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Compact extreme ultraviolet source at megahertz pulse repetition rate with a low-noise ultrafast thin-disk laser oscillator

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We present a compact extreme ultraviolet (XUV) source based on high-harmonic generation (HHG) at 2.4 MHz pulse repetition rate driven from the compressed output of a mode-locked thin-disk laser (TDL) oscillator. The system generates very high peak intensities, which enable highly nonlinear frequency conversion reaching VUV/XUV energies. These sources significantly increase the signal-to-noise ratio and reduce measurement durations in many fields such as condensed matter physics. The pulse repetition rate is increased from kilohertz to megahertz with high average photon flux, while keeping the pulse energy sufficiently low to avoid space charge effects. The system uses a semiconductor saturable absorber mirror mode-locked Yb:YAG TDL delivering an average power of up to 70 W with subpicosecond pulses, which are efficiently compressed to sub-100 fs in a simple, single-stage compressor based on a Kagome-type hollow-core photonic crystal fiber. Focusing into a high-pressure xenon gas jet, we generate XUV radiation with up to $>5 \times 10^7$ photons/s on the 19th harmonic (23 eV). This HHG system is very compact, has low-noise performance comparable to standard ultrafast low-power laser oscillators, and provides a new tool for the study of attosecond dynamics in condensed matter physics.

OCIS codes: (320.0320) Ultrafast optics; (140.4050) Mode-locked lasers; (320.5520) Pulse compression; (060.5295) Photonic crystal fibers; (320.7090) Ultrafast lasers.

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1. INTRODUCTION

Current femtosecond and attosecond pulses in the vacuum ultraviolet (VUV) to extreme ultraviolet (XUV) spectral regime, generated using high-harmonic generation (HHG) [1], typically have energies of 1–10 nJ at around 1 kHz repetition rate with pulses as short as ≈70 attoseconds (i.e., 1 as $= 10^{-18}$ s) [2]. For many desired VUV/XUV measurements, one needs to increase the average photon flux rather than the pulse energy further [3]. This requires laser sources with proportionally higher repetition rates and a corresponding increase in average power. We have proposed to bridge this gap by the development of novel high-average-power, high-pulse-energy femtosecond laser oscillators [4] and for the first time applied them to high-field physics experiments with photoelectron imaging spectroscopy using only 1 μJ pulse energy from a diode-pumped, ultrafast, solid-state laser oscillator at 14 MHz pulse repetition rate [5]. In this Letter, we use more than one order of magnitude more pulse energy to demonstrate for the first time, to the best of our knowledge, the applicability of these high-power lasers for HHG at few-megahertz pulse repetition rate. One key benefit of this achievement to the extended research community is that this novel XUV source, based on a compact laser system, allows for measurements with significantly shorter acquisition times and much better signal-to-noise ratio due to the higher megahertz pulse repetition rate and the lower noise from laser oscillators compared to more complex multistage amplifier systems.

Such sources will make ultrafast VUV/XUV science available for a broad range of applications in time-resolved spectroscopy, frequency metrology, and high-spatial-resolution microscopy. For example, assuming the need for a 1000 by 1000 pixel CCD detector, where each pixel needs 1000 photons/s, leads to the requirement of $>10^9$ photons/s. At a wavelength of 54 nm (i.e., photon energy of ≈23 eV), this corresponds to an average power of only ≈4 nW, which scales linearly with wavelength. Furthermore, especially in attosecond condensed matter experiments [6,7], the pulse energy of low-repetition-rate systems very often has to be increased to a level where space-charge effects (i.e., electromagnetic forces between the generated charged particles within the interaction volume) start to blur the dynamics to be investigated—simply because the minimal detectable signal requires a certain average photon flux. Strongly correlated electron systems, with their complex physics, are one of the biggest challenges in condensed matter physics today. Thus, space-charge effects have been a serious limitation for time-dependent measurements in high-T_c superconductors in the VUV regime using angle-resolved photoemission spectroscopy [8]. Multi-megahertz repetition rate XUV sources would significantly help with these issues, as the average photon flux is distributed to orders of magnitude more pulses compared to systems with kilohertz pulse repetition rates. Nowadays, laboratory HHG expands the coherent electromagnetic spectrum from the infrared (IR) to the x-ray regime, generating photon energies from the VUV (≈10 eV) to the XUV (≈100 eV) and more recently even up to the soft x rays (≈1 keV) with, however, lower photon flux [9]. Efficient HHG poses stringent prerequisites on the driving sources: a demanding combination of high peak power (>100 MW) and short pulse duration (<100 fs) is required. Therefore, the main high-power IR lasers for driving HHG have remained Ti:Sapphire amplifier systems for a long time. These comparatively complex and costly laser systems, however, are limited in repetition rate to <10–20 kHz. In contrast, high-power, diode-pumped, Yb-doped laser systems scale much better in average power but at the expense of longer pulse durations. Thus high-flux, high-repetition-rate, compact XUV sources based on HHG have seen enormous progress in the past decade, closely linked to recent record average powers achieved with ultrafast IR driving sources based on Yb-doped, diode-pumped, chirped pulse fiber laser amplifiers [10], innoslab amplifiers [11], thin-disk multipass and regenerative amplifiers [12], and thin-disk oscillators [13,14]. Although these sources reach impressive levels, even surpassing the kilowatt average power milestone, the peak powers available still remain moderate compared to Ti:Sapphire amplifiers, due to the longer pulse durations limited by the standard Yb-doped gain media. So far, the most successful method to overcome this limitation has been cavity-enhanced HHG [15], where up to several 100s of μW per harmonic have already been demonstrated at multiple 10s of MHz repetition rate (see ▲ in Fig. 1). However, efficiently separating XUV and infrared (IR) photons while simultaneously creating only minor IR

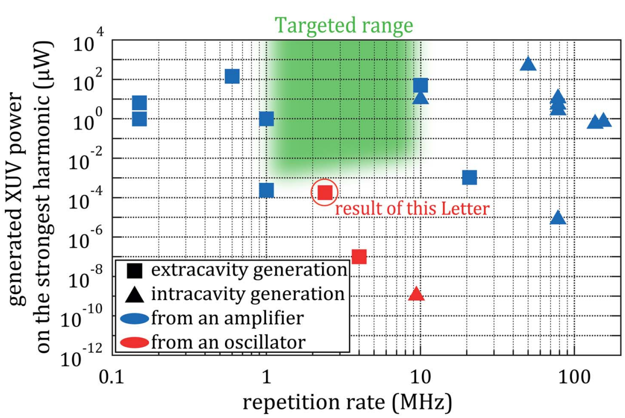


Fig. 1. Overview of the state of the art of XUV sources with repetition rates > 100 kHz. The generated power on the strongest harmonic is plotted versus the repetition rate of the source. Distinction can be made between intracavity generation, indicated by ▲(within an enhancement cavity or within a laser resonator), and extracavity generation consisting of a single pass into a gas jet, indicated by ■. In both cases, experiments can be driven by complex chains of amplifier systems (in blue) or with simple and compact oscillators (in red). The result presented in this Letter is highlighted in red and represents the highest XUV power generated directly from an oscillator, opening the way towards compact XUV sources in the near future. All references can be found in Supplement 1.

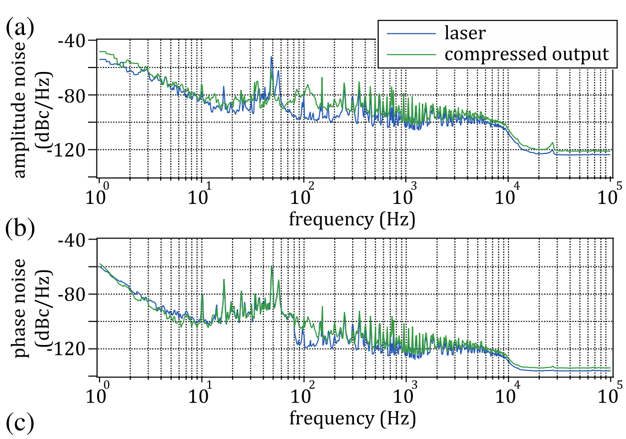
losses remains challenging. Recently, energies of up to 100 eV have been extracted thanks to pierced mirror designs [16]. Furthermore, this method of cavity-enhanced HHG is currently mainly focused on frequency metrology applications and only more recently on a few attosecond time dynamics studies, due to the difficulty of keeping the pulses short in the enhancement cavity, and the setups are complex and very alignment-sensitive. Other methods to enhance the efficiency of the HHG process have been tried such as HHG in waveguides [17,18], but so far the generated photon flux from these experiments at high repetition rates have remained insufficient for applications.

More recently, direct single-pass HHG driven by fiber chirped-pulse-amplification (CPA) systems has made great progress, showing that outstanding efficiencies can be reached even in the tight focusing regime [19,20], which is required for the moderate pulse energies available at these high repetition rates. In the most recent results, power levels of 100s of μW per harmonic (at 30 eV) with conversion efficiencies exceeding 10^{-6} have been demonstrated [21] (see Fig. 1). The driving system is still based on multistage amplifier systems, resulting in complex setups that are difficult to reduce in footprint. Furthermore, it still remains undemonstrated whether very-high-power CPA-based amplifier systems have sufficiently low noise properties for benefiting from the strongly reduced measurement times in sensitive detection schemes in many of the targeted experiments. Among the different technologies that reach sufficient average power and peak power for HHG, ultrafast diode-pumped, Yb-doped, thin-disk laser (TDL) oscillators [22] are particularly attractive: they provide similar performance to state-of-the-art amplifiers directly from more compact and low-noise tabletop oscillators. They also have the potential to stabilize the exact position of the electric field within the pulse envelope (i.e., the carrier envelope offset phase) [23] with very good performance [24], which is a crucial prerequisite for many of the targeted applications.

Here, we present the generation of high harmonics in a compact and simple setup directly driven by the compressed output of a mode-locked, TDL oscillator without further amplification. This result represents, to the best of our knowledge, the first demonstration of HHG directly driven by power-scalable, mode-locked thin-disk oscillators. Our driving source is a high-energy, mode-locked Yb:YAG TDL delivering pulses with a duration of 870 fs and pulse energy of 29 μJ. The oscillator operates at 2.4 MHz and at a center wavelength of 1030 nm. In order to reach sufficiently short pulses, we used a simple and efficient single-stage compression based on a gas-filled Kagome-type hollow-core photonic crystal fiber (HC-PCF) [25] providing pulses with 108 fs full-width at half-maximum (FWHM) with more than 70% of total efficiency. Focusing to a spot diameter of 22 μm ($1/e^2$) resulted in a peak intensity of $\approx 5.5 \times 10^{13}$ W/cm^2. The laser beam traversed through a xenon gas jet emitted by a 100 μm nozzle placed in the vicinity of the focus spot. With 1.5 bar of backing pressure, high harmonic radiation was generated up to the 25th harmonic (30 eV) with a maximum intensity on the 19th harmonic (23 eV) of $>5 \times 10^7$ photons/s, i.e., 0.18 nW. This average power flux is four orders of magnitude higher than in any previous HHG result driven by an oscillator [26,27]. The current source already provides sufficient flux for first coincidence detections or surface science experiments, for example. In addition, further improvement of this concept is expected in the near future by optimizing the pressure handling and by compressing the pulses further in order to further improve the phase-matching conditions [19]. We believe this result is the first step towards the next generation of compact XUV sources driven by tabletop oscillators, which would provide an outstanding low-noise, compact, and coherent XUV source.

2. SET-UP: LASER SOURCE AND PULSE COMPRESSION

Our oscillator is a diode-pumped Yb:YAG TDL using a semiconductor saturable absorber mirror for passive mode locking [28] with a similar design to the one which previously demonstrated the highest average power (up to 275 W) and pulse energy (up to 80 μJ) [22] compared to any ultrafast oscillator. For this experiment, we targeted more moderate average power levels for reliable pulse compression and first proof-of-principle demonstration in a high-power HHG beamline. The laser oscillator was set up in a medium vacuum environment, and up to 70 W of average power could be achieved with reliable daily operation. The pulses had a duration of 870 fs at a repetition rate of 2.4 MHz, corresponding to a pulse energy of 29 μJ and a peak power of 29 MW, and operated at a center wavelength of 1030 nm (more details of the laser oscillator can be found in Supplement 1). The noise of the laser was measured in free-running operation on the passively filtered fourth harmonic of the repetition rate using a state-of-the-art Signal Source Analyzer (Agilent, E5052, Fig. 2). Even though most of the noise of the laser output originates from the highly multimode (typically $M^2 > 200$) pump laser as described in detail in [24], we measured <0.60% [1 Hz–100 kHz] of amplitude noise, revealing that in free-running operation this laser does not exhibit more noise than any typical low-power, ultrafast, solid-state laser oscillator. Subsequent simple stabilization of



(c)

Frequence Range	Noise	Laser	Compressed Output
[1 Hz - 100 kHz]	Amplitude	0.57%	0.61%
	Phase	1.98 mrad	1.87 mrad
[100 Hz - 100 kHz]	Amplitude	0.25%	0.41%
	Phase	0.21 mrad	0.32 mrad

Fig. 2. Noise performance in free-running operation. The noise performance of the laser and of the compressed output was measured on the passively filtered fourth harmonic of the repetition rate using a state-of-the-art Signal Source Analyzer (SSA) (Agilent, E5052). (a) Amplitude and (b) phase noise power spectral density of the laser and of its compressed output. (c) Overview of the integrated amplitude and phase noise values (in %, respectively mrad).

the oscillator could reduce the noise level much further, potentially close to the quantum limit of our high-Q resonator.

Using this source for efficient HHG required additional external pulse compression to below 100 fs, as ultrafast diode-pumped, solid-state lasers with sufficient pulse energy at megahertz pulse repetition rates remain challenging in spite of many currently ongoing efforts [29]. We recently demonstrated that the most efficient method of external pulse compression compatible with a 100-W-class laser with peak powers in the range of 10s of MWs is Kagome-type HC-PCFs. This guiding mechanism [30,31] allows us to contain the laser mode into the fiber's hollow core with only minor overlap with the surrounding silica structure (<0.05%). With the latest development of Kagome fibers and the introduction of the hypocycloid core [32], such fibers have shown their outstanding potential for transmission of ultrashort pulses with high average powers [25] and mJ pulse energies [33]. Recently, we demonstrated for the first time a very efficient pulse compression scheme for a TDL to the sub-100 fs range in the 100 W average power level [25]. In the experiment presented here, we used a similar setup as in [25], with a 30 μm core diameter, hypocycloid-core, Kagome-type HC-PCF statically filled with 5 bar of krypton. At full launched power of 70 W, the spectrum was broadened and compressed to 108 fs FWHM using dispersive mirrors (more details of the compression setup are described in Supplement 1). We achieved an enhanced peak power of 105 MW for an average compressed power of 46 W, with a total footprint of the current setup (laser and compression stage) of only 160 × 120 cm. This could be potentially further reduced, for example, by including the compression setup in the vacuum chamber. Figure 2 demonstrates that the excellent low-noise beam properties provided by our oscillator output are maintained throughout the compression, as opposed to the findings in [34]. No significant additional noise was added by the compression despite its water cooling and its operation at 70 W of launched average power. The compressed output showed a remarkably low root-mean-square amplitude noise value of <0.60% [1 Hz–100 kHz] in free-running operation.

3. HIGH HARMONICS GENERATION

The compressed pulses were directly routed into a HHG chamber (see Fig. 3) and tightly focused with a lens to a spot diameter of 22 μm, leading to a calculated peak intensity of $\approx 5.5 \times 10^{13}\ \mathrm{W/cm^2}$. In the vicinity of the focal spot, which was placed in a subchamber with an ambient pressure of ≈1 mbar, we placed a gas nozzle with 100 μm hole diameter and 1.5 bar of xenon gas backing pressure. High harmonics were generated in the interaction region at an estimated gas pressure of 500 mbar. The XUV beam and the IR beam exit this subchamber through a 500 μm hole placed 5–7 mm after the focus. Our two-chamber system design allows us to compensate for the very high gas load required in such an experiment by creating a differential pumping stage between the subchamber and the main chamber. This allows the latter to stay at ambient pressure $<10^{-4}$ mbar for gas nozzle backing pressures of up to 10 bar. The beams were further routed by successive reflections on a fused-silica plate and a gold mirror used with an incidence angle of 56°. The IR beam was transmitted with more than 99% efficiency through the fused-silica plate onto a beam dump whereas around 5% of the XUV light was reflected, creating the necessary separation of XUV and high-average-power IR beam.

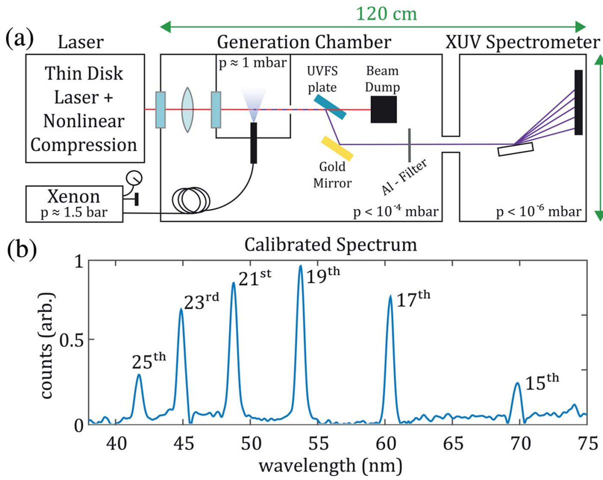


Fig. 3. Experimental setup of our high-harmonic generation (HHG) using the compressed output of a SESAM mode-locked, diode-pumped Yb:YAG TDL oscillator. (a) In the generation chamber, the compressed pulses are focused to a 22 μm spot diameter to reach an intensity of $\approx 5.5 \times 10^{13}$ W/cm^2. The XUV light generated is separated from the infrared light first via reflection on a fused silica plate at Brewster's angle and then at much less average IR power with an 400 nm Aluminum (Al) filter. (b) The XUV spectrometer (McPherson 248/310G) enables us to measure the spectrum of the XUV down to the 25th harmonic (30 eV). After calibration, we estimate an extraction from the subchamber of at least $\approx 5 \times 10^7$ photons/s on the 19th harmonic (23 eV), i.e., 0.18 nW.

After passing through a 400 nm thick aluminum (Al) filter, which blocked the residual IR light, the generated high harmonics were sent into an XUV spectrometer (McPherson 248/310G). Within the transmission band of the Al filter (18–80 nm), a distinct signal of several odd harmonics (15th to 25th) was detected. Based on the observed high harmonic cutoff frequency (25th harmonic), we confirmed that our intensity at the focus is close to 6×10^{13} W/cm^2 by calculations with the cutoff formula [35]. We used a calibrated electron multiplier (Photonis Magnum 5900) placed after the Al filter to estimate the total XUV flux extracted from the subchamber. Taking only into account the losses due to reflections on the silica plate and gold mirror, and due to transmission through the Al filter, we estimate that we have extracted at least $\approx 5 \times 10^7$ photons/s on the 19th harmonic (54 nm–23 eV), i.e, 0.18 nW from the subchamber. The total number of photons produced within the Al filter band corresponds to 3×10^8 photons/s (≈ 0.7 nW) and an efficiency of $\approx 1.5 \times 10^{-11}$, which is already an interesting flux to perform first surface science experiments. This is, to the best of our knowledge, the highest XUV flux ever produced by an oscillator-driven HHG source (see Fig. 1). We believe that, by further optimization, our achieved average photon flux can be significantly improved. Taking into account the propagation of the XUV light in the vicinity of the gas nozzle through a smeared out pressure gradient of approximately 500 mbar to a few mbar over a distance of a few mm, we believe that strong reabsorption occurs (resulting in an estimated transmission of $< 10^{-1}$–10^{-2}). We are therefore confident that using more directive gas nozzle designs and more efficient pumping schemes will allow us to increase the measured XUV extraction by several orders of magnitude in the near future. Furthermore, we are currently working on reducing the pulse duration and increasing the average power in the external pulse compressor, which should significantly increase the HHG efficiency [19].

4. CONCLUSION

In conclusion we have demonstrated, for the first time to our knowledge, HHG driven by the compressed output of a passively mode-locked, diode-pumped, TDL oscillator. By focusing the compressed pulses of our laser system (108 fs, 2.4 MHz) into a high-pressure xenon gas jet with peak intensities of $\approx 5.5 \times 10^{13}$ W/cm^2, high harmonic radiation reaching up to $> 5 \times 10^7$ photons/s on the 19th harmonic (23 eV) is generated. This proof-of-principle demonstration opens the door to compact oscillator-driven sources of XUV radiation at high repetition rates with excellent noise properties. Further ongoing steps to increase the flux of our novel source include better pressure handling and pulse duration reduction at even higher average power, in principle available from Yb:YAG thin-disk laserss. In the meantime, we believe the demonstrated source already provides an interesting starting point for a broad range of applications in which high-repetition-rate, low-noise XUV pulses are crucial, including ultrafast photoemission studies on solid surfaces or coincidence detection schemes in atomic and molecular physics.

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See Supplement 1 for supporting content.

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Compact XUV source at megahertz pulse repetition rate with a low-noise ultrafast thin-disk laser oscillator: supplementary material

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This document provides supplementary information to "Compact XUV source at megahertz pulse repetition rate with a low-noise ultrafast thin-disk laser oscillator", http://dx.doi.org/10.1364/optica.2.000980. We first describe in detail all the references used in the overview of the state-of-the-art of XUV sources with repetition rates > 100 kHz. In a second part, more details on the infrared laser source and compression set-up used in this work is given.

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1. Overview of XUV sources at high repetition rate

The photon energy and the reference of each point of the graph presented in Fig. 1 can be found in Fig. 1S.

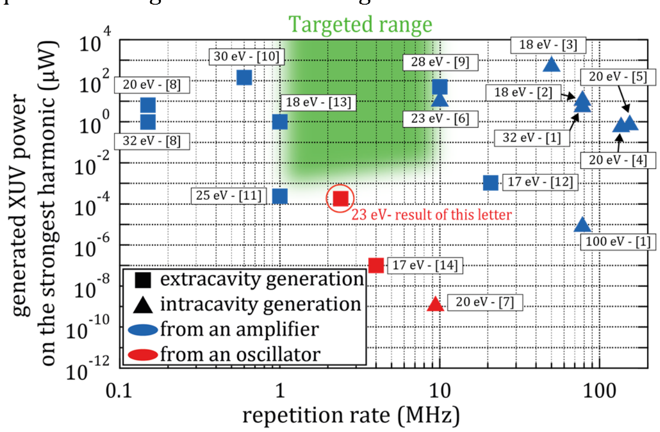

Fig. S1. Overview of the state-of-the art of XUV sources with repetition rates > 100 kHz with the related reference and photon energy. The generated power on the strongest harmonic order is plotted versus the repetition rate of the source. Distinction can be made between intracavity generation, indicated by ▲ (within an enhancement cavity [1-6] or within a laser resonator [7]), and extracavity generation consisting of a single-pass into a gas jet, indicated by ■ [8-14]. In both cases, experiments can be driven by complex multi-stage amplifier systems (in blue) or with simple and compact laser oscillators (in red).

The photon energy of the associated strongest generated harmonics is indicated on the graph. The result presented in this letter is highlighted in red and represents the highest XUV power generated directly from an oscillator, opening the way towards compact XUV sources in the near future.

2. Laser driving source and compression

Figure 2S shows the set-up of the SESAM-modelocked diode-pumped Yb:YAG thin disk laser oscillator and the single-pass Kagome hollow-core PCF compression stage. The laser cavity was built in a vacuum chamber operating at ≈ 100 mbar of Helium. Operating in a medium pressure Helium environment is beneficial to reduce the nonlinearity of the intracavity atmosphere in order to achieve stable soliton modelocking at high energy [15], but also relaxes thermal load on intracavity components compared to using lower air pressure. The use of a Herriott-type multipass cell (MPC) cavity extension inside the oscillator allowed us to reach an oscillator length of 63 m in a compact layout [16] (corresponding to a repetition rate of 2.4 MHz), while stable modelocking was started and stabilized by a high damage threshold SESAM [17] used as an end mirror in the laser cavity. Dispersive mirrors provided a total negative dispersion per roundtrip of ≈ -36’000 fs^2 to compensate for the nonlinear phase-shift of 150 mrad within the cavity and achieve soliton pulse formation [18]. In this configuration, we achieved up to 70 W of average power with reliable daily operation. The pulses had a duration of 870 fs at a repetition rate of 2.4 MHz, corresponding to a pulse energy of 29 µJ and a peak power of 29 MW, and operated at a center wavelength of 1030 nm.

The compression set-up was similar as in Ref. [19]. The fiber was a 7-cell 3-ring hypocycloid-core Kagome-type HC-PCF with a mode-field diameter of ≈ 30 μm and an extremely low loss figure of 180 dB/km at 1030 nm. The 67-cm fiber was embedded in water-cooled gas cells in which 5 bar of Krypton were statically applied in order to generate sufficient spectral broadening in the anomalous dispersion regime with only -5.4 fs^2/cm. At full launched power of 70 W, the spectrum was broadened from its initial 1.2 nm FWHM to more than 28 nm FWHM (see Fig . 2Sb). Using 22 bounces on dispersive mirrors, accounting for a total dispersion of -12 000 fs^2, the input pulses of 870 fs duration were compressed to a FWHM of 108 fs (Fig. 2Sc). Taking into account the pulse profile in the time domain, we estimate a percentage of 60% of the energy to be located in the main compressed peak, leading to an enhanced peak power of 105 MW for an average compressed power of 46 W.

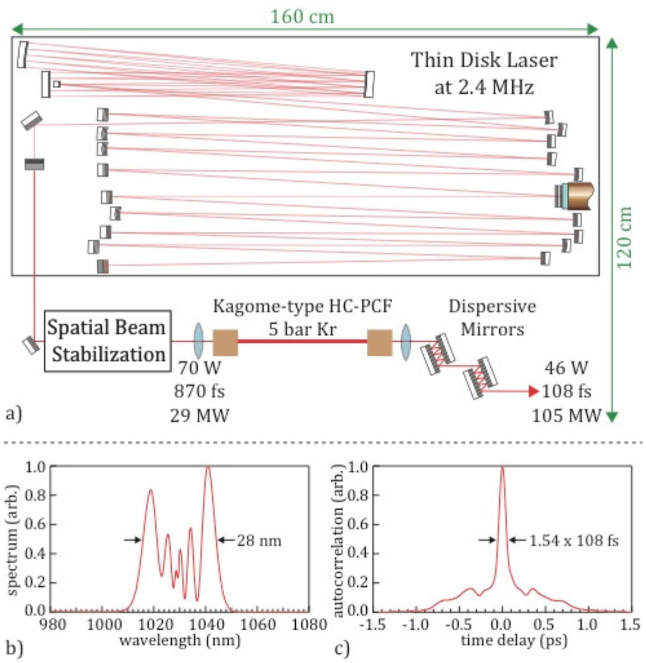

Fig. S2. Laser and nonlinear compression set-up used in our experiments. a) The Yb:YAG SESAM-modelocked thin disk laser was set up to deliver 70 W with 870 fs pulse duration at 2.4 MHz repetition rate, i.e. 29 μJ of pulse energy [15]. Subsequently, nonlinear pulse compression using a 67 cm long Kagome fiber with a mode-field diameter of 30 μm, filled statically with 5 bar of Krypton, leads to compressed pulse (b and c) reaching 108 fs with 60% of energy in the main peak, corresponding to 105 MW of peak power [19].

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