



High-power few-cycle Cr:ZnSe mid-infrared source for attosecond soft x-ray physics

VYACHESLAV E. LESHCHENKO,^{1,*}  BRADFORD K. TALBERT,¹ YU HANG LAI,¹ SHA LI,¹ YAGUO TANG,¹ STEPHEN J. HAGEMAN,¹ GREG SMITH,¹ PIERRE AGOSTINI,¹ LOUIS F. DIMAURO,¹ AND COSMIN I. BLAGA^{1,2}

¹Department of Physics, The Ohio State University, Columbus, Ohio 43210, USA

²Department of Physics, Kansas State University, Manhattan, Kansas 66506, USA

*Corresponding author: leshchenko.1@osu.edu

Received 30 March 2020; revised 25 May 2020; accepted 3 June 2020 (Doc. ID 393377); published 10 August 2020

The development of mid-infrared, high-energy pulses of extremely short pulse duration is of great importance to the strong-field physics community, as it enables extension of the high-harmonic cutoff towards the keV range of photon energies. Here we demonstrate a Cr²⁺:ZnSe laser amplifier delivering 7 mJ, 100 fs pulses at 1 kHz repetition rate and 2.4 μm central wavelength with excellent energy stability and beam quality. These pulses are post-compressed to 39 fs, 6.2 mJ, and 115 GW peak power in a novel nonlinear compression scheme. The laser system was used to generate coherent soft x-ray radiation up to 0.6 keV covering the entire water window, which is of high importance for bio-relevant applications. The obtained results open a wide range of applications in nonlinear optics, attosecond and strong-field physics, and remote sensing. © 2020 Optical Society of America under the terms of the [OSA Open Access Publishing Agreement](#)

<https://doi.org/10.1364/OPTICA.393377>

1. INTRODUCTION

Powerful femtosecond sources in the 2–10 μm spectral range are of high demand for multiple applications in strong-field and attosecond physics [1–4], molecular spectroscopy [5], medicine [6], free-space communication [7], remote sensing [8], and many other fields. There are two primary approaches to generate femtosecond optical radiation in this spectral range: 1) nonlinear frequency down-conversion of visible and near-infrared (NIR) pulses, and 2) direct lasing and amplification. Thus far, most of the applications have relied on different nonlinear frequency conversion techniques such as optical parametric amplification (OPA)/optical parametric chirped pulse amplification (OPCPA) [9–13], difference frequency generation (DFG) [14,15], which is a variation of OPA, and their cascaded variants [16]. The main drawback of these processes is the low conversion efficiency of ~ 0.01 –10% and relatively high complexity and cost of a system.

Cr- and Fe-doped chalcogenides have been long recognized as promising materials for the direct lasing and amplification in the 2–3 μm and 3–7 μm spectral ranges, correspondingly; however, they have been out of practical consideration for a long time due to the low maturity of the technology. Nonetheless, recent progress in the growth of Cr²⁺:ZnSe and in the development of powerful nanosecond pump sources in the Cr²⁺ absorption spectral range (1.6–2.1 μm) [17,18] enables the implementation of multi-Watt, multi-mJ laser systems. A detailed review of recent advances in

this field can be found elsewhere [17]. Recent highlights in the domain of ultrafast high-energy amplifiers include: 1) a Cr:ZnSe amplifier with 2.3 mJ, 88 fs, and 26 GW pulse duration at 1 kHz seeded with a DFG source [19], which represents the previous result with highest energy and peak power; 2) a Cr:ZnSe-only chirped pulse amplification system delivering 1 mJ, 184 fs, 5 GW pulses at 1 kHz [20]; and 3) a Fe:ZnSe amplifier with 3.5 mJ, 150 fs pulses centered at 4.4 μm [21], which is an important milestone for Fe:ZnSe technology. In the present work, we not only demonstrate the generation of pulses with higher energy (6.2–7 mJ), shorter pulse duration (39 fs), and significantly higher peak power of 115 GW at the same repetition rate of 1 kHz, but we also present a simple but powerful and scalable approach for nonlinear compression of amplified pulses to a few-cycle duration. In addition, the generation of soft x-ray harmonics in the entire water window is demonstrated, which is the first result of this kind using a direct output of a laser amplifier.

Here we report on a powerful and efficient alternative to the down-conversion sources in the mid-IR spectral range. It is a Cr:ZnSe laser system delivering 7 mJ, 101 fs pulses at 2.4 μm and 1 kHz repetition rate. The generated pulses are post-compressed to 39 fs, 6.2 mJ in a novel scalable nonlinear compression setup. In addition, we demonstrate the efficacy of Cr:ZnSe as a future solid-state, mid-IR laser driver of high-order harmonic sources operating in the water window soft x-ray range.

2. EXPERIMENTAL SETUP

The schematic layout of the experimental setup is shown in Fig. 1. The seed for the Cr:ZnSe chirped pulse amplifier (CPA) is generated in a homebuilt OPA based on the standard design with collinear amplification in beta barium borate (BBO) nonlinear crystals [22] and pumped with a homebuilt Ti:sapphire CPA system providing 80 fs, 4 mJ pulses at 1 kHz repetition rate. The signal for the first OPA stage is generated via white light continuum generation by focusing a small portion ($\sim 0.2\%$) of the OPA pump in a sapphire (Al_2O_3) window. The “idler” OPA output with spectrum centered at $\sim 2.4\ \mu\text{m}$ and pulse duration of 80 fs is stretched with a Martínez type stretcher [23] (600 lines/mm, 57° angle of incidence, $-30\ \text{cm}$ effective grating separation) to about 300 ps duration. The stretched pulses are amplified in a three-pass Cr:ZnSe amplifier. The amplified pulses are compressed in a Treacy type compressor [24] and sent to a nonlinear post-compression setup for further broadening and shortening.

The amplifier employs a Cr:ZnSe polycrystalline rectangular gain element (PGE-Cr-ZnSe, IPG Photonics Inc) with $48 \times 12 \times 5.9\ \text{mm}$ dimensions (length $\times$ width $\times$ height), which is anti-reflection (AR) coated for $1.9\text{--}3\ \mu\text{m}$ and has 95% single pass pump absorption at full pump power. A polycrystalline gain element is used because it is available in larger sizes and with higher and more uniform dopant concentrations than single crystals, while physical, spectroscopic, and laser characteristics of poly- and single crystals are almost identical [17]. Cr:ZnSe has a relatively large thermo-optic coefficient and corresponding thermal lensing effect [17]; therefore, a gain element with normal cut surfaces and an AR coating is used, since unlike a Brewster cut gain element, it is free of thermal induced astigmatism [25]. Moreover the AR coating of Cr:ZnSe has a damage threshold of about $1\ \text{J}/\text{cm}^2$, which is more than double the maximum fluence in the implemented setup. The properties of Cr:ZnSe have many similarities to the properties of Ti^{3+} :sapphire such as four-level energy structure, the spectral gain bandwidth supporting few-cycle pulses, about $5\ \mu\text{s}$ radiative lifetime, and high quantum efficiency at room temperature [17,26], which enables efficient operation of a Cr:ZnSe laser with a standard water cooled heatsink. Therefore, the crystal is water-cooled at 18°C .

The gain element is double-side-pumped with two Ho:YAG (Ho-doped yttrium aluminum garnet) Q-switched lasers (HLPN-2090-25-20-25, IPG Photonics Inc). Each pump delivers up to 17 mJ pulse energy at $2.09\ \mu\text{m}$ wavelength and 1 kHz repetition rate with about 30 ns pulse duration. Both pump beams have Gaussian profiles with 3.5 mm diameter on the e^{-2} intensity level in front of the Cr:ZnSe crystal. The corresponding peak fluence at maximum pump energy (17 mJ) is about $350\ \text{mJ}/\text{cm}^2$. The seed beam has a slightly larger diameter of about 3.8 mm in order to ensure good energy extraction efficiency. The angle between the two pump beams is set to about 0.6° ($\pm 0.3^\circ$ to the normal to the crystal surface), which is just enough for their reliable spatial separation on the 0.6 m distance between the crystal and the pump-in-coupling mirror (which corresponds to the 1.2 m overall size of the bow-tie setup). The angles between seed and pump beams were designed as small as possible in order to keep good overlap between them in a relatively long gain medium and to minimize spatial inhomogeneities caused by the spatial walk-off. The angle to the normal to the crystal surface is about 0.9° on the first and second passes and about 0.6° on the last pass. The smaller angle is chosen for the last pass in order to facilitate better energy extraction efficiency and higher beam quality. Between the passes, the beam is directed with flat multilayer dielectric mirrors (BBHR-5009CI-2.2-2.6) from Lambda Research Optics Inc. Note that the free-space propagation between passes in the implemented nonimaging setup acts as a low-pass spatial filter [27] removing potential small-scale inhomogeneities in the gain element transmission and/or doping concentration and improving the output beam quality. The diameter of the amplified beam on second and third passes stays close to the pump beam diameter, which indicates that the focal length of the thermal lens in the gain element is larger than $\sim 3\text{--}5\ \text{m}$ according to the analysis based on the ray transfer matrices formalism for Gaussian beams (see Supplement 1 for details). Note also that the Rayleigh length of the seed beam of about 4 m is larger than the entire beam path in the amplifier, which is 2 m from the first to the last crystal pass. Thus, at lower pump power and correspondingly smaller thermal lens, amplified beam size is also almost the same as the pump beam size through the entire propagation in the amplifier (see Supplement 1 for detailed analysis). Therefore, the implemented nonimaging setup

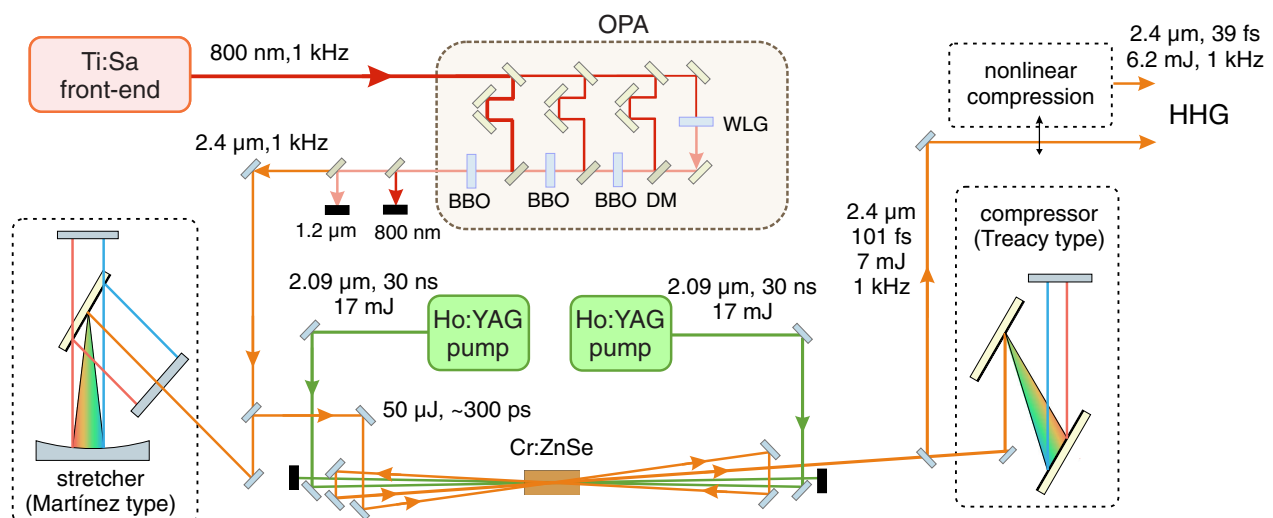


Fig. 1. Schematic layout of the experimental setup. Ti:Sa, titanium sapphire; OPA, optical parametric amplifier; BBO, beta barium borate; WLG, white-light generation; DM, dichroic mirror; HHG, high-harmonic generation.

is absolutely stable and safe to operate at any pump power available in the performed experiments. However, imaging between passes might be necessary at higher pump power or fluence when the thermal lens is stronger.

3. EXPERIMENTAL RESULTS

A. Amplification and Compression

The amplifier boosts the seed (stretched OPA output) energy from 60 μJ to 8.2 mJ (8.2 W), as measured with a water-cooled thermal power detector UP50N-50 H-W9 from GENTEC-EO. The power after first and second passes is 0.5 W and 3 W, respectively, so the small signal gain per pass calculated from the gain on the first pass is 10. The amplified spontaneous emission (ASE) has a negligible contribution to the output signal, as the detected power does not exceed the power meter detection limit of about 1 mW (here a more sensitive power meter UP19K was used) when the seed is blocked. Note that the estimated nonlinear phase (B-integral) accumulated in the amplifier is 0.7 (see Supplement 1 for evaluation details), which is low enough to prevent nonlinear distortions of amplified pulses. The dependence of the amplified pulse energy on the pump energy is shown in Fig. 2(a). In saturation, the amplifier has 34% slope efficiency and 25% optical-to-optical efficiency. Adding an extra (fourth) pass increases output energy and the conversion efficiency by a factor of 1.2 to 30% optical-to-optical efficiency; however, the beam quality degradation introduced due to thermal distortions in the gain element significantly affects the beam quality (M^2 degrades from ~ 1.2 to ~ 1.6), which causes problems with the following beam delivery and focusing. Therefore, the three-pass configuration is chosen as an optimum for the focused on-target pulse intensity. The achieved ~ 10 W output power is the maximum output power with practically acceptable beam quality that can be

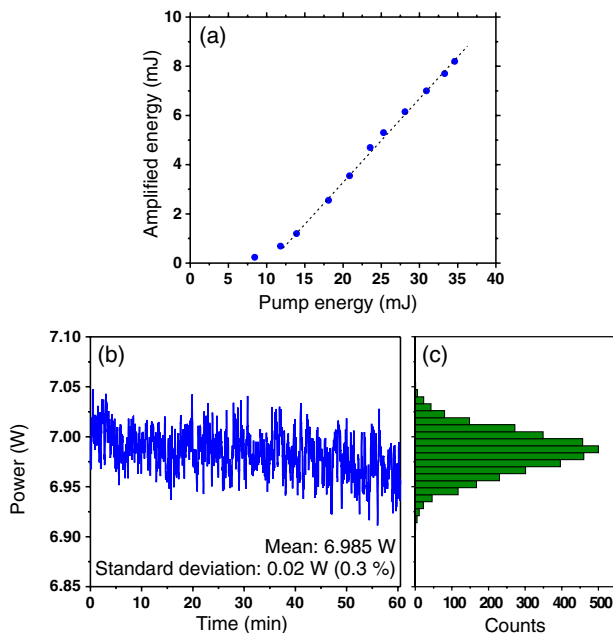


Fig. 2. Output power. (a) Dependence of the energy of amplified pulses (before the compressor) on the pump energy. The black dashed line corresponds to the linear fit in the region above the threshold and corresponds to the 34% slope efficiency. (b) Stability measurement of the output power after the compressor. (c) Histogram of the power measurement shown in (b).

obtained in a standard Cr:ZnSe gain element [28]. It is limited by thermo-optical properties (such as thermal conductivity, thermal expansion, and thermal lensing parameter) of Cr:ZnSe, which are worse compared to Ti^{3+} : sapphire [17,26], for example. Thermal distortions can be significantly reduced by scanning beams across a gain element, which can be realized with a spinning ring gain element [28] that enables >100 W output power. However, this approach is beyond the scope of this paper.

After amplification the beam is magnified with a reflective telescope (not shown in Fig. 1) to 18 mm in order to prevent damage of the gratings and nonlinear effects after the compressor. The amplified pulses are compressed in a Treacy type compressor with gold coated holographic diffraction gratings (Spectrogon, 600 lines/mm, 57° angle of incidence, 30 cm grating separation) to 101 fs as measured using a second-harmonic generation frequency-resolved optical gating (SHG-FROG) setup. It was noticed during FROG alignment that the instability of the SHG spectra integrated over 3 ms did not exceed 1%, which is a clear indication of better than 1% shot-to-shot energy instability at the fundamental wavelength. The measured and reconstructed FROG traces are presented in Fig. 3. The measured full width at half maximum (FWHM) pulse duration of 101 fs (12.6 optical cycles) is near the transform limited (TL) value of 86 fs (10.8 optical cycles). The residual spectral phase is caused mostly by the uncompensated fourth- and higher-order dispersion of the gain material, which cannot be compensated for in the present dispersion control scheme because the optimization of the angle of incidence and the gratings separation in the compressor allow only compensation of second and third dispersion orders. This conclusion is based on simulations of the dispersion in the stretcher–amplifier–compressor setup. Using the stretcher and compressor parameters specified above and the dispersion of ZnSe [29] with $48 \times 3 = 144$ mm of material length, the residual uncompensated fourth-order dispersion after the compressor is $5.8 \times 10^6 \text{ fs}^4$ when the compressor is perfectly compensating for the combined second- and third-order dispersion of the stretcher and ZnSe. It results in stretching a 86 fs pulse to 95 fs. Thus, the uncompensated part of the fourth-order dispersion of the gain

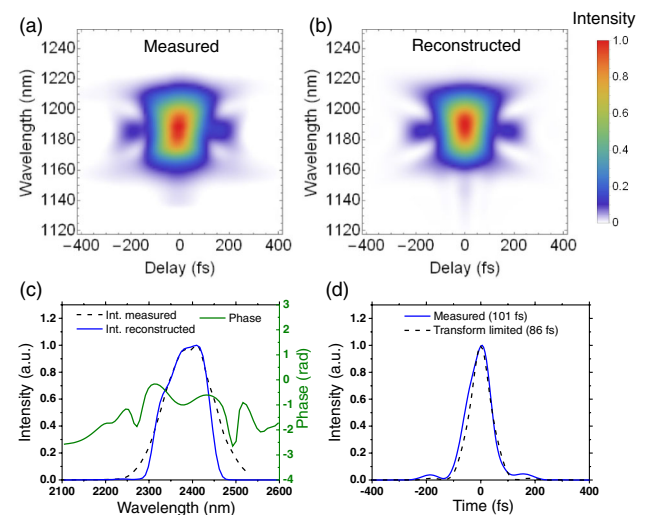


Fig. 3. Compression results. (a) Measured FROG trace. (b) Reconstructed FROG trace. The reconstruction error is 0.003. (c) Measured and reconstructed spectra and spectral phase. “Int.” in the legend stands for intensity. (d) Temporal intensity profile of the pulse.

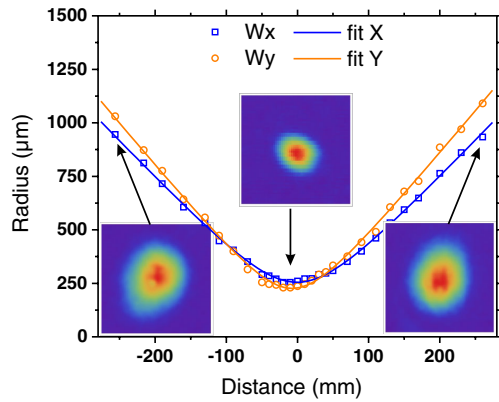


Fig. 4. Beam profile and M^2 measurement results with $M_x^2 = 1.18$ and $M_y^2 = 1.19$ at the compressor output.

material is the major contribution to the difference between compressed and TL pulse durations. Note that the seed pulse before the stretcher is well compressed and has about 20 times smaller amplitude of higher than third-order components in the spectral phase (see temporal characterization in Supplement 1); thus, its contribution to the uncompensated phase is negligible. In the future, the residual phase can be compensated for by adding a pair of chirped mirrors or a programmable spectral phase shaper. The energy of the compressed pulses is 7 mJ (the average power is 7 W), which corresponds to 85% compressor transmission. The system demonstrates an excellent power stability of 0.3% [Fig. 2(b)]. The measured energy and the temporal profile imply 62 GW peak power. The system delivers a high-quality beam of nearly Gaussian mode with an M-squared parameter of about 1.2 (Fig. 4) on the compressor output.

B. Nonlinear Post-Compression

Since a large number of strong-field and attosecond applications benefit from few-cycle mid-IR pulse duration and isolated attosecond pulses, the ability to generate pulses shorter than 100 fs is important. Although the gain material supports few times broader spectral bandwidth [17], a significant part of it (namely, >2500 nm) is within the air (mostly water and CO_2) absorption range [19], which requires the entire system to be in vacuum or purged with nitrogen, for example. A more practical and elegant way to achieve few-cycle operation is to limit the amplifier spectrum to the water transmission window (<2500 nm) and perform a nonlinear post-compression of the amplified pulses right in front of an experiment, which limits the air path of the broadband pulse to a minimum and excludes any significant absorption.

There are several well-developed techniques for the nonlinear broadening and post-compression of high-energy pulses of duration from tens of femtoseconds to a few picoseconds; among them, hollow-core fibers (HCFs) [30–32], multi-pass cells [33–35], and a thin plate or multiple thin plates [10,36–40] are the most common approaches. The majority of the nonlinear broadening and compression experiments have been done with Ti:sapphire, Yb:YAG, or Yb:fiber systems with wavelengths in the normal dispersion spectral range, and there are just a few realizations in the mid-IR spectral range [10,39,40]. The advantage of the ~ 2.4 μm wavelength is the anomalous dispersion regime for a broad range of transparent materials (i.e., CaF_2 , YAG, sapphire), which enables

soliton-like pulse compression with no additional dispersion management [10,39] such as chirped mirrors. Due to this attractive feature as well as the compactness and simplicity, the approach similar to the multiple thin plates compression scheme, which is typically called multiple-plate supercontinuum generation (MPSC) [36,37], was chosen for the present work. Despite some similarities to MPSC, our approach is easily scalable to multi-mJ energy, as experimentally demonstrated in the following, while MPSC has been limited to a few 100's μJ thus far [36,37]. In addition, compared to a single-substrate nonlinear compression demonstrated in the mid-IR spectral range at a multi-mJ energy level [39], our approach offers much better spatial homogeneity and beam quality while preserving simplicity and compactness.

The implemented scheme for nonlinear compression is shown in Fig. 5(a). In a standard MPSC realization [36,37], a set of thin plates is arranged around the focus of the beam; however, since a focused pulse with multi-mJ energy would ionize air and create a filament, the direct implementation of this approach can be done only in vacuum. Therefore, we have developed an alternative scheme based on a set of AR-coated CaF_2 lenses, which has a few key features. First, the suggested waveguide-like periodic sequence of negative and positive lenses can mimic the HCF behavior [41] and spatially homogenize the output beam, as was demonstrated in bulk-media-based multi-pass cells [42,43], whereas a single-substrate compression inherently results in spatially inhomogeneous output (see, e.g., Fig. 5 in the supplement of [39]) that is caused by different amounts of the nonlinear phase accumulated in different parts of the beam due to the Gaussian intensity profile, which is a fairly nonuniform intensity distribution as opposed to a top-hat one. Note that the pulse intensity and the corresponding

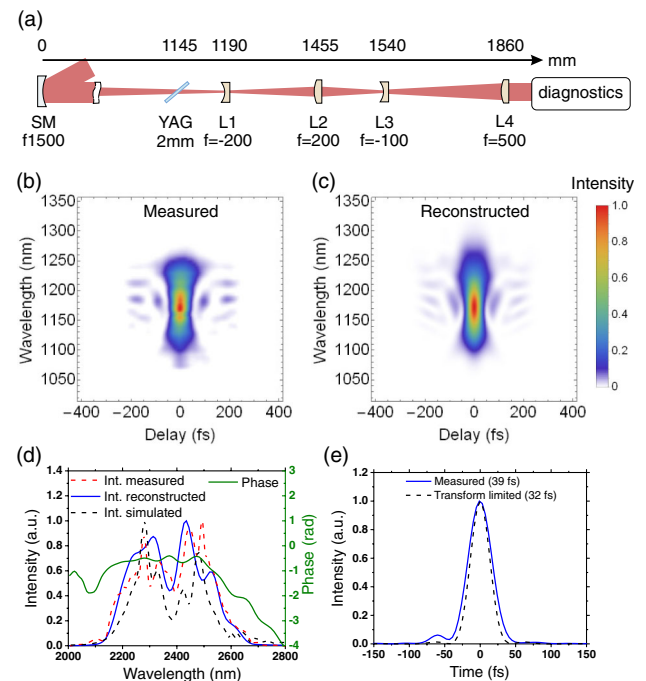


Fig. 5. (a) Nonlinear compression setup. SM, spherical mirror; L, lens (all lenses are made of CaF_2 with about 3 mm central thickness; L1 has -200 mm focal length, L2: 200 mm, L3: -100 mm, L4: 500 mm); all dimensions are in mm. (b) Measured FROG trace. (c) Reconstructed FROG trace. The reconstruction error is 0.005. (d) Measured, reconstructed, and simulated spectra and measured spectral phase. (e) Temporal pulse intensity profile.

nonlinear phase shift in the plano-concave lenses L1 and L3 is 25–64 times larger than in the plano-convex lenses L2 and L4, since the beam size is five to eight times smaller on the defocusing lenses (L1 and L3). Thus, plano-convex lenses are responsible for refocusing, but they do not make a significant contribution to broadening, and therefore can be replaced with focusing mirrors if necessary. Second, broadening in plano-concave lenses also slightly improves spatial homogeneity, since substrates are thinner in the middle of the beam with the highest intensity and thicker to the edges of the beam. However, for this effect to play a significant role, the focal lengths of a lens would have to be much shorter [38], which would result in strong chromatic aberrations. Third, CaF_2 has about a three times smaller nonlinear refractive index as well as group delay dispersion (GDD) than YAG [44], so the about 3 mm thick lenses used are equivalent to about 1 mm thick YAG substrates. Thus, the scheme can be effectively considered as a set of multiple thin plates.

The proposed post-compression setup demonstrates reproducible and stable performance. It enables significant broadening of the amplified pulses, increasing the spectrum width by a factor of about 2.5 [Fig. 5(d)]. Accordingly, the broadened pulses have 2.5 times shorter duration of 39 fs (4.8 optical cycles), as measured with the SHG-FROG diagnostics setup without any additional dispersion management such as chirped mirrors. Experimental results are in a good agreement with numerical simulations [Fig. 5(d) (black curve)], which were performed using a 1D + time split-step code following an approach similar to [45] (see Supplement 1 for more details) with the experimentally measured temporal profile (Fig. 3) used as the input pulse. M^2 of the compressed pulse slightly degrades to about 1.5 (see details in Supplement 1), which still corresponds to a very good beam quality. The average power measured after the post-compression setup is 6.2 W (89% transmission) with the same 0.3% power stability as the input pulses. Note that the broadening is very tolerable to small power changes as shown in Fig. 6. 10% input pulse energy variation (namely, reducing from 7 mJ to 6.2 mJ) barely changes the output pulse duration, which results in a very reproducible and reliable day-to-day operation.

Figure 7 demonstrates a significant improvement in spatial homogeneity compared to previous compression results of multi-mJ mid-IR pulses [39], which was one of the goals of the implemented setup. The obtained results show a tolerable increase in the pulse duration by a factor of 1.5 (from 39 fs to 60 fs) between the center and the edge of the beam. Whereas in [39], the increase in pulse duration was more than a factor of two, which is also consistent with our experimental results in a similar setup based on

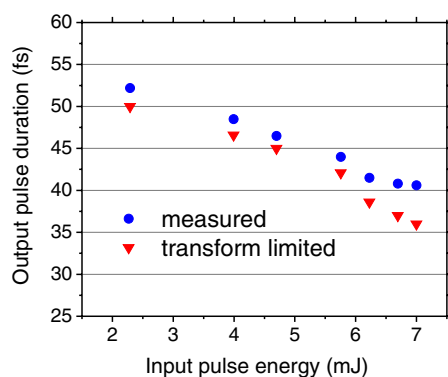


Fig. 6. Dependence of the pulse duration after the nonlinear compression setup on the input pulse energy.

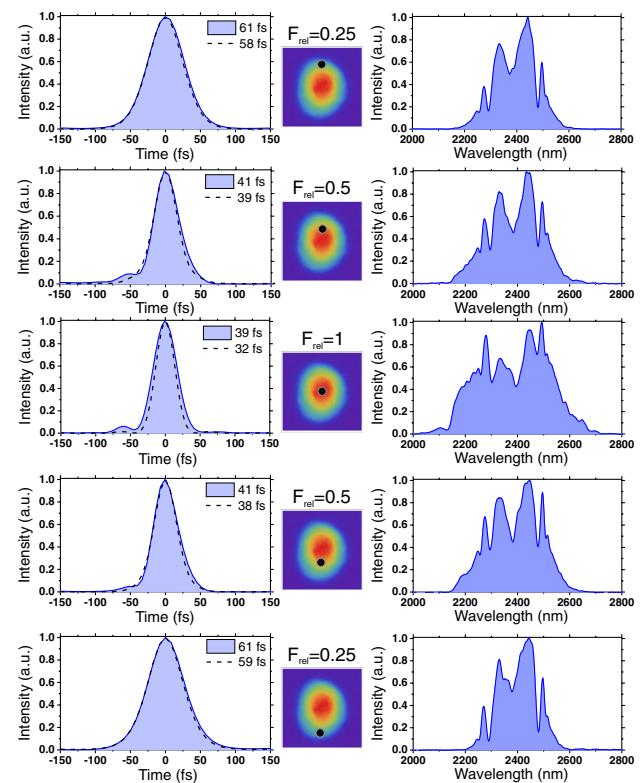


Fig. 7. Homogeneity of the nonlinear compression. The measured dependence of the spectrum and pulse duration across the beam profile by selecting a small part of the beam with an aperture indicated as a black circle in the middle row. F_{rel} is the fluence in the aperture relative to the peak fluence. Left row: temporal pulse profile retrieved from the measured SHG-FROG; blue: measured pulse, black dashed line: transform limited pulse. Right row: measured spectrum. Note that the measured beam properties are radially symmetrical, and the measured left-right scan is almost identical to the presented up-down scan, so it is not shown.

the broadening in a couple of YAG plates (see Supplement 1 for more details). Note that the YAG substrate in the implemented setup [Fig. 5(a)] was experimentally found to improve pulse compression from 50 fs without it to 39 fs with it and not to affect the spatial homogeneity, unlike the case without lenses presented in Supplement 1. There are no significant drawbacks of adding a substrate before the setup because the waveguide-like set of lenses homogenizes the beam, as discussed above. Despite the discussed inhomogeneity, the pulse duration measured at each point is smaller than the pulse duration before the nonlinear compression, which results in the increase in peak power to about 115 GW, with the spatial inhomogeneity being taken into account.

C. Soft X-Ray High-Harmonic Generation

For high-harmonic generation (HHG) experiments, the output of the grating compressor with 101 fs pulse duration is sent to a HHG vacuum chamber located about 10 m away from the system output. The beam with 18 mm diameter is focused with a plano-convex CaF_2 lens with a 400 mm focal length on a gas jet created by a pulsed piezo-valve (ACPV3 from AmsterdamPiezoValve) with a 0.5 mm inner diameter. The position of the gas jet relative to the beam focus and the backing pressure of the valve are optimized for the highest total flux. The HHG chamber is connected to a spectrometer chamber with a residual gas pressure below 10^{-7} mbar,

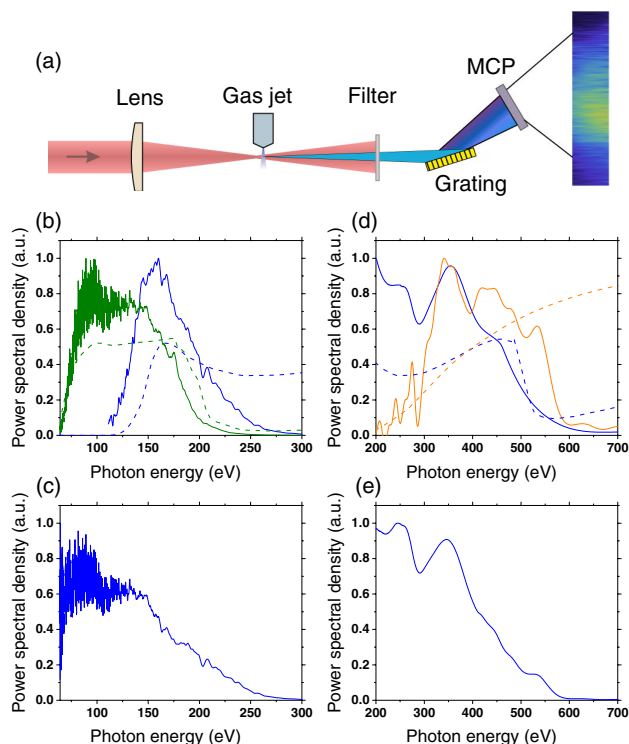


Fig. 8. HHG results. (a) Schematic layout of the HHG experimental setup. (b) Spectra generated in Ar and measured with Zr (green solid curve) and Sn (blue solid curve) filters. The corresponding filter transmission is shown with dashed lines. (c) Combined results from (b) after correction for the filter transmission. (d) Spectra generated in Ne and measured with Sn (blue solid curve) and Al (orange solid curve) filters. The corresponding filter transmission is shown with dashed lines. (e) Combined results from (d) after correction for the filter transmission.

which is maintained using a differential pumping scheme. The residual infrared radiation is blocked with a 200 nm thick metal foil filter (Al, Zr, or Sn). The HHG spectra are measured with a home-built flat-field spectrometer based on a variable line spacing concave grating (2400 l/mm, part #001-0450 from Hitachi). The generated harmonics are detected with a set of a microchannel plates (MCPs) and a phosphor plate, which is imaged on a complementary metal oxide semiconductor camera. The spectrometer is calibrated on the absorption edges of Al, Si, Zr, and Sn and by the position of harmonics where they are resolved.

Figure 8 shows HHG spectra measured in argon and neon with 60 s integration time. The on-target mid-IR energy is 2 mJ and 3.2 mJ for Ar and Ne, respectively, which is controlled with an iris before the lens and optimized to maximize the soft x-ray flux. At a higher pulse energy, phase matching of the HHG process is affected by the excessive ionization fraction of atoms and corresponding free-electron plasma dispersion contribution [46]. Therefore, increasing the focal length will enable utilizing the full available energy in future experiments. With argon at 2.5 bar valve backing pressure, the generated HHG spectrum extends in the soft x-ray region up to about 280 eV. The peak intensity estimated from the cutoff is 160 TW/cm^2 , which is in a good agreement with 190 TW/cm^2 estimated in the beam waist using the Fraunhofer diffraction theory and the experimental focusing geometry. Neon enables significantly higher HHG cutoff due to higher ionization saturation intensity related to higher ionization potential compared to argon. In neon, the HHG spectrum spans

the entire water window spectral region and extends up to 600 eV. This cutoff implies 350 TW/cm^2 on-target pulse peak intensity using the “ $3.17U_p + I_p$ ” law [47,48], which is in a good agreement with 450 TW/cm^2 as estimated in the beam waist from the focusing geometry. The optimum gas jet position is slightly behind the beam waist, so the on-target intensity should be slightly lower than the maximum in-waist value. The spectral dip in the measured spectrum [Fig. 8(e)] around the carbon K-edge at ca. 280 eV might originate from the beamline contamination. The absolute HHG flux was not measured due to the lack of a calibrated measurement device in the present realization of the experimental setup, but a lower bound of the HHG flux at the spectrometer entrance estimated from the registered counts is 5 pW in the presented spectral range. The estimate does not include beam clipping on the spectrometer entrance slit nor some possible losses on the differential pumping irises; thus, the HHG flux available for experiments should be higher.

Further extension of the HHG cutoff is feasible using helium with even higher ionization potential and by increasing the focused intensity, which can be easily done using full pulse energy and a shorter focal length and/or expanding the beam size. However, it is not feasible in the present setup because the valve does not allow the creation of pressure high enough to phase match HHG in helium [12,46]. To do so, a high pressure cell is required, which will be implemented in future studies.

4. CONCLUSION

We have demonstrated a $\text{Cr}^{2+}:\text{ZnSe}$ laser amplifier delivering 7 mJ, 100 fs pulses at 1 kHz repetition rate and $2.4 \mu\text{m}$ central wavelength with excellent energy stability and beam quality. These pulses were post-compressed to 39 fs, 6.2 mJ, and 115 GW peak power in a novel nonlinear compression scheme introduced here. It enables reliable post-compression in a compact and simple setup even at the multi-mJ pulse energy level without any additional dispersion management due to soliton-like regime in the anomalous dispersion range. The system was successfully applied to soft x-ray generation in the water window up to 0.6 keV. In addition to HHG applications, due to the very good pulse quality and mid-IR wavelength, the demonstrated system will enable efficient (significantly more efficient compared to $\sim 1 \mu\text{m}$ [49]) pumping of a sub-mJ-level $\sim 6\text{--}12 \mu\text{m}$ OPA, which will advance strong-field physics into the IR regime. An architecture similar to the one presented here can be used to generate pulses in the $3\text{--}4 \mu\text{m}$ spectral range using Fe^{2+} -doped chalcogenides [17] and KTiOAsO_4 (KTA)-based OPA for seed generation [9,10]. Moreover, the demonstrated architectures of both the amplifier and the post-compression scheme are scalable to higher repetition rates under the same average power and, providing the availability of larger crystals and more energetic pump sources, to higher pulse energies and peak powers, which will open a wide range of applications in nonlinear optics, attosecond and strong-field physics, and remote sensing.

Funding. Air Force Office of Scientific Research (MURI FA9550-16-1-0013); Department of Energy, Office of Science (DE-FG02-04ER15614).

Acknowledgment. The authors gratefully acknowledge Andrew Piper, Timothy Scarborough, and Qiaoyi Liu for helpful discussions. Special thanks are due to Zenghu Chang for his support and fruitful discussions of the amplifier design.

Disclosures. The authors declare no conflicts of interest.

See Supplement 1 for supporting content.

REFERENCES

- P. Agostini and L. F. DiMauro, "Atoms in high intensity mid-infrared pulses," *Contemp. Phys.* **49**, 179–197 (2008).
- C. I. Blaga, J. Xu, A. D. DiChiara, E. Sistrunk, K. Zhang, P. Agostini, T. A. Miller, L. F. DiMauro, and C. D. Lin, "Imaging ultrafast molecular dynamics with laser-induced electron diffraction," *Nature* **483**, 194–197 (2012).
- T. Popmintchev, M.-C. Chen, D. Popmintchev, P. Arpin, S. Brown, S. Ališauskas, G. Andriukaitis, T. Balčiūnas, O. D. Mücke, A. Pugžlys, A. Baltuška, B. Shim, S. E. Schrauth, A. Gaeta, C. Hernández-García, L. Plaja, A. Becker, A. Jaron-Becker, M. M. Murnane, and H. C. Kapteyn, "Bright coherent ultrahigh harmonics in the keV x-ray regime from mid-infrared femtosecond lasers," *Science* **336**, 1287–1291 (2012).
- J. Li, X. Ren, Y. Yin, K. Zhao, A. Chew, Y. Cheng, E. Cunningham, Y. Wang, S. Hu, Y. Wu, M. Chini, and Z. Chang, "53-attosecond x-ray pulses reach the carbon K-edge," *Nat. Commun.* **8**, 186 (2017).
- F. K. Tittel, D. Richter, and A. Fried, "Mid-infrared laser applications in spectroscopy," in *Solid-State Mid-Infrared Laser Sources*, I. T. Sorokina and K. L. Vodopyanov, eds. (Springer, 2003), pp. 458–529.
- B. Jean and T. Bende, "Mid-IR laser applications in medicine," in *Solid-State Mid-Infrared Laser Sources*, I. T. Sorokina and K. L. Vodopyanov, eds. (Springer, 2003), pp. 530–565.
- J. J. Liu, B. L. Stann, K. K. Klett, P. S. Cho, and P. M. Pellegrino, "Mid and long-wave infrared free-space optical communication," *Proc. SPIE* **11133**, 1113302 (2019).
- P. Werle, F. Slemr, K. Maurer, R. Kormann, R. Mücke, and B. Jänker, "Near- and mid-infrared laser-optical sensors for gas analysis," *Opt. Laser Eng.* **37**, 101–114 (2002).
- A. G. Ciriolo, M. Negro, M. Devetta, E. Cinquanta, D. Faccialà, A. Pusala, S. De Silvestri, S. Stagira, and C. Vozzi, "Optical parametric amplification techniques for the generation of high-energy few-optical-cycles IR pulses for strong field applications," *Appl. Sci.* **7**, 265 (2017).
- F. Lu, P. Xia, Y. Matsumoto, T. Kanai, N. Ishii, and J. Itatani, "Generation of sub-two-cycle CEP-stable optical pulses at 3.5 μm from a KTA-based optical parametric amplifier with multiple-plate compression," *Opt. Lett.* **43**, 2720–2723 (2018).
- U. Elu, T. Steinle, D. Sánchez, L. Maidment, K. Zawilski, P. Schunemann, U. D. Zeitner, C. Simon-Boisson, and J. Biegert, "Table-top high-energy 7 μm OPCPA and 260 mJ Ho:YLF pump laser," *Opt. Lett.* **44**, 3194–3197 (2019).
- J. Pupeikis, P.-A. Chevreuril, N. Bigler, L. Gallmann, C. R. Phillips, and U. Keller, "Water window soft x-ray source enabled by 25 W few-cycle 2.2 μm OPCPA at 100 kHz," *Optica* **7**, 168–171 (2020).
- B. E. Schmidt, N. Thiré, M. Boivin, A. Laramée, F. Poitras, G. Lebrun, T. Ozaki, H. Ibrahim, and F. Légaré, "Frequency domain optical parametric amplification," *Nat. Commun.* **5**, 3643 (2014).
- H. Liang, P. Kroger, Z. Wang, H. Park, T. Kroh, K. Zawilski, P. Schunemann, J. Moses, L. F. DiMauro, F. X. Kärtner, and K.-H. Hong, "High-energy mid-infrared sub-cycle pulse synthesis from a parametric amplifier," *Nat. Commun.* **8**, 141 (2017).
- J. Zhang, K. Fai Mak, N. Nagl, M. Seidel, D. Bauer, D. Sutter, V. Pervak, F. Krausz, and O. Pronin, "Multi-mW, few-cycle mid-infrared continuum spanning from 500 to 2250 cm^{-1} ," *Light. Sci. Appl.* **7**, 17180 (2018).
- Q. Wang, J. Zhang, A. Kessel, N. Nagl, V. Pervak, O. Pronin, and K. F. Mak, "Broadband mid-infrared coverage (2–17 μm) with few-cycle pulses via cascaded parametric processes," *Opt. Lett.* **44**, 2566–2569 (2019).
- S. B. Mirov, I. S. Moskalev, S. Vasilyev, V. Smolski, V. V. Fedorov, D. Martyshkin, J. Peppers, M. Mirov, A. Dergachev, and V. Gapontsev, "Frontiers of mid-IR lasers based on transition metal doped chalcogenides," *IEEE J. Sel. Top. Quantum Electron.* **24**, 1–29 (2018).
- K. K. Yu, X. M. Duan, D. X. Zhang, B. Q. Yao, and Y. J. Shen, "A 1 kHz, 30 mJ Ho:YAG master oscillator and power amplifier resonantly double-end-pumped by Tm:YLF lasers," *Laser Phys.* **23**, 065001 (2013).
- X. Ren, L. H. Mach, Y. Yin, Y. Wang, and Z. Chang, "Generation of 1 kHz, 2.3 mJ, 88 fs, 2.5 μm pulses from a ZnSe chirped pulse amplifier," *Opt. Lett.* **43**, 3381–3384 (2018).
- E. Slobodchikov, L. R. Chieffo, and K. F. Wall, "High peak power ultrafast Cr:ZnSe oscillator and power amplifier," *Proc. SPIE* **9726**, 972603 (2016).
- E. Migal, A. Pushkin, B. Bravy, V. Gordienko, N. Minaev, A. Sirotkin, and F. Potemkin, "3.5-mJ 150-fs Fe:ZnSe hybrid mid-IR femtosecond laser at 4.4 μm for driving extreme nonlinear optics," *Opt. Lett.* **44**, 2550–2553 (2019).
- K. R. Wilson and V. V. Yakovlev, "Ultrafast rainbow: tunable ultrashort pulses from a solid-state kilohertz system," *J. Opt. Soc. Am. B* **14**, 444–448 (1997).
- O. Martínez, "3000 times grating compressor with positive group velocity dispersion: application to fiber compensation in 1.3–1.6 μm region," *IEEE J. Quantum Electron.* **23**, 59–64 (1987).
- E. Treacy, "Optical pulse compression with diffraction gratings," *IEEE J. Quantum Electron.* **5**, 454–458 (1969).
- N. Rimgton, S. Schieffer, W. A. Schroeder, and B. K. Briceken, "Thermal lens shaping in brewster gain media: a high-power, diode-pumped laser," *Opt. Express* **12**, 1426–1436 (2004).
- I. T. Sorokina, "Cr²⁺-doped II–VI materials for lasers and nonlinear optics," *Opt. Mater.* **26**, 395–412 (2004).
- V. N. Ginzburg, A. A. Kochetkov, A. K. Potemkin, and E. A. Khazanov, "Suppression of small-scale self-focusing of high-power laser beams due to their self-filtration during propagation in free space," *Quantum Electron.* **48**, 325–331 (2018).
- I. Moskalev, S. Mirov, M. Mirov, S. Vasilyev, V. Smolski, A. Zakrevskiy, and V. Gapontsev, "140 W Cr:ZnSe laser system," *Opt. Express* **24**, 21090–21104 (2016).
- B. Tatian, "Fitting refractive-index data with the Sellmeier dispersion formula," *Appl. Opt.* **23**, 4477–4485 (1984).
- T. Nagy, S. Hädrich, P. Simon, A. Blumenstein, N. Walther, R. Klas, J. Buldt, H. Stark, S. Breikopf, P. Jójárt, I. Seres, Z. Váralyay, T. Eidam, and J. Limpert, "Generation of three-cycle multi-millijoule laser pulses at 318 W average power," *Optica* **6**, 1423–1424 (2019).
- G. Fan, T. Balčiūnas, T. Kanai, T. Flöry, G. Andriukaitis, B. E. Schmidt, F. Légaré, and A. Baltuška, "Hollow-core-waveguide compression of multi-millijoule CEP-stable 3.2 μm pulses," *Optica* **3**, 1308–1311 (2016).
- B. E. Schmidt, A. D. Shiner, P. Lassonde, J.-C. Kieffer, P. B. Corkum, D. M. Villeneuve, and F. Légaré, "CEP stable 1.6 cycle laser pulses at 1.8 μm ," *Opt. Express* **19**, 6858–6864 (2011).
- L. Lavenue, M. Natile, F. Guichard, Y. Zaouter, X. Delen, M. Hanna, E. Mottay, and P. Georges, "Nonlinear pulse compression based on a gas-filled multipass cell," *Opt. Lett.* **43**, 2252–2255 (2018).
- M. Kaumanns, V. Pervak, D. Kormin, V. Leshchenko, A. Kessel, M. Ueffing, Y. Chen, and T. Nubbemeyer, "Multipass spectral broadening of 18 mJ pulses compressible from 1.3 ps to 41 fs," *Opt. Lett.* **43**, 5877–5880 (2018).
- P. Russbueltdt, J. Weitenberg, J. Schulte, R. Meyer, C. Meinhardt, H. D. Hoffmann, and R. Poprawe, "Scalable 30 fs laser source with 530 W average power," *Opt. Lett.* **44**, 5222–5225 (2019).
- C.-H. Lu, Y.-J. Tsou, H.-Y. Chen, B.-H. Chen, Y.-C. Cheng, S.-D. Yang, M.-C. Chen, C.-C. Hsu, and A. H. Kung, "Generation of intense supercontinuum in condensed media," *Optica* **1**, 400–406 (2014).
- M. Seo, K. Tsendsuren, S. Mitra, M. Kling, and D. Kim, "High-contrast, intense single-cycle pulses from an all thin-solid-plate setup," *Opt. Lett.* **45**, 367–370 (2020).
- P. Lassonde, S. Mironov, S. Fourmaux, S. Payeur, E. Khazanov, A. Sergeev, J.-C. Kieffer, and G. Mourou, "High energy femtosecond pulse compression," *Laser Phys. Lett.* **13**, 075401 (2016).
- V. Shumakova, P. Malevich, S. Ališauskas, A. Voronin, A. M. Zheltikov, D. Faccio, D. Kartashov, A. Baltuška, and A. Pugžlys, "Multi-millijoule few-cycle mid-infrared pulses through nonlinear self-compression in bulk," *Nat. Commun.* **7**, 12877 (2016).
- M. Hemmer, M. Baudisch, A. Thai, A. Couairon, and J. Biegert, "Self-compression to sub-3-cycle duration of mid-infrared optical pulses in dielectrics," *Opt. Express* **21**, 28095–28102 (2013).
- S. N. Vlasov, E. V. Kuposova, and V. E. Yashin, "Spectral broadening and compression of high-intensity laser pulses in quasi-periodic systems with Kerr nonlinearity," *Quantum Electron.* **42**, 989–995 (2012).

42. J. Schulte, T. Sartorius, J. Weitenberg, A. Vernaleken, and P. Russbueltd, "Nonlinear pulse compression in a multi-pass cell," *Opt. Lett.* **41**, 4511–4514 (2016).
43. K. Fritsch, M. Poetzlberger, V. Pervak, J. Brons, and O. Pronin, "All-solid-state multipass spectral broadening to sub-20 fs," *Opt. Lett.* **43**, 4643–4646 (2018).
44. P. Kabaciński, T. M. Kardaś, Y. Stepanenko, and C. Radzewicz, "Nonlinear refractive index measurement by SPM-induced phase regression," *Opt. Express* **27**, 11018–11028 (2019).
45. A. Kessel, S. A. Trushin, N. Karpowicz, C. Skrobol, S. Klingebiel, C. Wandt, and S. Karsch, "Generation of multi-octave spanning high-energy pulses by cascaded nonlinear processes in BBO," *Opt. Express* **24**, 5628–5637 (2016).
46. T. Popmintchev, M.-C. Chen, A. Bahabad, M. Gerrity, P. Sidorenko, O. Cohen, I. P. Christov, M. M. Murnane, and H. C. Kapteyn, "Phase matching of high harmonic generation in the soft and hard x-ray regions of the spectrum," *Proc. Natl. Acad. Sci. USA* **106**, 10516–10521 (2009).
47. M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L'Huillier, and P. B. Corkum, "Theory of high-harmonic generation by low-frequency laser fields," *Phys. Rev. A* **49**, 2117–2132 (1994).
48. A. Shiner, C. Trallero-Herrero, N. Kajumba, B. Schmidt, J. Bertrand, K. T. Kim, H.-C. Bandulet, D. Comtois, J.-C. Kieffer, D. Rayner, P. Corkum, F. Légaré, and D. Villeneuve, "High harmonic cutoff energy scaling and laser intensity measurement with a 1.8 μm laser source," *J. Mod. Opt.* **60**, 1458–1465 (2013).
49. Q. Cao, F. X. Kärtner, and G. Chang, "Towards high power longwave mid-IR frequency combs: power scalability of high repetition-rate difference-frequency generation," *Opt. Express* **28**, 1369–1384 (2020).