Carrier-envelope phase stabilization of a terawatt level chirped pulse amplifier for generation of intense isolated attosecond pulses

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Abstract: We demonstrate the first carrier-envelope phase (CEP)-stabilized chirped pulse amplification system with pulse peak-powers in the terawatt regime. The system, which eventually is intended to be used in the generation of isolated attosecond pulses, consists of two consecutive multipass amplification stages. The first amplification stage is a commercial CEP-stable kHz system including a single 13-pass amplifier reaching a pulse energy of 2.3 mJ. Pulses are picked after the first stage at a repetition rate of 50 Hz and are further amplified in a 5-pass power-amplifier to pulse energies that reach up to 80 mJ before compression. After compression the pulse energy is 35mJ at a pulse duration of 32 fs, signifying a peak power of 1.1 terawatt. Peak-powers exceeding 1.5 TW should easily be achievable by improving the efficiency of the grating compressor. The CEP-stability of the terawatt system is demonstrated by single shot measurements of the residual CEP jitter at the full repetition rate and show an excellent root-mean-square value of 315 mrad.

OCIS codes: (020.2649) Strong field laser physics; (300.6530) Spectroscopy, ultrafast; (190.7110) Ultrafast nonlinear optics; (140.3280) Laser amplifiers; (140.7090) Ultrafast lasers; (140.7240) UV, EUV, and X-ray lasers

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References

1. M. Hentschel, R. Kienberger, C. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, "Attosecond metrology," Nature **414**, 509–513 (2001).
2. H. R. Telle, G. Steinmeyer, A. E. Dunlop, J. Stenger, D. H. Sutter, and U. Keller, "Carrier-envelope offset phase control: A novel concept for absolute optical frequency measurement and ultrashort pulse generation," Appl. Phys. B **69**, 327–332 (1999).
3. J. Reichert, R. Holzwarth, T. Udem, and T. W. Hansch, "Measuring the frequency of light with mode-locked lasers," Opt. Com. **172**, 59–68 (1999).
4. A. Baltuska, T. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, C. Gohle, R. Holzwarth, V. S. Yakoviev, A. Scrinzi, T. W. Hansch, and F. Krausz, "Attosecond control of electronic processes by intense light fields," Nature **421**, 611–615 (2003).
5. E. Goulielmakis, Z.-H. Loh, A. Wirth, R. Santra, N. Rohringer, V. S. Yakovlev, S. Zherebtsov, T. Pfeifer, A. M. Azzeer, M. F. Kling, S. R. Leone, and F. Krausz, "Real-time observation of valence electron motion," Nature **466**, 739–743 (2010).

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6. G. Sansone, F. Kelkensberg, J. Perez-Torres, F. Morales, M. Kling, W. Siu, O. Ghafur, P. Johnsson, M. Swoboda, E. Benedetti, F. Ferrari, F. Lepine, S. Zherebtsov, I. Znakovskaya, A. L'Huillier, M. Ivanov, M. Nisoli, F. Martin, and M. Vrakking, "Electron localization following attosecond molecular photoionization," Nature **465** 763–766 (2010).
7. A. L. Cavalieri, N. Muller, T. Uphues, V. S. Yakovlev, A. Baltuska, B. Horvath, B. Schmidt, L. Blumel, R. Holzwarth, S. Hendel, M. Drescher, U. Kleineberg, P. M. Echenique, R. Kienberger, F. Krausz, and U. Heinzmann, "Attosecond spectroscopy in condensed matter," Nature **449**, 1029–1032 (2007).
8. E. Goulielmakis, M. Schultze, M. Hofstetter, V. S. Yakovlev, J. Gagnon, M. Uiberacker, A. L. Aquila, E. M. Gullikson, D. T. Attwood, R. Kienberger, F. Krausz, and U. Kleineberg, "Single-cycle nonlinear optics," Science **320**, 1614–1617 (2008).
9. G. Sansone, E. Benedetti, F. Calegari, C. Vozzi, L. Avaldi, R. Flammini, L. Poletto, P. Villoresi, C. Altucci, R. Velotta, S. Stagira, S. De Silvestri, and M. Nisoli, "Isolated single-cycle attosecond pulses," Science **314**, 443–446 (2006).
10. L. A. A. Nikolopoulos and P. Lambropoulos, "Multichannel theory of two-photon single and double ionization of helium," J. Phys. B **34**, 545–564 (2001).
11. Y. H. Jiang, A. Rudenko, J. F. Perez-Torres, O. Herrwerth, L. Foucar, M. Kurka, K. U. Kuhnel, M. Toppin, E. Plesiat, F. Morales, F. Martin, M. Lezius, M. F. Kling, T. Jahnke, R. Dorner, J. L. Sanz-Vicario, J. van Tilborg, A. Belkacem, M. Schulz, K. Ueda, T. J. M. Zouros, S. Dusterer, R. Treusch, C. D. Schroter, R. Moshammer, and J. Ullrich, "Investigating two-photon double ionization of d-2 by xuv-pump-xuv-probe experiments," Phys. Rev. A **81**, 051402 (2010).
12. C. P. Hauri, W. Kornelis, F. W. Helbing, A. Heinrich, A. Couairon, A. Mysyrowicz, J. Biegert, and U. Keller, "Generation of intense, carrier-envelope phase-locked few-cycle laser pulses through filamentation," Appl. Phys. B **79**, 673–677 (2004).
13. F. Ferrari, F. Calegari, M. Lucchini, C. Vozzi, S. Stagira, G. Sansone, and M. Nisoli, "High-energy isolated attosecond pulses generated by above-saturation few-cycle fields," Nat. Phot. **4**, 875–879 (2010).
14. H. Mashiko, S. Gilbertson, C. Q. Li, S. D. Khan, M. M. Shakya, E. Moon, and Z. H. Chang, "Double optical gating of high-order harmonic generation with carrier-envelope phase stabilized lasers," Phys. Rev. Lett. **100**, 103906 (2008).
15. H. Mashiko, S. Gilbertson, C. Li, E. Moon, and Z. Chang, "Optimizing the photon flux of double optical gated high-order harmonic spectra," Phys. Rev. A **77**, 063423 (2008).
16. X. M. Feng, S. Gilbertson, H. Mashiko, H. Wang, S. D. Khan, M. Chini, Y. Wu, K. Zhao, and Z. H. Chang, "Generation of isolated attosecond pulses with 20 to 28 femtosecond lasers," Phys. Rev. Lett. **103**, 183901 (2009).
17. S. Gilbertson, Y. Wu, S. D. Khan, M. Chini, K. Zhao, X. Feng, and Z. Chang, "Isolated attosecond pulse generation using multicycle pulses directly from a laser amplifier," Phys. Rev. A **81**, 043810 (2010).
18. C. Siedschlag, H. G. Mller, and M. J. J. Vrakking, "Generation of isolated attosecond pulses by two-color laser fields," Las. Phys. **15**, 916–925 (2005).
19. T. Pfeifer, L. Gallmann, M. J. Abel, P. M. Nagel, D. M. Neumark, and S. R. Leone, "Heterodyne mixing of laser fields for temporal gating of high-order harmonic generation," Phys. Rev. Lett. **97**, 163901 (2006).
20. E. J. Takahashi, P. Lan, O. D. Mücke, Y. Nabekawa, and K. Midorikawa, "Infrared two-color multicycle laser field synthesis for generating an intense attosecond pulse," Phys. Rev. Lett. **104**, 233901 (2010).
21. E. Takahashi, Y. Nabekawa, T. Otsuka, M. Obara, and K. Midorikawa, "Generation of highly coherent submicrojoule soft x rays by high-order harmonics," Phys. Rev. A **66**, 021802 (2002).
22. N. M. Naumova, J. A. Nees, I. V. Sokolov, B. Hou, and G. A. Mourou, "Relativistic generation of isolated attosecond pulses in a λ^3 focal volume," Phys. Rev. Lett. **92**, 063902 (2004).
23. S. V. Bulanov, N. M. Naumova, and F. Pegoraro, "Interaction of an ultrashort, relativistically strong laser pulse with an overdense plasma," Phys. Plasmas **1**, 745–757 (1994).
24. Y. Nomura, R. Horlein, P. Tzallas, B. Dromey, S. Rykovanov, Z. Major, J. Osterhoff, S. Karsch, L. Veisz, M. Zepf, D. Charalambidis, F. Krausz, and G. D. Tsakiris, "Attosecond phase locking of harmonics emitted from laser-produced plasmas," Nat. Phys. **5**, 124–128 (2009).
25. T. Baeva, S. Gordienko, and A. Pukhov, "Relativistic plasma control for single attosecond x-ray burst generation," Phys. Rev. E **74**, 065401 (2006).
26. P. Heissler, R. Hrlein, M. Stafe, J. Mikhailova, Y. Nomura, D. Herrmann, R. Tautz, S. Rykovanov, I. Fldes, K. Varj, F. Tavella, A. Marcinkevicius, F. Krausz, L. Veisz, and G. Tsakiris, "Toward single attosecond pulses using harmonic emission fromsolid-density plasmas," Appl. Phys. B **101**, 511–521 (2010).
27. J.-F. Hergott, O. Tcherbakoff, P.-M. Paul, P. Demengeot, M. Perdrix, F. Lepetit, D. Garzella, D. Guillaumet, M. Comte, P. D. Oliveira, and O. Gobert, "Carrier-envelope phase stabilization of a 20 w, grating based, chirped-pulse amplified laser, using electro-optic effect in a linbo3 crystal," Opt. Express **19**, 19935–19941 (2011).
28. N. Ishii, R. Butkus, A. Baltuska, E. Goulielmakis, M. Uiberacker, R. Kienberger, T. Fuji, V. S. Yakovlev, V. Smilgevicius, R. Danielius, A. Piskarskas, and F. Krausz, "Toward a terawatt few-optical-cycle driver laser for attosecond spectroscopy," in "Ultrafast Phenomena XIV," , vol. 79 of *Springer Series in Chemical Physics*, T. Kobayashi, T. Okada, T. Kobayashi, K. A. Nelson, S. Silvestri, F. Schfer, J. Toennies, and W. Zinth, eds. (Springer Berlin Heidelberg, 2005), 8–12.

#155262 - $15.00 USD Received 26 Sep 2011; revised 20 Oct 2011; accepted 20 Oct 2011; published 22 Nov 2011

29. A. Renault, D. Z. Kandula, S. Witte, A. L. Wolf, R. T. Zinkstok, W. Hogervorst, and K. S. E. Eikema, "Phase stability of terawatt-class ultrabroadband parametric amplification," Opt. Lett. **32**, 2363–2365 (2007).
30. I. Thomann, E. Gagnon, R. J. Jones, A. S. Sandhu, A. Lytle, R. Anderson, J. Ye, M. Murnane, and H. Kapteyn, "Investigation of a grating-based stretcher/compressor for carrier-envelope phase stabilized fs pulses," Opt. Express **12**, 3493–3499 (2004).
31. Z. Chang, "Carrier-envelope phase shift caused by grating-based stretchers and compressors," Appl. Opt. **45**, 8350–8353 (2006).
32. C. Q. Li, E. Moon, and Z. H. Chang, "Carrier-envelope phase shift caused by variation of grating separation," Opt. Lett. **31**, 3113–3115 (2006).
33. C. Q. Li, E. Moon, H. Mashiko, C. M. Nakamura, P. Ranitovic, C. M. Maharjan, C. L. Cocke, Z. H. Chang, and G. G. Paulus, "Precision control of carrier-envelope phase in grating based chirped pulse amplifiers," Opt. Express **14**, 11468–11476 (2006).
34. E. Gagnon, I. Thomann, A. Paul, A. L. Lytle, S. Backus, M. M. Murnane, H. C. Kapteyn, and A. S. Sandhu, "Long-term carrier-envelope phase stability from a grating-based, chirped pulse amplifier," Opt. Lett. **31**, 1866–1868 (2006).
35. C. Iaconis and I. A. Walmsley, "Self-referencing spectral interferometry for measuring ultrashort optical pulses," IEEE J. Quantum Electron. **35**, 501–509 (1999).
36. M. Mehendale, S. A. Mitchell, J. P. Likforman, D. M. Villeneuve, and P. B. Corkum, "Method for single-shot measurement of the carrier envelope phase of a few-cycle laser pulse," Opt. Lett. **25**, 1672–1674 (2000).
37. M. Kakehata, H. Takada, Y. Kobayashi, K. Torizuka, Y. Fujihira, T. Homma, and H. Takahashi, "Single-shot measurement of carrier-envelope phase changes by spectral interferometry," Opt. Lett. **26**, 1436–1438 (2001).
38. C. Li, E. Moon, H. Wang, H. Mashiko, C. M. Nakamura, J. Tackett, and Z. Chang, "Determining the phase-energy coupling coefficient in carrier-envelope phase measurements," Opt. Lett. **32**, 796–798 (2007).
39. C. Li, E. Moon, H. Mashiko, H. Wang, C. M. Nakamura, J. Tackett, and Z. Chang, "Mechanism of phase-energy coupling in f-to-2f interferometry," Appl. Opt. **48**, 1303–1307 (2009).
40. G. Steinmeyer, "personal communication," .
41. S. Koke, C. Grebing, B. Manschwetus, and G. Steinmeyer, "Fast f-to-2f interferometer for a direct measurement of the carrier-envelope phase drift of ultrashort amplified laser pulses," Opt. Lett. **33**, 2545-2547 (2008).
42. T. Fordell, M. Miranda, A. Persson, and A. L'Huillier, "Carrier-envelope phase stabilization of a multi-millijoule, regenerative-amplifier-based chirped-pulse amplifier system," Opt. Express **17**, 21091–21097 (2009).

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1. Introduction

In the last decade there have been great advances in the field of attosecond science using high harmonic generation (HHG) to produce attosecond pulse trains (APTs) and isolated attosecond pulses (IAPs). Although IAPs were already demonstrated [1] and used in experiments prior to the development of the first CEP-stable laser amplifier, the controlled experimental generation and use of IAPs made a tremendous step forward in 2003, when, extending the pioneering CEP-stabilization of nano-Joule level ultrashort pulse oscillators [2, 3], the first CEP-stable laser amplifier was accomplished [4]. This method opened up new possibilities in the study and control of electron dynamics in atoms [5], molecules [6] and condensed matter [7]. The most popular measurement scheme to investigate such ultrafast phenomena are pump-probe experiments where the system under investigation absorbs one (or a few) photons from a pump laser source and one (or a few) photons from a probe laser source. Such schemes are so far very difficult to realize using XUV light, since the conversion efficiency from IR photons to XUV photons is rarely better than 10^{-6} for APTs and even less for IAPs which have been limited to pulse energies of pJ - nJ level [8, 9]. Considering the low cross-section for non-linear processes in the XUV spectral region ($10^{-49} - 10^{-52}$ cm^{4}s for two photon XUV ionization [10]), the XUV intensities would have to exceed 10^{13} W/cm^2, in order to produce a measurable two-photon signal. Sofar this has restricted attosecond science to XUV-IR pump-probe experiments where the optical cycle of the IR pulse is used as an attosecond clock. When increasing the attosecond pulse energy close to the µJ level XUV-XUV pump-probe experiments would come into reach. The general feasibility of such experiments has recently been demonstrated at the free-electron laser facility in Hamburg (FLASH), where microjoule level 38 eV pulses with a duration of 30 fs were focused to intensities of $10^{13} - 10^{14}$ W/cm^2 to probe the ultrafast nuclear wavepacket motion of D_2^+ [11].

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To reach such high pulse energies for IAPs from table top systems however, development in two areas is needed. On the one hand, the efficiency of IAP production techniques has to be improved while loosening the requirements imposed on the driving laser pulse duration. On the other hand the pulse energies of the driving laser sources have to be increased while maintaining the obligatory laser parameters such as pulse duration and particularly CEP-stability. In this article progress in the latter field will be presented by the successful implementation of an amplifier system that delivers CEP-stable femtosecond pulses with terawatt level peak-powers at 50 Hz repetition rate.

It is nonetheless important to note that the two areas of development are strongly linked, since laser pulses in the TW regime cannot directly be applied to the traditional techniques of IAP generation. To put the technical work presented in this article into the correct context, an overview of current developments in the field of attosecond pulse production is given prior to presenting the results on the TW CEP-stable amplifier. The overview will focus in particular on the scalability of these techniques to higher energies of the driver pulse.

2. Isolated attosecond pulses from high energy laser sources

The first method that demonstrated the production of IAPs and that is still extensively used to this day consists of using few-cycle driver pulses from a chirped pulse amplification (CPA) system compressed in hollow core fibres [8] or filamentation [12] stages and spectral filtering of the cut-off harmonics generated via HHG in a gas target. Pulse durations as short as 80 as have been demonstrated using this technique [8].

Even though in recent experiments Ferrari and coworkers [13] demonstrated that the conversion efficiency into IAPs can be significantly increased by using CEP-stable few-cycle laser pulses with peak intensities which are higher than the gas saturation intensity, scaling to higher driving pulse energies is experimentally extremely demanding. Post-compression techniques to achieve intense few-cycle laser pulses are limited to input energies below 10 mJ. Additionally the strong drop of harmonic intensity in the cut-off region leads to very low conversion efficiencies from IR photons to XUV photons. Consequently the resulting XUV photon flux is only on the order of 10^5 photons / pulse (for $\sim$80 eV photons) [8].

Another technique that uses few-cycle laser pulses for the generation of IAPs is polarization gating (PG) [9] which uses two co-propagating pulse replicas with counter rotating circular polarization that have a small temporal overlap. In the region of overlap the electric fields of the two replicas add up to a linearly polarized gate. If the gate width is restricted to a single half cycle of the driving field, IAPs can be produced. Sansone et. al. [9] demonstrated pulses with 130 as duration and a photon flux of $1.25 \cdot 10^7$ photons / pulse (39 eV photons) using this technique.

At the same time PG lays the groundwork for a number of techniques that try to circumvent the need for few-cycle driving pulses opening up the possibility of generating IAPs directly from multi-cycle driver pulses. Such systems with pulse durations up to 30 fs are nowadays widely available and can reach energies even in the multi-Joule level.

The first advancement towards using longer driver pulses was made with double optical gating (DOG) [14], which additionally exploits the asymmetry in the driving laser field introduced by adding a small fraction of second harmonic radiation to the fundamental PG light field. The asymmetry in the driver field limits attosecond pulse production to every full cycle of the field, thus allows the use of a larger gate width and additionally suppresses ionization from the leading edge of the driving pulse. Consequently the use of laser pulses with durations of up to 12 fs has been demonstrated to produce IAPs which reached pulse energies of 6.5 nJ at 39 eV in Argon and 170 pJ at 50 eV in Neon, i.e. corresponding to photon fluxes of approximately 10^9 photons / pulse and $2 \cdot 10^7$ photons / pulse respectively [15] before filtering with an aluminum mirror. The DOG technique was further refined by introducing its generalized form (GDOG) [16] which uses elliptically polarized light instead of circularly polarized lightfields as in PG and DOG. In this way ionization from the leading edge of the driving laser pulse could further be reduced allowing for the first time the use of laser pulses directly from a laser amplifier with pulse durations up to 28 fs to produce IAPs from Argon with a duration of 160 as and a pulse energy of 170 pJ at 40 eV ($2.6 \cdot 10^7$ photons / pulse) [17]. Nevertheless at higher driver energies one will have to deal with an increased ionization which calls for further development of new phase-matching techniques.

Another approach to generating IAPs from multi-cycle driver pulses is the heterodyne mixing of two laser fields which was proposed for the first time by Siedschlag and coworkers [18], who predicted the production of isolated attosecond pulses from driving pulses with a duration of up to 30 fs. They further investigated phase-matching conditions and the necessity of a stabilized CEP for efficient IAP generation. Similar theoretical results were also presented in [19]. Experimental results from combining 800 nm, 30 fs laser pulses with weak 1300 nm, 40 fs pulses show the generation of continuous spectra in the XUV region, that can lead to the formation of IAPs [20]. This scheme further reduces the ionization in the leading edge of the pulse by two orders of magnitude compared to DOG and GDOG. Conversion efficiencies of 10^{-5} are expected. Using appropriate driving laser sources and phase-matching techniques this approach eventually could lead to IAPs from HHG in gas with energies in the microjoule range [21].

All of the sofar demonstrated techniques for IAP production rely on HHG in gaseous media. However when using plasmas generated from solid targets as a non-linear medium, conversion efficiencies of up to ten percent are expected [22]. In this case ionization changes from being a limitation in the process of HHG to being an inherent necessity for the process. At intensities when the magnetic field of the laser electromagnetic field becomes relevant, the plasma surface generated by the leading edge of the pulse begins to oscillate parallel to the laser propagation direction acting as a mirror for the incoming radiation that moves with relativistic velocities. This can lead to HHG and attosecond bunching of the reflected radiation [23]. In 2009 the first APT from solid target HHG was observed [24] and there have been a number of schemes proposed for the production of IAPs, most of which require the use of few-cycle laser pulses. Nonetheless the use of polarization gating techniques seem feasible for the production of IAPs even when multi-cycle driver pulses are used [25]. Recent measurements [26] support this notion.

For all mentioned techniques of IAP production, the stabilization of the CEP is mandatory if one wants to produce attosecond pulses with consistent temporal shape and photon flux for all laser pulses. While the restrictions on the values of CEP jitter to produce IAP have also been loosened by the newer generation techniques [14, 16, 20], a larger CEP jitter will nonetheless generally result in strong energy fluctuations of the produced IAP. In XUV-XUV pump-probe experiments, due to the non-linear nature of the processes involved, a good pulse-to-pulse stability is, however, necessary to reduce the noise in the final measurement. Energetic light sources with low CEP jitter hence are still an undeniable prerequisite for high quality non-linear XUV experiments. The system demonstrated in this publication presents an ideal source of such IR pulses.

When scaling CPA systems to higher pulse energies, the repetition rate of the laser system is usually limited to a few tens of Hertz. However, all of today's commercially available CEP-stabilized laser systems are operated at pulse repetition rates in the kHz range and pulse energies of not more than $\sim$ 10 - 20 mJ [27]. There are multiple groups that work on developing CEP-stable terawatt light sources relying on optical parametric CPA (OPCPA) [28]. To the knowledge of the authors, the only successful implementation of CEP stabilization in such a system so far was achieved by Renault and coworkers [29], resulting in 7.6 fs, 15.5 mJ (2

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TW) IR pulses. It should be noted, however, that the CEP stable construction of terawatt level OPCPA systems is technically challenging and the short pulse durations, as described in the paragraphs above, is not an obligatory precondition for production of IAPs anymore. The techniques to build terawatt level CPA systems on the other hand are well established and with the system presented in this paper a major step towards low repetition rate, even higher energy systems with very high quality CEP-stability has been demonstrated.

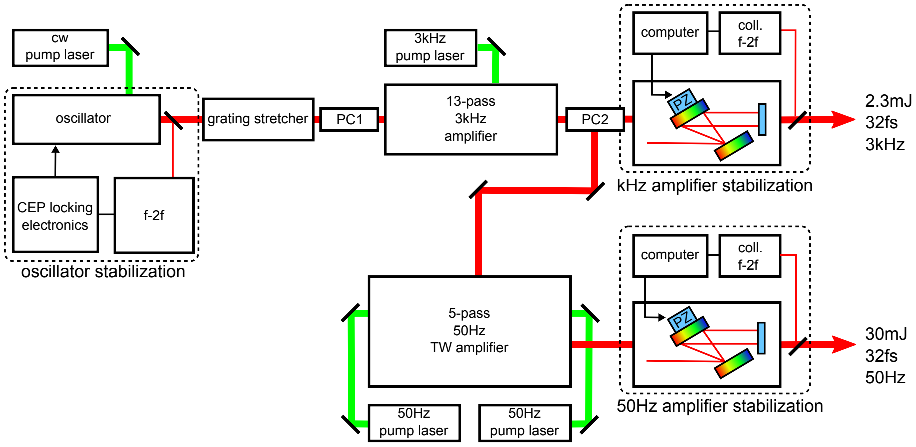

Fig. 1. Schematic drawing of the complete laser system including the elements for CEP-stabilization. The oscillator is stabilized using a piezo actuated tilt mirror [30] in combination with Menlo XPS f-2f interferometer and PLL-electronics. The output of both amplifier stages is stabilized by controlling the grating separation in the grating compressors [31–33]. The second amplification stage amplifies pulses at 50 Hz to the TW level while maintaining the same residual phase error as in the kHz-arm.

3. The laser setup

A schematic overview of the complete laser system is depicted in Fig. 1. It comprises two beam lines which will further be referred to as the kHz-arm and the 50 Hz-arm of the laser system. The kHz-arm consists of a commercial CEP-stabilized Ti:Sapphire based CPA system running at 3 kHz repetition rate (upper half of Fig. 1, KMLabs Dragon). The 50 Hz-arm is an additional power-amplifier seeded by amplified pulses of the kHz-amplifier which are picked at 50 Hz (lower half of Fig. 1) while the pulses at the remaining 3 kHz (every 60th pulse missing) further propagate through the original pulse compressor and can be used for experiments. Amplified pulses from the 50 Hz-arm are compressed in a separate grating compressor. Details of the implementation are discussed below.

The nJ seed-pulses are produced from a prism-based ultrashort pulse oscillator (KMLabs Griffin). An f-2f interferometer (Menlo XPS) including a photonic crystal fibre to generate the octave spanning spectrum and Menlo locking electronics (Menlo XPS-E) ensure CEP-stability by feeding an error signal back to a piezo actuated tilt mirror inside the oscillator cavity [3,30]. The resulting root-mean-square (rms) in-loop phase jitter is 104 mrad when measured over 40 milliseconds at 8 MHz bandwidth.

The CEP-stabilized oscillator seed pulses are stretched to approximately 220 ps in a grating based stretcher. After the pulses have been picked at a repetition rate of 3 kHz they are amplified in a single stage 13-pass cryogenically cooled Ti:Sapphire amplifier [34] which is pumped by a single diode-pumped 75W Nd:YLF laser (Photonics Industries, DM60). The amplified pulses reach an energy of 3.8 mJ, are routinely compressed to 30 fs and leave the compressor with an energy of 2.4 mJ. The CEP-stabilization of the kHz-arm was originally achieved by measuring the relative CEP with a commercial collinear f-2f interferometer and software (Menlo APS800) and feeding back to the grating separation using a piezo actuator inserted in the compressor translation stage [33]. In this configuration the rms CEP-jitter is around 300 mrad. The kHz-amplifier breadboard is rather compact and contains the stretcher, kHz-pulse-picker, kHz-amplifier and compressor.

To implement the power-amplifier a second pulse picker was inserted in the beam path before the compressor of the kHz-arm, picking the 3.8 mJ pulses at 50 Hz. The additional losses introduced to the kHz-arm by the pulse picker are only small and a pulse energy of 2.3 mJ after the kHz-compressor is maintained. There is also no measurable impact of the pulse picker on the CEP of the transmitted pulses.

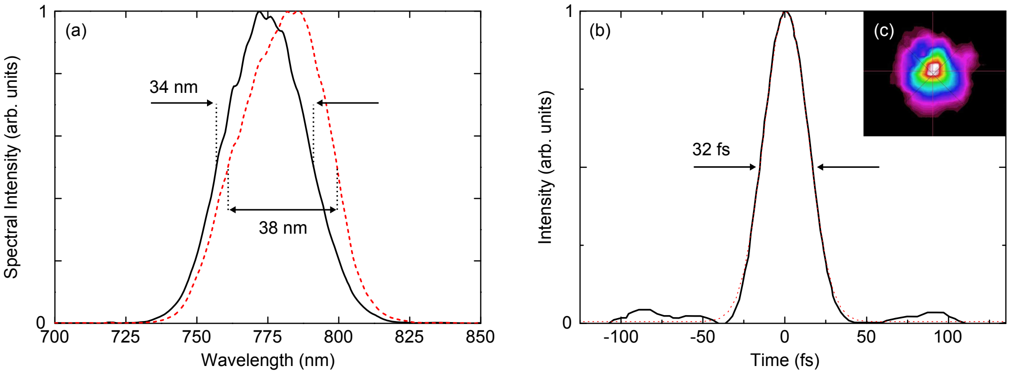

Fig. 2. Characteristics of the compressed 2 TW peak-power pulses. (a) The spectral profiles of the laser pulses before (black solid line) and after (dashed red line) amplification and their respective FWHM bandwidth. (b) The black curve shows the temporal pulse profile measured with SPIDER. The profile was fitted with a gaussian curve (dashed red line) which gives a FWHM duration of 32 fs. (c) The inset shows the spatial beam profile in the farfield as measured after a 1m focal length lens. It exhibits a slightly non-gaussian structure.

The picked pulses for the 50 Hz 5-pass power-amplifier are resized to a beam diameter of 4 mm. In view of the importance of the spatial beam quality in the 5-pass power-amplifier for achieving optimal energy extraction and a high final pulse energy, a pinhole spatial filter was inserted after the pulse picker. This comes at the expense of a reduction of the input energy into the power-amplifier to 1.4 mJ, however allows a higher final pulse energy from the amplifier crystal. The improved energy extraction is essential for the CEP-stabilization as will be clarified in the course of the article.

The 50 Hz power-amplifier is built up on a separate (90 x 180 cm^2) breadboard containing the pinhole setup, power-amplifier, compressor, two pump-lasers and an f-2f interferometer. For optical pumping two flashlamp-pumped Nd:YAG lasers are used (Litron, NanoTRL 320-50 dual head version). Each pump-laser can reach pulse energies of 160 mJ at a wavelength of 532 nm. The Ti:Sapphire crystal in the power-amplifier is water-cooled and pumped from both sides with each pump-laser respectively. At maximum pumping levels the amplified pulses can reach energies of >80 mJ. Routinely the output of the pump-lasers is slightly lower due to aging of the flash lamps and the crystal is effectively pumped with only around 120 mJ on each surface of the crystal. In this case the energies of the infrared pulses routinely reach 75 mJ indicating an optical efficiency of 31%. Thermal lensing inside the gain medium is compensated with several diverging lenses in between the individual passes.

Before compression, the 50 Hz amplified pulses are telescoped up to a 1/e diameter of 30 mm to prevent damage to the compression gratings. The spectral bandwidth after the kHz-amplifier is maintained in the 50 Hz power-amplifier with only a red-shift of the central wavelength by 10nm (see Fig. 2a). SPIDER (spectral phase interferometry by direct electric field reconstruction [35]) measurements indicated a pulse duration of 32 fs (see Fig. 2b). Due to the low individual efficiencies of the gratings (82%) the overall efficiency of the 50 Hz grating compressor is presently below 45%. The compressed pulses therefore only have an energy of 35 mJ corresponding to peak-powers of over 1.1 TW. By changing the gratings in the compressor energies exceeding 50 mJ should be easily feasible.

4. CEP-stabilization of the terawatt amplifier

In the course of the work the original Menlo APS f-2f setup of the kHz arm was rebuilt into a purely reflective setup to allow easier alignment and more flexible adjustment for higher fringe visibility by directly focusing the copropagating f and 2f beams [36, 37] onto the entrance slit of the spectrometer. A second equivalent system was set up to be used with the new 50 Hz amplifier stage. White-light generation was achieved by focusing the leaked light of a backside-polished mirror after the compressor into a 2 mm thick sapphire plate. The second harmonic of the long wavelength part of the white-light was generated in a 1 mm thick BBO crystal cut at a phase matching angle for 960 nm. The resulting spectral interference pattern which indicates a delay between the pulses of approximately 190 fs was detected using a USB-spectrometer (Thorlabs CS100).

At the same time the original Menlo software was replaced by a Labview-based homebuilt stabilization software, that allows online real single shot measurements as well as detailed settings for the PID feedback loop. The feedback in both compressors is achieved using a piezo actuators (Thorlabs Piezoelectric Actuator, Max Displacement 9.1 µm, 3.5 x 4.5 x 10 mm) inserted into the grating translation stages and the respective piezo controllers (Thorlabs MDT694A). The required voltages were directly written to the controller via the RS232 interface of the controller, allowing higher feedback frequency than in the original setup.

The first step to implement the CEP-stabilization of the 50 Hz-amplifier was to measure the isolated impact of the newly set up grating compressor [30], bypassing the 50 Hz power-amplifier and only running the oscillator CEP-stabilization. The recorded phase after the 50 Hz-compressor was compared against the phase measured after the original kHz-compressor. The fast phase jitter was comparable after both compressors, however the slow phase drifts measured behind the kHz-compressor were not visible in the measurements behind the 50 Hz compressor. We conclude therefore that most of the slow phase drifts of the kHz-system are accumulated only in the compressor setup whereas the stretcher has no measurable effect on the slow drift of the CEP. We suspect that this is due to thermal fluctuations as well as direct thermal load on the kHz-compressor. The fluctuations are not visible in the 50 Hz-compressor as the thermal load without pumping is negligible.

Taking into account the influence of thermal drifts introduced by the amplifier and the pump-lasers, the 50 Hz-amplifier was set up in such a way as to leave ample room in between the individual components. Additionally, compressor and f-2f interferometer were separated by vertical metal walls inside the amplifier housing and all walls were covered with foam to prevent acoustic noise from coupling into the CEP. The two pump-lasers were effectively decoupled from the breadboard by placing 25 mm thick sorbothane sheets beneath them. However it turned out that the pump-lasers initially introduced strong thermal fluctuations causing CEP jumps which could not be compensated by the slow feedback loop. Accordingly the pump-lasers were as well separated with metal walls from the amplifier section and it was ensured that there was

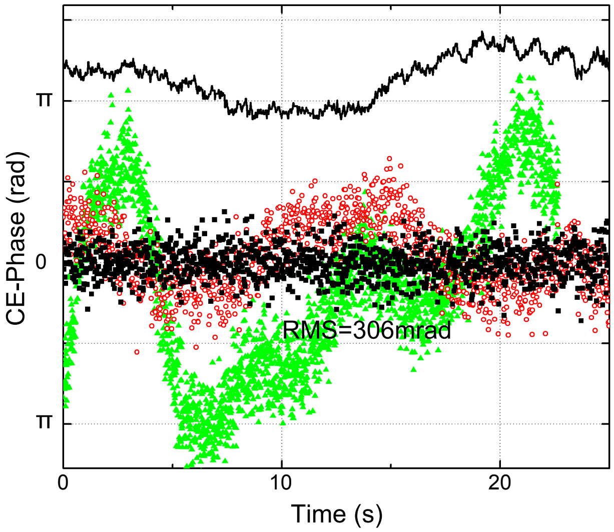

Fig. 3. Short term CEP measurements showing the stabilized (black dots) phase and the necessary compensating phase (black line) that is introduced in the grating compressor. For comparison typical cases of the free-running CEP evolution in the kHz arm (green triangles) and in the 50 Hz arm (red circles) are depicted as well. These measurements were not taken simultaneously and therefore one cannot draw any conclusions on correlation between the free running evolution in the two arms.

enough air circulation in the pump-laser section to remove the excess heat from the rest of the breadboard housing.

Figure 3 shows the CEP-drift for the amplified 50 Hz-arm in the stabilized case (black squares) and in the free-running case (red circles). For comparison a trace of the kHz-arm in the free running case is also plotted (green triangles). In both free-running cases, only the oscillator CEP-stabilization was active. In agreement with the before mentioned observation, the 50 Hz-arm shows a slow drift with significantly lower amplitude compared to the kHz-arm. This is due to the direct thermal load on the compressor gratings, as mentioned before. In the kHz-compressor the dissipated heat is by a factor 2 - 3 higher than in the compressor after the TW-amplifier. Additionally the thermal expansion coefficient of the copper grating substrates used in the kHz-compressor ($17 \cdot 10^{-6}$/°C) is significantly higher then those of the BK-7 gratings ($7 \cdot 10^{-6}$/°C) used in the TW-compressor.

In the first amplification attempts the optical efficiency was rather low with values of around 20-25%. As a result the pulse-to-pulse energy fluctuations of the amplified pulses were large. This can easily translate into instabilities in the CEP measurement mostly during the strongly non-linear white-light generation process [38, 39]. As a result the measured CEP-stability only reached values of above 700 mrad (see Fig. 4 blue trace). In order to achieve a better stability of the pulse energy and therefore also of the CEP measurement, we adjusted the amplifier to be operated at higher saturation levels. At efficiency levels of 31 % the phase-jitter is reduced by a factor of more than two to values around 300 mrad in single pulse measurements. Results of a long term measurement are shown in Fig. 4. The rms-stability of both pump-lasers is 0.56% and of the amplified pulse in this configuration is 0.6%.

Nonetheless even at this value the achievable CEP stability appears to be limited by shot noise resulting from the low photon numbers in the relevant spectral ranges produced in the white-light generation process. This assumption is supported by two facts: First the histograms plotted

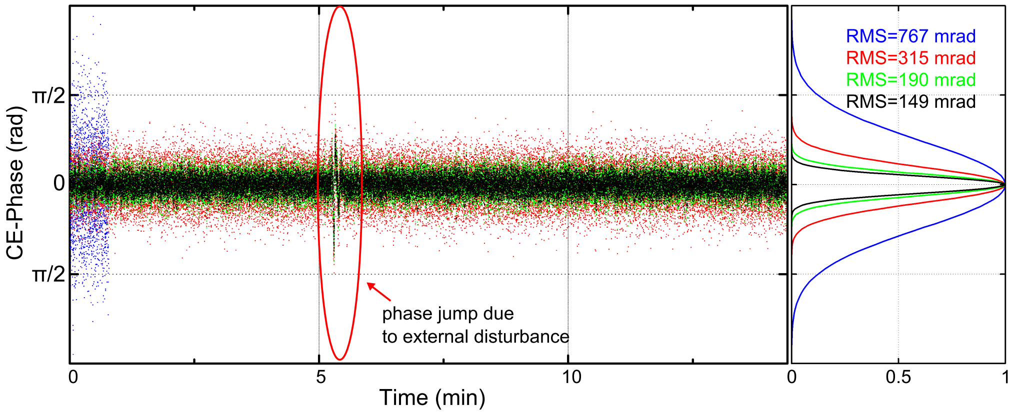

Fig. 4. Measurement of the CEP stability over an extended time interval. The red curve shows the CEP single shot CEP measurements of the 50 Hz TW-amplifier in the optimal configuration. The dots in green and black show the same measurement when averaging over three shots and five shots respectively. Histograms for all three cases are shown on the right with the respective rms error values of 315 mrad, 190 mrad and 149 mrad. For comparison a short measurement of the phase jitter is included in the case of low extraction efficiency (21%) (blue dots).The respective rms value is 767 mrad.

in Fig. 4 and the resultant rms-values follow almost perfectly a Gaussian statistic, indicative for a white noise limitation. If however the oscillator would be a limiting factor at this point, one would expect a non-Gaussian statistic [40]. Second, the analysis of the frequency spectra of the residual phase error (not plotted here) revealed similar indications. A clear reduction of the slow modulations up to a frequency of 0.3 Hz was visible comparing the free-running against the stabilized case. Higher frequencies however were already covered by the average shot noise level.

The high shot noise level finds its origin in the extremely low spectral densities in the 2f arm of the interferometer. Considering that the white-light generation in the Sapphire plate is limited to only a few microjoule of input energy, the effectively detected photon number N in the frequency-doubled part of the spectrum might be as low as 10^4, taking into account conversion efficiency into the IR, the doubling efficiency and losses in the spectrometer. The achievable phase resolution between consecutive laser pulses is then limited to a value of $\Delta\phi_{CE} = \pi/(N/2)^{1/2}$ [41], i.e. values between 40 - 50 mrad. These numbers however hold only for a perfect detection system. In the case of a fast CCD chip the resultant value could be higher by up to one order of magnitude [40] and thus lie at similar levels as those measured in our f-2f setup.

The situation could be tremendously improved by increasing the photon numbers in the relevant frequency range. Using super-continuum generation in noble gases would allow input energies in the mJ range with high conversion efficiencies in the super-continuum generation process. A realistic increase in the relevant photon number by a factor of 10 or more would hence improve the shot noise by more than an order of magnitude. With the laser system presented in this chapter, the pulse energies to run a noble gas filled fibre setup which would generate the necessary white-light, would only amount to 2 - 3 % of the total output energy of the system, and should therefore be the next implementation step.

Even at shot noise limitations the final CEP-stability of the system is still strongly dependent on external environmental influences. Especially mechanical vibrations seem to play a major role, i.e. the difference of rms-error values of the CEP can be as high as 150 mrad when comparing measurements taken during the day and after working hours. Since the laser systems are supplying experimental setups that are not connected to the optical table, floating the table is not an option. We expect that the rms value of the system could still improve when moving to a more stable environment.

It should be noted that while most publications on CEP results of amplifier systems are based on averaged measurements, the values presented in this paper are real single shot measurements at the full repetition rate of the laser system. As was shown by [41], single shot measurements at the full repetition rate of the laser system are necessary to determine the real phase stability of a system. The demonstrated rms-values of the residual phase jitter are therefore excellent values and rank among the best values measured so far for such laser systems [27,29,41,42]. To make the results comparable with other published data, the long term single shot measurement in Fig. 4 is also shown for cases when a moving average over three and five pulses was applied. As expected, the rms-value of the phase jitter in these cases is reduced dramatically to 190 mrad and 149 mrad respectively.

5. Conclusion

In this article we present a terawatt level femtosecond laser system, which is perfectly suited for the generation of IAP using the techniques reviewed in the first part of the paper. The demonstrated pulse properties of the amplified pulses are a pulse duration of 32 fs and a peak-power of 1.1 TW at 50 Hz repetition rate. It was shown that CEP-stabilization in this CPA system can be achieved with a residual phase jitter which competes with the best stabilization results published so far.

To our knowledge the presented results show the highest pulse energy reached for CEP-stabilized femtosecond pulses to date, and opens up the possibility to produce IAPs with unprecedented photon fluxes. This is an important step towards experimental conditions that allow experimental researchers to perform XUV-XUV pump-probe experiments.

Further it paves the way towards even more energetic driver pulses with stable CEP. Nowadays CPA systems with repetition rates in the 10 Hz regime and several Joule of pulse energy are commercially available. From the demonstrated measurements we conclude that scaling to higher energies should be straight forward, as long as the pulse repetition period is short compared to the slow thermal fluctuations introduced in the compressor and the pulse-to-pulse energy instabilities can be maintained at a low level.

The availability of high pulse energies might additionally promote the use of super-continuum generation in fibres for the use with the f-2f interferometer. This could potentially allow a significant increase in the phase sensitivity in the f-2f measurement and therefore should be further investigated in the future use of such a laser system.

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