Characterization of ultrashort double pulses and their application in attosecond physics

PhD thesis

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Introduction

Light is an indispensable part of our life, yet we cannot touch it directly. Light allows us to perceive objects, creatures or our fellow beings, in other words, it allows us to see. Light is one of the fundamental natural elements, similarly to water or oxygen, however it is not a vital element. It is in fact electromagnetic radiation the mediating particles of which are the photons, which behave either as waves or particles, depending on the investigated physical effect. Electromagnetic radiation can most simply be characterized by the frequency of the mediating photons, or by the corresponding wavelength, and can be divided into different subgroups based on the "colour" of the mediating particles. In other words, the broad spectrum of electromagnetic radiation represents light having different coloured photons.

During biological evolution, the human eye, or more precisely the photosensitive visual organ called retina, has evolved in a way to be able to detect photons only from a certain wavelength range. In other words, our eye is sensitive only to photons of specific colours. This section of the electromagnetic spectrum is commonly called "visible light". Our visual organ is completely insensitive to the electromagnetic radiation outside of this wavelength region, which can even pose danger, as our body could be exposed to radiation without the realization of the potential hazard. Such electromagnetic waves come from outer space, for instance from the Sun, but fortunately injurious interstellar radiation is completely filtered out by the more than 10 km thick atmosphere of the Earth, so they cannot harm organisms or the human body. By taking into account all types of electromagnetic radiation, they cover broad spectral range, starting from cosmic and gamma rays with wavelengths of only a few picometres, through the region of X-rays and visible light, up to the domain of infrared and the radio waves reaching the micro- and several metres long wavelengths, respectively. Although the human eye is not capable of sensing such radiation, some of them have numerous well-known and very useful everyday applications, such as medical imaging devices, thermal cameras, routers used for wireless internet connection, or mobile phones, which is probably an integral part of our lives.

Lasers, the name of which comes from an acronym (Light Amplification by the Stimulated Emission of Radiation), also emit electromagnetic waves, however, with characteristics different from those of the applications summarized above. Because of the exceedingly advantageous and useful attributes of laser radiation, they are applied in almost all areas of life, for example in barcode scanners, modern printers, in the automotive industry, or during vascular or eye surgeries. Besides the colour of emitted photons, lasers can also be classified based on the temporal characteristics of their radiation. According to this categorization, we can distinguish two major types: lasers that have continuous wave radiation, and lasers which emit pulse trains. Lasers belonging to the first group are most often used for machining metals, while pulsed lasers are frequently utilized in research areas where ultrafast processes are investigated, since the temporal duration of the emitted pulses can go down to the femtosecond (1 fs = 10^{-15} s) range. This time period compares to a second as a second compares to ~ 32 million years. This means that we have such a short radiation that spans one millionth of one billionth of a second. For this reason, we are able to trace changes in charge migration. However, if we take into account the colour of laser photons as well, the temporal duration of the pulses cannot be less than one full optical cycle of the corresponding radiation. Accordingly, there exist a physical limit, which prevents the production of arbitrary short laser pulses.

Due to this limitation, scientists have developed a method, called high-order harmonic generation using femtosecond lasers to produce coherent radiation in the extreme ultraviolet wavelength region. The technique is based on the nonlinear frequency up-conversion of the driving laser, therefore the radiation produced in this way has shorter wavelength as well as higher frequency than the driving laser source. Consequently, the pulse duration limit mentioned previously can be overcome, and the production of shorter pulses down to the attosecond (1 as =

10^{-18} s) range becomes feasible. This paves the way towards the experimental examinations of processes occurring on *as* time scale, e.g. electron dynamics or other molecular and atomic processes.

Scientific background

In the following sections I will give a detailed overview about the most important characteristics of laser pulses, present a pulse reconstruction technique, and describe one of the experimental applications of ultrashort laser pulses, high-order harmonic generation (HHG).

Description of ultrashort laser pulses

In the mathematical description of ultrashort laser pulses the Fourier transform establishes the connection between the complex electric field and the complex spectrum. For the sake of easier mathematical handling, it is worth dividing the complex spectrum to positive and negative frequency terms, and in addition, it is worth expressing the electric field with real functions (real time-dependent amplitude and phase). In the most common cases, the spectral amplitude has distribution only around a well-defined angular frequency value, called central angular frequency. By using this angular frequency and the complex temporal envelope function, which can be described by the real temporal envelope and time-dependent phase, the electric field can be expressed. Thereby the formula describing the laser pulse's electric field can be written in a form which leads to simpler mathematical handling, and from which the physical effects acting on an ultrashort laser pulse can be easily deduced.

For example, the distorting effects on the laser pulse's temporal electric field variation are strongly related to the spectral phase. To describe these phenomena, the spectral phase must be expanded to Taylor series around the central angular frequency, where each phase-derivative is responsible for different pulse distortions. These include the carrier-envelope phase (CEP), which represents a phase shift between the peak of the pulse envelope and the electric field maximum, and the group delay (GD) that shifts the pulse in time only, without modifying the pulse shape itself. Additionally, the next phase derivative is called group delay dispersion (GDD), which extends the electric field and changes the instantaneous frequency. Here we must also mention third-order dispersion (TOD), whose main effect is to produce side-pulses with decreased intensity beside the main peak. These pulse-distorting effects must be determined before the experimental usage of laser pulses, otherwise the expected results will not be representative, or we will get a different experimental outcome.

Besides the above mentioned pulse-distorting effects, double pulse structures are used in numerous experimental applications, which sometimes positively, sometimes negatively affect the outcome of the experiment. From the mathematical point of view, the same formulas can be applied as for a single pulse. Because of the spectral interference of the two constituents of the double-pulse structure, the spectral amplitude changes when the temporal delay between them is varied. Furthermore, if the time delay is comparable to the temporal duration of the laser pulse, interference will occur directly between the electric fields, resulting in a complex intensity substructure.

The laser field envelope can be experimentally measured by various laser pulse reconstruction methods, from which numerous techniques have gained routine applications in laser laboratories worldwide. The most widespread of these methods are Frequency Resolved Optical Gating (FROG), Spectral Phase Interferometry for Direct Electric field Reconstruction (SPIDER), and Self-Referenced Spectral Interferometry (SRSI). In the following, I will describe in more detail the SRSI method, since this is indispensable to fully understand my motivation and results.

Self-referenced spectral interferometry

The self-referenced spectral interferometry pulse characterization technique utilizes the cross-polarized wave (XPW) generation process to create a reference pulse for the characterization of the unknown laser pulse. XPW is a frequency-conserving third-order nonlinear process, which is particularly sensitive to the polarization state of the driving pulse. Therefore, the measurement setup must contain an input polarizer, which assures the linear polarization state. The replica pulse, required for pulse characterization, is produced by a birefringent fused silica plate, after which the two pulses are orthogonally polarized, and delay τ is introduced between them. The two pulses propagate collinearly. After the fused silica plate, the reference pulse is generated by the p-polarized pulse in a BaF_2 crystal, while the s-polarized pulse goes through the crystal without any nonlinear interaction. Then, an output polarizer assures that the reference and the measured pulses have the same polarization state, thereby they can interfere. Finally, the interference pattern of the two pulses, or in other words the spectral interferogram is recorded by a spectrometer, having a broad spectral range. The measured interferogram contains the information needed for pulse reconstruction.

In the first step in the evaluation process of the spectral interferogram we need to inverse Fourier transform the given signal, thereby we obtain the temporal interferogram, which contains three well-separated peaks. There is one at the delay of 0 fs, and in addition, two peaks appear at positive and negative τ delay instants, respectively. By numerically filtering the peaks at the delay of 0 fs and $+\tau$, and by Fourier transforming them back to spectral domain, the spectral amplitude of both the measured and the reference pulses can be expressed by two analytical formulas. However, for complete temporal pulse characterization the calculation of the spectral phases is also required, which can be determined from the measured interferogram by an iterative phase retrieval algorithm. The input parameters of the algorithm are the previously calculated spectral amplitudes, and using the spectral interferogram, we must estimate the phase. The algorithm fine-tunes the input spectral phase in each iteration cycle, which converges to the final spectral phase after a few iterations. As a final step, the spectral phase can be extracted, which can be used for complete temporal pulse reconstruction.

Nevertheless, this iterative phase retrieval algorithm inaccurately determines the spectral phase which corresponds to a double-pulse structure, since in this case the spectral phase contains phase jumps, or even disruptions at the positions where the spectral amplitude drops to zero because of spectral interference between the two constituting pulses. Therefore, this results in a discontinuous spectral phase, which is challenging to retrieve by the iterative phase retrieval algorithm.

High-order harmonic generation

As I already mentioned in the Introduction section, femtosecond-long pulsed lasers are used to drive the high-order harmonic generation (HHG) process. During HHG, a nonlinear frequency upconversion of the driving laser radiation is carried out, thereby broadband, coherent, extreme ultraviolet radiation can be produced. Because of the symmetry properties of the process, spectral peaks appear in the spectrum of the produced radiation at the positions of odd multiples of the driving laser's central angular frequency. These spectral peaks with fixed, equidistant separation are called harmonics. The distance between the peaks is twice the central angular frequency of the driving laser, therefore the central photon energy of the distinct harmonics cannot be arbitrarily varied, since this feature is unambiguously determined by the spectral properties of the driving laser.

The HHG process can most simply be described by the three-step model. The model summarizes the harmonic generation phenomenon with three simple steps: (1) Firstly, the supposed atomic Coulomb potential is distorted by the interacting strong laser field in such a way that the bound electron is able to break free via tunnelling through the potential barrier. As a result, the motion of the freed electron is influenced only by the laser's electric field, the atomic potential does not affect it. (2) Then the laser's electric field accelerates the electron, and during motion it gains energy from the driving field. If the direction of the field reverses, the motion of the moving electron is oriented towards the ionic core. (3) While approaching the core, the electron recombines with the ionized nucleus, and simultaneously the energy gained from the laser's electric field is released in the form of a high energy extreme ultraviolet photon. These three steps occur every half-cycle of the driving laser field, which results in extreme ultraviolet radiation. This way the most important characteristics of the HHG process can be described, and the most important features of the harmonic dipole spectrum can be presented. However, to investigate harmonic generation in more detail, quantum mechanical approaches are required.

To be able to take into account quantum mechanical effects during HHG, the time-dependent Schrödinger equation must be solved. However, the numerical solution of this equation is complicated and exceptionally time-consuming even in case of a 1-dimensional model. The calculation can be significantly simplified by applying the so-called strong-field approximation, which means the following assumptions: (i) During light-matter interaction, the contribution of excited states can be neglected, and only the ground state of the atom takes part in the ionization process. (ii) The electron population of the ground state cannot be depleted. (iii) The ionized electron can be investigated as a free particle, and its motion is influenced only by the laser's electric field. In this case the effect of the Coulomb potential on the electron's motion can be neglected. By applying these assumptions, the single atom dipole spectrum can be expressed analytically. Since this approximation was first described by Lewenstein and co-workers, it is called Lewenstein integral. The numerical evaluation of the Lewenstein integral is relatively simple, thus the HHG process can be studied by considering the quantum mechanical effects as well.

Results

I. I have improved an already existing femtosecond laser pulse characterization technique, called SRSI to be able to reconstruct femtosecond double-pulse structures. I have shown that by manipulating the temporal interferogram, a spectral phase correction term can be extracted. The combination of this phase correction term and the originally retrieved spectral phase gives an improved final phase for complete pulse reconstruction. I verified the improved method by simulations, and I successfully reconstructed the temporal shape of experimentally produced double pulses. I determined the deviations of the retrieved spectral phases, obtained both by the original and the improved methods, from the expected spectral phase, and by this I quantitatively demonstrated the level of improvement of the technique on a wide range of amplitude ratios of double pulses [T1].

II: I have investigated theoretically the impact of the temporal double-pulse structure on the high-order harmonic generation process. I have proposed to use these electric field structures to control the spectral characteristics of the generated high-order harmonics. I have demonstrated with simulations that the central photon energy of the harmonics can be directly tuned, and that the tuning range broadens with increasing harmonic order. Moreover, I have shown that spectral narrowing of harmonics can also be achieved by increasing of the temporal delay between the driving interfering pulses [T2].

III: I successfully produced double pulses under experimental conditions in noncollinear geometry by utilizing a custom-made split-and-delay unit. I showed that the high-order harmonics generated using these pulse structures inherit the central angular frequency from the driving laser spectrum, thereby the central photon energy of harmonics can be controlled by changing the delay between the driving laser pulses. I demonstrated that the tuning range of certain harmonic orders can reach the photon energy of the fundamental laser field. Moreover, I measured that the delay-dependent spectral bandwidth of the driving laser manifests its effects in the bandwidth evolution of the generated high-order harmonics. With the measurements, I revealed that regardless of the spectral shape of a single pulse originating from the laser source, spectral characteristics of the generated harmonics can be controlled by using double pulses [T3].

Own publications

T - publications closely related to the thesis
O - other scientific publications

[T1] **L. Gulyás Oldal**, T. Csizmadia, P. Ye, N. G. Harshitha, M. Füle, and A. Zaïr, “Double-pulse characterization by self-referenced spectral interferometry,” *Appl. Phys. Lett.*, vol. 115, no. 5, p. 051 106, 2019. DOI: 10.1063/1.5089959.

[T2] **L. Gulyás Oldal**, T. Csizmadia, P. Ye, N. G. Harshitha, A. Zaïr, S. Kahaly, K. Varjú, M. Füle, and B. Major, “Generation of high-order harmonics with tunable photon energy and spectral width using double pulses,” *Phys. Rev. A*, vol. 102, p. 013 504, 2020. DOI: 10.1103/PhysRevA.102.013504.

[T3] **L. Gulyás Oldal**, P. Ye, Z. Filus, T. Csizmadia, T. Grósz, M. De Marco, Z. Bengery, I. Seres, B. Gilicze, P. Jójárt, K. Varjú, S. Kahaly, and B. Major, “All-optical experimental control of high-harmonic photon energy,” *Phys. Rev. Applied*, vol. 16, p. L011001, 2021. DOI: 10.1103/PhysRevApplied.16.L011001.

[O1] P. Ye, T. Csizmadia, **L. Gulyás Oldal**, H. N. Gopalakrishna, M. Füle, Z. Filus, B. Nagyillés, Z. Divéki, T. Grósz, M. Dumergue, P. Jójárt, I. Seres, Z. Bengery, V. Zuba, Z. Várallyay, B. Major, F. Frassetto, M. Devetta, G. D. Lucarelli, M. Lucchini, B. Moio, S. Stagira, C. Vozzi, L. Poletto, M. Nisoli, D. Charalambidis, S. Kahaly, A. Zaïr, and K. Varjú, “Attosecond pulse generation at eli-alps 100 khz repetition rate beamline,” *J. Phys. B: At. Mol. Opt. Phys.*, vol. 53, no. 15, p. 154 004, 2020. DOI: 10.1088/1361-6455/ab92bf.

[O2] D. You, K. Ueda, M. Ruberti, K. L. Ishikawa, P. A. Carpeggiani, T. Csizmadia, **L. Gulyás Oldal**, H. N. G, G. Sansone, P. K. Maroju, K. Kooser, C. Callegari, M. D. Fraia, O. Plekan, L. Giannessi, E. Allaria, G. D. Ninno, M. Trovò, L. Badano, B. Diviacco, D. Gauthier, N. Mirian, G. Penco, P. R. Ribič, S. Spampinati, C. Spezzani, S. D. Mitri, G. Gaio, and K. C. Prince, “A detailed investigation of single-photon laser enabled auger decay in neon,” *New J. Phys.*, vol. 21, no. 11, p. 113 036, 2019. DOI: 10.1088/1367-2630/ab520d.

[O3] T. Grósz, A. P. Kovács, K. Mecseki, **L. Gulyás**, and R. Szipőcs, “Monitoring the dominance of higher-order chromatic dispersion with spectral interferometry using the stationary phase point method,” *Opt. Commun.*, vol. 338, pp. 292–299, 2015, ISSN: 0030-4018. DOI: https://doi.org/10.1016/j.optcom.2014.10.047.

[O4] T. Grósz, **L. Gulyás**, and A. P. Kovács, “Advanced laboratory exercise: Studying the dispersion properties of a prism pair,” in *ETOP 2015 Proceedings*, Optical Society of America, 2015, TPE32. [Online]. Available: http://www.osapublishing.org/abstract.cfm?URI=ETOP-2015-TPE32.

Conference presentations

* Presenting author; OP - Oral presentation; PP - Poster presentation

[OP1] P. Ye, **L. Gulyás Oldal***, T. Csizmadia, Z. Filus, T. Grósz, P. Jójárt, I. Seres, Z. Bengery, B. Gilicze, S. Kahaly, K. Varjú, and B. Major, “High-flux, 100-khz attosecond pulse train source driven by a high average-power laser beam,” in *Frontiers in Optics + Laser Science*, Online, 2021.

[OP2] **L. Gulyás Oldal***, P. Ye, Z. Filus, T. Csizmadia, T. Grósz, M. De Marco, and B. Major, “Spectrally tunable attosecond pulse generation,” in *Conference on Lasers and Electro-Optics/Europe — European Quantum Electronics Virtual Conferences*, Online, 2021.

[OP3] P. Ye, **L. Gulyás Oldal**, T. Csizmadia, Z. Filus, T. Grósz, M. De Marco, P. Jójárt, I. Seres, Z. Bengery, Z. Várallyay, B. Gilicze, S. Kahaly, K. Varjú, and B. Major*, “High-flux attosecond source at 100 khz repetition rate,” in *Conference on Lasers and Electro-Optics/Europe -– European Quantum Electronics Virtual Conferences*, Online, 2021.

[OP4] T. Csizmadia*, P. Ye, **L. Gulyás Oldal**, Z. Filus, T. Grósz, P. Jójárt, I. Seres, Z. Bengery, Z. Várallyay, B. Gilicze, S. Kahaly, K. Varjú, B. Major, and M. Füle*, “Firstly commissioned secondary source of eli-alps: The hr ghhg gas beamline,” in *7th ELI-ALPS User Workshop*, Szeged, Hungary, 2019.

[PP1] **L. Gulyás Oldal***, P. Ye, Z. Filus, T. Csizmadia, T. Grósz, M. De Marco, Z. Bengery, I. Seres, B. Gilicze, P. Jójárt, K. Varjú, S. Kahaly, and B. Major, “All-optical control of high-harmonic photon energy,” in *Frontiers in Optics + Laser Science*, Online, 2021.

[PP2] **L. Gulyás Oldal***, P. Ye, T. Csizmadia, T. Grósz, Z. Filus, and B. Major, “Generation of high-order harmonics with tunable photon energy and spectral width using double pulses,” in *ELI Summer School*, Szeged, Hungary, 2020.

[PP3] **L. Gulyás Oldal***, T. Csizmadia, P. Ye, N. G. Harshitha, A. Zaïr, and M. Füle, “High-harmonic generation in gaseous medium by double pulse forms,” in *7th International Conference on Attosecond Science and Technology*, Szeged, Hungary, 2019.

[PP4] T. Csizmadia*, **L. Gulyás Oldal**, P. Ye, N. G. Harshitha, M. Füle, and A. Zaïr, "Microscopic analysis of the intensity and wavelength dependence of quantum path interferences in high-order harmonic generation," in *7th International Conference on Attosecond Science and Technology*, Szeged, Hungary, 2019.

[PP5] N. G. Harshitha*, T. Csizmadia, P. Ye, M. Füle, **L. Gulyás Oldal**, E. Constant, and A. Zaïr, "Characterisation of attosecond pulse train using windowed rabbitt technique," in *7th International Conference on Attosecond Science and Technology*, Szeged, Hungary, 2019.

[PP6] Z. Filus*, T. Csizmadia, M. Füle, **L. Gulyás Oldal**, N. G. Harshitha, P. Ye, B. Nagyillés, Z. Divéki, T. Tímár-Grósz, M. Dumergue, A. Zaïr, S. Kahaly, and K. Varjú, "High harmonic generation gas cell for high repetition rate secondary sources," in *7th International Conference on Attosecond Science and Technology*, Szeged, Hungary, 2019.

[PP7] M. Füle*, P. Ye, T. Csizmadia, N. G. Harshitha, **L. Gulyás Oldal**, B. Nagyillés, Z. Divéki, T. Tímár-Grósz, M. Dumergue, Z. Filus, P. Jójárt, I. Seres, V. Zuba, Z. Bengery, M. Devetta, F. Frassetto, B. Moio, M. Lucchini, M. Nisoli, L. Poletto, S. Stagira, C. Vozzi, G. Sansone, K. Osvay, S. Kahaly, K. Varjú, and A. Zaïr, "First attosecond beamline operational at eli-alps," in *7th International Conference on Attosecond Science and Technology*, Szeged, Hungary, 2019.

[PP8] T. Csizmadia*, **L. Gulyás Oldal**, P. Ye, N. G. Harshitha, M. Füle, and A. Zaïr., "Double pulse characterisation by self-referenced spectral interferometry," in *ELI Summer School*, Szeged, Hungary, 2018.

[PP9] **L. Gulyás Oldal***, T. Csizmadia, P. Ye, N. G. Harshitha, M. Füle, and A. Zaïr, "Double pulse characterisation by self-referenced spectral interferometry," in *Erice Attosecond School - The Frontiers of Attosecond and Ultrafast X-ray Science*, Erice, Sicily, Italy, 2019.

[PP10] N. G. Harshitha*, T. Csizmadia, **L. Gulyás Oldal**, P. Ye, M. Füle, K. Varjú, G. Sansone, and A. Zaïr, "Characterisation of attosecond pulse train using rabbitt method," in *Laser Plasma Summer School*, Salamanca, Spain, 2018.