High-energy, phase-stable, ultrabroadband kHz OPCPA at 2.1 µm pumped by a picosecond cryogenic Yb:YAG laser

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Abstract: We report on a kHz, mJ-level, carrier-envelope phase (CEP)-stable ultrabroadband optical parametric chirped-pulse amplifier (OPCPA) at 2.1-µm wavelength, pumped by a high-energy, 14 ps, cryogenic Yb:YAG pump laser, and its application to high-order harmonic generation (HHG) with Xe. The pre-amplifier chain is pumped by a 12-ps Nd:YLF pump laser and both pump lasers are optically synchronized to the signal pulse of the OPCPA. An amplified pulse energy of 0.85 mJ was obtained at the final OPCPA stage with good beam profile. The pulse is compressed to 4.5 optical cycles (<32 fs) with a spectral bandwidth of 474 nm supporting 3.5 optical cycles. The CEP stability was measured to be 194 mrad and the super-fluorescence noise is estimated to be ~9%. First HHG results are demonstrated with Xe showing significant cutoff extension to >85 eV with an efficiency of ~10−10 per harmonic, limited by the maximum gas pressure and flow into the chamber. This demonstrates the potential of this 2.1-µm source for scaling of photon energy and flux in the water-window range when applied to Ne and He at kHz repetition rate.

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OCIS codes: (190.4970) Parametric oscillators and amplifiers, (140.3615) Lasers, ytterbium, (320.7110) Ultrafast nonlinear optics.

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1. Introduction

High-order harmonic generation (HHG) with long-wavelength driver pulses [1–9] has been regarded as a promising source of high-flux coherent extreme ultraviolet/soft X-ray (XUV) radiation with photon energies approaching the keV regime via cutoff extension. However, the single-atom response in the HHG efficiency unfavorably scales with driving wavelength (λ−5 for the cutoff region and λ−6 for the plateau region), mostly governed by quantum diffusion of electron trajectories. This unfavorable scaling makes phase matched and absorption limited generation important for practical use, in order to avoid an additional efficiency drop. In general, mJ-level pulse energy is necessary to avoid the phase mismatch induced by the Guoy phase shift. Recently, phase-matched HHG with covering the water window range was experimentally demonstrated [8] using a 10-Hz, 2.4-mJ, 2-μm optical parametric amplifier (OPA) pumped by a Ti:sapphire chirped-pulse amplification (CPA) laser. The generated XUV photon number per second over 1% bandwidth, however, is still as low as

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~6x10^7, limiting potential applications. Increasing the repetition rate to the kHz-range is a clear path towards increasing XUV photon flux by 2 orders of magnitude. More recently, sub-mJ few-cycle kHz OPA sources at around 1.8 µm, pumped by a kHz CPA Ti:sapphire laser, have been demonstrated with the help of external pulse compression techniques [10,11] for application to HHG experiments.

Optical parametric chirped-pulse amplification (OPCPA) [12] is generally regarded as a more energy-scalable method than OPA, but the generation of >mJ energy from OPCPA at kHz repetition rates is nontrivial due to the lack of suitable pump sources. Fuji et al. [13] pioneered a kHz, few-cycle OPCPA system at 2.1-µm wavelength using an intra-pulse difference-frequency generation (DFG) seed and a ~50-ps Nd:YLF pump laser. They obtained output energies of up to 80 µJ, which was limited by excessive super-fluorescence (SF). Gu et al. [14] extended this work and demonstrated amplification up to 0.74 mJ with 11 mJ of total pump energy. To increase the DFG energy and thereby overcome the SF problem, they employed a complex high-energy seed source based on a CPA Ti:sapphire laser and a hollow-fiber external compressor. More recently, Moses et al. [15] developed a 0.2-mJ 3-cycle OPCPA system at 2.2-µm wavelength and showed that the SF noise can be efficiently suppressed using careful optimization that avoids amplifier over-saturation [16], without using a high-energy CPA seed laser. A total pump energy of 4 mJ from a 12-ps Nd:YLF CPA chain at 1 kHz repetition rate was used. Preliminary experiments on HHG with the 0.2-mJ, 2.2-µm OPCPA source and further theoretical studies confirmed that further energy scaling is necessary. In this paper, we present an upgrade of this OPCPA system using a new ps pump laser based on power-scalable cryogenic Yb:YAG technology. The system delivers 1 mJ-level kHz-repetition-rate pulse trains for experimental demonstration of high-flux long-wavelength HHG, geared towards keV photon energy.

Since the energy scaling of such OPCPA systems at kHz repetition rate is ultimately limited by the power and energy handling of Nd:YLF laser technology, new pump laser technologies based on Yb-doped laser materials are now being rapidly developed. Metzger et al. [17] developed a 25-mJ ~1-ps Yb:YAG CPA laser at 3 kHz based on thin-disk laser technology to avoid thermal problems. On the other hand, cryogenic solid-state laser technology provides a straightforward way of energy and average power scaling [18–22]. Recently, Hong et al. [23] have demonstrated 40 mJ amplification of ps pulses at 2 kHz from a cryogenic Yb:YAG CPA chain. Compared to the thin-disk regenerative amplifier (RGA) [17], the cryogenic Yb:YAG RGA has the distinctive advantage of using a much lower number of roundtrips (<15 vs. ~150) thanks to the higher single-pass gain, which is crucial for stable operation of RGAs without bifurcation and reliable optical timing synchronization with the OPCPA seed pulses.

In this work, the energy scaling of a 2.1-µm kHz ultrabroadband OPCPA system is achieved using a high-energy, ps cryogenic Yb:YAG pump laser [23], optically synchronized to the signal. We maintained a lower energy Nd:YLF pump laser for the first two OPCPA stages as pre-amplifiers to minimize the modification of the former system [15], which operates stably and is already optimized for SF suppression. For power amplification, the final OPCPA stage was pumped by the novel high-energy cryogenic Yb:YAG laser.

The paper is organized as follows. In section 2, we describe the experimental details of the new pump laser and optical synchronization scheme. In section 3, the OPCPA experiments and amplification results will be discussed. We present the first experimental result of HHG using this source in section 4 and conclude in section 5.

2. Pump lasers and optical synchronization

The ultrabroadband 2.1-µm OPCPA system operating at 1 kHz repetition rate consists of a master oscillator with a DFG stage, a Nd:YLF pump laser, a cryogenic Yb:YAG pump laser, and three OPCPA stages followed by a compressor. Figure 1 shows the optical synchronization scheme of the OPCPA stages with two pump lasers and the CPA schematic of each pump laser. An octave-spanning Ti:sapphire oscillator at 85 MHz (Octavius-85M, IdestaQE, Inc.) generates passively CEP-stabilized seed pulses at 2.1 µm by intra-pulse DFG...
in a 2-mm-long MgO-doped periodically poled congruent lithium niobate (MgO:PPLN) crystal with a poling period, \( \Lambda \), of 13.1 \( \mu \)m. The 650 nm and 940 nm portions of the Ti:Sapphire spectrum are mixed together to generate ultrabroadband (~900 nm wide at ~20 dB) spectra around 2.1\( \mu \)m. The 1-\( \mu \)m portion (1010-1060 nm) of the oscillator spectrum with an average power of ~3 mW is amplified by a ytterbium-doped fiber amplifier (YDFA) to ~20 mW for seeding two pump lasers.

![Diagram](image)

Fig. 1. Schematic diagram of a kHz 2.1-\( \mu \)m ultrabroadband OPCPA pumped by Nd:YLF and cryogenic Yb:YAG CPA lasers, optically synchronized to the OPCPA signal. (YDFA, ytterbium-doped fiber amplifier; CFBG/CVBG, chirped fiber/volume Bragg grating; MLD, multi-layer dielectric; RGA, regenerative amplifier)

The first pump source is a 4-mJ, 12-ps, Nd:YLF CPA chain, composed of a chirped fiber Bragg grating (CFBG) stretcher, YDFA, RGA followed by two multi-pass slab amplifiers, and a diffraction grating compressor [15]. This laser pumps the first and second OPCPA stages (pump 1 in Fig. 1). The CFBG has a chirp rate of ~440 ps/nm and the pulse duration after the RGA is ~110 ps over 0.25 nm of bandwidth. Since the required pump energy for the first two stages of the OPCPA was only ~1.9 mJ, the Nd:YLF RGA was operated slightly below saturation, which helps avoid possible optical damage under long-term operation while slightly losing in energy stability. We maintained the energy at ~3.5 mJ after the multipass amplification and compression, which was split into the first and second OPCPA stages with an appropriate ratio using a variable beam splitter while the unused energy was dumped.

The second pump source is a high-energy, 14 ps, cryogenic Yb:YAG CPA chain, which was slightly modified from the original setup seeded by a Yb-doped fiber oscillator [23]. The transmitted portion of the CFBG at 1047 nm, containing a broad spectrum at around 1029 nm, is seeded into the Yb:YAG laser. The stretcher is composed of a chirped volume Bragg grating (CVBG) pair with a chirp rate of ~100 ps/nm per bounce. The spatial chirp of the CVBGs (Ondax, Inc.) has been significantly reduced by stressing the middle of the gratings [24]. After 8 bounces off the CVBGs, we obtained ~560 ps positively chirped pulses with 0.7 nm bandwidth at 1029 nm, as shown in Figs. 2(a) and (b). This stretcher replaced the previously used CFBG [23] with a chirp rate of 900 ps/nm because its large group delay ripple prevented clean compression to below 20 ps. Figure 2(b) was measured using a fast photodiode with a rise time of 27 ps and a sampling oscilloscope. Two YDFAs before and after the CVBG stretcher ensure high enough pulse energy (~100 pJ) for seeding of the cryogenic Yb:YAG RGA. Even though the output energy of the RGA can be higher than 10 mJ, it was fixed to ~5 mJ at 1 kHz for long-term stable operation without optical damage, where the pump power was 45 W from a fiber-coupled cw laser diode at 940 nm. The spectral
bandwidth was reduced to 0.24 nm, corresponding to ~190 ps pulse duration after the RGA. The beam from the RGA was delivered to a double-pass cryogenic Yb:YAG amplifier with two crystals in an evacuated chamber. We limited the maximum energy from this amplifier to 20 mJ at 1 kHz with a relatively large pump beam size (~3.6 mm in Gaussian diameter) also for damage-free long-term operation, as in the RGA. Our experimental and theoretical investigations uncovered that the gain guiding effect along with the thermal lensing reduced the beam size at each pass, leading to optical damage at high-energy operation. The RGA output beam is expanded using a 1:2 telescope and set to be diverging to partially compensate for this gain guiding effect at the cost of efficiency such that the beam size of the unamplified pulse after the double-pass amplifier is ~4.8 mm in diameter. As the maximum pump power approaches 320 W for 20 mJ (20 W) of amplified energy (power), the beam size decreases to ~3.1 mm in diameter. According to our simulation, the energy scaling is feasible with a proper telescope and 4-pass amplification without increasing the pump power.

![Diagram](image)

Fig. 2. Stretching and compression of picosecond pulses: (a) 8-pass CVBG stretcher, (b) positively chirped, stretched pulse shape with ~560 ps over ~0.7 nm of spectral bandwidth, and (c) autocorrelation trace of compressed 14.2-ps pulses with 0.25 nm of spectral bandwidth after an MLD grating compressor.

The pulse is compressed using a multi-layer dielectric (MLD) grating pair, where the gratings have a groove density of 1752 lines/mm and a diffraction efficiency of 95%. The separation between two MLD gratings was set to ~2.75 m with multiple bounces off dielectric mirrors to achieve a compact setup. A throughput efficiency of 75% was achieved and a compressed pulse duration of 14.2 ps was measured using a picosecond autocorrelator, as shown in Fig. 2(c). The transform-limited pulse duration supported by the output spectral bandwidth of 0.24 nm is 7.5 ps. The pedestal in the autocorrelation trace goes to zero at around +/−40 ps and the energy shed to the pedestal is estimated to be ~10%. The group delay ripple in the CVBGs seems to induce the pedestal, limiting better compression. The compressed ~14-ps pulse is delivered to the third OPCPA stage after a waveplate and a polarizer for energy control and a delay stage for timing overlap with the OPCPA signal. The maximum energy at the third OPCPA stage is 13 mJ after all optics.

The use of a single mode-locked Ti:sapphire master oscillator providing all optical seed synchronization ensures the timing between pump and signal pulses at all OPCPA stages without high frequency jitter. The timing overlap was achieved by properly choosing the pickup and dumping times of both RGAs and by fine adjusting the optical delay lines for all three pulses (signal and two pump pulses).
3. OPCPA setup and amplification result

3.1 Configuration of the OPCPA system

The optical layout of the OPCPA system pumped by two CPA lasers is illustrated in Fig. 3. The majority of the OPCPA setup of the first two stages is the same as in Ref [15], but the following modifications have been made: 1) The pulse duration is further stretched to ~14 ps after the second stage by a 30-mm silicon block to optimize its match to the pump pulse duration of the Yb:YAG laser. 2) The second stage output energy was increased to ~25 µJ. 3) Aiming for multi-mJ amplification, the periodically poled crystals are not suitable in the third stage due to the limited aperture size available to us. Thus, we used a 5-mm-long type-I β-barium borate (BBO) crystal (θ = 21.4°) instead of periodically poled stoichiometric lithium tantalate (MgO:PPSLT) at the cost of phase-matching bandwidth. Nevertheless, our calculation showed that the supported phase-matching bandwidth of the 5-mm-long BBO crystal is more than 350 nm at 20 GW/cm² pump intensity with a maximum conversion efficiency of 12% for Gaussian beams and pulses in space and time, respectively.

The 2.1-μm seed pulses with ~2 pJ at 85 MHz and a spectral bandwidth of ~500 nm at −10 dB, generated by intra-pulse DFG in MgO:PPLN, are stretched to ~5 ps with a 30-mm-long silicon block and then amplified to 2.5 µJ at 1 kHz in the first OPCPA stage (OPA1 in Fig. 3) based on a 2-mm-long MgO:PPLN (Λ = 31.0 μm) which is pumped by 0.5 mJ of energy from the Nd:YLF laser. The pulses are then stretched to ~9 ps full width at half maximum (FWHM) using an acousto-optic programmable dispersive filter (AOPDF; Dazzler, Fastlite) which also compensates for the high-order dispersion terms to achieve high-quality pulse compression. The second OPCPA stage based on a 2-mm-long MgO:PPSLT crystal, pumped by ~1.4 mJ of energy, amplifies the 2.1 μm pulses to 25 µJ with a spectral bandwidth of 405 nm in full width at half maximum (FWHM) as shown in Fig. 4(a). The pulses are further stretched to ~14 ps (FWHM) in the second 30-mm-long silicon block with a Fresnel reflection loss of ~50% for better matching to the pump pulse duration of the cryogenic Yb:YAG laser at the third stage.
3.2 High-energy amplification and pulse compression

The third OPCPA stage was optimized by varying the pump and signal beam sizes at the BBO crystal while monitoring the output energy and the relative SF noise. At the end, the pump beam size was set to be ~1.4 mm in FWHM while the seed beam size was set to ~4.0 mm in FWHM, which was much larger than the pump beam size for efficient SF suppression and for transfer of the high-quality pump beam mode to the signal beam. The corresponding pump intensity was ~40 GW/cm² at 13 mJ of energy. The angle between the pump and the seed was minimized to <2° to maximize phase matching bandwidth. After optimization of temporal and spatial overlaps and incidence angle of the pump beam into the BBO crystal, we obtained a maximum energy of 0.85 mJ with a pump energy of 13 mJ, indicating a conversion efficiency of 6.5%. The amplified spectrum from the third OPCPA stage is represented as the solid line in Fig. 4(a). The spectral bandwidth of 474 nm FWHM centered at 2.1 µm supports a transform-limited pulse duration of 24.5 fs or 3.5 optical cycles, as shown in Fig. 4(b). An output bandwidth broader than that of the input pulse coming from the second stage (405 nm) indicates gain saturation at the third stage. The bandwidth of 474 nm is also broader than the calculated phase matching bandwidth of ~350 nm, which is attributed to the broadband seed from the second stage, the gain saturation at the third stage, and the slightly detuned phase-matching angle. The valley in the central part of the spectrum is already found in the second stage spectrum, also due to a slightly detuned grating period. This valley enhances the wings in the time domain, as found in the transform-limited pulse shape of Fig. 4(b) and later in the compressed pulse shape of Fig. 4(c).

The pulse compression was achieved using two Brewster-angle Suprasil 300 blocks with a total path length of 620 mm. We finely adjusted the AOPDF to minimize the pulse duration and suppress the pedestal while monitoring interferometric autocorrelation traces. Figure 4(c) shows the compressed pulse with a duration of 31.7 fs or 4.5 optical cycles, which is 1.3 times longer than the transform-limited value. The throughput efficiency of the compressor is ~95% with a metallic (silver) mirror as a retro-reflector, but it can be enhanced to almost 100% if longer Suprasil 300 blocks are used without any mirrors.

The dispersion compensation scheme of the entire OPCPA system up to fourth-order is described in Table 1. Two silicon blocks and the AOPDF positively chirp the pulses with a
positive group-delay dispersion (GDD) value at 2.1 µm to stretch the pulse to ~14 ps, while the Suprasil 300 fused silica block compresses the pulses by providing the proper negative GDD value. Here, TeO$_2$ is the material of the AOPDF and CaF$_2$ is the material used in the telescope optics. The major contributing dispersion terms are represented in bold. As indicated by this table, the AOPDF can finely compensate for the high-order dispersion terms (third and fourth orders) as well as the GDD term. In the experiment, the set values of the AOPDF optimal for pulse compression were found to be 2.15x10$^4$ fs$^2$, 4.40x10$^5$ fs$^3$, and 1.70x10$^6$ fs$^4$, respectively, which agree very well with the design values in Table 1.

| Element       | path length (mm) | GDD (fs$^2$) | TOD (fs$^3$) | FOD (fs$^4$) |
|---------------|------------------|--------------|--------------|--------------|
| Silicon       | 60               | 4.68x10$^4$  | 5.87x10$^7$  | 3.16x10$^9$  |
| Suprasil 300  | 620              | -7.21x10$^4$ | 3.31x10$^5$  | -1.71x10$^6$ |
| TeO$_2$ (o-axis) | 45             | 4.140        | 3.01x10$^5$  | -1.65x10$^5$ |
| CaF$_2$       | 15               | -380         | 1.950        | -8.250       |
| AOPDF         | -                | 2.16x10$^5$  | -4.22x10$^6$ | 1.85x10$^6$  |
| Total         | -                | -0           | -0           | -0           |

3.3 Beam profile, CEP and noise characteristics

The near-field beam profile after the compressor, measured using a pyroelectric charge-coupled device (CCD), has a clean fundamental mode as shown in Fig. 5(a), where the Gaussian diameter of the beam is ~6 mm. There is no beam quality degradation found in the compressor. The $M^2$ value, not measured here, is expected to be <1.5 based on a knife-edge measurement in our previous work [15].

We characterized the stability and noise characteristics of the amplified pulses to identify the energy portion occupied by the amplified SF. First, the shot-to-shot energy fluctuation of the amplified 2.1-µm pulses is measured to be 4.1% (rms). This is about 3 times higher than the measured 1.5% for the former 0.2-mJ system in Ref [15]. The stability degradation naturally comes along with the increased SF noise in higher-gain amplification. In addition, the stability of the pump laser system is slightly worse because the Nd:YLF RGA is operating slightly under saturation and the overall complexity of using the two pump lasers is also higher. Second, the shot-to-shot intensity fluctuations, calculated by halving the corresponding intensity fluctuations of low-conversion second harmonic generation (SHG) of the 2.1-µm pulses, are 3.9% (rms). Third, the shot-to-shot fluctuations of the unseeded SF are ~23%. Based on the three stability parameters given above, we can estimate the SF noise to be ~9% from a simple statistical treatment [15]. This means that the signal energy in the main pulse is ~0.77 mJ out of 0.85 mJ. Compared to the parameters in the similar work by Gu et al. [14], i.e., 9% energy fluctuation and 20% SF noise with 0.74 mJ of compressed pulse energy, our system demonstrates twice better noise performance with a slightly higher compressed pulse energy, but with a longer pulse duration due to the limited phase matching bandwidth of BBO (vs. PPLN) in the last stage. There is another indication proving that this comparison is reasonable. The energy ratio between the amplified pulses and unseeded amplified SF was ~2.5:1 or better in our case, whereas it was only ~1.1:1 in the work of Gu et al. [14]. The smoother amplified spectra of our OPCPA system is also an indirect indication of the lower SF noise.

It has already been well proven that intra-pulse DFG generates passively CEP-stabilized pulses [25]. To confirm the CEP-stable operation of our OPCPA system, we measured the shot-to-shot CEP jitter using an f-3f spectral interferometer where the third-harmonic generation and the self-phase modulated signal is self-referenced with a fixed time delay in the spectral domain, as shown in Fig. 5(b). For practical reasons, this spectrogram was obtained using the pulses from the second OPCPA stage (~25 µJ) after compression. This was
done to take advantage of using the well aligned $f$-$3f$ spectral interferometer setup used in another experiment [26], because the compressor for the 0.85-mJ pulses is located in another room in front of the HHG setup. A shorter length of Suprasil 300 was used for compression for the second stage pulses without using the second silicon block, as in Ref [15]. The measured CEP jitter is 195 mrad in 30 s, revealing good CEP stability of the passively stabilized pulses after amplification.

![Near-field beam profile after the compressor with a Gaussian beam diameter of ~6 mm (a) and the $f$-$3f$ spectral interferograms (single shot) over 30 seconds, revealing a CEP stability of 194 mrad. The CEP stability was measured after the second stage followed by a different compressor for convenience.](image)

**Fig. 5.** Near-field beam profile after the compressor with a Gaussian beam diameter of ~6 mm (a) and the $f$-$3f$ spectral interferograms (single shot) over 30 seconds, revealing a CEP stability of 194 mrad. The CEP stability was measured after the second stage followed by a different compressor for convenience.

### 3.4 Challenges for long-term stable operation

Continuous operation of the cryogenically cooled amplifier, using evaporation of the liquid nitrogen from the Dewar, was achieved by installing an automatic refilling system. We achieved more than 4 hours of continuous operation at maximum pump power.

Another important issue is the thermal pointing drift of the Yb:YAG laser, induced by the high power laser pump diodes. A small amount of angular drift of the Yb:YAG amplifier output beam can drop the OPCPA output power by tens of % because of the long beam path in the MLD-grating compressor and the concomitant delay change deteriorating the timing overlap. Both the RGA and the multipass amplifier require about 20 minutes of warm-up time to obtain stable pointing. Residual beam pointing fluctuations are taken out by an active pointing stabilizer system with a bandwidth of >100 Hz to improve the pointing stability on both short and long time scales. Technical issues related to thermal problems will become even more important as further average power scaling is pursued.

### 4. First HHG experiment using the 2.1-µm OPCPA

The ultimate goal of this OPCPA development is to achieve high-flux HHG in the water-window region at kHz repetition rates, as mentioned in the introduction. As a first step to show the feasibility of achieving this goal, we started HHG experiments in Xe, a heavy noble gas with a low saturation intensity of just above $10^{14}$ W/cm². Even though He and Ne are more suitable for HHG with high photon energy, they have relatively high saturation intensity and require a high pressure of ~10 bars at the interaction region [8]. Since this high pressure is not yet available in our current HHG setup based on a pulsed gas jet operating at kHz repetition rates, Xe is an appropriate gas to start with.

The 2.1-µm OPCPA output was delivered into our HHG vacuum chamber and focused onto the gas jet using an $f = 200$ mm CaF₂ lens. The energy and focal spot were controlled together using an iris before the vacuum chamber. The HHG signal was first detected with an Al-coated XUV photodiode (AXUV100) after another Al filter with 500 nm of thickness allowing ~20% of transmission over the Al transmission window (20–70 eV). The photodiode
signal was magnified using a low-noise electronic amplifier to significantly improve the detection sensitivity because the HHG efficiency was expected to be about 2 orders-of-magnitude lower than that driven by 800-nm pulses according to the wavelength scaling. The strongest HHG signal was observed at the peak intensity of \((1-2)\times10^{14}\ \text{W/cm}^2\) at \(\sim200\ \text{mbar}\) of Xe. Since the HHG signal level monotonically increased with pressure, we used the maximum pressure available at the pressure regulator, indicating that the HHG efficiency was limited by the gas pressure. The backing pressure was about 2 bars in this experiment.

![Fig. 6. HHG spectrum driven by the kHz, 2.1-μm OPCPA using a Xe gas jet. The blue line is measured with an Al filter showing the Al edge at \(\sim70\ \text{eV}\) while the red line is measured with a Zr filter showing the cutoff at \(\sim85\ \text{eV}\).](image)

After confirming the generation of XUV photons in the Al transmission window, we measured the spectrum using our XUV spectrometer [27] with Al and Zr filters, where the thickness of the Zr filter was 400 nm. Figure 6 clearly shows the high-order harmonics up to \(\sim85\ \text{eV}\) (\(>149\)th harmonic, \(<14\) nm), where the Al edge at 70 eV is well identified. The cutoff extension to \(\sim85\ \text{eV}\) in Xe is already significant, compared to HHG in Xe driven by our 35-fs, 800-nm driver which is \(\sim35\ \text{eV}\). The achieved cutoff energy is not high enough to satisfy the \(\lambda^2\) scaling considering the ionization potential of Xe, 12.1 eV, but the phase matching cutoff, scaled with \(\lambda^{1.6-1.7}\) [5], for our experimental conditions is in fact calculated to be \(\sim94\ \text{eV}\), indicating near phase-matched HHG in our experiment.

The HHG efficiency per harmonic is estimated to be \(\sim10^{-10}\), which can be improved with higher pressure. However, this efficiency is already consistent with the wavelength scaling of the HHG efficiency starting from \(10^{-2}-10^{-8}\) at 90 eV with an 800-nm driver in Ne or He [27]. In the following attempt, we also observed HHG spectra with Kr and a systematic experimental study is under way. Further scaling of photon energy towards the water window region at a kHz repetition rate will be possible with a high-pressure gas jet or a gas cell filled with He or Ne, as demonstrated recently with a 10-Hz OPA system [8].

5. Conclusion

We demonstrated a mJ-level ultrabroadband 2.1-μm OPCPA operating at 1 kHz repetition rate, pumped by a novel high-energy cryogenic Yb:YAG CPA chain and a Nd:YLF CPA-chain. High order harmonics were generated in Xe with an extended cutoff wavelength of 85 eV. Optical synchronization of the OPCPA signal and two pump laser pulses was achieved using an octave-spanning Ti:sapphire master oscillator and several fiber pre-amplifiers. We obtained 0.85 mJ of output energy from the third OPCPA stage with a SF noise of \(-9\%\) and compressed the pulse duration to <32 fs or 4.5 optical cycles with good beam profile.
Dispersion control was achieved up to fourth-order dispersion and CEP stability was confirmed using an $f$-3$f$ spectral interferometer.

In our first HHG experiment driven by this 2.1-$\mu$m OPCPA source, we detected and maximized XUV signals generated in Xe using an amplified XUV photodiode over the Al window and then measured the high harmonic spectrum. Extension of cutoff energy beyond 85 eV was demonstrated with an efficiency of $\sim 10^{-30}$ per harmonic, indicating that phase matched HHG was achieved. Our result shows the feasibility of further scaling of photon energy and flux using an ultrabroadband CEP-stable kHz OPCPA system as a next-generation source for high-field physics and attosecond science.

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