Few-cycle pulse generation in the mid-infrared and THz spectral domains

PhD Thesis Booklet

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2022

1. Introduction

The history of nonlinear optics started with the theory of two-photon absorption, which was predicted in 1931, by Maria Goeppert Mayer in her PhD thesis. However, the scientific community had to wait another thirty years for the first experimental evidence. The construction of the first laser (light amplification by stimulated emission of radiation) in 1960 by T. H. Maiman, made it possible to develop light sources with sufficient intensity for nonlinear optics. W. Kaiser and P. A. Franken demonstrated two-photon absorption and second harmonic generation (SHG) in 1961.

Shortly after the discovery of SHG, the sum frequency generation (SFG) was demonstrated in 1962 by M. Bass and R. C. Miller. This nonlinear optical process allowed the development of tunable light sources in the ultraviolet (UV) spectral domain. Another important nonlinear optical phenomenon is difference frequency generation (DFG), which was demonstrated in 1963 by A. W. Smith. This nonlinear optical process allowed the development of tunable light sources in the infrared (IR) spectral domain. Another nonlinear optical phenomenon is optical parametric amplification (OPA), which was presented in 1965 by S. A. Akhmanov and J. A. Giordmaine. This nonlinear optical process is able to effectively multiply/amplify the photons in the IR spectral domain.

G. Mourou and D. Strickland have made important contribution to nonlinear optics by the development of chirped pulse amplification (CPA). CPA allowed for the construction of laser systems with peak powers in the Terawatt (TW) and Petawatt (PW) ranges. The next important development step in the history of nonlinear optics came in 1992, when A. Dubietis, G. Jonusauskas and A. Piskarskas combined the OPA and the CPA phenomena and developed the optical parametric chirped pulse amplification (OPCPA) scheme. This technology made available light sources with even more diverse spectral ranges and improved the spectral tunability of these light sources.

One of the current trends in few-cycle pulse development is to move towards longer central wavelengths, to the mid-infrared (MIR) spectral region, where OPCPA sources are routinely built nowadays. The advantage of a longer carrier wavelength in these systems is the ponderomotive potential. The secondary light source pumped with MIR laser systems produce higher cut-off photon energy for high harmonic generation (HHG) and generate higher intensities for Terahertz (THz) pulse generation. Pulse duration can be further shortened by subsequent self-phase modulation (SPM) in bulk solid-state media followed by re-compression. A possible solution for the re-compression is the use of dispersive mirrors (DMs), which can be designed to have an arbitrary spectral phase.

THz covers the longwave infrared (LWIR) and the far infrared (FIR) spectral domains. Besides the fact that natural THz radiation surrounds us as background radiation, there are several ways to generate coherent THz sources. An important scheme, which is the subject of my doctoral dissertation, is the two-color ionization in gases. This experimental approach results in spectrally broad and coherent THz radiation, which is a useful tool for THz time-domain spectroscopy (THz-TDS). The THz radiation source is the transverse electron current (TEC) built up inside plasma, which is a renewable secondary source due to its generating medium: air. Several scientific studies have shown that the efficiency of this type of THz generation increases with the carrier wavelength of the fundamental pulse due to the ponderomotive potential.

2. Scientific background

The different spectral domains of the electromagnetic spectrum can be classified according to the physical, chemical, and biological effects of the electromagnetic waves. The main topic of my doctoral dissertation focuses on the infrared (IR) spectral domain, which wavelength range starts at 0.7 μm and ends at 1 mm. This spectral domain was first recognized by Sir William Herschel in 1800. The IR spectral domain has the following subdomains: the near infrared (NIR) (0.7 μm – 1.4 μm), the shortwave infrared (SWIR) (1.4 μm – 3 μm, the mid-infrared (MIR) (3 μm – 8 μm), the LWIR (8 μm – 15 μm) and the FIR (15 μm – 1 mm). The IR spectrum has numerous useful applications in spectroscopy, metrology, climatology, astronomy, and defense. IR spectroscopy is a very useful method when it comes to the identification, quantification, and characterization of materials. THz spectroscopy has several advantages: for example, most packaging materials, such as paper, cardboard, and textile, are transparent in this spectral range. Furthermore, many materials, such as explosives, narcotics, and medicines, have unique spectral characteristics in the THz spectral domain, which allows for their identification.

In linear optics, the dielectric polarization of the material is a linear function of the applied electric field. When the applied electric field is in the order of 5.14×10^{11} V/m, the second term of the dielectric polarization cannot be neglected anymore. There are four different second order nonlinear optical processes: optical rectification (OR), SHG, SFG and DFG. However usually only one of them is intense enough to generate a significant number of photons. The energy and the impulse conversion determine which process is the dominant. When the applied electric field is in the order of 2.64×10^{23} V/m, the third term of the dielectric polarization cannot be neglected anymore. The most important third order nonlinear optical process: third harmonic generation, four wave mixing (FWM), self-focusing and self-phase modulation.

THz pulse generation from two-color pulses ionized plasma is a simple and renewable, secondary sources for THz spectroscopy. In this method, the source of radiation is the transverse electron current (TEC) inside the laser induced plasma. The most important step in this process is the formation of an asymmetric electric field, which accelerates free electrons in one direction more efficiently than in the other direction, resulting in a non-vanishing TEC build-up inside the plasma. The two simplest options to achieve it is the use of few-cycle or/and two-color pulses. The two-color pulses are created through the combination of the fundamental beam and its second harmonic (SH). The combined electric fields produce tunnel, whereupon the free electrons accelerate. There are two main, but conceptually different explanations of the physical processes for THz pulse generation from gases: the FWM-OR and the TEC model. Although both descriptions explain most of the experimental results, it has been observed that TEC works better in the low frequency domain (<10 THz), therefore the numerical simulations presented in my work used this model.

The are several measurement techniques for the spectral phase characterization of optical elements. Spectrally resolved interferometry (SRI) is one of the most widely used technique. In most cases, it utilizes a two-arm interferometer, illuminated by a broadband light source and detected by a high spectral resolution spectrometer. With the appearance of spectrometers providing high spectral resolution, it became clear that the most accurate evaluation methods are the ones based on the Fourier transformation (FT). In the next step we need to apply the inverse FT, which connects the spectral and temporal domains of the electric fields. In the following step we need to filter out the peak around $+\tau$, which is the time delay between the two arms of the interferometer. The final step is the FT, where we obtain a complex result. The amplitude is the spectral profile, while the angle is the spectral phase. The evaluation of the conventional SRI and the SH-SRI is the same as described. The only difference is that the recovered spectral phase of the SH-SRI is two times the actual spectral phase.

3. Applied methods

During my research I have developed experimental arrangements and numerical simulations. These are summarized in the following paragraphs.

The optical layout of the SRI is based on the Mach-Zehnder interferometer, where the input beam is divided into two roughly equal parts by a CaF_2 beam splitter. The sample and reference arms both contain three unprotected flat gold mirrors (GMs). The two beams are combined by the same type of CaF_2 beam splitter as the first one. Note that both the sample and the reference beams underwent one transmission and one reflection on the CaF_2 beam splitters, which nullifies the spectral phase difference between the two arms in case of an empty interferometer. After combination, the beams focused on the entrance slit on a MIR spectrometer (Fastlite – Mozza), which had a spectral resolution of 5 cm-1 (~2.5 nm).

The optical layout of the SH-SRI is based on the same interferometer as the SRI. However, this time the output went through an SHG stage. The combined beams were focused by a 150 mm focal length, CaF_2 plan-convex lens with broadband antireflection (BBAR) (2-5 μm) coating into a 100 μm thick, silver gallium sulfide (AgGaS2 or AGS) crystal (θ=39°, φ=45°) placed at the focus for SHG. Both the fundamental and SH beams collimated by another 150 mm focal length CaF_2 lens with BBAR (1.4-1.7 μm) coating at the SH wavelength. Thereafter, the fundamental and the SH beams were separated by a DcM. Finally, the SH beams were sent to a NIR optical spectrum analyzer (Yokogawa – AQ6375B), which spectral resolution is 0.05 nm. In both cases the evaluations are based on the FT method.

The postcompression setup driven by the MIR OPCPA system at ELI-ALPS. The pulse (130 μJ, 42 fs in FWHM, 100 kHz) was focused in air with a concave spherical GM, which had a –1000 mm radius of curvature to guarantee a sufficiently large spot size in the focus without plasma development in air. The chosen ROC value was a good compromise between broadening and stability. Behind the focal plane of the focusing GM, thin plates were positioned at normal incidence along the beam propagation axis for SPM. The spectrally broadened beam was collimated with, CaF_2 BBAR (2–5 μm) plan-convex lens having a focal length of 250 mm. After collimation, the beam was sampled with a specially coated sapphire sampler window having 7% reflectance between 2.2 μm and 4.2 μm, and nearly zero GDD and TOD for the reflected beam. I used the reflected beam for the spectrum and the CEP measurements. The BaF_2-Si double plate combination proved to be the most advantageous choice. With the help of three DMs aligned at 10°, 25° and 25° angle of incidences, respectively, and a 5 mm thick CaF_2 plate, the laser pulses were compressed down to 19.6 fs FWHM, which is less than two optical cycles at this central wavelength. After recompression, I had an average power of 8.24 W. The pulse duration (19.6 fs) was measured by a locally developed, all-reflective SHG frequency resolved optical gating (FROG).

I developed the numerical model the simulate the THz pulse generation in nitrogen, which starts with the SHG with the slowly varying amplitude approximation. First, the nonlinear crystal is split into several thin slices, where the linear and nonlinear wave equations are solved for a short distance, sequentially for each thin slice. It was used for the calculations of the energies, the spectral intensity and the pulse phase of the SH pulse. this illustrative numerical simulation. In the next step, I calculated the focusing of the beams based on the ABCD law. In this numerical simulation, to avoid any unwanted absorption during propagation, the full beam path was considered to be in dry nitrogen, while dispersion was assumed to follow on the generalized Sellmeier equation. In the focus, the tunnel ionization rate was calculated based on the Ammosov-Delone-Krainov (ADK) formula. Once the electrons are free, they are accelerated in the asymmetric electric field. The time dependent velocity of the free electrons is calculated next. After both the density and velocity of the free electrons are known, the transverse electron current (TEC) was calculated. THz radiation is in the low angular frequency part of the spectrum emitted by the TEC.

4. New scientific results.

My most important scientific results are summarized as follows:

I: I developed a spectrally resolved interferometer assisted by second harmonic generation. Thanks to this innovation, I was able to overcome the limited spectral resolution of commercially available spectrometers in the mid infrared spectral domain. I achieved this by transferring the spectral phase information in the near infrared spectral domain, where commercial spectrometers with higher spectral resolution are more common. I verified the newly developed second harmonic assisted spectrally resolved interferometer with materials with well-known spectral phase. I also compared it with the conventional spectrally resolved interferometer in terms of root mean square deviation and standard deviation, and it was found to perform better in both terms. Using this technique, I was able to measure the group delay spectrum of the newly manufactured dispersive mirror pair designed for postcompression in the mid infrared spectral domain, and I found good agreement with the designed value. [T1]

II: I used the dispersive mirror pair characterized in the first thesis point and developed the postcompression stage of the mid infrared laser system at ELI ALPS Research Institute. I tested several available materials for self-phase modulation. Barium fluoride, potassium bromide, and silicon were found to be best in terms of transmission and spectral broadening. I also tested different combinations of these materials, and the barium fluoride and silicon pair was found to have the highest potential peak power. I was able to compress the pulse duration almost down to the Fourier transform limit with the help a calcium fluoride window and three dispersive mirrors. I managed to shorten the pulse duration, which was originally five optical cycles (~50 fs) to less than two optical cycles (~20 fs) during postcompression. During my work, another aim was to increase the peak intensity of the system. Here I achieved a 30.3% increment. [T1]

III: I investigated the possibility of using the mid infrared system at ELI ALPS Research Institute to generate terahertz pulses in nitrogen plasma with two-color pulses. With numerical simulation I investigated the efficiency improvement in terahertz pulse generation with mid and longwave infrared two-color pulses compared to near infrared two-color pulses with the same input pulse parameters. I investigated the spectral domain from 2.15 μm to 15.15 μm and tested different thicknesses of the nonlinear crystal. I found that the scaling law of the terahertz pulse generation increases with increasing nonlinear crystal thickness. From this I concluded that terahertz pulse generation is very sensitive to second harmonic generation. Furthermore, I investigated the scaling law with different cutoff frequencies and found that it slowly increases with decreasing cutoff frequency. From this I concluded that the lower frequency part of the terahertz spectrum is more sensitive to the driving wavelength. Finally, I found that terahertz radiation generated with mid infrared (3.3 μm) and longwave infrared (7.3 μm) is by one and two orders of magnitude more intense than the terahertz radiation generated with near infrared (0.8 μm) two-color pulses. [T2]

IV: I investigated the relative phase tuning ability of thin dielectric plates with mid infrared, two-color pulses during terahertz pulse generation in nitrogen plasma. My aim was to determine which material is best suited for fine-tuning the relative phase between the two-color pulses, and thus also for optimizing the peak intensity of the terahertz pulse. I investigated terahertz pulse generation after several different materials, which are transparent in the mid infrared spectral domain. I also simulated the terahertz pulse generation after different material thicknesses and angles of incidence. I found that the best candidates are different fluorides, especially lithium and calcium fluoride. These materials have negative group velocity dispersion, and thanks to this they are not just able to control the relative phase between the two-color pulses but also improve the temporal overlap between them, which further increases the efficiency of terahertz pulse generation. [T2]

V: I investigated terahertz pulse generation in nitrogen plasma as a function of pulse duration and polarization angle. I identified three distinctly different regions, where theo optimal pulse parameters for the most efficient terahertz pulse generation are different. The first or conventional region is where the two-color pulse scheme is dominant above 3.2 optical cycles (34.1 fs at 3.2 μm). The second or unconventional region is, where the one-color pulse scheme is dominant below 1.7 optical cycles (18.1

fs at 3.2 μm). The last or semi-conventional/transitional region is, where the two-color pulse scheme is still dominant, but the sign of the relative phase is also determinative. In the conventional region, two-color pulses with the opposite sign of relative phase generate terahertz pulses with the same absolute peak power, but this is not true in the transitional region. In addition, I found that the lower boundary of the conventional region increases with increasing central wavelength, while the upper boundary of the unconventional region decreases with increasing central wavelength. [T3]

VI: I investigated the carrier to envelope and the relative phases effect on terahertz pulse generation as a function of pulse duration and polarization angle. As the pulse duration gets shorter, terahertz pulse generations moves away from the conventional to the unconventional scheme. That is why the sensitivity of the terahertz pulse generation to the carrier to envelope phase increases, while sensitivity to the relative phase decreases with decreasing pulse duration. As the pulse duration decreases, the need for the second harmonic pulse also decreases, which also reduces the effect of the relative phase. As the pulse duration increases, the number of individual optical cycles increases, which reduces the difference between them, and consequently diminishes the impact of the carrier to envelope phase. Furthermore, I also investigated the effect of the polarization angle of the fundamental pulse on these sensitivities. I found that the switching point, the point where the two sensitivities are equal, increases with a decreasing polarization angle. As the polarization angle decreases, the efficiency of second harmonic generation also decreases, which reduces the importance of the relative phase. This, in turn, increases the position of the switching point. [T3]

5. Magyar nyelvű összefoglaló

A tudományos munkám során a néhány optikai ciklusból álló impulzusok keltését vizsgáltam a közép és a távoli infravörös tartományokban. A hosszabb központi hullámhossz egyik előnye a ponderomotoros erő, ami egy a hullámhossz négyzetével arányos erőhatás. Ennek köszönhetően, a hosszabb központi hullámhosszon működő rendszerekkel gerjesztett másodlagos források például nagyobb levágási fotonenergiát biztosítanak a magasharmonikus-keltés során vagy nagyobb intenzitást eredményeznek terahertzes impulzus generálás során. A THz-es sugárzásnak számos előnye van a spektroszkópia területén: például sok anyagnak, például robbanóanyagok, kábítószerek és gyógyszerek egyedi spektrális jellemzőkkel rendelkezik a THz-es spektrális tartományban, ami lehetővé teszi azonosításukat.

A munkám során kifejlesztettem egy frekvenciakétszerezéssel segített, spektrálisan bontott interferométert. Ennek köszönhetően képes voltam megkerülni a közép infravörös spektrális tartományban elérhető spektrométerek limitált spektrális felbontásán. Ezt úgy értem el, hogy a spektrális fázisinformációt átvittem a közeli infravörös spektrális tartományba, ahol a jobb spektrális felbontással kapható spektrométerek érhetőek el.

A munkám során megépítettem az ELI ALPS Kutatóintézet közép infravörös rendszerének a posztkompressziós fokozatát. Az eredetileg öt optikai ciklusból álló impulzust sikerült két optikai ciklus hosszúságúra rövidíteni a posztkompresszió során. Munkám során a csúcsintenzitást is sikerült növelni.

A munkám során numerikus szimuláció segítségével megvizsgáltam a központi hullámhossz, az impulzushossz, a polarizációs szög, a vivő-burkoló fázis és a relatív fázis THz-es impulzuskeltésre gyakorolt hatásait. A központi hullámhossz vizsgálata során azt tapasztaltam, hogy a középinfravörös impulzussal egy, míg a hosszúhullámú infravörös impulzussal két nagyságrenddel intenzívebb a keltett THz-es impulzus, mint a közeli infravörös impulzussal esetén. Az impulzushossz vizsgálata során azt tapasztaltam, hogy három tartomány figyelhető meg, ahol a THz-es impulzuskeltés optimális impulzusparaméterei eltérőek. A fázisok vizsgálata során azt tapasztal, hogy az impulzushossz csökkenésével a vivő-burkoló fázis hatása növekszik, míg a relatív fázis hatása csökken.

6. Publications

Publications related to the thesis:

[T1] **R. Flender**, A. Borzsonyi, V. Chikan, “Phase-controlled, second-harmonic-optimized terahertz pulse generation in nitrogen by infrared two-color laser pulses”, Journal of the Optical Society of America B 37(6) 1838-1846 (2020)
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[T2] **R. Flender**, M. Kurucz, T. Grosz, A. Borzsonyi, U. Gimzevskis, A. Samalius, D. Hoff, B. Kiss, “Dispersive mirror characterization and application for mid-infrared postcompression”, Journal of Optics 23(6), 065501 (2021)
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[T3] **R. Flender**, A. Borzsonyi, V. Chikan, “The role of asymmetry in few-cycle, mid-IR pulses during THz pulse generation”, Journal of Optics 24(4), 045502 (2022)
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Further scientific publications:

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[F2] C. Vass, B. Kiss, **R. Flender**, Z. Felhazi, P. Lorenz, M. Erhardt, K. Zimmer, “Comparative study on grating fabrication in transparent materials by TWIN-LIBWE and ultrashort pulsed ablation techniques”, Journal of Laser Micro/Nanoengineering 10(1), 38-42 (2015)
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Társszerzői nyilatkozat

Alulírott nyilatkozom arról, hogy Flender Roland *„Few-cycle pulse generation in the mid-infrared and THz spectral domains"* című doktori értekezésének T1, T2, T3, T4, T5 és T6 tézispontjaiban szereplő, az alábbi cikkekben közösen publikált eredmények elérésében a jelölt szerepe meghatározó volt. Ezeket az eredményeket korábban nem használtam tudományos fokozat megszerzésére, és ezt a jövőben sem teszem.

T1-T2. R. Flender, M. Kurucz, T. Grosz, A. Borzsonyi, U. Gimzevskis, A. Samalius, D. Hoff, B. Kiss, "*Dispersive mirror characterization and application for mid-infrared post-compression*", Journal of Optics 23(6), 065501 (2021)
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T5-T6. R. Flender, A. Borzsonyi, V. Chikan, "*The role of asymmetry in few-cycle, mid-IR pulses during THz pulse generation*", Journal of Optics 24(4), 045502 (2022)
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