Fiber-amplifier-pumped, 1-MHz, 1-µJ, 2.1-µm, femtosecond OPA with chirped-pulse DFG front-end

YIZHOU LIU,1,2,3 PETER KROGEN,3 KYUNG-HAN HONG,3 QIAN CAO,1,2 PHILLIP KEATHLEY,3 AND FRANZ X. KÄRTNER1,2,3,4,*

1Center for Free-Electron Laser Science, DESY, Notkestraße 85, D-22607 Hamburg, Germany
2Physics Department, University of Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany
3Research Laboratory of Electronics, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, Massachusetts 02139, USA
4The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, Hamburg 22761, Germany
*franz.kaertner@cfel.de

Abstract: High-repetition-rate, high-power, few-cycle mid-infrared lasers with carrier-envelope phase (CEP) stabilization are ideal driving sources for studying strong-field nonlinear processes, such as strong-field driven electron emission, solid-state high-harmonic generation, and nonlinear microscopy. Here, we report on a 1-MHz, 1-µJ, femtosecond, 2.1-µm optical parametric amplifier (OPA), pumped by a Yb-doped fiber chirped-pulse amplifier (CPA) and seeded by a chirped-pulse difference-frequency generation (DFG) front-end providing positively chirped 2.1-µm signal pulses. The home-built multi-stage 1030-nm Yb-doped fiber CPA pump laser generates >55-µJ near-transform-limited (245-fs) pulses at 1 MHz repetition rate using a novel 4-pass all-fiber stretcher/front-end for careful dispersion/spectral management. The chirped-pulse DFG scheme is achieved by wave-mixing the 1030 nm pump pulse with a dispersive wave at 645–735 nm generated in a photonic crystal fiber, allowing passive CEP stability of the 2.1-µm pulses. The 2.1-µm pulse is amplified to 1 µJ in a two-stage dispersion-managed optical parametric amplifier (OPA) with a pump energy of ~21 µJ resulting in 95-fs pulses with nice beam profile without additional pulse compression. Multi-µJ, sub-30 fs pulses can be obtained at full pump energy and additional dispersion compensation. The fiber-amplifier-based mid-infrared OPA can be directly applied to high-harmonic generation in solids and optical-field-driven nanophotonic devices and is a compact front-end for a future high-power, high-repetition-rate, long wavelengths CEP-stabilized source for gas-phase high-order harmonic generation.

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1. Introduction

In recent years, due to its particular importance of strong-field light-matter interactions, demands for high repetition rate few-cycle CEP-stabilized laser sources have rapidly increased, resulting in tremendous technological advancements. High-repetition-rate (MHz-level), high-power laser sources with few-cycle µJ-level pulses can significantly enhance the statistics in strong-field applications by increasing data acquisition rates and therefore improving the signal-to-noise ratio (SNR). For example, attosecond time-resolved spectroscopy using high-harmonic generation (HHG) and space-charge-limited ultrafast photoelectron spectroscopy can benefit from high-repetition-rate sources [1–5]. Solid-state HHG can also greatly benefit from high-repetition-rate pump lasers because it requires only sub-µJ-level pump pulses [6,7]. For ultrafast optical-field-driven electron emission from nanostructures, the applicable pulse energy is limited by damage of the devices, making a high repetition rate necessary for detecting the photo-electron current with a good SNR [10–14]. Furthermore, few-cycle mid-infrared (MIR) laser sources with MHz-level repetition rates are ideal for studying optical tunnelling from metal and semiconductor nanostructures.
because the ponderomotive potential scales with the wavelength squared and the Keldysh parameter is reduced for longer wavelengths at a given laser intensity [10,11]. High-repetition-rate near-infrared and MIR pulse trains have also been used to improve the SNR for the detection of electrons and excitons in graphene [8,9]. Therefore, the development of high-power MIR sources with high repetition rate is very important for these emerging research areas.

High-power, few-cycle MIR sources based on optical parametric amplifiers (OPAs) and optical parametric chirped-pulse amplifiers (OPCPAs) have been routinely implemented at various repetition rates and average powers for strong-field applications [15–24]. Average power scaling is relatively straightforward with the OPA/OPCPA technique because the parametric process does not involve energy storage in a nonlinear crystal and the residual absorption of pump/signal and idler beams in a nonlinear crystal is typically low. Compared to mJ-level, few-cycle MIR OPA/OPCPAs with kHz repetition rates, there are unique challenges with realizing MIR OPA/OPCPAs with MHz-level or sub-MHz-level repetition rate due to the higher pump power in a tight focusing geometry which induces thermal effects. In refs [16,17], few-cycle µJ-level OPCPAs at 800 nm wavelength pumped by thin disk laser amplifiers or fiber chirped-pulse amplifiers (CPAs) were pushed in repetition rate to the range from 500 kHz to 1 MHz. In ref [23], a 100-µJ, 2-µm OPCPA pumped by a ~1-µm Yb-doped slab amplifier (Innoslab) was demonstrated at a repetition rate of 100 kHz. Previously a 3.2-µm OPCPA producing 9-cycle pulses with a pulse energy of 1.2 µJ at a repetition rate of 100 kHz [24] was also demonstrated. Nevertheless, there is still a lack of robust and easy-to-implement high-power few-cycle µJ MIR OPA/OPCPA with a repetition rate in the MHz range. In addition, CEP stabilization is important for many strong-field applications in the few-cycle regime. The source discussed here is based on a passively CEP-stabilized architecture using difference-frequency generation (DFG) from a common source, which is a well-established technique for obtaining CEP-stable optical pulses without using electronic feedback or a CEP-stable laser oscillator [25–32]. The combination of DFG and OPA/OPCPAs has been an excellent choice for generating high-repetition-rate, passively CEP-stable, few-cycle optical pulses [27–32].

A proper pump laser is a key for developing a few-cycle OPA/OPCPA. Fiber amplifiers are suitable for average power scaling of moderately energetic pulses at high repetition rates with a compact architecture [33–38]. Furthermore, they ensure excellent long-term power stability and output beam quality, both of which are critical for pumping OPA/OPCPA systems. In this paper, we report on a Yb-doped fiber-amplifier-pumped, femtosecond, 2.1-µm OPA generating 1-µJ pulses with an excellent beam profile at a 1-MHz repetition rate. For pumping the MIR OPA system, we developed a 1.03-µm, 1-MHz Yb-doped fiber CPA with an all-fiber stretcher and front-end. A chirped-pulse DFG scheme, based on wave-mixing of the 1.03-µm pump pulse and the dispersive wave created during supercontinuum generation in a photonic-crystal highly nonlinear fiber (HNF) in the wavelength range 645-735 nm, was employed to provide passively CEP-stable MIR (2.1-µm) seed pulses. Here, we demonstrate a dispersion-managed MIR OPA, which compresses the pulses during the amplification process without the need for a pulse compressor. In section 2, we describe the construction of the 1.03-µm fiber CPA system with an all-fiber stretcher to generate >55-W (>55 µJ), 1-MHz, 250-fs laser pulses amplified by an aero-gain rod-type fiber amplifier. In section 3, we describe the 1-MHz, 1-µJ, 95-fs, 2.1-µm dispersion-managed OPA system and its chirped-pulse DFG seed with a passively CEP-stable architecture. Finally, conclusions and outlook are discussed in section 4.

2. 1 MHz, 55 µJ, 1 µm Yb-doped CPA pump source

We have developed a 1.03-µm Yb-doped all-fiber CPA for both providing the 2.1-µm seed signal and pumping the 2.1-µm OPA stages. We demonstrate that an all-fiber stretcher built with polarization-maintaining single-mode fiber (PM SMF) and dispersion-compensating
fiber (DCF) can realize the full compensation of up to third-order dispersion (TOD) and partial compensation of fourth-order dispersion (FOD) when using a grating compressor. Using this combination of PM SMF, DCF and a grating pair we compress the ~800 ps chirped pulses to <300 fs. The CPA source consists of a mode-locked Yb-doped fiber laser, dispersion management system, and multi-stages of amplification (Fig. 1). The oscillator was a home-built, 28-MHz, 1-μm fiber oscillator based on nonlinear polarization evolution (NPE) [44–48]. The output power of the oscillator was decreased to 1 mW using a polarizing beam splitter (PBS) and a half wave-plate (HWP), to avoid nonlinear phase accumulation in the DCF. As shown in Fig. 2(a), this seed pulse was stretched to ~800 ps in an all-fiber four-pass stretcher. Shown in Fig. 2(b), the stretcher contains 150 m of PM980 fiber ($\beta_2 = 0.023$ ps$^2$, $\beta_3 = 0.00046$ ps$^3$/m), a PM circulator, a fiber PBS, a fiber polarization controller, 66-m long section of non-PM DCF (OFS stretcher-fiber FemtoComp, $\beta_2 = 0.12$ ps$^2$, $\beta_3 = -0.00096$ ps$^3$/m), a non-PM Yb-doped amplifier, and a Faraday mirror. The lengths of the PM980 fiber and DCF were selected to compensate the group-delay dispersion (GDD) and TOD and nearly compensate the FOD together with the grating-based compressor after the power amplifier. An Yb-doped fiber amplifier was inserted in the stretcher to compensate the 10 dB insertion loss of the stretcher and suppress the amplified spontaneous emission (ASE) in the subsequent amplifiers. A Faraday mirror is used at one end of the DCF which is non-PM to compensate for the polarization rotation induced in the 66 m length of fiber and to introduce a 90-degree polarization rotation per round trip through the DCF. By carefully releasing the coiling stress and tuning the fiber polarization controller on the DCF, a 20 dB polarization extinction ratio is achieved. Additional stretching is achieved in the DCF using a fiber PBS and a fiber mirror, which results in the 4-pass configuration (2 roundtrips). The stretched pulse is finally isolated using a PM circulator and then sent to the subsequent amplifiers. The B-integral accumulated in the stretcher is estimated to be ~1 rad, while providing 31 ps$^2$ of total GDD with 1 mW (36 pJ at 28 MHz) to seed subsequent amplifiers.
The 1-mW, 28-MHz, stretched seed pulse train is amplified by a two-stage PM SM Yb-doped fiber pre-amplifier to 100 mW (~3.6 nJ). Then the repetition rate of the amplified pulses is decreased to our target repetition rate, 1 MHz, for the power amplification using an acousto-optic modulator (AOM). The three-stage power amplifier chain employs three different Yb-doped gain fibers: a 0.5-m-long PM SM gain fiber (CorActive YB-501PM), a 4-m-long, 25-μm core-diameter, PM double-cladding gain fiber (Nufern PLMA-YDF-25/250), and a 0.8-m-long, 85-μm core-diameter, rod-type gain fiber (NKT Aerogain-ROD). We are able to achieve an average output power of >90 W with 38°C of pump diode working temperature at 1 MHz. For a long-term operation, we set the output power at 55 W with 25°C of pump diode working temperature. The B-integral for the amplifier chain is conservatively estimated to be ~4 rad at 55 W and ~6 rad at 90 W of output power.
Since the gain bandwidth of the rod-type gain fiber of the final amplifier is in the range of 1030-1040 nm and the effective gain bandwidth of the PM double-cladding gain fiber is also above 1030 nm, the grating compressor is optimized at this wavelength range and therefore the spectral components below 1030 nm are more susceptible to high-order spectral phase distortion. By removing the short wavelength component using a long-pass filter (LPF), we were able to observe a better pulse profile after compression. Furthermore, shaping the spectrum more parabolic way makes the curve of the phase more flat for easier compression [49]. To check the influence of spectral contents on the pulse compression, we calculate the compression quality without and with the LPF, as shown in Figs. 3(a) and (c), respectively. With the original spectrum from the laser oscillator (Fig. 3(b)) the non-negligible pedestals remain after compression, which reduce peak power of the compressed pulse to ~66% of the calculated peak power of the TL pulse. On the other hand, by using a sharp LPF to eliminate the wavelength range below 1030 nm like Fig. 3(d), we confirm that the compressed pulse could nearly match the TL pulse as shown in Fig. 3(c). The peak power of the compressed filtered pulse is as high as 93% of the TL pulse.

For the experimental implementation, we carefully chose the length of gain fibers and coatings of the elements to effectively use them as a LPF at 1030 nm and to optimize the compression. Figure 4(a) shows the spectra of the oscillator and the final output of the amplified 1030-1040 nm pulses with the effective LPF at 1030 nm. The throughput efficiency of the reflective grating compressor (1480 groves/mm) is 95%. Figure 4(b) shows the measured intensity autocorrelation trace of the compressed pulse and the Gaussian fitting result. After careful optimization of the grating spacing and incidence angle, we obtained the shortest pulse duration at 48° of angle of incidence and 0.899 m of normal distance between two gratings. The measured duration of autocorrelation trace was 345 fs in full-width at half-maximum (FWHM), corresponding to a duration of 245 fs in FWHM with a Gaussian pulse. Based on the spectrum of the compressed laser pulse, the calculated TL pulse duration is 190 fs. We estimate ~75% of the power contained in the central peak of the pulse. The small pedestals in the autocorrelation are attributed to the residual FOD, B-integral, and mode beating effects in the power amplifier. We also obtained an excellent near-field beam profile, shown as inset of Fig. 4(b).

![Fig. 4. (a) The pulse spectrum at the output of the 1-MHz amplifier and (b) a measured autocorrelation trace of the amplifier pulses indicating a pulse width of 245 fs FWHM. Gaussian fit to the measured pulse in red.](image)

3. 1 MHz, 2.1 µm OPA with chirped-pulse DFG seed

We have built a 1-MHz µJ-level 2.1-µm degenerate OPA [20,21] pumped by the 1-MHz 55-µJ 1.03-µm Yb-doped CPA system. As illustrated in Fig. 5, the 2.1-µm OPA consists of three main parts: (1) a chirped-pulse DFG stage for CEP-stable 2.1-µm signal generation, (2) the
first-stage OPA for pre-amplification in MgO:PPLN and (3) the second-stage OPA for power amplification in β-barium borate (BBO).

The DFG scheme with the pump and signal from a common source is a well-established architecture for generating passively CEP-stable pulses [16–33]. Intrapulse DFG is the simplest method of realizing this method, but it requires an ultrabroadband laser unlike our 1.03-μm Yb-doped fiber CPA. Therefore, we implemented an additional beam line to generate a coherent dispersive wave from a photonic-crystal fiber (PCF) and then wave-mixed the dispersive wave with the original beam for chirped-pulse DFG, which inherently preserves the CEP stability. To generate ~2.1 μm (DFG idler) with 1.03 μm (DFG signal), we need to have a dispersive wave at ~690 nm (DFG pump). We used a 4-cm-long 3.2-μm-core-diameter PCF (NKT, HNF 945) with a zero-dispersion wavelength at 945 nm for this purpose. About 20 mW of the 1.03-μm laser power was coupled into HNF 945 to generate a dispersive wave at 645-735 nm with 1 mW of output power. The dispersive wave in the visible was positively chirped after propagating through the HNF 945 fiber [39–43]. After mode-matching, the beam diameters for both the 690-nm and 1.03-μm beam were set to ~0.8 mm before the nonlinear crystal. A 1-mm-thick type-I BBO crystal cut normal at DFG phase matching angle, θ = 20.5° was used for the chirped-pulse DFG process. The polarization directions of the 1.03-μm beam (ordinary) and the 690-nm beam (extraordinary) were set to satisfy the type-I phase matching condition to generate the idler beam at 2.1 μm (ordinary). Both the 690-nm beam (DFG pump) and 1.03-μm beam (DFG signal) are collinearly focused using an f = 35 mm lens into the BBO crystal to generate positively chirped 2.1-μm pulses (idler beam). About 1.05 W of the 1.03-μm beam was used to drive the chirped-pulse DFG stage and generate ~3 μW (or 3 pJ) broadband 1-MHz, 2.1-μm pulses. While the passive CEP stability of 2.1 μm pulse is physically ensured in the DFG process, an additional active feedback loop could enhance the stability because the timing jitter between the DFG pump and signal arms can be induced by mechanical stability, thermal drift, and amplitude-to-phase noise.

![Fig. 5. Schematic of the cascaded 1-MHz, 2.1-μm OPA system. CM: curve mirror, DM: dichroic, DL: delay line, TFP: thin film polarizer. These two inserts show the beam profile of the amplified 2.1-μm signal beams.](image)
Fig. 6. (a) The spectrum of the 690-nm dispersive wave, (b) the spectrum of the 1-µm pump pulses, (c) the spectrum of generated broadband 2.1-µm signal pulses during the chirped-pulse DFG in the 1 mm thick BBO crystal, (d) numerically calculated spectrum of the chirped pulse DFG in the 1 mm thick BBO crystal.

The optical spectrum of the 690-nm dispersive wave (DFG pump), 1.03-µm pulse (DFG signal), and resulting 2.1-µm pulse (DFG idler) is shown in Figs. 6(a), (b), and (c), respectively. The 2.1-µm spectrum supports a TL duration of 20 fs. The dispersive wave has a spectral bandwidth of 90 nm in FWHM centered at 690 nm which is phase matched from the zero dispersion wavelength of 945 nm with the HNF 945 fiber. The phase matching wavelength for DFG is ~2070 nm. The average power of the generated 2.1-µm DFG idler beam was ~3 µW, corresponding to ~1% of conversion efficiency from 690 nm to 2.1 µm from DFG. The measured spectrum of the 2.1-µm signal pulse is shown in Fig. 6(c) matching well with the simulated Gaussian spectrum shown in Fig. 6(d). The FWHM of the 2.1-µm signal pulse spectrum was ~300 nm. The pulse duration of the generated positively chirped 2.1-µm pulse was calculated to be ~192 fs.

Since the 2.1-µm signal and 1.03-µm pump beams for OPA are relatively short with 192 fs and 245 fs, respectively, unlike OPCPA a highly dispersive stretcher and compressor setup is not necessary. Instead, both of OPA stages were designed to be dispersion managed. The MgO:PPLN crystal and BBO crystal can provide negative chirp to the 2.1 µm pulses during the amplification process to compensate the positive chirp experienced from the chirped-pulse DFG process. MgO:PPLN is known to have one of the highest nonlinear coefficients, $d_{eff} = 14$ pm/V, and therefore suitable for broad phase matching bandwidth and high gain amplification [43], while the damage threshold is relatively low. Therefore, a thin MgO:PPLN is an excellent choice for a high-gain OPA stage as a pre-amplifier. For the first OPA stage, the spot size of the 2.1-µm signal beam and the 1.03-µm pump beam was 100 µm and 130 µm in diameter, respectively, at the 3-mm-long MgO:PPLN crystal. The horizontal polarization was used for both the pump beam and signal beam, satisfying the (type-0) quasi-phase matching condition. The difference of incidence angle between the 1.03-µm pump and 2.1 µm signal beams was designed to be 1.9° for spatial separation. Temporal overlap and spatial overlap
were ensured using a fast photodetector, a pin hole and a beam profiler. Using $\sim 1.5$ W ($\sim 1.5 \mu\text{J}$) of the 1.03-µm pump power in the MgO:PPLN pre-amplifier, we amplified the 2.1 µm signal pulses to $\sim 22$ nJ at 1 MHz. The beam profile of the 2.1-µm signal beam after the signal amplifier is shown as an insert in Fig. 5 next to PPLN. The pump-to-signal efficiency was $\sim 1.6\%$.

For the second stage of OPA we used a BBO crystal as the damage threshold of the BBO crystal is much higher than that of the MgO:PPLN crystal and BBO can handle a much higher aperture size for energy and average-power scaling. The 2.1-µm, 22-nJ pulses were amplified to the µJ level at 1 MHz in a 4-mm-thick type-I BBO crystal. The polarization directions of the 2.1-µm signal beam (ordinary) and the 1.03-µm pump beam (extraordinary) were set to fulfill the type-I phase matching condition. The 22-nJ, 2.1-µm beam was set to $\sim 300$ µm in diameter inside the crystal using an $f = 150$ mm CaF$_2$ lens, while the 1.03-µm pump laser was focused with an $f = 500$ mm curved silver mirror to 350 µm of spot size in diameter inside the crystal. The difference of incident angle of these two beams was 1.5°. Since we were close to the damage threshold of the crystal, only 21 W of the femtosecond 1-µm pump laser, or about half of the available pump power, was used to amplify the 20-nJ, 2.1-µm signal pulses to $\sim 1$ µJ. The pump-to-signal efficiency was $\sim 5\%$. The beam profile at the output port is shown as an inset in Fig. 5. For further energy scaling of the 2.1-µm pulses, the remaining 20 W of pump power can be used to amplify the pulse energy up to the multi-µJ level.

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**Fig. 7.** (a) Full-wavelength-range spectrum includes the 690-nm dispersive wave (red), 1035-nm pump (green), 2.1-µm signal (blue) and 2.1-µm amplified output (black). (b) The measured IAC of the amplified pulse (red curve) and the calculated IAC to fit the measured results (blue curve) assuming a linearly chirped pulse with the measured 2.1-µm spectrum after amplification in (a). (c) The superfluorescence spectrum of MgO:PPLN OPA measured after blocking the 2.1-µm seed at 2 W pump power. (d) Amplified signal average power versus pump average power. Inset shows beam profile of the output signal beam.
We measured the spectral and temporal characteristics of amplified pulses. In Fig. 7(a) various spectra along the MIR generation are shown. Amongst them the amplified 2.1-\(\mu\)m signal pulses (black curve), generated 2.1 \(\mu\)m signal pulses from the chirped pulse DFG (blue curve), the 690 nm dispersive wave (red curve) and the 1.03-\(\mu\)m pump pulse (green curve). There is a spectral narrowing effect in the two-stage OPA. Since the 2.1-\(\mu\)m signal pulse (192 fs) is shorter the 1.03-\(\mu\)m pump pulse (245 fs), we don’t expect a bandwidth loss caused by the pulse width mismatch. For comparison with the amplified 2.1-\(\mu\)m signal spectrum, the superfluorescence spectrum from the 3-mm-thick MgO:PPLN of the first stage OPA is shown in Fig. 7(c), which is a direct indication of the gain bandwidth. Spectral narrowing is already observed after the first-stage OPA which must be caused by the limited OPA gain bandwidth of MgO:PPLN. Considering the limited phase matching bandwidth and the MIR transmission loss inside the BBO crystal, we believe wavelengths longer than 2.3 \(\mu\)m were not efficiently amplified in the two OPA stages. Wavelengths outside the transmission window of the BBO crystal were largely inhibited in the first stage amplifier. Therefore, we did not notice any thermal issues caused by absorption in the second amplifier. The chirp of the 2.1-\(\mu\)m pulse was flipped from positive to negative in the two-stage OPA due to the material dispersion, so that the chirped pulse was slightly over-compressed during the amplification process. Figure 7(b) shows the measured interferometric auto-correlation (IAC) trace (red curve) of the final 2.1-\(\mu\)m output pulses and the calculated IAC trace (blue curve) when fitting the IAC of a linearly chirped pulse with the measured 2.1-\(\mu\)m spectrum of the black curve in Fig. 7(a). From this measurement and the fitting we can infer that the output pulse has a width of 94.5 fs with \(\pm\)1200 fs\(^2\) of residual GDD. The pulse is expected to be compressed to \(\pm\)1.2 times TL pulse duration, 24.5 fs, if de-chirped by the propagation through a 1.2-mm-thick Si window. As shown earlier in [22], the \(\mu\)J-level, sub-30-fs, 2.1-\(\mu\)m pulses can be further self-compressed to sub-two-cycle duration with nonlinear self-compression in the CaF\(_2\) for the application to phase-sensitive strong-field experiments with nano-structures in few-cycle regime. The 2.1-\(\mu\)m output power versus 1.03-\(\mu\)m pump power is shown in Fig. 7(d), where we demonstrate >1 W output power or \(1 \mu\)J output energy at 1 MHz with an excellent beam profile. Even without full compression, we generated sub-100-fs, 1-\(\mu\)J, 1-MHz pulses, well-suited for many applications requiring high-power, high-repetition-rate, ultrabroadband infrared optical pulses.

4. Conclusion

We demonstrated a 1-MHz, 50-W, 250-fs, 1-\(\mu\)m pump laser as an all-fiber CPA system with dispersion management and spectral content management. Chirped-pulse DFG was then used to generate passively-CEP-stable, 1-MHz, 2.1-\(\mu\)m seed pulses. Using a two-stage dispersion-managed OPA system, we amplified the 2.1-\(\mu\)m pulses to the \(\mu\)J-level while maintaining an excellent beam profile and measured an output pulse duration of \(\sim\)95 fs. We note that this source has the potential to be extended even further into the MIR region with shorter pulse durations [22,50]. The high-repetition-rate, high-power, few-cycle, passively-CEP-stable, MIR laser source has a wide range of scientific applications in several emerging areas, such as improving the signal to noise ratio in attosecond time-resolved spectroscopy using high-harmonic generation (HHG), avoiding space charge effects in photoelectron spectroscopy [1–5], increasing solid-state HHG photon flux [6,7], electron dynamics and exciton excitation in graphene [8,9], and optical-field-driven electron emission from nanostructures [10–14].

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