# Carrier-envelope-phase-stable, 1.2 mJ, 1.5 cycle laser pulses at 2.1 μm

**Yunpei Deng,[1,*] Alexander Schwarz,[1] Hanieh Fattahi,[1] Moritz Ueffing,[1] Xun Gu,[1] Marcus Ossiander,[1,2] Thomas Metzger,[1] Volodymyr Pervak,[3] Hideki Ishizuki,[4] Takunori Taira,[4] Takayoshi Kobayashi,[5,6] Gilad Marcus,[1,7] Ferenc Krausz,[1] Reinhard Kienberger,[1,2] and Nicholas Karpowicz[1]**

[1]*Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, Garching 85748, Germany*
[2]*Department of Physics, Technische Universität München, James Franck Strasse, Garching 85748, Germany*
[3]*Ludwig-Maximilians-Universität München, Am Coulombwall 1, Garching 85748, Germany*
[4]*Institute for Molecular Science, 38 Nishigonaka, Myodaiji, Okazaki 444-8585, Japan*
[5]*Advanced Ultrafast Laser Research Center, University of Electro-Communications, 1-5-1 Chofugaoka, Chofu, Tokyo 182-8585, Japan*
[6]*Japan Science and Technology Agency (JST), Core Research for Evolutional Science and Technology (CREST),*
*5 Sanbancho, Chiyoda-ku, Tokyo 102-0075, Japan*
[7]*The Benin School of Engineering and Computer Science, The Hebrew University of Jerusalem, Jerusalem 91904, Israel*
**Corresponding author: yunpei.deng@mpq.mpg.de*

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We produce 1.5 cycle (10.5 fs), 1.2 mJ, 3 kHz carrier-envelope-phase-stable pulses at 2.1 μm carrier wavelength, from a three-stage optical parametric chirped-pulse amplifier system, pumped by an optically synchronized 1.6 ps Yb:YAG thin disk laser. A chirped periodically poled lithium niobate crystal is used to generate the ultrabroad spectrum needed for a 1.5 cycle pulse through difference frequency mixing of spectrally broadened pulse from a Ti:sapphire amplifier. It will be an ideal tool for producing isolated attosecond pulses with high photon energies. © 2012 Optical Society of America

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Isolated soft x-ray 50–150 eV attosecond pulses are currently ideal tools to investigate dynamics with atomic temporal resolution and nanometer spatial resolution [1]. Typically, attosecond pulses are produced via high harmonic generation (HHG) in noble gases, driven by intense carrier-envelope phase (CEP) stabilized few-cycle laser pulses at a wavelength around 800 nm [2,3]. In order to investigate processes with even higher temporal resolution or study the dynamics of core electrons, higher photon energies and shorter attosecond pulses are needed. Both of these can be obtained using lasers with longer central wavelengths. From the perspective of a single atom's response to the field, increasing wavelength allows for an extension of the cutoff photon energy at the same intensity, described in atomic units by $E_{\text{cut-off}} \cong I_p + 3.17\frac{I\lambda^2}{16\pi^2 c^2}$ where $I$ is the laser intensity and $I_p$ is the ionization potential of the atom [4]. This comes with the trade-off of reduced photon flux due to increased dispersion of the electron wave packet as it takes more time to re-collide with the parent ion. However, recent work has suggested that this decrease in the single-atom efficiency may be compensated by more favorable phase matching, and thus great interest in long-wavelength sources remains [5–8]. So far, few-cycle, high-intensity IR optical parametric amplifier (OPA) and optical parametric chirped-pulse amplifier (OPCPA) sources have been reported with central wavelengths near 1.8 μm [9,10], 2 μm [11–13], 3 μm [14], and 4 μm [15]. To generate an isolated attosecond pulse by HHG, a sub-two-cycle pulse is highly desirable. Currently, multicycle mid-infrared pulses generated by OPA can be spectrally broadened and compressed to few-cycle durations by self-phase modulation in hollow fibers, but with scalability limited by the transmission efficiency of the fiber. In this Letter, we report a 2.1 μm OPCPA laser system pumped by a Yb:YAG thin-disk amplifier. This system produces 1.2 mJ, CEP-stable, 10.5 fs (1.5 optical cycle) pulses, which are the shortest millijoule level pulses ever generated in this spectral range. It will be an ideal tool for generating isolated attosecond pulses with much higher photon energies. It also can be scaled straightforwardly with pump energy to reach terawatt powers.

In order to achieve millijoule energies and near-single-cycle pulses with an OPCPA system, (i) a broad bandwidth seed and (ii) an amplification bandwidth spanning nearly one octave are needed.

To meet the first requirement, we generate a broadband seed by difference-frequency generation (DFG) in a chirped MgO-doped periodically poled lithium niobate (PPLN) crystal, which gives an octave-spanning spectrum by matching the quasi-phase-matching conditions to the evolving envelope of the laser pulse as it passes through the crystal, providing a wider phase matching region than unchirped PPLN due to the increased range of $k$ vectors determined by the varying grating period [16]. To obtain the input pulse for the DFG process, a 30 μJ 25 fs 800 nm pulse (4% of the output of a commercial Femtopower Pro multipass Ti:sapphire amplifier) is coupled into a hollow-core fiber filled with 5-bar-pressure krypton gas, to be spectrally broadened by means of self-phase-modulation, providing spectral components from ~500 to ~1050 nm. The spectrally broadened pulse is temporally compressed with chirped mirrors, and the low- and high-frequency components of the pulse are subsequently mixed in chirped PPLN with a quasi-phase-matching period changing linearly from 8 to 11 μm, generating a broadband DFG signal centered around 2.1 μm, from <1.5 to >2.8 μm. The DFG process

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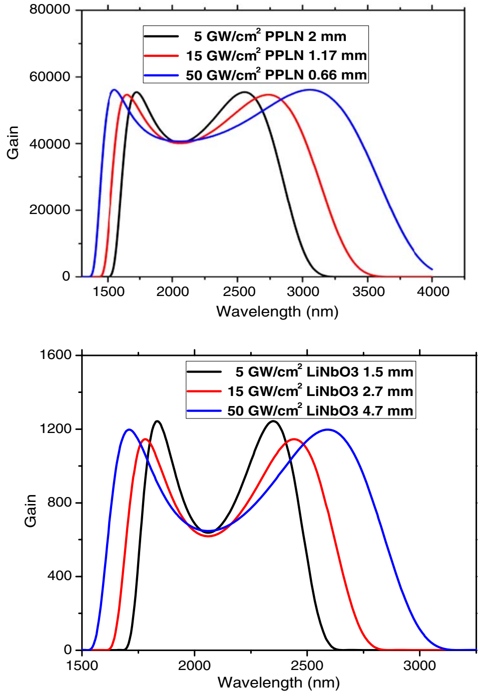

Fig. 1. (Color online) Calculated unsaturated gain profiles of PPLN, and $\mathrm{LiNbO_3}$, with pump intensities corresponding to optimal safe operating conditions with 1.6, 25, and 160 ps pump durations, ($50\ \mathrm{GW/cm^2}$, $15\ \mathrm{GW/cm^2}$ and $5\ \mathrm{GW/cm^2}$, respectively) with crystal lengths set such that the gain at the central wavelength is constant. The damage threshold is $\sim 100\ \mathrm{GW/cm^2}$ for $\mathrm{LiNbO_3}$ with 1.6 ps pulses at 1030 nm.

ensures CEP stability [17,18], which is preserved in the subsequent OPA processes.

For broadband amplification of a picojoule-level seed pulse to the millijoule level, an amplification factor of about $10^8$ is needed and gain narrowing becomes an issue. The OPA gain spectral narrowing is primarily a consequence of the phase mismatch ($\Delta kL$), where $\Delta k$ is the wavevector mismatch between signal, pump, and idler, and $L$ is the crystal length. The phase mismatch is a function of the crystal length. With a shorter crystal, the OPA gain bandwidth can be wider, but the gain also depends on the crystal length as proportional to $\alpha\sqrt{I} * L$ In order to support this bandwidth with a nearly constant gain level, shorter crystal lengths and higher pump intensities are needed [19]. The intensity cannot be increased arbitrarily due to the damage threshold of the nonlinear crystals, but this threshold increases with the inverse of the square root of the pulse duration. Therefore, with a shorter pump pulse combined with a thinner crystal, the OPCPA system can achieve more gain bandwidth at the same gain level. With a 1.6 ps pulse, the pump intensity can be more than $50\ \mathrm{GW/cm^2}$. This pump intensity can support a high gain with a wide bandwidth close to one octave, as shown in Fig. 1.

The full schematic of the OPCPA system is shown in Fig. 2. The pump laser is a home-built, 1.6 ps, 1030 nm Yb:YAG thin-disk regenerative amplifier delivering pulses up to 20 mJ at 3 kHz repetition rate [20]. To optically synchronize the pump pulse and seed pulse, the pump laser is seeded by the same Ti:sapphire oscillator (Femtolasers Rainbow). The oscillator pulse energy within the gain bandwidth of Yb:YAG at 1030 nm, diverted for optical seeding of the regenerative amplifier, is 2 pJ. With 1.6 ps pump pulses, 1–2 mm bulk $\mathrm{LiNbO_3}$ crystals are expected to deliver high parametric gain with an amplification bandwidth approaching an octave. On the other hand, the synchronization of pump and seed pulses is more delicate. To account for this, we used a spectrally resolved cross-correlation technique [21] combined with active stabilization, which allows us to obtain an RMS timing jitter of 24 fs, 1.5% of the pump pulse duration.

To ensure the broadest possible amplification bandwidth, three nearly degenerate OPA stages are used. The signal and pump beams are crossed at a small angle (~3°) to spatially separate them after amplification. 350 μJ of the pump laser pulse is used to amplify the seed with a 400 μm beam diameter (FWHM) in the first stage, a 2 mm thick PPLN crystal with 29.9 μm poling period. The output signal energy is 15 μJ in this stage. 15% of the pump beam is then used with 2 mm beam diameter in the second stage, which again consists of a 2 mm thick PPLN crystal with 29.9 μm period. The third stage employs a 1.5 mm thick MgO-doped $\mathrm{LiNbO_3}$ crystal. The signal energy is boosted to 1.2 mJ using the remaining 14 mJ

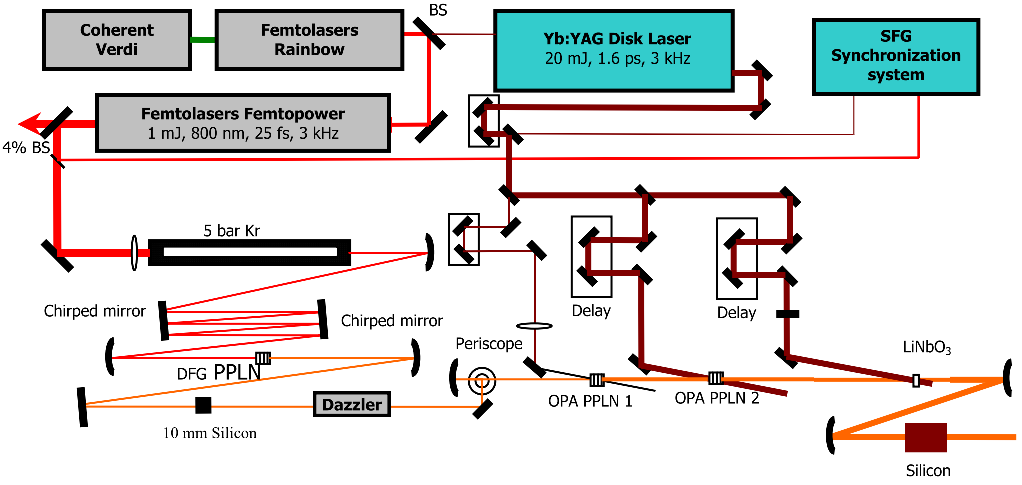

Fig. 2. (Color online) Scheme of the IR OPCPA.

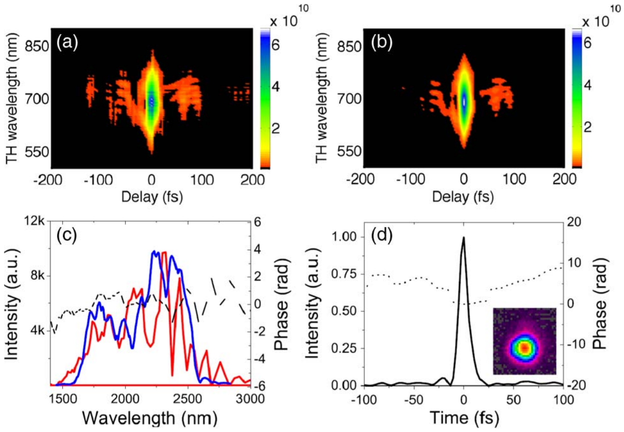

Fig. 3. (Color online) THG FROG measurement results of the compressed 10.5 fs pulse. (a) Measured FROG trace, (b) retrieved FROG trace, (c) measured (blue) and retrieved (red) spectral intensity and phase (dashed black), and (d) measured temporal intensity and phase. Inset: measured spatial intensity profile after the third stage.

pump pulse with 4 mm beam diameter. Superfluorescence accounts for less than 5% of the total output, as determined by the unseeded output power. PPLN could support much more bandwidth than bulk $LiNbO_3$ crystal for the same gain, but is not currently available with apertures large enough to support the third OPA stage.

In order to extract the maximum energy from the pump pulse while maintaining bandwidth, the durations of the seed and pump pulses should be matched. In this laser system, the seed pulse is stretched to 700 fs by a Dazzler acousto-optic pulse shaper. A 10 mm silicon bulk is placed before the Dazzler to positively chirp the pulse and match the laser pulse duration to the Dazzler crystal length, in order to achieve the maximum diffraction efficiency. The amplified IR pulse is recompressed by a 1.5 mm thick silicon wafer. The compressed and amplified pulse of the OPCPA system is characterized by a home-made third-harmonic-generation (THG) frequency-resolved optical gating (FROG) apparatus, as shown in Fig. 3. Using the Dazzler, the pulse is compressed to 10.5 fs at FWHM, which is very close to the Fourier limit of 10 fs. At 2.1 μm, this pulse duration is around 1.5 cycles. Such a short, millijoule-level pulse is an ideal tool for producing high photon energy, isolated attosecond pulses via HHG. Furthermore, as shown in the previously reported f–to-3f nonlinear interferometry measurement [11], the CEP of the amplified signal is stable, which is also important for HHG.

In summary, we have demonstrated the generation of CEP-stable, 1.5 cycle (10.5 fs), 1.2 mJ pulses at 2.1 μm carrier wavelength and 3 kHz repetition rate, from a broadband OPCPA system pumped by a 1.6 ps short pulse duration laser.

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## References

1. F. Krausz and M. Ivanov, Rev. Mod. Phys. **81**, 163 (2009).
2. E. Goulielmakis, V. S. Yakovlev, A. L. Cavalieri, M. Uiberacker, V. Pervak, A. Apolonski, R. Kienberger, U. Kleineberg, and F. Krausz, Science **317**, 769 (2007).
3. I. P. Christov, M. M. Murnane, and H. C. Kapteyn, Phys. Rev. Lett. **78**, 1251 (1997).
4. J. L. Krause, K. J. Schafer, and K. C. Kulander, Phys. Rev. Lett. **68**, 3535 (1992).
5. J. Tate, T. Auguste, H. G. Muller, P. Salieres, P. Agostini, and L. F. DiMauro, Phys. Rev. Lett. **98**, 013901 (2007).
6. T. Popmintchev, M.-C. Chen, D. Popmintchev, P. Arpin, S. Brown, S. Alisauskas, G. Andriukaitis, T. Balciunas, O. D. Mucke, A. Pugzlys, A. Baltuska, B. Shim, S. E. Schrauth, A. Gaeta, C. Hernandez-Garcia, L. Plaja, A. Becker, A. Jaron-Becker, M. M. Murnane, and H. C. Kapteyn, Science **336**, 1287 (2012).
7. V. S. Yakovlev, M. Ivanov, and F. Krausz, Opt. Express **15**, 15351 (2007).
8. G. Marcus, W. Helml, X. Gu, Y. Deng, R. Hartmann, T. Kobayashi, L. Strueder, R. Kienberger, and F. Krausz, Phys. Rev. Lett. **108**, 023201 (2012).
9. B. E. Schmidt, A. D. Shiner, P Lassonde, J. C. Kieffer, P. B. Corkum, D. M. Villeneuve, and F. Légaré, Opt. Express **19**, 6858 (2011).
10. C. Li, D. Wang, L. Song, D. Liu, P. Liu, C. Xu, Y. Leng, R. Li, and Z. Xu, Opt. Express **19**, 6783 (2011).
11. T. Fuji, N. Ishii, C. Y. Teisset, X. Gu, T. Metzger, A. Baltuska, N. Forget, D. Kaplan, A. Galvanauskas, and F. Krausz, Opt. Lett. **31**, 1103 (2006).
12. X. Gu, G. Marcus, Y. Deng, T. Metzger, C. Teisset, N. Ishii, T. Fuji, A. Baltuska, R. Butkus, V. Pervak, H. Ishizuki, T. Taira, T. Kobayashi, R. Kienberger, and F. Krausz, Opt. Express **17**, 62 (2009).
13. K.-H. Hong, S.-W. Huang, J. Moses, X. Fu, C.-J. Lai, G. Cirmi, A. Sell, E. Granados, P. Keathley, and F. X. Kärtner, Opt. Express **19**, 15538 (2011).
14. C. J. Fecko, J. J. Loparo, and A. Tokmakoff, Opt. Commun. **241**, 521 (2004).
15. G. Andriukaitis, T. Balčiūnas, S. Ališauskas, A. Pugžlys, A. Baltuška, T. Popmintchev, M.-C. Chen, M. M. Murnane, and H. C. Kapteyn, Opt. Lett. **36**, 2755 (2011).
16. C. Heese, C. R. Phillips, L. Gallmann, M. M. Fejer, and U. Keller, Opt. Lett. **35**, 2340 (2010).
17. A. Baltuska, T. Fuji, and T. Kobayashi, Phys. Rev. Lett. **88**, 133901 (2002).
18. T. Fuji, A. Apolonski, and F. Krausz, Opt. Lett. **29**, 632 (2004).
19. I. N. Ross, P. Matousek, G. H. C. New, and K. Osvay, J. Opt. Soc. Am. B **19**, 2945 (2002).
20. T. Metzger, A. Schwarz, C. Y. Teisset, D. Sutter, A. Killi, R. Kienberger, and F. Krausz, Opt. Lett. **34**, 2123 (2009).
21. A. Schwarz, M. Ueffing, Y. Deng, X. Gu, H. Fattahi, T. Metzger, M. Ossiander, F. Krausz, and R. Kienberger, Opt. Express **20**, 5557 (2012).