Coherent Soft-X-Ray Pulses at Multi MHz Repetition Rates Using Enhancement Cavities

E. Fill, I. Pupeza, S. Holzberger, H. Carstens, N. Lilienfein, A. Apolonski and F. Krausz

Abstract Enhancement cavities are passive optical resonators which allow increasing the power of multi-MHz pulse trains by orders of magnitude. Generating high harmonics from enhanced pulses of an Yb-laser ($\lambda = 1040$ nm) coherent XUV pulses with wavelengths down to 11.5 nm (108 eV) were obtained at a repetition rate of 78 MHz. Harmonics were coupled out of the cavity by a small hole in a mirror. With up to 5 kW of fundamental laser power in the cavity, average powers of 5.4 μW and 8 pW were measured at 38 and 11.5 nm respectively. New developments include scaling of fundamental laser powers in a cavity up to 670 kW and efforts aiming at the generation of attosecond pulses with multi-MHz repetition rates.

1 Introduction

The basic principle of an enhancement cavity involves coherent stacking of ultrashort pulses until the loss in the cavity is equal to the rate of input coupling [1–4]. A generic enhancement cavity is a ring resonator consisting of an input coupling mirror (typical reflectivity 99.5 %) and several other mirrors with much higher reflectivity. Two of these are curved with a radius of curvature and a separation chosen for providing a stable Gaussian beam circulating in the cavity.

The resonance conditions require that the roundtrip time of the cavity be matched to the repetition rate of the seed laser and that the phase of the input pulses be exactly synchronized to the phase of the circulating pulse. If the power reflectivity R of the input coupling mirror equals the round trip power transmission of the cavity the cavity is said to be "impedance matched". On resonance and with a lossless input coupling mirror (power transmission $T = 1 - R$) the enhancement factor is given by

$$e = 1/(1 - R). \tag{1}$$

Typical values for the repetition rate are around 100 MHz but cavities have been operated at 10 MHz, and up to 1 GHz. For the enhancement factor a value of a few

E. Fill (✉) · I. Pupeza · S. Holzberger · H. Carstens · N. Lilienfein · A. Apolonski · F. Krausz
Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Str. 1, 85748 Garching, Germany
e-mail: ernst.fill@mpq.mpg.de

Ludwig-Maximilians-Universität München, Am Coulombwall 1, 85748 Garching, Germany

J. Rocca et al. (eds.), *X-Ray Lasers 2014*, Springer Proceedings in Physics 169,
DOI 10.1007/978-3-319-19521-6_23

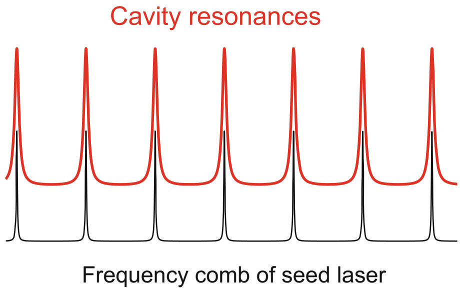

Fig. 1 Resonance conditions of an enhancement cavity in the frequency domain. The comb lines of the seed laser must be exactly aligned to the cavity resonances

hundred is routinely obtained but in special cases values of up to 10,000 have been demonstrated. However, for such a high enhancement the cavity becomes sensitive to small disturbances and its operation is demanding.

It is instructive to consider the situation in the frequency domain, as shown in Fig. 1. The resonance conditions require that the frequency comb lines of the seed laser exactly match the cavity resonances and that the carrier envelope offset frequency is equal to that of the cavity. The first condition is maintained by generating an error signal and locking the repetition rate of the seed laser by a piezoelectrically actuated mirror to the cavity. The carrier offset frequency is in most cases more stable and is manually adjusted.

2 Generating Harmonics in an Enhancement Cavity

Coherent harmonic pulses at multi-MHz repetition rates constitute a frequency comb in the XUV. This was first demonstrated by beating the third harmonic of a Ti:sapphire laser with the third harmonic generated by a nonlinear crystal [3]. In another experiment heterodyne beating between the third harmonic coupled out of an enhancement cavity and the fifth harmonic of a laser at 1064 nm was recorded [2]. Recently low-order harmonics have been used in direct frequency comb spectroscopy of argon and krypton resonance lines [5].

For harmonic generation a gas jet is placed at the focus between the curved mirrors. Intensities exceeding 10^{14}W/cm^2 are easily generated. Usually a so called "end-fire nozzle" consisting of a tube with a small opening at its end is used for producing the gas jet.

Harmonic radiation is emitted collinearly with the circulating fundamental pulses and therefore has to be coupled out of the cavity. Several methods have been used for this purpose, such as a Brewster window [3] or a nano-grating etched into a mirror placed after the focus [6]. These methods carry the risk of damage to these components and have the disadvantage of a low output coupling efficiency for the higher harmonics.

In our experiment output coupling was established by means of a small hole in the mirror after the focus [7]. This method had been suggested in combination with

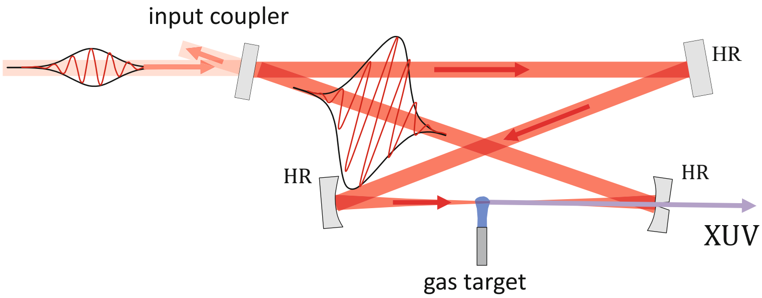

Fig. 2 Schematic layout of harmonic generation experiment. Input is a 175 or 57 fs pulse train with a 78 MHz repetition rate. A gas nozzle is placed close to the focus. Output coupling is realised through a small hole in the mirror following the focus. Focusing is asymmetric with curved mirror radii of 100 and 300 mm for argon and 38 and 150 mm for neon. XUV diagnostics is a grating followed by a calibrated diode or a grazing incidence spectrometer with a CCD camera on the Rowland circle

a donut-shaped mode circulating in the cavity, which avoids the hole [8]. However, we use the normal TEM_{00} mode and make the hole small enough so losses are below 0.5 % and the cavity can be operated with an enhancement factor of about 200.

Hole output coupling has the following advantages:

- Increased output coupling efficiency at shorter wavelengths
- No dispersion introduced
- Polarization independent
- Harmonics are collinear.

3 Experimental Results

A schematic layout of the experiment is shown in Fig. 2. The seed laser was an Yb-laser emitting a power of up to 57 W with a pulse duration of 175 fs. By focusing the pulse into a nonlinear fiber and shortening it by chirped mirrors pulses with duration down to 27 fs could be obtained [9].

First experiments were carried out with argon, using mirrors with radii of curvature of 100 and 300 mm and a focal spot size of 20 μm. Plasma generated in the focus severely affected the performance of the cavity. Dynamic ionization during pulse propagation resulted in a blue shift of the circulating pulse reducing the spectral overlap with the seed pulses. The situation was much improved when seeding the cavity with shorter pulses. The bandwidth of the cavity mirrors was not wide enough for enhancing 27 fs pulses. However, seeding with 57 fs pulses the intensity in the focus could be increased from 6×10^{13}W/cm^2 to 8×10^{13}W/cm^2. Higher order harmonics and higher power could be generated in this way (Fig. 3a). With a calibrated diode a power of 5.3 μW was measured at a wavelength of 38.5 nm

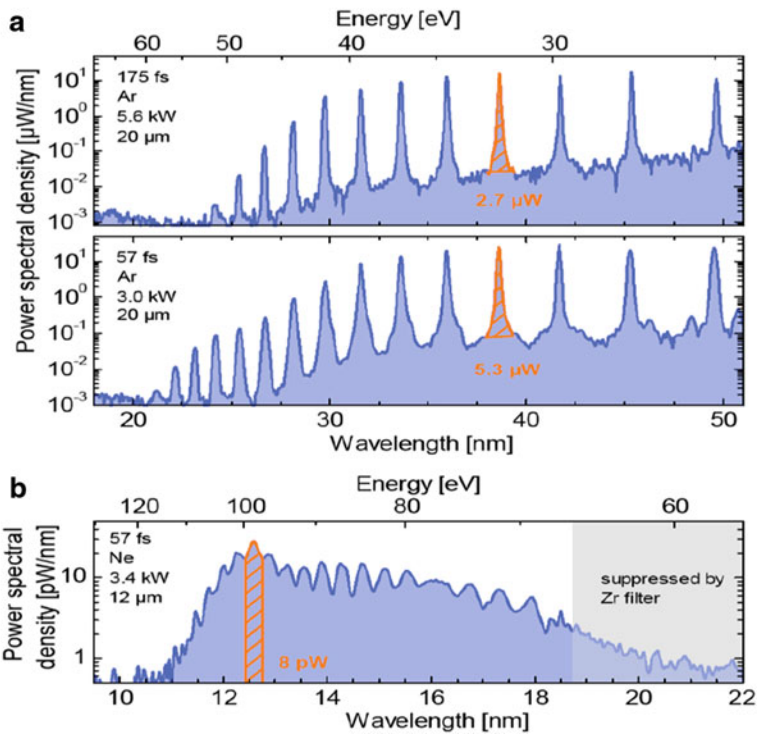

Fig. 3 Harmonic spectra obtained with argon (**a**) and neon (**b**). In argon, shorter pulses generate higher harmonics and more harmonic power. In neon a smaller focal spot size (12 μm instead of 20 μm) generates harmonics with wavelengths down to 11.5 nm. Figure reproduced from ref. [7] with permission

(27th harmonic). To generate higher harmonics intensities exceeding 10^{14} W/cm^2 are required. For this purpose the cavity mirrors were changed to radii of 38 and 150 mm, generating a 12 μm focal spot size. With 5 kW of average power in the cavity a harmonic spectrum as shown in Fig. 3b was obtained.

The data recorded by the CCD camera demonstrate spatial coherence of the generated harmonics. In Fig. 4 the horizontal axis is the spectrally dispersed coordinate and the vertical axis signifies the spatial dimension. For relatively low order harmonics the spatial pattern exhibits diffraction lobes and can well be simulated by a plane wave model. The higher order harmonics display a central core and a pedestal attributed to short and long quantum path harmonics [10].

4 New Developments

Recent projects conducted by the enhancement cavity group at MPQ/LMU include power scaling to MW-range circulating powers and the enhancement of few-cycle pulses in the cavity. Appropriately filtered harmonics are expected to generate isolated attosecond pulses.

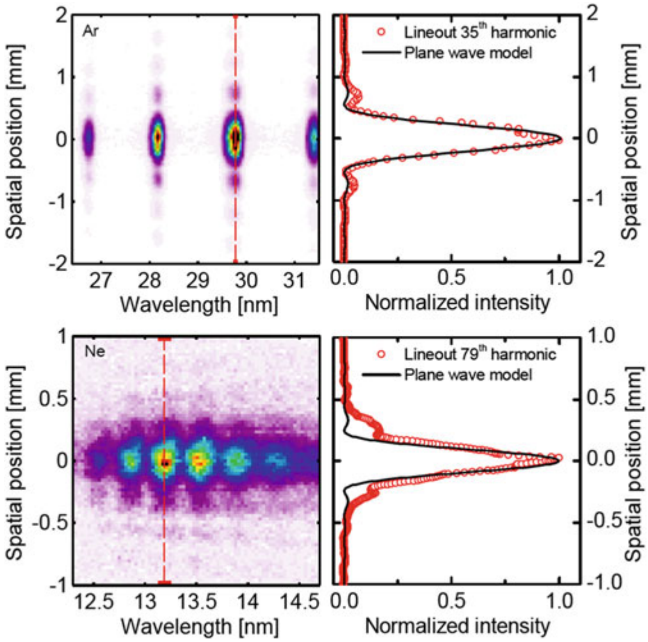

Fig. 4 Two-dimensional recording of harmonics demonstrating spatial coherence of *low*-order harmonics and short-long quantum path composition of *high*-order harmonics. Figure reproduced from ref. [7] with permission

4.1 Further Power Scaling

One of the goals of this research is to generate more powerful harmonics and to extend their spectral range to shorter wavelengths, possibly to the water window. For this purpose a new seed laser was installed, capable of delivering up to 420 W of power at a pulse duration of 250 fs and a repetition rate of 250 MHz. With such a high seed power two limitations are met in the enhancement cavity:

- Damage of the mirrors induced by the high intensity
- Thermal effects due to the high circulating power.

To circumvent these problems a new cavity was designed, which exhibits a considerably larger spot size on the mirrors. The cavity is operated close to the limits of stability, where the spot size of an optical resonator increases significantly. However, in turn an increased alignment instability is expected (Fig. 5).

In theory and experimentally it was found that at one of the stability edges, alignment of the cavity was very sensitive to small disturbances, whereas at the other (with the curved mirrors close to a confocal arrangement) alignment was straightforward. Operating the cavity close to that stability limit the illuminated area on the mirrors was increased by a factor of 15, allowing correspondingly higher circulating powers without the risk of mirror damage [11].

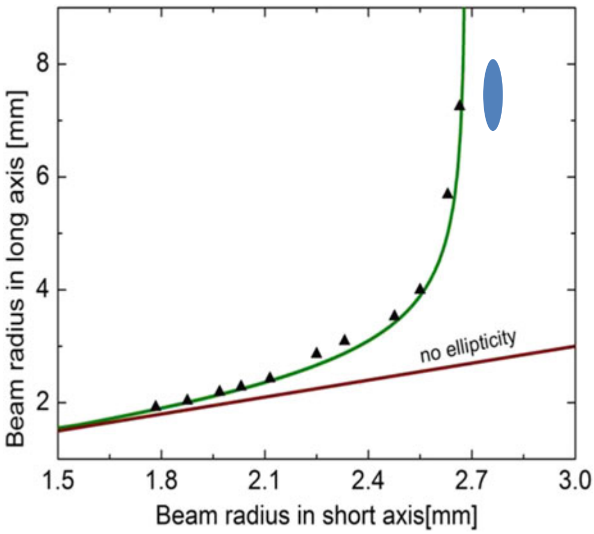

Fig. 5 Beam radius in long *axis* (*sagittal*) vs. beam radius in short *axis* (*tangential*). The diagram shows the increased ellipticity of the beam profile, resulting from oblique incidence on the curved mirrors. The experimental data (*triangles*) follow well the theoretical curve (*green*). The curve with no ellipticity would result for 0° angles of incidence

When running the cavity with high power thermal effects on the mirrors come into play. Absorption of the radiation in the first layers of the mirrors results in a temperature increase and a change in the radius of curvature. Using ultralow-expansion (ULE) glass for the mirrors and a coated sapphire substrate for the input coupler a circulating power of 670 W at a pulse duration of 10 ps could be realized [12]. For 250 fs pulses a circulating power of 400 kW was achieved, limited by mirror damage. Higher damage threshold mirrors allowing close to MW circulating power at a short pulse duration are being developed.

4.2 Generation of Few-Cycle Pulses and Attosecond Harmonics

One of the grand goals of this research is the generation of isolated attosecond pulses (IAPs) at multi-MHz repetition rates. In many investigations with low yield acquisition times could be dramatically reduced using such a source. Examples are angle resolved photoemission spectroscopy (ARPES) [13], and cold target recoil ion momentum spectroscopy (COLTRIMS) [14]. In these experiments the signal yield is limited by space charge effects, resulting in hours of acquisition time with kHz repetition rate lasers.

For generating IAPs the fundamental radiation must consist of few cycle pulses [15]. Their harmonic spectrum does not show isolated peaks but is continuous. To

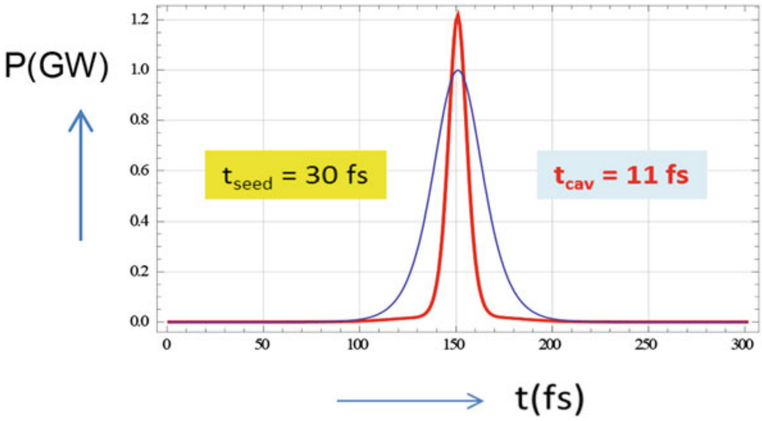

Fig. 6 Generating a soliton in an enhancement cavity. The 30 fs seed pulse is reduced to a 11 fs circulating pulse, shown in *red*. The parameters used in the simulation are: linear enhancement factor 200, seed peak power 5 MW, *second* order dispersion $\beta_2 = -1.2\text{fs}^2$, nonlinear medium: 100 μm sapphire plate. The seed pulse is scaled by the linear enhancement factor

achieve this goal a soliton may be launched in the enhancement cavity by introducing a Kerr medium (with a second order nonlinearity) and negative second order dispersion [16]. Numerically solving the "driven nonlinear Schrödinger equation" it appears that a soliton with significantly shorter pulse duration than that of the driver pulse can be generated in the cavity (Fig. 6). However, for obtaining an IAP, further pulse shortening must be achieved, possibly by applying a gating technique

References

1. Jones, R.J., Ye, J.: Femtosecond pulse amplification by coherent addition in a passive optical cavity. Opt. Lett. **27**, 1848–1850 (2002)
2. Gohle, C., et al.: A frequency comb in the extreme ultraviolet. Nature **436**, 234–237 (2005)
3. Jones, R.J., et al.: Phase coherent frequency combs in the vacuum ultraviolet via high-harmonic generation inside a femtosecond enhancement cavity. Phys. Rev. Lett. **94**, 193201 (2005)
4. Mills, A.K., et al.: XUV frequency combs via femtosecond enhancement cavities. J. Phys. B At. Mol. Opt. Phys. **45**, 142001–142023 (2012)
5. Cingöz, A., et al.: Direct frequency comb spectroscopy in the extreme ultraviolet. Nature **482**, 68–71 (2012)
6. Yost, D.C., Schibli, T.R., Ye, J.: Efficient output coupling of intracavity high-harmonic generation. Opt. Lett. **33**, 1099–1101 (2008)
7. Pupeza, I., Holzberger, S., et al.: Compact high-repetition-rate source of coherent 100 eV radiation. Nat. Photon **7**, 608–612 (2013)
8. Moll, K.D., Jones, R.J., Ye, J.: Output coupling methods for cavity-based high-harmonic generation. Opt. Express. **14**, 8189–8197 (2006)
9. Eidam, T., et al.: 57 W, 27 fs pulses from a fiber laser system using nonlinear compression. Appl. Phys. **92**, 9–12 (2008)

10. Zaïr, A., et al.: Quantum path interferences in high-order harmonic generation. Phys. Rev. Lett. **100**, 143902 (2008)
11. Carstens, H., et al.: Large-mode enhancement cavities. Opt. Express **21**, 11606–11617 (2013)
12. Carstens, H., et al.: Megawatt-scale average-power ultrashort pulses in an enhancement cavity. Opt. Lett. **39**, 2595–2598 (2014)
13. Damascelli, A., Hussain, Z., Shen, Z.: Angle–resolved photoemission studies of the cuprate superconductors. Rev. Mod. Phys. **75**, 473–541 (2003)
14. Ulrich, J., et al.: Recoil-ion and electron momentum spectroscopy: reaction-microscopes. Rep. Prog. Phys. **66**, 1463–1545 (2003)
15. Brabec, T., Krausz, F.: Intense few-cycle laser fields: frontiers of nonlinear optics. Rev. Mod. Phys. **72**, 545–591 (2000)
16. Kalashnikov, V.L.: Femtosecond pulse enhancement in an external resonator: impact of dispersive and nonlinear effects. Appl. Phys. **92**, 19–23 (2008)