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Report on the improvement of conversion efficiency and photon energy from HHG in gases

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Deliverable Nature	
R = Report, P = Prototype, D = Demonstrator, O = Other	R
Dissemination Level	
PU = Public PP = Restricted to other programme participants (incl. the Commission Services) RE = Restricted to a group specified by the consortium (incl. the Commission Services) CO = Confidential, only for members of the consortium (incl. the Commission Services)	PU

A. Abstract / Executive Summary

Experimental and theoretical studies have been performed to optimize high-harmonic generation for higher output power and shorter wavelengths. The techniques investigated include longer drive laser wavelength, shorter and higher energy drive pulses, and quasi phase matching. Several new laser sources have been realized, leading to the highest reported flux of 500 eV photons, and demonstrations of X-ray and attosecond spectroscopy.

B. Deliverable Report

1 Introduction

High-harmonic generation (HHG) is an important technique to generate coherent extreme ultraviolet (XUV) and soft-X-ray light pulses (0.1 - 100 nm), which can be used for a myriad of applications such as (ultrafast) spectroscopy and microscopy. The HHG process is based on focusing powerful ultrashort laser pulses (tens of femtoseconds in duration, or shorter) into a gaseous medium (typically a noble-gas). The high electromagnetic field of the optical pulses distorts the Coulomb potential of the electrons in the medium in such a way that electrons can tunnel out of the atoms. The electric field of the light then accelerates the free electrons, so that they acquire energy from the light field. When the electron reverses direction (because the electric field does so periodically), the electron can be accelerated towards the atoms it came from, and recombine again while releasing the excess energy in the form of a high-energy photon. This process can repeat each half-cycle of the fundamental driving field, leading to a periodic burst of high-energy photons. Due to this periodicity (twice the fundamental light frequency), the output spectrum consists of (odd) harmonics of the fundamental frequency. The energy of the photons depends quadratically on the wavelength of the driving laser: a longer wavelengths accelerates the electron for a longer time, so that it reaches a higher energy and can emit higher energy photons (although at lower efficiency). More laser power also increases the yield of the HHG process and the highest energy. The HHG yield also depends strongly on phase-matching: the contributions of HHG light from atoms in different parts of the medium must be kept in phase so that it adds up coherently and leads to a higher output. In general the driving laser and the generated HHG light have a different phase velocity. One can compensate that by adjusting the pressure of the medium, adjust the propagation conditions geometrically, or perform quasi-phase-matching: periodically modulate the driving field or the medium in such a way that no light is produced from regions of the medium where the HHG light is out of phase with the driving light. Because HHG is competing with ionization it also helps to use shorter laser pulses (as short as 4 fs). These aspects have been investigated experimentally and theoretically by the contributing partners to enhance the energy of the photon and/or the overall efficiency of the HHG process.

2 Objectives

Improvement of conversion efficiency and photon energy from HHG in gasses, using different strategies: use of longer drive wavelengths, quasi-phase matching schemes (modulating either the laser field of by employing a new periodic nozzle), and by increasing the laser power or shorting of the laser pulses to reduce ionization. The theoretical/numerical efforts are mainly focused on the study of the high-order harmonic generation process beyond the conventional procedure, using non-homogeneous laser fields and resonant atomic systems. Also improvement of the XUV beam quality is pursued.

3 Work performed / results / description

MPQ implemented a differentially pumped gas-filled hollow core fiber system to improve frequency broadening of 2 mJ / 20 fs laser pulses from a Titanium:Sapphire amplifier /

booster system. These pulses were compressed to < 4 fs at a pulse energy of 1 mJ. Using these pulses as a driver for High-order Harmonic generation (HHG), MPQ was able to extend the cut off of Extreme Ultraviolet (XUV) radiation to 145 eV with enough flux to perform attosecond streaking experiments at surfaces and layered systems [1,2].

ICFO has successfully implemented a high pressure effusive target for ponderomotive shifting of high harmonic generation. They used their sub-2-cycle 1 kHz source for HHG at backing pressures up to 10 bar and demonstrated coherent harmonic emission up to the oxygen K-shell edge of 534 eV [3]. The source signifies the highest flux in the water window [4,5] and was used to demonstrated x-ray absorption fine structure spectroscopy (XAFS) on a solid state target [4,5].

STFC-CLF has also constructed a few-cycle source in the infrared (12 fs, 0.4 mJ, 1.8 micron) using hollow fibre compression of the idler from an OPA, which will be available for facility users from 2016. QUB has designed a new multi-jet target for HHG using quasi-phase matching [6], This should enable enhanced output at wavelengths where phase matching cannot be achieved by conventional methods.

LLAMS built a new pump laser operating at 300 Hz with a 50 ps pulse length and maximum pulse energy of 140 mJ at 1064 nm and ~90 mJ at 532 nm [7]. With this pump laser a chirped pulse optical parametric amplifier was recently realized producing up to 15 mJ (before compression) with a bandwidth of 720 nm - 920 nm (Fourier limit < 10 fs). When fully optimized it is expected to generate sub-10 fs ~1TW pulses at 300 Hz, providing a high power source for HHG.

LENS studied the enhancement of the conversion efficiency and the possible control of the spectral features of medium-order harmonics by exploring different interaction geometries and using gas regions with modulated density profiles.

CEA LIDyL has improved spatial quality of an IR laser beam by means of modal filtering. It combines standard spatial and modal filtering, using propagation in a hollow core fiber acting as a waveguide. With this simple and robust technique, IR beam quality is notably improved close to TEM00 mode. High-harmonic generation in rare gas shows enhancement of the efficiency and XUV beam quality [8].

At CLPU numerical simulations were carried out on high-order harmonic generation using non-homogeneous laser fields and resonant atomic systems to investigate optimization of the XUV yield, and the results were published in a series of papers [9, 10, 11].

4 Conclusions

This workpackage has been successfully completed; multiple new laser systems have been realized that have extended the energy range of HHG (even reporting a record high output at 500 eV) or the energy per pulse at lower photon energy (e.g. 145 eV, enabling attosecond physics) and improved XUV beam quality. Also the foundation has been laid for further improvements in the future by realization of optimized laser systems for HHG, special nozzles for quasi-phase matching, and theoretical/numerical investigations into the optimum conditions for generating high energy photons at a high yield.

5 References/Publications

(Publications resulting from the JRA need to indicate DOI and whether open access will be/is granted (yes/no). Please, remember the Laserlab-Europe, EC-GA 284464 acknowledgement.)

1. S. Neppl, R. Ernstorfer, A.L. Cavalieri, C. Lemell, G. Wachter, E. Magerl, E.M. Bothschafter, M. Jobst, M. Hofstetter, U. Kleineberg, J.V. Barth, D. Menzel, J.

Burgdörfer, P. Feulner, F. Krausz, R. Kienberger: „Direct observation of electron propagation and dielectric screening on the atomic length scale",, Nature **517**, 342 (2015). doi:10.1038/nature14094. No open access.

2. C. Lemell, S. Neppl, G. Wachter, K. Tőkési, R. Ernstorfer, P. Feulner, R. Kienberger, and J. Burgdörfer: Real-time observation of collective excitations in photoemission, Phys. Rev. B **91**, 241101(2015). DOI: http://dx.doi.org/10.1103/PhysRevB.91.241101. No open access.
3. http://www.opticsinfobase.org/abstract.cfm?uri=CLEO_QELS-2014-FTu3B.4
4. S.L. Cousin, F. Silva, S. Teichmann, M. Hemmer, B. Buades, J. Biegert, "High flux table-top soft X-ray source driven by sub-2-cycle, CEP stable, 1.85 µm 1 kHz pulses for carbon K-edge spectroscopy", Opt. Lett. **39**, 5383 (2014). No open access, doi: 10.1364/OL.39.005383
5. S.L. Cousin, F. Silva, S. Teichmann, M. Hemmer, J. Biegert, "Molecular fine structure from water-window coherent soft-X-rays", Optics and Photonics News, 12, 58 (2014), Optics in 2014 Highlight (access for OSA members).
6. Hage, A; Landgraf, B; Taylor, M; et al., "New design of a multi-jet target for quasi phase matching", Rev.Sci. Instr. **85**, 103105 (2014). No open access. http://dx.doi.org/10.1063/1.4897269 .
7. D.W. E. Noom, S. Witte, J. Morgenweg, R. Altmann, K.S.E. Eikema, "High energy, high repetition rate picosecond pulses from a quasi-CW diode pumped Nd:YAG system", Opt. Lett. **38**, 3021-3023 (2013). Open access, doi: 10.1364/OL.38.003021
8. B. Mahieu, D. Gauthier, M. Perdrix, X. Ge, W. Boutu, F. Lepetit, F. Wang, B. Carré, T. Auguste, H. Merdji, D. Garzella, O. Gobert, "Spatial quality improvement of a Ti:Sapphire laser beam by modal filtering", Appl. Phys. B **118**, 47–60 (2015), DOI 10.1007/s00340-014-5953-4, No open access; we apologize that Laserlab-Europe has been forgotten is the acknowledgements.
9. Marcelo Ciappina, Jose Antonio Pérez-Hernández, Tahir Shaaran, Maciej Lewenstein, *"*Coherent XUV generation driven by sharp metal tips photoemission*", Euro. Phys. J. D **68**:172 (2014) . No open access, doi: 10.1140/epjd/e2014-50060-4*
10. M.F. Ciappina, J. A. Pérez-Hernández, A.S. Landsman, T. Zimmermann, M. Lewenstein, L. Roso and F. Krausz, "Carrier-Wave Rabi-Flopping Signatures in High-Order Harmonic Generation for Alkali Atoms", *Phys. Rev. Lett. **114**, 143902 (2015) No open access, http://dx.doi.org/10.1103/PhysRevLett.114.143902*
11. M.F. Ciappina, J.A. Pérez-Hernández, L. Roso, A. Zaïr and M. Lewenstein, "High-order harmonic generation driven by plasmonic fields: a new route towards the generation of UV and XUV photons?", *J. Phys.: Conf. Ser.* **601** 012001 (2015) doi:10.1088/1742-6596/601/1/012001 (Open Access)