Ph.D. Thesis Booklet

Advanced metrology for few-cycle mid-IR pulses

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Szeged
2021

I. Introduction

In recent decades, we have been witnessing the rapid development of mid-IR laser sources. This revolution has been fueled by the numerous applications for mid-IR radiation such as strong-field physics in solid-state media, time-resolved spectroscopy, remote sensing, just to name a few. One of the promising applications for high intensity mid-IR pulses is attosecond science. Attosecond pulse generation is based on high-harmonic generation (HHG) driven by a few-cycle driving laser pulse. HHG is a strongly nonlinear process during which a target is ionized by an intense laser pulse, generating free electrons. Due to the sign change of the oscillating optical electric field, the returning electron recombines with the parent ion generating harmonic radiation. The maximum achievable photon energy depends on the ponderomotive energy, the quiver energy of the ionized electron in the laser field, which scales quadratically with the driving wavelength. As a results, a high wavelength driving laser can produce a broader HHG spectrum, which allows for the generation of even shorter attosecond pulses. However, high harmonic generation efficiency falls drastically with increasing wavelength. To generate a broad HHG spectrum with sufficient yield, few-cycle, high energy, CEP stable mid-IR systems are required.

Today, the most common solid state lasers use neodymium (Nb^{3+}), ytterbium (Yb^{3+}), or titanium (Ti^{3+}) dopants, emitting in the near infrared wavelength range. Femtosecond mid-IR lasers started to appear only at the turn of the century. Common laser components such as dispersive mirrors, high-reflective coatings, acousto-optic modulators, etc. have less favorable parameters compared to their near-IR counterparts. They are also more expensive, and are manufactured only by a few companies. Another issue is that the assortment of detection and diagnostic devices for mid-infrared lasers is also limited. The aforementioned near-IR femtosecond lasers can use detection technologies based on silicon, the most common material in the semiconductor industry. However, silicon is not suitable for mid-infrared detection, so InAsSb, or HgCdTe compounds are used as base materials for semiconductor detectors, which generally increases production costs. Furthermore, detector arrays or cameras for mid-infrared radiation are prohibitively expensive. For this reason, a large number of diagnostic tools for femtosecond mid-infrared lasers utilize workaround methods. As part of my research, my aim was to expand the palette of these diagnostic tools and develop cost-effective simple methods for the mid-IR spectral region.

In my dissertation three methods are presented, developed for ultrafast mid-infrared lasers. For the majority of the experiments the MIR laser system of ELI-ALPS was employed. This laser system generates 4-cycle laser pulse around 3.2 μm central wavelength at 100 kHz repetition rate. The average power of the laser is 14 W (140 μJ pulse energy), which corresponds to 3.3 GW peak power for a laser pulse.

The experiments discussed in my dissertation were performed in the laboratories of ELI-ALPS Laser Research Institute (ELI-HU Non-Profit Ltd.). A preliminary experiment was performed for the TOUCAN method at the University of Bordeaux with the supervision of Prof. Eric Cormier. The TIPTOE method (tunneling ionization with a perturbation for the time-domain observation of an electric field) was developed at the Gwangju Institute of Science and Technology, which provided us with a useful reference measurement for the temporal characterization of the post-compressed pulses.

II. Scientific background

Mid-infrared radiation

Light can be described as a transverse electromagnetic wave with attributes, that can be calculated from the wave equation. Primarily, it can be described by its wavelength or frequency, which can be the basis to assign light to different subcategories. The human eye is only sensitive to a very narrow wavelength region of light – called visible light – between roughly 400 and 700 nm. Longer wavelength light waves are called infrared radiation, which can be subdivided into further categories: near-infrared, mid-infrared, long-wavelength infrared and far infrared. This categorization is based on the wavelength dependence of microscopic processes during light-matter interaction. The consequence of this fundamental difference is that near-infrared optics and detectors are mostly unusable for the mid-IR.

Carrier-envelope phase

The temporal distribution of the electric field of a laser pulse can be described as the product of the envelope of the pulse and the oscillating carrier wave. The phase difference between the carrier and the envelope changes from pulse to pulse, and it is called carrier-envelope phase and abbreviated as CEP. When the laser pulse is sufficiently short (only a few-cycle long), then the CEP value can dramatically influence the temporal distribution of the electric field of the pulse, consequently the outcome of extreme nonlinear effects (ionization, recombination). For this reason, it is necessary to monitor the CEP changes, or to stabilize the CEP of the pulse train. Several methods have been developed in the last two decades to measure the shot-to-shot change in CEP. The single-shot CEP measurement techniques can be divided into two categories, depending on whether they are based on nonlinear interferometry or ionization. The advantage of interferometric methods is that they only require small peak power pulses. On the other hand, ionization based methods have the benefit that single-shot measurements can be performed at high repetition rates (>10 kHz) too. The TOUCAN method presented in my dissertation combines the benefits of these two groups, it is capable of the single-shot measurement of low peak power pulses at high repetition rates.

Spectral phase-shift measurement

When light is polychromatic, the solution to the wave equation can be presented as the superposition of different monochromatic waves. The optical energy in pulses emitted by pulsed laser sources is concentrated in short bursts, which allows for higher peak intensities compared to continuous lasers. These pulses can be similarly divided into their spectral components, which make up the spectrum of the pulse. The dispersion phenomenon stems from the velocity difference between these monochromatic wave components, which results in different phase shifts of these waves; it stretches or compresses the pulse in time. The spectral phase of optical elements (mirrors, windows, fibers, gratings, etc…) can be measured by several techniques. One of the most easily implementable and straightforward of them is spectrally resolved interferometry, or SRI for short. Spectral measurement is required for the recording of an SRI interferogram, where the spectral span of the recovered information is limited by the spectrum of the coherent light source. The dual-band SRI method, presented in my dissertation, is a modified version of the SRI method. This method is suitable to determine the spectral phase shift of optical elements at the fundamental and SH (second harmonic) frequency ranges in parallel from an interferogram recorded exclusively at the SH frequency region.

Post-compression

Ultrafast CEP stable mid-IR laser pulses can most often be generated with the OPCPA (optical parametric chirped pulse amplification) technology. The pulse duration of these laser pulses is limited, among other things, by the spectral bandwidth of the nonlinear crystals used for amplification. The resulting pulses can further be compressed in time using the post-compression method. Post-compression is based on spectral broadening resulting from a nonlinear optical process called self-phase modulation. This can be achieved in several media, waveguides, gas-filled cells, or in thin transparent plates. After this process, the shortest pulse duration can be achieved by completely compensating for the dispersion. In my dissertation I present the spectral broadening and temporal compression of pulses from the MIR laser system of ELI-ALPS, using thin-plate post-compression. With this method, the mid-IR pulses were compressed to close to half of the pulse duration of the original pulses, which was proven by two independent measurement techniques.

III. New scientific results

My most important scientific achievements are summarized in the following thesis points:

I. I have developed an interferometric delocalized CEP measuring method, named TOUCAN, which is capable of single-shot measurements of few μJ energy pulses at higher repetition rates than ever before. The device was experimentally verified using the 100 kHz repetition rate MIR laser system. Shot-to-shot pulse energy change and temperature fluctuations as potential sources of measurement error were examined, and I have concluded that they have minimal effect on the accuracy of CEP measurements in the case of the examined laser. I have also shown that time jitter between the trigger and the detected photodetector signals significantly decreases the accuracy of the measurement. I have circumvented this issue by creating an additional interference in the detected signal and removed the effect of time jitter via post-processing. The analysis of the CEP noise showed overwhelmingly high frequency noise components, which proves that CEP measurement and stabilization at full repetition rate is necessary to further reduce CEP noise in the MIR laser system. [T1]

II. I have developed a technique called dual-band SRI to characterize the spectral phase shift of optical elements at the fundamental and SH frequency bands of an ultrafast laser source simultaneously from interferograms recorded exclusively at the SH frequency band. The technique was first examined by theoretical description and numerical simulation, and was then tested experimentally. I built a dual-band SRI device, and performed measurements using 9 μJ energy pulses from the MIR laser system. The spectral phase shift of well-known optical materials was measured, which verified both the technique and the device. Furthermore, I have proven that a single measured interferogram holds double the information necessary for the complete reconstruction of the spectral phase shift at both bands, by employing and comparing different evaluation methods. By using the wave theory description of dual-band SRI, I examined further extensions of the method to measure more than two frequency bands simultaneously. [T2]

III. I have designed and built a thin-plate post-compression setup for the spectral broadening and compression of pulses from the MIR laser system. Overall, compression to 23 fs FWHM pulse duration was achieved from the 50 fs 100 kHz repetition rate mid-IR pulse train. I found that the combinations of YAG and Si materials are ideal for spectral broadening thanks to their linear and nonlinear optical properties. This was verified experimentally by using a 2 mm thick YAG and a 1 mm Si crystal plate, where intensities in the materials were fine-tuned by moving the plate positions with respect to the focusing mirror. Recompression to 23 fs pulse duration was achieved in a 3.3 mm thick CaF2 bulk, proven by the measurement of two distinct diagnostic devices, SH-FROG and TIPTOE. Long-term pulse-to-pulse energy, power, spectrum and CEP stability of the output was monitored for 8 hours continuously, which proved that it can be utilized for long-term experiments. [T3]

IV. Methods of investigation

During my research I have employed several experimental methods and devices. These are summarized in the following points.

I. The TOUCAN method is based on nonlinear interferometry and dispersive Fourier transform. The dispersion parameters of the fiber necessary for the dispersive Fourier transform were determined using the spectrally modulated signal of the 2f-to-f interferometer. Spectral intensity distribution was measured by an optical spectrum analyzer before coupling into the fiber. After propagation in the fiber, the temporally stretched signal was transformed into an electric signal by a photodetector, and it was recorded by an oscilloscope. The same oscilloscope and photodetector were employed during the TOUCAN measurement as well. At different set CEP values the recorded spectra and temporal distributions were used the determine the dispersion parameters of the fiber using an iterative algorithm. I have performed a reference measurement for the delocalized CEP using a commercial CEP measuring device (Fringeezz, Fastlite) at 10 kHz repetition rate. During the comparison of the CEP values, the CEP data recorded at 100 kHz with the TOUCAN method were decimated (every tenth datapoint was kept). The corresponding CEP datapoints were plotted on a 2D scatter plot, and I have performed linear fitting on these points as well. Furthermore, I have calculated the circular correlation between the corresponding data and compared their fluctuations. To measure the temporal jitter of the TOUCAN method, a Fabry-Peron etalon was used to create a stationary interference. The CEP sensitive and CEP insensitive interferences were separated using the Fast Fourier transform (FFT) numerical method. Afterward, the error of the measured delocalized CEP stemming from the time jitter was corrected numerically. The effect of intensity-phase coupling was determined by calculating the linear-circular correlation between the measured delocalized CEP value and the maxima of a mid-IR photodetector signal. The determination of the dispersion parameters with an iterative algorithm, the evaluation and comparison of the measured CEP values, and the implementation of the FFT algorithm were all performed in MATLAB environment.

II. The dual-band SRI method I have developed is based on the SRI and the SH-SRI (second harmonic assisted spectrally resolved interferometry) methods. To realize the dual-band SRI method, both Mach-Zehnder and Michelson type interferometers were used. The spectrograms were recorded by an optical spectrum analyzer. The separation of different frequency interferences and the evaluation of the phase was achieved by the FFT numerical method. The numerical simulation of the method and the evaluation software were developed in MATLAB environment.

III. For the temporal compression of the pulse of the MIR laser, I used the thin-plate post-compression method and a bulk compressor. The spectral broadening was measured with a mid-IR spectrometer. The pulse length was measured with two devices, one home-made SH-FROG and a TOUCAN device. The long-term stability of the energy and the average power were measured by a mid-IR photodetector and a power meter, respectively. The long-term CEP stability was measured by the Fringeezz measuring device. The Strehl ratio was calculated form the near-field distribution measured by a wavefront sensor. The thermal changes on the thin plates were monitored by a thermal camera.

V. Publications

Publications related to the thesis

[T1] M. Kurucz, S. Toth, R. Flender, L. Haizer, B. Kiss, B. Persielle, E. Cormier, "Single-shot CEP drift measurement at arbitrary repetition rate based on dispersive Fourier transform", Optics Express 27(9), 13387-13399 (2019) DOI: 10.1364/OE.27.013387

[T2] M. Kurucz, R. Flender, T. Grosz, A. Borzsonyi, B. Kiss, "Simultaneous spectral phase shift characterization in two frequency bands", Optics Communication 500 127332 (2021). DOI: https://doi.org/10.1016/j.optcom.2021.127332

[T3] M. Kurucz, R. Flender, L. Haizer, R. S. Nagymihaly, W. Cho, K. T. Kim, S. Toth, E. Cormier, B. Kiss, "2.3-cycle mid-infrared pulses from hybrid thin-plate post-compression at

7 W average power”, Optics Communication 472 126035 (2020) DOI: 10.1016/j.optcom.2020.126035

Further scientific publications

[F1] S. Tóth, R. Flender, B. Kiss, M. Kurucz, A. Andrianov, L. Haizer, E. Cormier, K. Osvay, “Comparative study of an ultrafast, CEP-stable dual-channel mid-IR OPCPA system”, Journal of the Optical Society of America B 36(12) 3538-3546 (2019). DOI: 10.1364/JOSAB.36.003538

[F2] R. Hollinger, D. Hoff, P. Wustelt, S. Skruszewicz, Y. Zhang, H. Kang, D. Würzler, T. Jungnickel, M. Dumergue, A. Nayak, R. Flender, L. Haizer, M. Kurucz, B. Kiss, S. Kühn, E. Cormier, C. Spielmann, G. G. Paulus, P. Tzallas, M. Kübel, “Carrier-envelope-phase measurement of few-cycle mid-infrared laser pulses using High Harmonic Generation in ZnO”, Optics Express 28(5) 7314-7322 (2020). DOI: 10.1364/OE.383484

[F3] Y. Deng, Z. Zeng, P. Komm, Y. Zheng, W. Helml, X. Xie, Z. Filus, M. Dumergue, R. Flender, M. Kurucz, L. Haizer, B. Kiss, S. Kahaly, R. Li, G. Marcus, “Laser-induced inner-shell excitations through direct electron re-collision versus indirect collision”, Optics Express 28(16) 23251-23265 (2020). DOI: 10.1364/OE.395927

[F4] C.T. Holló, K. Miháltz, M. Kurucz, A. Csorba, K. Kránitz, I. Kovács, Z.Z. Nagy, and G. Erdei. “Objective quantification and spatial mapping of cataract with a Shack-Hartmann wavefront sensor”, Scientific Reports 10(1), 1-10 (2020). DOI: 10.1038/s41598-020-69321-3

[F5] C. Medina, D. Schomas, N. Rendler, M. Debatin, L. Ben Ltaief, Z. Filus, B. Farkas, R. Flender, L. Haizer, B. Kiss,

M. Kurucz, B. Major, S. Toth, F. Stienkemeier, R. Moshammer, T. Pfeifer, S. R. Krishnan, A. Heidenreich, and M. Mudrich "Single-shot electron imaging of dopant-induced nanoplasmas", New Journal of Physics 23(5), 053011 (2021). DOI: 10.1088/1367-2630/abf7f9

[F6] F. Haniel, H. Schroeder, S. Kahaly, A. Nayak, M. Dumergue, S. Mondal, Z. Filus, R. Flender, M. Kurucz, L. Haizer, B. Kiss, D. Charalambidis, M. F Kling, P. Tzallas, and B. Bergues "Saturating Multiple Ionization in Intense Mid-Infrared Laser Fields", New Journal of Physics. 23 (2021) DOI: 10.1088/1367-2630/abf583

[F7] R. Flender, M. Kurucz, T. Grosz, A. Borzsonyi, U. Gimzevskis, A. Samalius, D. Hoff, and B. Kiss, "Dispersive mirror characterization and application for mid-infrared post-compression", Journal of Optics 23(6), 065501 (2021). DOI: 10.1088/2040-8986/abf88e

[F8] M. Kübel, P. Wustelt, Y. Zhang, S. Skruszewicz, D. Hoff, D. Würzler, H. Kang, D. Zille, D. Adolph, G. G. Paulus, A. M. Sayler, M. Dumergue, A. Nayak, R. Flender, L. Haizer, M. Kurucz, B. Kiss, S. Kühn, B. Fetić, and D. B. Milošević, "High-Order Phase-Dependent Asymmetry in the Above-Threshold Ionization Plateau", Phys. Rev. Lett. 126(11), 113201 (2021). DOI: 10.1103/PhysRevLett.126.113201

Conference presentations

(O) - oral presentation, (P) - poster presentation, Bold* - Presenting author.

[O1] **M. Kurucz***, Sz. Toth, R. Flender, L. Haizer, B. Kiss, B. Perseille, E. Cormier, "Dispersive Fourier transform based single-shot CEP drift measurement at arbitrary repetition rate", CLEO/Europe 2019, Munich, Germany, 23–27 June 2019. DOI: 10.1109/CLEOE-EQEC.2019.8871557

[O2] **Sz. Tóth***, R. S. Nagymihály, A. Andrianov, B. Kiss, R. Flender, M. Kurucz, L. Haizer, E. Cormier, K. Osvay, “Conceptual study of a 1 kHz 10 mJ-class mid-IR OPCPA system with thermal aspects”, UFO, Bol, Croatia, 6–11 October 2019. URL: https://ultrafastoptics2019.engin.umich.edu/schedule/full-program/

[O3] **K. Osvay***, A. Borzsonyi, H. Cao, V. Chvykov, E. Cormier, R. Flender, P. Jojart, M. Kalashnikov, B. Kiss, M. Kurucz, N. Khodakovskiy, R. Lopez-Martens, R. S. Nagymihaly, V. Pajer, S. Toth, “Few cycle, phase controlled laser developments for ELI-ALPS”, CLEO 2020, San Jose, California, USA, 11–15 May 2020. URL: www.cleoconference.org/

[O4] **A. Borzsonyi***, E. Cormier, R. Lopez-Martens, M. Kalashnikov, B. Kiss, P. Jojart, J. Csontos, S. Toth, N. Khodakovskiy, R. Nagymihaly, R. Flender, M. Kurucz, I. Seres, Z. Varallyay, K. Varju, G. Szabo, “Operation experiences and further developments of the few-cycle, high average power lasers of ELI-ALPS”, ASSL, OSA Virtual Event, 13–16 October 2020. URL: https://www.osapublishing.org/conference.cfm?meetingid=1&yr=2020

[O5] **M. Kurucz***, R. Flender, L. Haizer, R. S. Nagymihaly, E. Cormier, B. Kiss, “Sub-two-cycle pulses in the mid-IR based on thin plate compression at high average power”, HILAS, OSA Virtual Event, 16–20 November 2020. URL: https://www.osapublishing.org/conference.cfm?meetingid=119&yr=2020

[O6] **M. Kurucz***, S. Tóth, J. Csontos, B. Kiss, E. Cormier, "Every single-shot CEP drift detection for near-infrared lasers with modified TOUCAN method", HILAS, OSA Virtual Event, 16–20 November 2020. URL: https://www.osapublishing.org/conference.cfm?meetingid=119&yr=2020

[O7] **M. Kurucz***, R. Flender, T. Grosz, A. Borzsonyi, U. Gimzevskis, A. Samalius, D. Hoff, and B. Kiss "High resolution mid-IR spectrally resolved interferometry", SPIE Virtual Event, 17–23 April 2021. URL: https://spie.org/conferences-and-exhibitions/optics-and-optoelectronics/programme

[O8] **M. Kurucz***, S. Toth, J. Csontos, B. Kiss, and E. Cormier. "Every single-shot CEP drift detection for near-infrared lasers with a modified TOUCAN method", CLEO/Europe-EQEC, 21–25 June 2021. URL: https://www.cleoeurope.org/wp-content/uploads/2021/06/cleo_2021_advance_programme.pdf

[P1] **M. Kurucz***, Á. Börzsönyi, M. Kovács, R. Nagymihály, K. Osvay, "Az SZTE TeWaTi femtoszekundumos lézerrendszer vivő-burkoló fáziscsúszásának mérése és stabilizálása," Magyar Fizikus Vándorgyűlés, Szeged, Hungary, 24–27 August 2016. URL: http://titan.physx.u-szeged.hu/fizikus_vandorgyules_2016/node/6#poszterek

[P2] **M. Kurucz***, Sz. Tóth, R. Flender, L. Haizer, B. Kiss, B. Perseille, E. Cormier, "High-accuracy single-shot CEP noise measurement at arbitrary repetition rate", UFO, Bol, Croatia, 6–11 October 2019. URL: https://ultrafastoptics2019.engin.umich.edu/schedule/full-program/

[P3] **M. Kurucz***, R. Flender, L. Haizer, B. Kiss, R. S. Nagymihaly, Sz. Tóth, E. Cormier, "Two-cycle pulses in the mid-IR based on hybrid thin plate compression at high average power”, ICEL, Prague, Czech Republic, 21–25 October 2019. URL: https://indico.eli-beams.eu/event/334/page/142-welcome

[P4] **M. Kurucz***, Sz. Tóth, R. Flender, L. Haizer, B. Kiss, B. Perseille, E. Cormier, “High-accuracy single-shot CEP noise measurement at arbitrary repetition rate”, ICEL, Prague, Czech Republic, 21–25 October 2019. URL: https://indico.eli-beams.eu/event/334/page/142-welcome

[P5] **M. Kurucz***, R. Flender, T. Grosz, A. Borzsonyi, U. Gimzevskis, A. Samalius, D. Hoff, and B. Kiss, “High resolution spectrally resolved interferometry in the mid-IR”, CLEO/Europe-EQEC, 21–25 June 2021. URL: https://www.cleoeurope.org/wp-content/uploads/2021/06/cleo_2021_advance_proramme.pdf