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Extreme ultraviolet light source at a megahertz repetition rate based on high-harmonic generation inside a mode-locked thin-disk laser oscillator

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We demonstrate a compact extreme ultraviolet (XUV) source based on high-harmonic generation (HHG) driven directly inside the cavity of a mode-locked thin-disk laser oscillator. The laser is directly diode-pumped at a power of only 51 W and operates at a wavelength of 1034 nm and a 17.35 MHz repetition rate. We drive HHG in a high-pressure xenon gas jet with an intracavity peak intensity of 2.8×10^{13} W/cm^2 and 320 W of intracavity average power. Despite the high-pressure gas jet, the laser operates at high stability. We detect harmonics up to the 17th order (60.8 nm, 20.4 eV) and estimate a flux of 2.6×10^{8} photons/s for the 11th harmonic (94 nm, 13.2 eV). Due to the power scalability of the thin-disk concept, this class of compact XUV sources has the potential to become a versatile tool for areas such as attosecond science, XUV spectroscopy, and high-resolution imaging.

OCIS codes: (190.7110) Ultrafast nonlinear optics; (190.2620) Harmonic generation and mixing; (140.7090) Ultrafast lasers; (140.4050) Mode-locked lasers.

Focusing intense femtosecond pulses into a gas target enables the generation of higher harmonics of the fundamental laser frequency [1]. To drive this highly nonlinear process, the femtosecond laser has to deliver peak intensities above 10^{13} W/cm^2. Standard HHG systems typically rely on Ti:sapphire chirped pulse amplifiers, which usually operate at kilohertz repetition rates and average powers of a few watts. The conversion efficiency to the high-harmonic radiation being very low [2,3] (below 7.5×10^{-5} for a single harmonic [4]); this results in very low average power of the generated extreme ultraviolet (XUV) radiation. This is an issue for applications such as high-resolution imaging, because it strongly affects measurement speed and resolution [5]. Furthermore, the low repetition rate is a challenge for many measurements, for example, experiments in which the energy or momentum of photoelectrons has to be precisely measured. Increasing the repetition rate from kilohertz to megahertz can avoid space charge effects and strongly decrease measurement time [6]. Moreover, XUV sources operating at kilohertz repetition rates are not suitable for direct frequency comb spectroscopy. Due to this large scientific potential, the last years have seen a tremendous increase in research efforts targeting HHG with high photon flux at a megahertz repetition rate. So far, two main directions have been investigated. The most obvious one is to increase both the average power and the repetition rate of the driving laser system, so that sufficiently high peak intensities can be obtained to drive single-pass HHG at megahertz repetition rates. This was demonstrated for the first time in 2009 with a high-power fiber chirped-pulse-amplifier (FCPA) system as the driving laser [7]. Record-high megahertz HHG power levels have recently been achieved using coherently combined FCPAs, in combination with temporal pulse compression in gas-filled hollow-core fibers [8]. Ultrafast high-power slab amplifiers also enabled HHG at a megahertz repetition rate, however, so far at significantly lower XUV power levels [9]. An alternative to amplifier-based systems is single-pass HHG driven by ultrafast thin-disk laser (TDL) oscillators [6,10] which was demonstrated in 2015 [11]. The second direction is based on enhancement cavities for ultrashort pulses, which enabled HHG at megahertz repetition rates already in 2005 [12,13]. The development of powerful FCPA-based frequency combs [14] pushed this technique further,

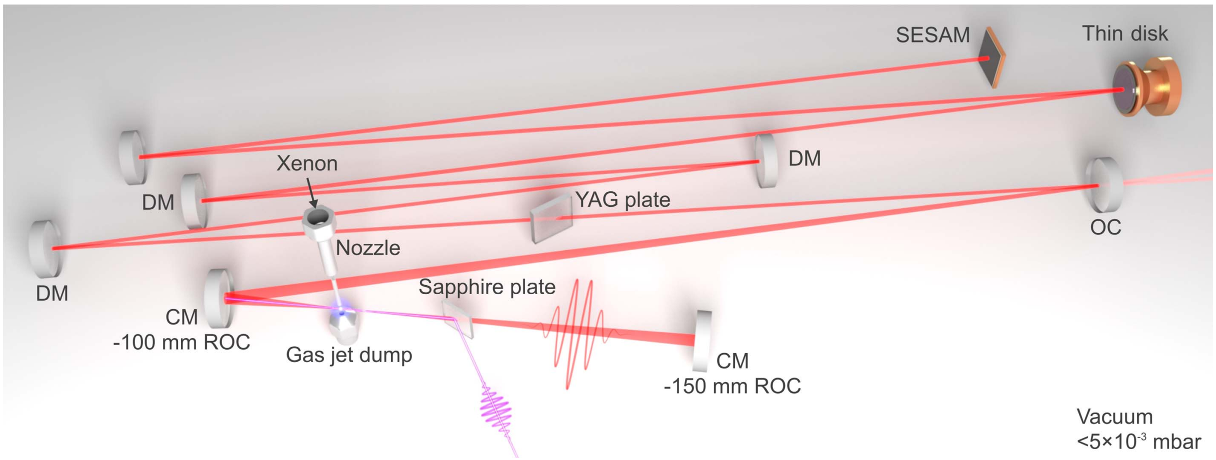

Fig. 1. Illustration of the experiment's principle (DM, dispersive mirror; CM, concave mirror; ROC, radius of curvature; OC, output coupler).

which has resulted in the generation of an XUV photon flux up to several hundred microwatts in a given harmonic [15] and allowed, e.g., the generation of photon energies exceeding 100 eV at a repetition rate of 250 MHz [16]. However, its experimental realization is highly complex. Stable coupling of femtosecond pulses from an amplified frequency comb into a high-finesse resonator containing the HHG interaction is challenging. In addition, the requirements on the phase stability of the driving laser system are very demanding.

Placing the HHG interaction directly inside a mode-locked oscillator is a simpler approach that does not require any input matching of ultrashort pulses. Instead, the circulating femtosecond pulse can adapt to the present cavity nonlinearities and the dispersion. In 2012, the feasibility of this concept was demonstrated using a Ti:sapphire laser oscillator [17], but only at an intracavity average power of 10 W. Due to thermal effects and nonlinearities, ultrafast lasers using bulk crystals, such as Ti:sapphire, are severely limited in average power. This is not the case for ultrafast TDL oscillators. Here the gain medium has the shape of a thin disk which is mounted onto a heat sink and used in reflection with a large beam diameter [10,18]. The thin-disk geometry enables efficient cooling, thus limiting thermal aberrations, and strongly reduces nonlinearities in the gain element. Ultrafast TDL oscillators achieve the highest average power and pulse energy of any mode-locked laser oscillator technology, both intra- and extra-cavity [19–21]. Recently, ultrafast TDL oscillators based on Yb-doped gain materials achieved pulse durations as short as 30 fs directly emitted from the oscillator [22,23]. Furthermore, they can operate with low noise, and carrier-envelope offset (CEO) frequency stabilization has been achieved [24,25], showing their suitability for frequency comb applications.

In this Letter, we report on the first HHG inside an ultrafast TDL oscillator. Our system is based on a diode-pumped $Yb:Lu_2O_3$ thin disk, and mode-locked with a semiconductor saturable absorber mirror (SESAM) [26]. We evaluate the laser noise properties and compare operation with and without HHG process. Even though we inject xenon gas at high backing pressure into an intracavity focus, we do not observe any instabilities of the mode-locked laser. Our experiment shows that ultrafast TDL oscillators are well suited for extreme intracavity nonlinear optics experiments such as HHG.

The resonator, TDL head, and gas target are placed in a compact vacuum chamber with dimensions of 80 × 160 cm^2. An illustration of the experimental setup is shown in Fig. 1. The vacuum chamber is evacuated to a pressure of $\sim 10^{-4}$ mbar with two turbomolecular pumps. As a gain element, we use a wedged 200 μm thick $Yb:Lu_2O_3$ disk that is mounted onto a water-cooled diamond heat sink. The disk is pumped by a fiber-coupled diode at the zero-phonon line at 976 nm. It is used as a folding mirror in the standing-wave cavity. A SESAM inserted as an end mirror enables mode locking. It has 1.6% modulation depth, 0.3% nonsaturable losses, and a saturation fluence of 47.5 $\mu J/cm^2$. The beam radii on the SESAM and on the disk are 0.95 and 1.15 mm, respectively. A 4 mm thick YAG plate is placed at a Brewster's angle for enforcing linear p-polarization and introduces sufficient self-phase modulation (SPM) for soliton mode locking. The SPM is balanced by three dispersive mirrors (DMs). The total intracavity group delay dispersion is $-3000\ fs^2$ per roundtrip. An output coupler (OC) with a transmission of 0.7% is used as a folding mirror. A 12 μm radius intracavity focus is created between two concave mirrors (CMs) with 100 and 150 mm radii of curvature (ROC), the latter one being used as an end mirror. To extract the generated XUV light, we placed a sapphire plate with a thickness of 250 μm at a Brewster's angle for the laser wavelength 2 cm behind the focus. This outcoupling method has been extensively used in cavity-enhanced HHG [12,13]. Without gas, the laser generates 264 fs pulses at a repetition rate of 17.35 MHz with an intracavity average power of 320 W at a pump power of 49 W. Its intracavity peak power of 62 MW leads to a peak intensity of $\sim 2.7 \times 10^{13}\ W/cm^2$ at the focus. The corresponding intensity autocorrelation trace, optical spectrum, and radio frequency (RF) spectrum are shown in Figs. 2(a)–2(c). At higher pump power levels without the HHG process, we observed mode locking instabilities, most likely due to operation close to the roll-over of the SESAM reflectivity, in combination with the finite gain bandwidth of the gain material [27].

We use a quartz nozzle with a ~100 μm opening diameter for gas delivery into the intracavity focus. In order to keep the chamber pressure below 5×10^{-3} mbar while a high-pressure gas jet is used, a gas jet dump is placed below the nozzle [28]. When the xenon gas jet is emitted into the focus,

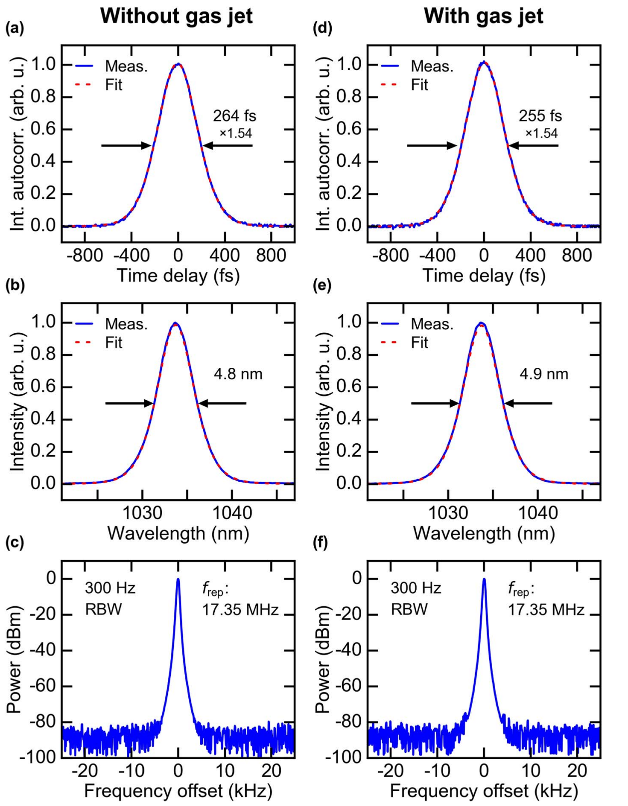

Fig. 2. Comparison of the TDL oscillator output parameters (a)–(c) without a high-pressure gas jet and (d)–(f) with a high-pressure gas jet. (a), (d) Intensity autocorrelation traces; (b), (e) optical spectra, and (c), (f) RF spectra.

HHG is observed and detected with a channel electron multiplier (CEM, Photonis Magnum 5900). The XUV light is directed by an unprotected gold mirror to a wavelength-calibrated monochromator (Acton VM-502) equipped with a 1200 g/mm iridium-coated grating. The slits' width was set for a 3.4 nm spectral resolution. We acquired the XUV spectra with and without a 0.2 μm thick aluminum filter to check the validity of our measurement.

We use 3.4 bar of backing pressure in the nozzle which leads to a pressure at the laser focus estimated to ∼400 mbar. At this gas pressure, the average output power of the laser drops slightly, and we increase the pump power from 49 to 51 W to achieve the same intracavity average power of 320 W as without gas jet. The TDL oscillator with HHG operates with slightly shorter 255 fs pulses. Its intracavity peak power is 64 MW, which leads to a peak intensity of $\sim 2.8 \times 10^{13}$ W/cm^2 at the focus. The corresponding intensity autocorrelation trace, optical spectrum, and RF spectrum are shown in Figs. 2(d)–2(f). High harmonics with orders up to the 17th (60.8 nm, 20.4 eV) are detected, in accordance with predictions from the cutoff formula [29]. The acquired spectra are shown in Fig. 3. Harmonics below the 11th order (94 nm, 13.2 eV) were not detected, most likely due to reabsorption in xenon for the 9th harmonic and the low quantum efficiency of our detector in the spectral range corresponding to the 7th harmonic and above it. Using the measured spectra and an additional measurement of the XUV flux with the CEM in all the detected harmonics before the monochromator without

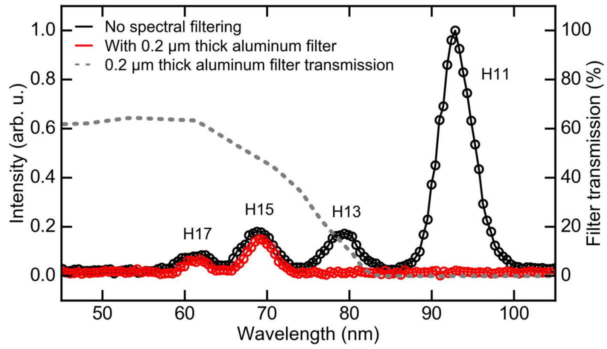

Fig. 3. Measured spectra of the generated XUV light. The full spectrum is plotted in black, while the spectrum filtered by a 0.2 μm thick aluminum foil is plotted in red. The theoretical transmission of the aluminum filter is shown for reference and plotted as a gray dashed line.

an aluminum filter, we estimate the average power and photon flux generated at the focus for the 11th harmonic with a similar method as the one described in Ref. [11]. A very conservative estimation results in a generated flux $\gtrsim 2.6 \times 10^8$ photons/s. This corresponds to an average power $\gtrsim 0.55$ nW and a conversion efficiency $\gtrsim 1.7 \times 10^{-12}$ with respect to the intracavity average power and $\gtrsim 1.1 \times 10^{-11}$ with respect to the diode pump power.

To evaluate if there is any laser perturbation induced by the plasma generated during the HHG process, we compare the transverse beam quality and laser noise with and without HHG. The laser operates in both cases in a fundamental transverse TEM_{00} mode with an M^2 factor <1.02. The noise of the TDL oscillator output was measured in free-running operation on the passively filtered 4th harmonic of the repetition rate using a phase noise analyzer (Rohde & Schwarz FSWP). The measured power spectral densities of the amplitude and phase noises are shown in Fig. 4. Although our vacuum chamber is connected to two turbomolecular pumps and we did not optimize our opto-mechanical components for high stability, we achieve an integrated relative intensity noise over a large frequency range (1 Hz–1 MHz) of only 0.78% and 0.76% with and without gas, respectively. The phase noise integrated in the same frequency range amounts to 1.33 and 1.25 mrad at 17.35 MHz with and without gas, respectively. Both turbomolecular pumps were running at their maximum speeds during these measurements, and the nozzle's backing pressure during the

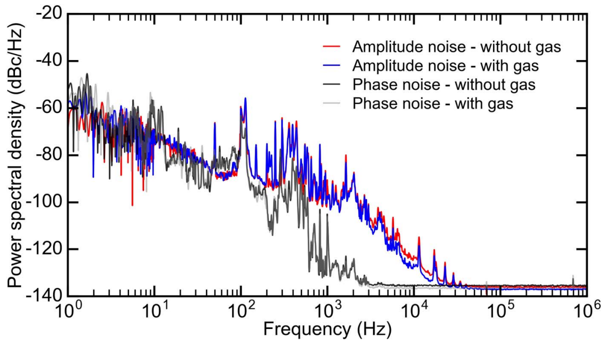

Fig. 4. Amplitude and phase noise measurements of the mode-locked TDL oscillator output in free-running operation with and without gas.

measurement with gas was 3.4 bar as during the XUV light spectra acquisition. Our laser noise is comparable to the typical values of free-running ultrafast TDL oscillators [11,30]. We therefore expect that CEO stabilization of a TDL oscillator can also be achieved with an intracavity HHG process.

In conclusion, we have reported the first intracavity HHG inside a mode-locked TDL oscillator, generating XUV light down to a wavelength of 60.8 nm (17th harmonic, 20.4 eV) at a repetition rate of 17.35 MHz. The conversion efficiency and flux of this proof-of-principle experiment are limited by the long pulse duration and the moderate peak power. However, TDL oscillators are power-scalable and have already been operated at 10 times higher intracavity peak power [31]. Moreover, we recently generated 35 fs pulses with 73 MW intracavity peak power from a Yb:Lu_2O_3 Kerr lens mode-locked TDL oscillator operating in air [23]. The corresponding optical spectrum was three times larger than the gain spectrum. We expect that by using materials with a broader gain bandwidth such as Yb:CALGO [22], substantially shorter pulse durations should be within reach of ultrafast TDL oscillators. Therefore, further optimization of laser parameters to reach performances similar to state-of-the-art HHG systems at the megahertz repetition rate [8,16] appears feasible. In combination with phase-matching optimization of the HHG process [3,32], this should significantly increase the conversion efficiency and allow for generating higher-energy photons. Furthermore, more efficient extraction schemes [33–35] should significantly increase the XUV flux available in future experiments. Our approach of HHG inside a TDL oscillator can lead to a novel class of coherent XUV light sources, which combines efficient megahertz repetition rate operation at a high XUV flux with a compact and portable design. Such systems will be highly attractive for driving a large number of applications ranging from high-resolution imaging to XUV spectroscopy and attosecond science.

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