $\mu$ m. Two gas inlets are arranged orthogonally to the laser waveguide, which is flooded with helium. After the capillary a set of thin metal filters and a pair of dichroic mirrors are used to separate the soft x-ray

radiation from the fundamental laser light. The home-built spectrometer located at the end of the beamline consists of a focusing mirror and a transmission gold grating with a groove density of 1000 lines/mm. Alternatively the spectrometer could be removed in order to record the HH intensity beam profile. For FEL seeding with HHG the HH selective filters are in principle not needed since the undulator itself acts as a wavelength selective filter which is resonant at the FEL emitting wavelength. Consequently the results presented here take into account the real HHG spectrum and power distribution at the target position and are corrected for the transmission curves of the metal filters and the quantum efficiency of the CCD camera.

## RESULTS

Under the experimental conditions described above a maximum laser intensity of  $7 \cdot 10^{14} \text{ W/cm}^2$  is achieved at the waveguide entrance port. A typical spectrum of fully phase-matched HH is presented in Fig. 2 for He. The spectral HH shape becomes flattop for a gas pressure of 200 mbar. The flattop-like spectrum is a signature for fully phase matched HHG for all harmonic order  $q$ . The harmonic plateau starts to cut off around 140 eV and is limited by the spectrometer optics.

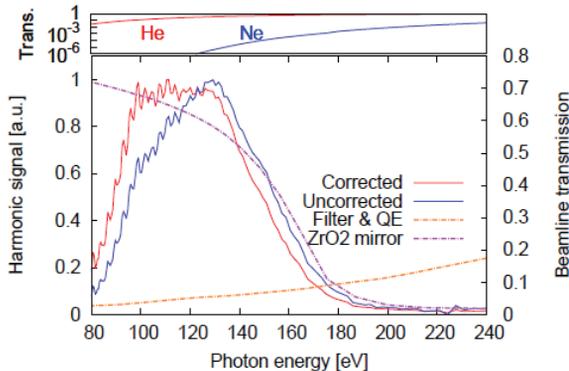


Figure 2: HHG in He in fully phase-matched conditions. The HH is flattop between 100 to 140 eV. The high energy cut-off is limited by the spectrometer optics. (from ref [8])

Fully phase-matched HHG across such a broad energy range and independent of the harmonic order is possible thanks to the use of a capillary waveguide. In the waveguide there are three main contributions of phase mismatch  $\Delta k$  between the fundamental and the HH beam, being the waveguide, the neutral gas and the free electrons. The mismatch is described by the following equation

$$\Delta k = \frac{qu_{11}^2\lambda_L}{2\pi a^2} - qP \left[ \frac{2\pi}{\lambda_L}(\Delta\delta) - \eta \left\{ \frac{2\pi}{\lambda_L}(\Delta\delta) + N_{atm}r_e\lambda_L \right\} \right] \quad (1)$$

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

Efficient HHG requires the ionization fraction of the laser-ionized gas to be below the critical value  $\eta_{cr}$ ,

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

which is defined as the ionization fraction for which phase mismatch caused by the neutral gas is compensated by the corresponding contribution from the free electrons. Fully phase-matched harmonics are generated if  $\eta \leq \eta_{crit}$ . In fact, it is the gas pressure which allows to match the (negative) mismatch of the neutral atoms with the (positive) mismatch of the capillary and free electrons and to achieve a  $\Delta k=0$  for all  $q$ . The fraction of ionization  $\eta$  is defined by the applied laser intensity in the capillary. In our experiment we estimated an ionization ratio of approximately 1% for HHG in He (Fig. 3a, red line).

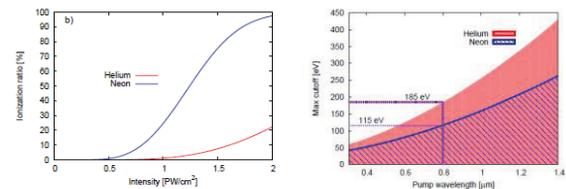


Figure 3: (a) critical ionization ratio in dependence of the laser intensity. In our experiment  $0.7 \text{ PW/cm}^2$  is reached. (b) cut-off photon energies in He and Ne for fully phase-matched HHG in a waveguide. (from ref [8])

According to equation (1) the maximum HH cut off energy in He and Ne for fully phase matched HHG in a capillary is 185 eV and 115 eV, respectively (Fig. 3b) using a Ti:Sapphire laser emitting at  $0.8 \mu\text{m}$  wavelength. In our experiment we could not observe such high photon energies (Fig. 2) due to limitations in our spectrometer.

Shown in figure 4 is the absolute HH photon numbers per second for different gas pressures plotted at the resolution defined by the CCD of the spectrometer (1 pixel corresponds to 1% bandwidth at 160 eV and 0.5% bandwidth at 80 eV, respectively). At a pressure of 200 mbar the previously mentioned

flattop like spectrum is achieved with an integrated photon fluence of  $\approx 10^{10}$  photons/second. In our experiment, increasing the pressure beyond 200 mbar was not feasible due to the limited capabilities of the vacuum pumping system.

The observed signature of fully phase-matched HHG suggests the use of even more elongated waveguides in order to enhance the absolute photon numbers and conversion efficiency further. Under our experimental conditions of full phase-matching the coherence length is expected to be significantly longer than the 33-mm-long

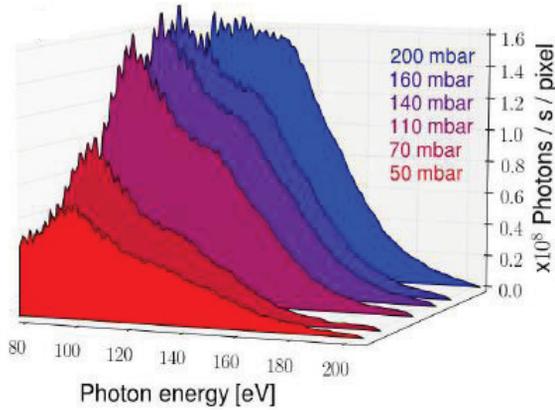


Figure 4: High harmonic spectra recorded in helium at different pressures. 1 pixel corresponds to 1% (0.5%) bandwidth at 160 eV (80 eV). (from ref [8])

capillary. Studies with elongated waveguides will be performed in near future.

The experimentally observed high photon flux indicates the potential of this source for seeding an FEL. For successful seeding, the HH peak power needs to be above the FEL shot noise by typically 2 orders of magnitude [9]. Naturally the shot noise of the FEL increases with the resonant photon energy while HHG conversion efficiency drastically decreases. Currently, state of the art HHG sources do therefore not allow efficient seeding at 20 nm and below.

In order to verify if the HH peak power is sufficient for efficient seeding we estimated the shot noise  $P_{sn}$  according to [10]:

$$P_{sn} = 6\sqrt{\pi}\rho_{3D}^2 \frac{P_b}{N\sqrt{\log\left(\frac{N}{\rho_{3D}}\right)}} \quad (2)$$

with  $P_b$  the electron beam peak current,  $N_e$  the number of electron per radiation wavelength and  $\rho$  the FEL Pierce parameter. We estimated the shot noise using the following electron beam parameters: normalized emittance of  $\epsilon_n=0.43$  mm-mrad, average beta function  $\beta=10$  m, peak current  $P_b=2.7$  kA, kinetic energy of 2.1 GeV and an electron energy spread of 250 keV. Those values are close to the future SwissFEL soft x-ray

facility covering 1-7 nm. The undulator period is 40 mm and the undulator parameter K is 1.2.

Shown in Figure 5 is the peak power for the harmonics calculated for central photon energies between 80 and 180 eV with a bandwidth of 1%. At phase-matched conditions a peak power slightly above 200 W is achieved at 140 eV under consideration of a bandwidth of  $\pm 0.7$  eV.

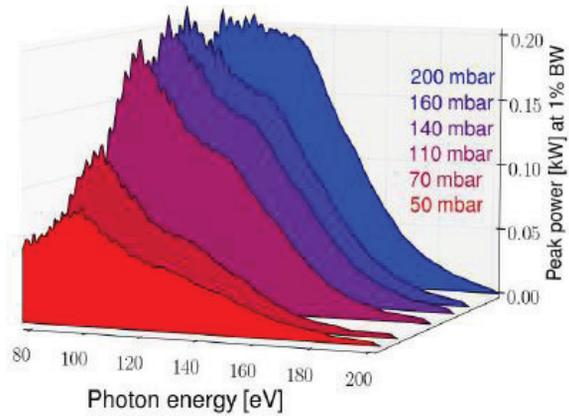


Figure 5: High-harmonic peak power calculated from the experimental measurements shown in Fig 4, for a 1% bandwidth at the corresponding photon energy. (From ref [8]).

In figure 6 the HH peak power is compared to the FEL shot noise calculated by eq. (2). With the presently used Ti:sapphire system and HH waveguide the HH peak power is at best about a factor of 2 above the FEL shot noise level at around 120 eV (i.e. 10 nm). Further up-scaling of the HH peak power seems feasible by combining the presented capillary approach with even longer waveguides and with a Ti:sapphire laser system delivering up to 100 mJ at 100 Hz. Such lasers are now commercially available and should allow significantly better seed-to-SASE power ratio than presented here.

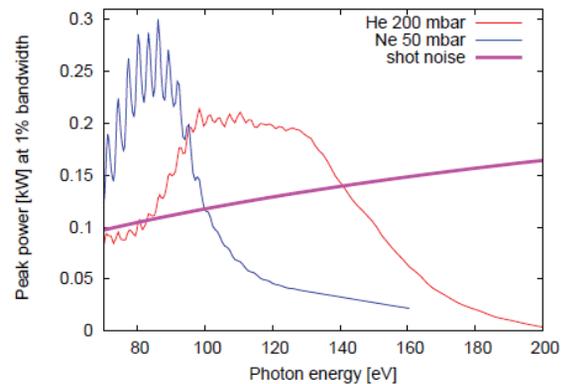


Figure 6: Calculated FEL shot noise and HH peak power spectrum for harmonics produced in He and Ne (from ref [8])

## CONCLUSION

In conclusion, we reported on fully phase-matched high-harmonic generation by use of a 6 mJ Ti:sapphire laser system and an elongated waveguide for HHG. The HH source delivers up to  $10^{10}$  photons per second in a range of 80-160 eV. Phase-matching has been explored in dependence of gas pressure. Under optimized conditions a HH peak power of 0.2 kW is achieved within a 1% bandwidth up to photon energies of 140 eV surpassing the FEL shot noise by a factor of 2. At increased laser power and by use of elongated waveguides the approach presented here offers the potential to achieve sufficient HH peak power for seeding FELs at significantly shorter wavelengths than state of the art.

## ACKNOWLEDGMENT

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## REFERENCES

- [1] G. Lambert et al. "Injection of harmonics generated in gas in a free electron laser providing intense and coherent extreme-ultraviolet light," *Nature Physics*, 4(4):296-300 (2008)
- [2] S. Ackermann et al. "Optimization of HHG seeding at FLASH II," FEL conference, Nara, Japan (2012)
- [3] T. Togashi et al. "Extreme ultraviolet free electron laser seeded with high-order harmonic of Ti:sapphire laser," *Opt. Express* 19(1):317-324 (2011)
- [4] M. Zepf et al. "Bright quasi phase matched soft x-ray harmonic radiation from argon ions," *Phys. Rev. Lett.* 99 (14):143901 (2007)
- [5] A.L. Lytle et al. "Quasi phase matching and characterization of high-order harmonic generation in hollow waveguides using counterpropagating light," *Opt. Express* 16:6544-6566 (2008)
- [6] L.Z. Liu et al. "Quasi phase matching of high-order harmonic generation using polarization beating in optical waveguides," *Phys. Rev. A* 85, 053823 (2012)
- [7] I.J. Kim et al. "Highly efficient high harmonic generation in an orthogonally polarized two-color laser field," *Phys. Rev. Lett.* 94, 243901 (2005)
- [8] F. Ardana-Lamas et al., "Spectral characterization of fully phase-matched high harmonics generated in a hollow waveguide for free electron laser seeding," *New J. Physics* 15, 073040 (2013)
- [9] G. Lambert et al. "Seed level requirement for improving the temporal coherence of a free electron laser," *EPL* 88, 54002 (2009)
- [10] M. Xie "Exact and variational solutions of 3D eigenmodes in high gain FELs," *NIM A*, 445 59 (2000)