# Vacuum ultraviolet frequency combs generated by a femtosecond enhancement cavity in the visible

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We present the first (to our best knowledge) femtosecond enhancement cavity in the visible wavelength range for ultraviolet frequency comb generation. The cavity is seeded at 518 nm by a frequency-doubled Yb fiber laser and operates at a peak intensity of $1.2 \times 10^{13}$ W/cm$^2$. High harmonics of up to the ninth order (~57 nm) are generated in an intracavity xenon gas jet. Intracavity high harmonic powers of several milliwatts for the third harmonic order and microwatts for the fifth harmonic order prove the potential of the "green cavity" as an efficient ultraviolet frequency comb source for future spectroscopic experiments. A limiting degradation effect of the cavity mirrors is avoided by operating at a constant oxygen background pressure. © 2012 Optical Society of America

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The vacuum ultraviolet (VUV) wavelength region (10–200 nm) is spectroscopically still rather unexplored due to the lack of suitable direct laser sources. Coherent VUV radiation is available via high harmonic generation (HHG) from ultrashort pulses. The required laser intensities of $>10^{13}$ W/cm$^2$ may be reached by using a high-power laser system [1], inside a femtosecond enhancement cavity (fsEC), or by reducing the pulse repetition rate maintaining the time-averaged power. The latter method, standard for many years, is not suitable for optical frequency metrology [2] because of the corresponding small mode spacing. With cavity-based systems, harmonics down to 40 nm have successfully been generated, mainly motivated by the prospect of two-photon precision frequency comb spectroscopy in the VUV [3–5]. VUV direct frequency comb spectroscopy of an electric dipole transition has recently been demonstrated [6].

Because typical conversion efficiencies of HHG are on the order of $10^{-7}$ or less, different approaches have been pursued to optimize the output power of first-generation cavity-assisted VUV sources, which were based on Ti:sapphire seed lasers [3,6,7]. The most common one by now is to use high-power Yb-doped fiber laser systems that provide several tens of watts for seeding the fsEC [8–11]. With these systems, the achievable intracavity powers on the order of several kilowatts cause undesirable effects that are difficult to avoid or to control because they result from the high intensities (i) on the cavity mirrors [9], (ii) in the gas target [11,12], and (iii) on the Brewster plate output coupler [3]. Therefore, our approach for obtaining higher VUV output powers is to improve the conversion efficiency itself. Recent investigations of the VUV power scaling claim that the conversion efficiency scales with $\sim\lambda^{-6.3\pm1.1}$ in Xe for the plateau harmonics, thus making the driving wavelength $\lambda$ the most crucial experimental parameter for an optimized VUV output [13]. Several single-pass HHG experiments with kHz repetition rate systems found that shorter driving wavelengths yield higher conversion efficiencies [14], albeit with reduced cutoff energy.

In this Letter, we present the first cavity-assisted VUV source based on a fsEC in the visible wavelength region. As shown in Fig. 1, an Yb-doped fiber oscillator emitting 130 mW of average power at a center wavelength of 1040 nm and a repetition rate of 128 MHz seeds an Yb fiber amplifier based on a chirped pulse amplification scheme that has been described in detail in [15]. After amplification, 10 W of average power at a pulse duration of 130 fs are available for frequency-doubling in a 1 mm thick BBO crystal, yielding 155 fs pulses centered at 518 nm with an average power of 5 W. The green light seeds an impedance matched ring cavity consisting of four plane and two concave ($R = 50$ mm) highly reflecting mirrors with low group delay dispersion (<0.15 fs$^2$ over more than the full spectral bandwidth of 14 nm centered at 518 nm, transmission of the input coupler $T_{IC} \sim 0.5\%$). For a constructive

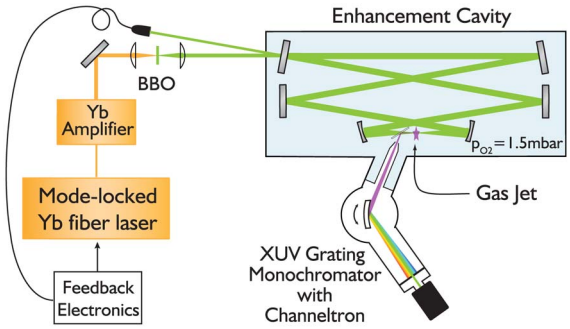


Fig. 1. (Color online) Experimental setup for the VUV frequency comb generation. The output of an amplified, frequency-doubled pulsed fiber laser is injected into a fsEC. The tight cavity focus enables HHG in a Xe gas jet. A thin fused silica plate placed at Brewster's angle extracts the VUV light, which is characterized with a VUV monochromator and a Channeltron.

 

build-up, the resonator length of the laser is actively stabilized to the fsEC with the Pound–Drever–Hall method. The carrier-envelope offset frequency of the laser is manually controlled for stable operation and an optimized VUV output. A 100 $\mu$m thin fused silica plate placed behind the focus at Brewster's angle for the fundamental wavelength 518 nm (~55.6°) extracts the generated VUV out of the fsEC.

Figure 2(a) shows the incident laser spectrum (black solid line) and the intracavity spectra with the Brewster plate and both Brewster plate and gas jet (dashed blue and dotted violet curves, respectively). The group delay dispersion of our Brewster plate is ~8.5 fs$^2$ so that its chirping effect is rather negligible (measured intracavity pulse duration ~166 fs). The drop in power enhancement of about 20% with respect to the empty fsEC can be mostly attributed to the gas jet due to nonlinearities in the tight focus ($1/e^2$ intensity radius of $w_0 = 6.3$ $\mu$m). Limitations in the achievable power enhancement due to these nonlinearities have recently been observed by other groups [11,12].

In comparison with an IR cavity system, the reflectivity of the green cavity mirrors is lower due to the increased sensitivity of the shorter fundamental wavelength to imperfections of the dielectric mirror layers. We reach a power enhancement of 200 relative to the incident laser power of 250 mW (spatial mode matching 80%). Twice this value is obtained with the IR laser radiation using the best available mirrors [15]. The enhancement reduces with increasing input power [see Fig. 2(b)] and saturates at an intracavity power of 300 W with 4 W of injected power (5 W incident) into the empty fsEC with an oxygen ($O_2$) background pressure of 10 mbar. This background pressure is necessary to prevent an effect that is a commonly known phenomenon in the VUV range (see, e.g., [16,17]), but reported for the visible for the first time here to our knowledge: the combination of high intracavity powers (>40 W in our case) and pressures smaller than 1 mbar causes a quick degradation of the reflectivity of the fsEC mirrors, which affects the fsEC finesse within a few seconds [see Fig. 2c]. After the degradation, the fsEC can be stabilized onto higher order TEM modes only with a largely reduced intracavity power of about 5%–10% of the original value [see the insets of Fig. 2(c)]. The most common explanation for the deterioration of the mirror reflectivity in the VUV range under vacuum is that residual organic compounds are dissociated by the intense laser radiation to form a carbon layer on the mirror surface [17]. We suspect that the same mechanism also causes the degradation in our experiment. Increased scattering and a stained appearance of the mirror surfaces after the degradation support this assumption. A possible degradation of the Brewster plate transmission has not been investigated separately. An established technique to restore the mirror reflectivity is a treatment with $O_2$ that chemically removes the carbon compounds so that no additional cleaning is required. In our experiment, the mirror degradation is reversible when $O_2$ is supplied to the cavity mirrors while the fsEC is locked. Moreover, the degradation can be prevented completely by operating the fsEC at a permanent $O_2$ background pressure of 1.5 mbar. Because the generated VUV radiation is strongly absorbed by the $O_2$, a specially designed spectrometer (see Fig. 1) is used: a small

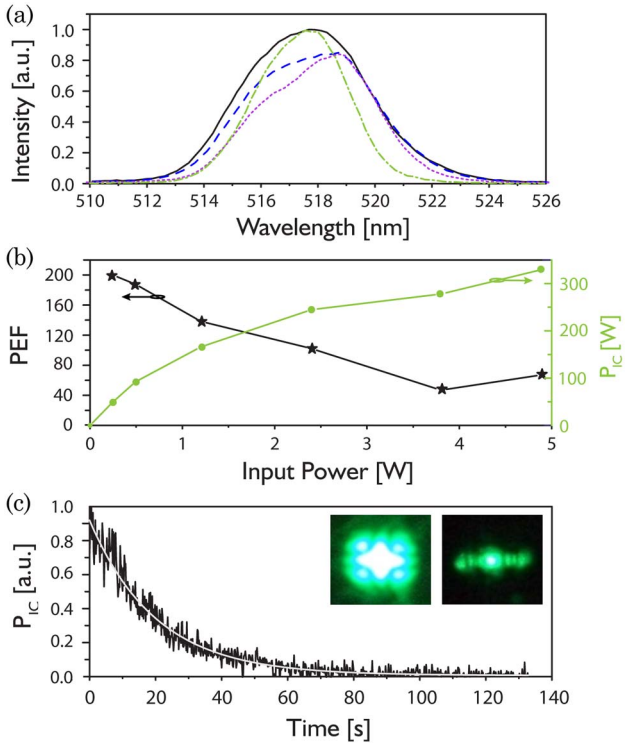


Fig. 2. (Color online) (a) Laser spectrum in front of the cavity (solid black curve) and intracavity spectra with Brewster plate (dashed blue curve) and both Brewster plate and gas jet (dotted violet curve). The green dashed–dotted curve shows the intracavity spectrum with detuned carrier-envelope offset for stable long-term operation. (b) Saturation of intracavity power in dependence of the incident laser power and respective power enhancement factor (PEF). (c) Degradation of intracavity power in vacuum. Insets, higher order TEM cavity modes after degradation. Data in (b), (c) are recorded with an empty cavity, i.e., no output coupler, no gas jet.

tube placed directly next to the outcoupling Brewster plate forms the entrance of the differentially pumped detection chamber (~$10^{-5}$ mbar), which comprises a VUV monochromator (Jobin Yvon LHT30) and a Channeltron (Photonis CEM4839) to characterize the generated VUV light. The required $O_2$ background leads to a stronger coupling of vibrations and acoustics to the fsEC so that the in-loop error signal of the cavity stabilization becomes noisier. Locking instabilities due to plasma interactions [7,8] in $O_2$ could be excluded by side investigations with $O_2$ as the HH generating medium. For long-time and robust operation (>45 min), the carrier-envelope offset of the laser was slightly detuned with a slightly narrower enhanced spectrum [green dashed–dotted curve in Fig. 2(a)] for a broader resonance that increases the range of the error signal. Under stable conditions, an intensity of $1.2 \times 10^{13}$ W/cm$^2$ at the cavity focus is achieved. This enables the generation of high harmonics up to the ninth order (~57 nm) in a Xe gas jet. The recorded spectrum is shown in Fig. 3. Because our Channeltron is not sensitive enough to detect the third harmonic, a photomultiplier is used instead. The ninth order has to be investigated separately because the $O_2$ absorption suppresses the weak signal in the overall wavelength scan. For this reason, the ninth harmonic is generated without the $O_2$ background and detected in the short time window before cavity

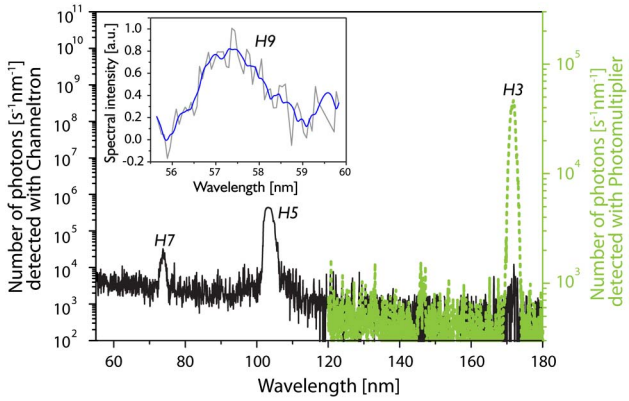


Fig. 3. (Color online) Measured spectrum of high harmonics coupled out of the enhancement cavity. The third harmonic was measured with a CsI-coated photomultiplier (dotted green curve scaled to match the Channeltron data) because the Channeltron was not sensitive enough in this wavelength region. Inset, weak signal from the ninth harmonic that had to be measured separately without the $O_2$ background due to its large absorption.

degradation sets in. Table 1 summarizes the generated ($P_{GEN}$) and outcoupled ($P_{EX}$) powers for the different harmonic orders. For the underlying absolute power calibration, the VUV monochromator is removed and the Channeltron detector is placed directly after the tube separating the two vacuum chambers with a Parylene foil coated with 300 nm of Mg (Lebow Co.) to maintain the $O_2$ pressure. The power calibration is performed by taking into account the Mg–Parylene transmission ($T_{Mg} \sim 10^{-4}$, measured at 104 nm), the transmission of the ~1.2 cm long path through $O_2$ [18], the reflectivity of the fused silica output coupler at Brewster's angle (calculated), the diffraction and detection efficiency of the monochromator (2%, assumed to be independent of wavelength), and the Channeltron (specified: H3, 0.001%; H5, 8%; H7, 10%; H9, 10%). Comparing the generated powers in THE detected harmonic orders 3 to 9 with the achieved intracavity power of 161 W yields conversion efficiencies of $10^{-5}$, $10^{-8}$, $10^{-10}$, and $10^{-11}$, respectively.

A proper comparison between the presented results and previous cavity-assisted sources driven by IR lasers is difficult because most experimental parameters differ. Apart from exploiting the favorable wavelength scaling of HHG, the main advantage of our system is that the low harmonic orders, which typically exhibit high conversion efficiencies, reach well into the VUV due to the shorter fundamental wavelength. Therefore, compared to the latest IR fsECs, less fundamental pulse energy is required to obtain a similar conversion efficiency for the same VUV wavelength. Moreover, due to the highly nonlinear nature of HHG, a small improvement of the intracavity power should enable the generation of a plateau in the high harmonic spectrum and hence lead to a dramatic improvement of the conversion efficiency for the higher harmonic orders. In addition, the bandwidth of the servo system (~30 kHz) could be increased by an intralaser cavity electro-optic modulator. To further increase the extracted power, carefully selected output couplers, which exhibit up to 1 order of magnitude higher reflectivities in the VUV (e.g., MgO [10]), could be used.

**Table 1. Summary of Generated and Outcoupled Powers: $P_{GEN}$ and $P_{EX}$, for the Different Harmonic Orders (HO)[a]**

| HO | $\lambda$[nm] | $P_{GEN}$ | $T_{O2}$ | $R_{FS}$ | $P_{EX}$ |
|---|---|---|---|---|---|
| Third | 172.7 | 6.5 mW | 0.55 | $8.8 \times 10^{-4}$ | 3.1 $\mu$W |
| Fifth | 103.6 | 3.9 $\mu$W | 0.70 | $4.4 \times 10^{-2}$ | 120 nW |
| Seventh | 74.0 | 150 nW | 0.58 | $5.6 \times 10^{-2}$ | 5.0 nW |
| Ninth | 57.6 | 5.2 nW | (0.32) | $7.3 \times 10^{-2}$ | 122 pW |

[a]$T_{O2}$ is the transmission through the 1.2 cm long path of oxygen at a pressure of 1.5 mbar and $R_{FS}$ is the reflectivity of the fused silica output coupler at Brewster's angle for 518 nm (see Fig. 1).

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