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Intra-oscillator high harmonic generation in a thin-disk laser operating in the 100-fs regime

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Abstract: We demonstrate that Kerr lens modelocking is well-suited for operating an ultrafast thin-disk laser with intra-oscillator high harmonic generation (HHG) in the 100-fs pulse duration regime. Exploiting nearly the full emission bandwidth of the gain material Yb:YAG, we generate 105-fs pulses with an intracavity peak power of 365 MW and an intracavity average power of 470 W. We drive HHG in argon with a peak intensity of ~$7 \cdot 10^{13}$ W/cm^2 at a repetition rate of 11 MHz. Extreme-ultraviolet (XUV) light is generated up to the 31st harmonic order (H31) at 37 eV, with an average power of ~0.4 μW in H25 at 30 eV. This work presents a considerable increase in performance of XUV sources based on intra-oscillator HHG and confirms that this approach is a promising technology for simple and portable XUV sources at MHz repetition rates.

1. Introduction

High harmonic generation (HHG) driven by ultrafast laser pulses in a noble gas target is the most common method for tabletop coherent extreme-ultraviolet (XUV) light sources. Driving this highly nonlinear process requires peak intensities in the range of 10^{13} - 10^{15} W/cm^2. Such high intensities are traditionally achieved using chirped pulse amplifier (CPA) systems based on Ti:sapphire bulk lasers. Due to thermal effects in the bulk gain material, these systems are limited in average power and typically operate with low kilohertz repetition rates in a single-pass configuration. The progress towards megahertz repetition rate XUV sources is of high scientific and technical interest, because it can strongly reduce acquisition times in imaging and pump-probe experiments, overcome limiting space-charge effects in photoelectron spectroscopy [1–4], and enable XUV frequency combs for spectroscopy applications [5,6].

Compared to Ti:sapphire based systems, Yb-based solid-state laser systems are much better suited for reaching the high pulse energies necessary for HHG combined with high repetition rates due to the lower quantum defect, the therefore lower thermal effects, and the more efficient direct diode pumping scheme. High-power fiber CPA systems have been achieving considerable success in this domain [7–9]. In 2015, an average power level of 50 μW in a single harmonic order at 28 eV and 10.7-MHz repetition rate was demonstrated [8]. This performance was reached with a driving laser that was based on the coherent combination of fiber chirped pulse amplification channels followed by a nonlinear temporal pulse compression in a gas filled photonic crystal fiber.

Another successful approach for HHG at megahertz repetition rates is based on placing the HHG gas target inside a passive femtosecond enhancement cavity (fsEC) [6,10], where the pulse enhancement provides sufficiently high intracavity peak power, even when operating at repetition rates of several hundred-megahertz. Because of the resonant enhancement, the power requirements of the driving laser are considerably reduced. Using this technology, XUV photon energies up to 94 eV with 1.3-nW average power in a single harmonic order at 250-MHz repetition

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rate were demonstrated [11]. A record XUV average power of ~2 mW in a single harmonic at 13 eV and 77-MHz repetition rate was reported in 2018 [12]. However, the experimental realization of fsECs is challenging: coherent coupling of ultrafast pulses into a high-finesse optical resonator containing the HHG process is very demanding. For example, the system presented in [12] operated at an enhancement factor of ~200. In order to reach such enhancement factors, total cavity roundtrip losses have to be lower than 1%, making fsECs very sensitive to any kind of losses. This presents a constraint for the achievable enhancement and the implementation of efficient XUV output coupling methods, which often add further losses.

The high peak and average powers required for efficient HHG are also achievable inside ultrafast mode-locked thin-disk laser (TDL) oscillators, opening the potential for single-stage XUV sources. Placing the gas target directly inside the cavity of a TDL oscillator simplifies the overall experimental setup, requiring neither coherent coupling into an actively stabilized external fsEC nor temporal pulse compression. The available gain per cavity roundtrip strongly reduces the sensitivity to cavity losses compared to fsECs. In high-power TDL oscillators, the gain can be further increased with the number of passes over the disk. For instance, with a double pass on the disk, the roundtrip gain can compensate for output coupling rates of about 15%, while still exploiting the full available gain bandwidth [13,14]. This allows for the implementation of efficient XUV output coupling mechanisms that introduce significant losses for the circulating intracavity pulse, such as a pierced mirror with an increased diameter of the through hole [15,16].

In a proof-of-principle experiment in 2017, we demonstrated intra-oscillator HHG in xenon, driven inside a 255-fs semiconductor saturable absorber mirror (SESAM) mode-locked Yb:Lu_2O_3 TDL [17]. We generated XUV radiation up to the 17th harmonic (H17) at 20 eV with an average power of ~0.55 nW in H11 at 13 eV. In the same year, another demonstration of intra-oscillator HHG based on a Kerr lens mode-locked (KLM) Yb:YAG TDL was reported in [18] and recently published in more detail in [19]. The system operated with a pulse duration of 610 fs at a peak power of 445 MW and a repetition rate of 3.1 MHz. XUV light was generated in neon up to 52 eV (H43) and in argon with an average power of 47 nW in H17 at 20 eV.

In this work, we present intra-oscillator HHG at significantly improved performance. We use a 105-fs KLM Yb:YAG TDL operating at 365 MW of intracavity peak power and 11-MHz repetition rate. We generate XUV in argon with up to 37 eV (H31) with an average power of ~0.4 μW in H25 at 30 eV. Figure 1 shows a comparison of our system to other ultrafast high intracavity peak power TDLs together with the evolution of the intra-oscillator HHG driving systems. The desired parameter range for efficient HHG is depicted by the green shaded corner, corresponding to gigawatt level peak powers and tens of femtosecond pulse duration [4,25].

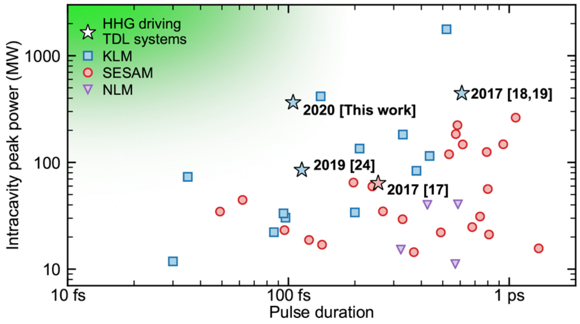

Fig. 1. Overview of ultrafast TDL oscillators based on SESAM, KLM, or nonlinear-mirror (NLM) modelocking with more than 10 MW of intracavity peak power. Stars with year show the evolution of HHG driving TDLs. The modelocking mechanism of HHG TDLs is indicated in the color code (blue for KLM; red for SESAM). References in [14,17–24].

However, a tradeoff between intracavity peak power and pulse duration for TDL oscillators utilizing different modelocking techniques can clearly be identified. The nearly instantaneous response of the Kerr effect and the high damage threshold of the components currently favor KLM for the combination of short pulse durations and high intracavity peak powers, making this approach most promising for intra-oscillator HHG.

2. Experimental setup

The TDL setup for intra-oscillator HHG is housed in a vacuum chamber with a footprint of $0.8 \times 1.6\ \mathrm{m}^2$ as shown in Fig. 2. The cavity design is based on the power-scaling approach for KLM TDLs presented in [26]. The laser is built using a commercially available TDL head designed for 36 passes of the pump through a ~100-μm thick Yb:YAG disk in order to achieve a high pump absorption. The disk is optically pumped at 969 nm with a fiber-coupled pump-diode on a 4.2-mm diameter pump spot. The diode delivers an average power up to 2 kW, but in the here described experiments, we only used a maximum pump power of 380 W. The cavity is folded twice over the disk, increasing the gain per cavity round trip. For KLM, an anti-reflection coated undoped 1-mm thick YAG plate is placed in the vicinity of an intracavity focus created by two concave mirrors (CM1, CM2) with 1-m radius of curvature (RoC). A water-cooled copper plate with a 3.6-mm diameter hole serves as hard aperture. Four dispersive mirrors introduce a total cavity roundtrip dispersion of -7000 fs^2. Mode-locked operation in vacuum is initialized by shaking one of the cavity mirrors mounted on a piezo stage. The cavity is extended by a telescope consisting of two concave mirrors (CM3, CM4) with RoC of 2 m and 3 m, respectively. One cavity end mirror serves as an output coupler with a transmission of 0.77%.

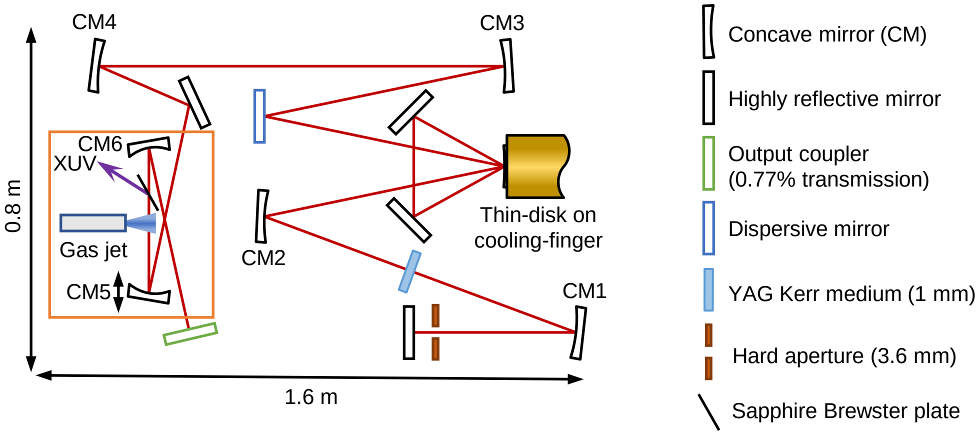

Fig. 2. Schematic of the Kerr lens mode-locked Yb:YAG thin-disk laser with a double pass on the disk and $4f$-extension with tight-focus for HHG, which is indicated with an orange box. CM1 and CM2, RoC of 1 m; CM3, RoC of 2 m; CM4, RoC of 3 m; CM5 and CM6, RoC of 200 mm.

A $4f$-extension consisting of two concave mirrors (CM5, CM6) with 200-mm RoC inserted in the output-coupling arm of the cavity creates a tight focus required to reach the high peak intensities for HHG. CM5 is placed on a piezo stage with a total travel range of 500 μm for fine-tuning of the cavity during mode-locked operation. Argon with a backing pressure of up to 10 bar can be injected into the cavity through a 50-μm opening diameter glass nozzle which is placed in the vicinity of the tight focus. A mass flow controller with a pressure sensor enables a precise delivery of the generation gas. A gas dump connected to the primary vacuum pump, placed on the opposite side of the tight focus than the nozzle, evacuates most of the gas, reducing the gas load on the turbomolecular pumps [27]. The gas nozzle is mounted on a motorized xyz-stage for fine-adjustment of the XUV generation point. To extract the generated XUV light and to ensure p-polarized operation of the TDL, a 325-µm thick sapphire plate is placed under Brewster's angle for the fundamental laser wavelength at a distance of ~15 mm from the tight focus. Due to the simplicity of the aforementioned XUV out-coupling mechanism, the method has been routinely used in fsECs [28–30]. However, the maximum reflectivity of the sapphire plate is limited to 17% at a wavelength of 50 nm (25 eV) and decreases strongly for shorter and longer wavelengths (see, e.g., [31]). The generated XUV light is directed by an unprotected gold mirror towards a spectrometer (248/310 McPherson). The XUV flux is measured with an aluminum coated AXUV100Al photodiode. To suppress any residual infrared light from reaching the photodiode, an additional 200-nm thick aluminum filter is inserted in front.

Two turbomolecular pumps provide a vacuum level which prevents significant reabsorption of the generated XUV light within the distance of ~20 cm separating the detector from the generation point. The cavity components experiencing the highest intensities, the anti-reflection coated YAG plate and sapphire plate, are purged with oxygen from both sides to prevent contamination during laser operation. Optical coatings have been designed inhouse and grown in our ion-beam sputtering coating facility.

3. Experimental results

Without injection of gas into the tight focus, our laser runs with an intracavity average power of 640 W, an intracavity peak power of 520 MW, and an intracavity pulse energy of 55 µJ at a pump power of 285 W. This performance is reached after optimizing the distance between CM5 and CM6 with the piezo stage in mode-locked operation. Intensity autocorrelation trace, optical spectrum, and radiofrequency spectrum are shown in Figs. 3(a) to 3(c). The optical spectrum of the 99-fs soliton pulse is centered at 1027.8 nm with a full width at half maximum bandwidth (FWHM) of 14.0 nm. We confirmed single pulse operation by a 180-ps scan in the autocorrelator and by observing the pulse train with an 18.5-ps-rise-time photodetector on a 40-GHz sampling oscilloscope.

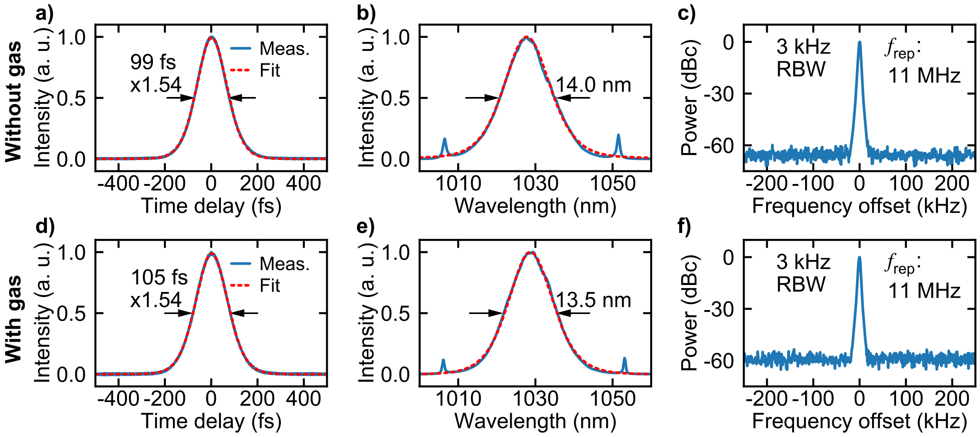

Fig. 3. Comparison of the thin-disk laser output parameters a) to c) without the injection of gas and d) to f) with injection of argon gas into the intracavity focus for HHG. a), d) Intensity autocorrelation traces with sech^2 fit. b), e) Optical spectra with spectral bandwidth and sech^2 fit. c), f) Radiofrequency spectra of the fundamental repetition rate (f_{rep}) with 3-kHz resolution bandwidth (RBW).

To initialize XUV generation, argon is injected into the tight focus of the laser cavity. In the current system, the gas plasma affects the laser: the position of CM5 has to be slightly shifted (< 500 µm) between operation with and without gas. We attribute this behavior to the lensing effect in the plasma since it can be compensated by this shift. With HHG, the laser operates with 105-fs pulse duration at an intracavity peak power of 365 MW and a pulse energy of 40 µJ. The intracavity average power is 470 W, which is obtained for a diode pump power of 180 W. Intensity autocorrelation trace, optical spectrum, and radiofrequency spectrum are shown in Figs. 3(d) to 3(f). Compared to operation without HHG, the optical spectrum shifts by 1 nm to a central wavelength of 1028.8 nm with a FWHM bandwidth of 13.5 nm.

The XUV optical spectrum attenuated by a 200-nm thick aluminum filter and generated in argon with a backing pressure of 3 bar is shown in Fig. 4. We generate up to H31 which corresponds to a wavelength of 33 nm and an energy of 37 eV. Utilizing the harmonic cut-off formula, the intracavity peak intensity in the tight focus is estimated to be $7{\cdot}10^{13}$ W/cm^2, corresponding to a focal radius of ~18 µm in mode-locked operation. A total generated XUV power of ~2 µW within the spectral range from H17 to H31 (20 - 37 eV) is conservatively estimated. We corrected by the tabulated values for the extraction efficiency of a sapphire plate (see, e.g. [31]), the reflectance of a gold mirror, and the transmission of a 200-nm thick aluminum filter. The absorption of XUV radiation in the gas background (oxygen: ~$9{\cdot}10^{-3}$ mbar; argon: ~$2.5{\cdot}10^{-3}$ mbar) to the detector is considerably small and therefore neglected. Assuming a flat spectral response of the XUV spectrometer, a generated average power of ~0.4 µW in H25 (30 eV) is estimated.

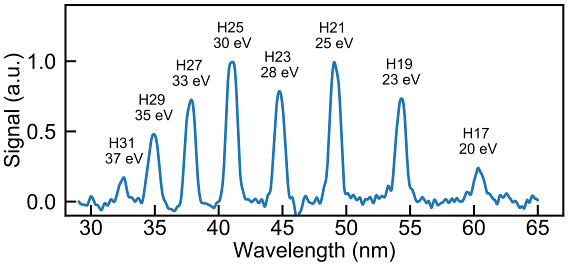

Fig. 4. Optical spectrum of XUV light generated in argon showing 17th to 31st harmonic. Peaks are labeled with harmonic order and corresponding photon energies. The XUV spectrum is attenuated by a 200-nm thick aluminum filter.

In the current study, thermal drift of uncooled cavity components limited operation to less than 15 minutes, which prevented a systematic optimization, especially with respect to the phase matching of the harmonics. Phase matching becomes increasingly difficult at repetition rates above ~10 MHz, due to a steady-state plasma accumulated in the generation volume [12]. Nevertheless, phase matching at even higher repetition rates has already been demonstrated, for instance, in single pass configuration at 10.7 MHz [8] and in a fsEC at 77 MHz [12]. After implementation of water-cooled mirror mounts in our cavity, which is currently in progress, we will investigate phase matching in detail. For this study, we plan to use an improved gas target similar to the system reported in [32].

4. Conclusion and outlook

We have demonstrated intra-oscillator HHG in a 105-fs KLM Yb:YAG TDL operating at 11-MHz repetition rate. In comparison to our previous result with XUV photon energies up to 20 eV (H17) and a total generated average power of ~0.55 nW in H11 (13 eV) in xenon [17], we now generate up to 37 eV (H31) with an average power of ~0.4 µW in H25 (30 eV) in argon, which is an increase in XUV flux of three orders of magnitude. Combined with the decreased pulse duration of the TDL by a factor of more than two, our result presents a considerable advance in TDL based intra-oscillator XUV sources [17,19].

Following the intracavity peak power scaling concept for KLM TDLs introduced by Brons *et al.* [26] in combination with shorter pulse durations by operating the laser in the strongly SPM-broadened regime [33,34], we expect further advancement of our system towards sub-100-fs operation with gigawatt intracavity peak powers. Due to the strong dependency of the HHG efficiency on peak intensity and pulse duration, this should lead to a substantial improvement of the performance of the system [4,25]. Further advances are expected by more efficient XUV extraction. Instead of a sapphire plate under Brewster's angle for the fundamental wavelength, a grazing incidence plate or a pierced mirror can be employed. With a grazing incidence plate, an XUV reflectivity up to 70% can be reached at 80° angle of incidence and fused silica as top layer of the anti-reflection coating for the fundamental laser-wavelength [31]. The pierced mirror method is especially suited for the efficient extraction of highly energetic XUV light [11,15]. In addition to an improved XUV extraction efficiency, a further reduction of the pulse duration of the driving TDL, reducing the level of steady-state plasma adversely affecting the TDL and improving phase matching of the generated harmonics, is also desirable [8,15].

We believe that our approach of HHG inside a TDL oscillator will lead to a novel class of single-stage coherent XUV light sources operating at MHz repetition rate. As most of the volume in our vacuum chamber remains empty, we expect that the footprint of our system can be strongly reduced with sufficient engineering, leading to a more compact and transportable design. We expect that such systems will soon operate at a performance comparable to state-of-the-art megahertz repetition rate HHG systems.

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Data availability. Data underlying the results presented in this paper are available in Ref. [35].

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