SUMMARY OF THE PHD THESIS

Analysis and applications of spectrally and spatially resolved interferometry

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Szeged, 2012

I. Introduction

Experiments with intense ultrashort pulses are one of the most intriguing and exciting fields of science nowadays. Countless applications can benefit from its research starting from biology, medical sciences to particle structure and nuclear fusion. Laser systems, which are capable of generating such pulses, are coming into use on a more and more widespread scale; moreover, their development is escalating towards the highest intensities and shortest pulse durations. The impact of this evolution can be felt closely even at the *University of Szeged*, where the *Extreme Light Infrastructure* (*ELI*) has been decided to construct recently. The aim of *ELI-ALPS (Attosecond Light Pulse Source)* is to produce few-cycle, phase-stabilized pulses having multi-terawatt (10^{12} W) peak power in order to generate attosecond (10^{-18} s) light signals, which are necessary to observe femtosecond (10^{-15} s) and faster phenomena. The generation and maintenance of their parameters during propagation of such pulses are significantly challenging and require precise diagnostic tools. One of the most powerful methods for this purpose is called *Spectrally and Spatially Resolved Interferometry* (*SSRI*), which is capable of measure the relative spectral phase of broadband light sources with exceptional accuracy.

The main purpose of present thesis is to reveal the advantages and limitations of the *SSRI* technique through several model calculations and experimental studies, which were carried out in the laser laboratories of the *Department of Optics and Quantum Electronics* of the *University of Szeged* (Szeged, Hungary), the *Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy* (Berlin, Germany) and the *Laboratory of Applied Optics* of the *École Polytechnique* (Palaiseau, France).

II. Preliminaries and objectives

The temporal shape of ultrashort pulses can be strongly affected during propagation. Fourier theorem declares that the shorter the pulse, the broader its spectrum is. Spectral components possess different velocity of propagation in the vast majority of transparent media, thus ones with shorter wavelength get behind, while longer wavelength components get ahead of the core of the pulse. This separation process increases the pulse duration and diminishes the peak intensity. Several diagnostic methods have been developed to monitor spatiotemporal parameters. Some of them are self-reference techniques (e.g. autocorrelation, *FROG*, *SPIDER*) in order to determine absolute values and therefore based on nonlinear processes, like second harmonic generation, which require intense beams and make the precision sensitive to alignment. On the contrary, linear techniques (e.g. *Spectrally Resolved Interferometry – SRI*, *Spectrally and Spatially Resolved Interferometry – SSRI*) are easy to assemble and use experimentally, moreover better accuracy can be achieved. These make *SRI* and *SSRI* powerful tools to measure relative spectral phase shift. After collinear *SRI* has been studied exhaustively already, it has become timely to investigate *SSRI* in details.

1. The first goal of my thesis was to thoroughly examine the *SSRI* method, particularly the influence of different parameters on the accuracy of measurement. I devised model calculations to determine the optimal parameters of the light source and the elements of the experimental set-up in order to minimize the standard deviation of measured *Group Delay Dispersion* (*GDD*) and *Third Order Dispersion* (*TOD*) results. Furthermore, I studied the effect of several error sources on the best achievable precision.

Spatiotemporal diagnostics are vital parts of *Chirped Pulse Amplification* (*CPA*) laser systems, since amplified pulses can be used effectively only when they are close to the Fourier transform limit. After leaving the compressor, pulses often travel a considerable amount of distance to the experimental target, and if they are propagating in atmospheric air, its linear and nonlinear dispersion effects should be taken into account. In several cases of more advanced laser systems, pulse compressors and beam lines until the target are built in vacuum chambers in order to avoid undesired phase defects. Still, the pressure dependent dispersion, more precisely the *Specific Group Delay* (*SGD*), the *Specific GDD* (*SGDD*) and the *Specific TOD* (*STOD*) of residual gas mixtures in vacuum systems should be considered during design of these beam lines.

2.a My aim was to measure the *SGDD* and *STOD* of air as a function of pressure within the range from 0.1 mbar to 1 bar by the means of *SSRI*. I laid down calculations to estimate the pressure limits of vacuum systems at given propagation lengths, where the elongation of pulse duration stays within 2% and 20%.

2.b I elaborated measurements to determine the specific dispersion coefficients of several gas mixtures at different concentrations. I plotted to analyze the correlation between my experimental results and the expected values from Lorentz-Lorenz theorem.

2.c I purposed to compute new coefficients of Sellmeier-type refractive index formulas of inert gases and nitrogen based on their measured *SGD, SGDD* and *STOD* values.

The intensity of laser pulses produced by *CPA* systems can easily reach over the threshold where nonlinear processes become significant. It could result self-focusing and self-phase modulation, which often lead damages on the surface of crystals and mirrors. On the other side, exciting phenomena can be generated, like high-harmonics, attosecond pulses, white light and extremely stable filaments. Most of these experiments are preformed in inert gas media, because of their special electron configuration. Several measurements on the nonlinear refractive index of atmospheric and inert gases have been published, yet there is no comprehensive data available on the pressure dependence of n_2 of inert gases.

3. My next objective was to develop and carry out *SSRI*-based experiments to reveal the pressure dependence of nonlinear refractive index of gases. With my experimental realization, I intended to measure the nonlinear indices of air, argon, neon, nitrogen and xenon between 0.1 mbar and 1 bar, and compare my results obtained at 1 bar with values available in literature sources.

Another substantial issue of the *CPA* lasers is the temporal contrast of the pulses, since parasitic pre- and post-pulses can entirely change the outcome of certain experiments. One of the most promising methods to improve the contrast is the so-called *cross-polarized wave (XPW)* generation. In this third-order nonlinear process, electromagnetic waves are generated with perpendicular polarization compared to the incident light. The conversion efficiency is proportional to the third power of temporal intensity; therefore the contrast of the pulses can be increased by several orders of magnitude. Moreover, besides the contrast enhancement, *XPW* generation has a number of useful properties, e.g. spectral broadening, temporal cleaning and easy experimental realizability. Unfortunately, the efficiency is fairly sensitive to the spectral phase, consequently, to the temporal compression of the incoming pulses; for this reason, practically transform limited pulses are required. Another problem might have arisen for experimental applications, if the *CEP* of the pulses has been altered by the *XPW* process. Theoretical predictions declare that the *CEP* should be preserved during the *XPW* generation, but experimental proof has yet to be demonstrated.

4. Based on two independent, *SSRI*-related experiments, I contrived to show that the *CEP* of *XPW* pulses depends exclusively on the *CEP* of the original pulses, but independent from the generation process itself.

The severe spatiotemporal difficulties of ever widely used *CPA* systems have made it necessary to develop methods for independent control of dispersion coefficients of ultrashort laser pulses. Among several ideas that have been suggested, the *AOPDF* devices are found to be one of the best solutions, which are based on the nonlinear interaction between light and accordingly formed acoustic waves propagating in a crystal. These sound waves are generated by an electronic signal in the radio frequency range and in this manner; it allows separate control of the dispersion coefficients between rather wide bounds. In spite of the widespread use of *AOPDF* devices, there is no comprehensive study available – besides one single conference proceeding – on the precision of the dispersion coefficients and possible spatiotemporal side effects. Since the front and rear sides of the acousto-optical crystal are designed to be non-parallel, one can get suspicious about the introduction of angular dispersion into the beam. Taken into account that after the acousto-optic diffraction, the spectral components propagate according to the extraordinary refractive index, and also that the length of the acoustic waves are depend on the chirp, the optical path length of the components can be different; and it can happen, that the angular dispersion of the diffracted pulses are related to the phase properties of the radio frequency signal.

5. My last goal was to investigate the accuracy of the manually preset material dispersion coefficients and residual angular dispersion of an *AOPDF*. I planned to compare the obtained results and their expected values based on my model calculations.

III. Methods of investigation

During my research, different methods, materials and devices were used. These are detailed in the following bullet points.

1. My evaluation algorithm for *SSRI* images was based on nonlinear function fitting by the means of the method of least squares. I compiled the code in *C++* language to ensure platform-independent utilization. This software was used throughout my doctoral studies.

The modeled and afterwards experimentally realized set-up of the *SSRI* method was based on the combination of a Mach-Zehnder interferometer and a two-dimensional imaging spectrograph. Femtosecond laser pulses at 800 nm central wavelength with at least 70 nm bandwidth were applied as light source in the model calculations and all further measurements. I used Monte-Carlo simulations to optimize the parameters of the experimental circumstances. I compared the effectiveness and accuracy of my code with the commonly used Fourier-based algorithm.

2.a The dispersion properties of air was measured in two steps. First, I determined the *SGDD* and the *STOD* of air at ambient pressure by using *SSRI* method. I set the difference of the arm lengths of the Mach-Zehnder interferometer exactly to integer times of the distance of subsequent pulses. In the second step, I inserted a gas tube into the sample arm, filled the tube with air and varied the pressure from 1 bar to 0.01 mbar. Since I could obtain only relative values from this latter measurement, I applied the results of the first experiment as a reference.

2.b With the same experimental set-up as in the second step of 2.a, I filled the tube with gas mixtures with known concentration. For the demonstration, I chose gases with low (He and Ne) or high dispersion (Xe) to mix with moderately dispersive molecular nitrogen.

2.c I constructed a regression algorithm to calculate Sellmeier coefficients with the use of refractive index values and phase derivatives. I applied this method to the combined data of recently determined dispersion properties of inert gases and nitrogen and refractive indices from several independent publications found in the literature.

3. In order to measure nonlinear refractive index of gases, I established experimentally a Mach-Zehnder interferometer with the beams of both arms propagated in the same gas chamber, but their intensity was split to a ratio of 10:1 by a partial reflector. I created an algorithm to extract the nonlinear indices at certain pressures from the difference in spectral phase due to the unevenly intense beams of the interferometer arms.

4. I executed two separated experiments to prove the preservation of *CEP* during *XPW* generation. The arms of the Mach-Zehnder interferometer were created by a polarization splitter: the sample arm contained the *XPW* pulses, while the fundamental beam propagated in the reference arm. In the first experiment, I recorded interference patterns at different orders of magnitude of exposure times to analyze the *CEP* stability through the visibility of the fringes. In the second one, I controlled the *CEP* of the *XPW* pulses in a known manner with the use of glass wedges and detected the *CEP* detunement interferometrically.

5. I performed full characterization of the material and angular dispersion properties of beams from different stages of a *CPA* laser system when propagating through an *AOPDF*

device. Material dispersion was measured with regular *SSRI* method. An inverted Mach-Zehnder interferometer was used to determine the phase front angular dispersion, and propagation direction angular dispersion was also quantified by focusing the beam on the slit of a two-dimensional imaging spectrograph. I applied a mechanical beam rotator to explore the angular dispersion along both horizontal and vertical axes.

IV. New scientific results

The summary of my scientific results are listed as the followings:

1. I have developed an evaluation method for extracting the spectral phase surface from two-dimensional *SSRI* images. I have determined the effect of the most frequent error sources on the accuracy of phase derivatives by the means of Monte-Carlo simulations [T1]. The noise of the *CCD* camera has been found to be the most critical. I have analyzed the effects of the computational roundings, visibility changes of interference fringes, bandwidth of the light source, inaccurate wavelength calibration, *CEP*-drift, Gaussian phase fronts and typical optical noises (e.g. diffraction). I have obtained that the achievable accuracy is 0.1 fs^2 in *GDD* and 3 fs^3 in *TOD*.

2.a I have measured the *SGD*, *SGDD* and *STOD* of air as a function of pressure from 0.1 mbar to 1 bar with the use of *SSRI* method. I have completed calculations to optimize the pressure of vacuum tubes of large laser systems in order to keep the elongation of the pulse duration under 2% or 20% [T2].

2.b I have applied the *SSRI* technique to determine the pressure dependence of the specific phase dispersion coefficients of gas mixtures between 0.1 mbar and 1 bar [T3]. Different concentrations of helium, neon and xenon in nitrogen have been investigated. My results showed exceptional correspondence with the expected values from the Lorentz-Lorenz theorem.

2.c I have laid down new coefficients of Sellmeier-type equations for the refractive index of helium, neon, krypton, xenon and nitrogen based on recently measured phase dispersion properties and earlier published refractive index values by independent authors [T3].

3. I have measured the pressure dependence of nonlinear refractive index of air, argon, neon, nitrogen and xenon between 0.05 mbar and 1 bar by the means of *SSRI* method and *CPA*-generated high intensity pulses [T4]. My findings were in great agreement with the expectations based on the literature.

4. I have proved the preservation of *CEP* of the *XPW* pulses during their generation by two *SSRI*-related, independent experiments [T5]. In the first one, I showed the independence of the visibility from the exposure time. In the second experiment, I controlled the *CEP* of the *XPW* pulses with a glass wedge. I have found that the *CEP* of the *XPW* pulses is defined unambiguously by the *CEP* of the original pulses and the position of the wedges.

5. I have characterized the accuracy of an *AOPDF* device in material dispersion and its residual angular dispersion using *SSRI* [T6]. I have obtained exceptional agreement within the measurement precision between the measured and preset *GDD* and *TOD* through several orders of magnitude. I have examined the residual angular dispersion of the *AOPDF* with two separated methods in both horizontal and vertical axes. I have found from both experiments that the angular dispersion depends on the operating temperature of the crystal; however, it is practically negligible in most experimental applications of *AOPDF* devices.

V. Publications

Peer reviewed journal publications related to the theses:

[T1] Á. Börzsönyi, A.P.Kovács, M.Görbe, K.Osvay, *"Advances and limitations of phase dispersion measurement by spectrally and spatially resolved interferometry,"* Optics Communications **281** (2008) 3051-3061.

[T2] K. Osvay, Á. Börzsönyi, A.P. Kovács, M. Görbe, G. Kurdi, M.P. Kalashnikov, "*Dispersion of femtosecond laser pulses in beam pipelines from ambient pressure down to 0.1 mbar,"* Applied Physics B **87** (2007) 457-461.

[T3] Á. Börzsönyi, Zs. Heiner, M.P. Kalashnikov, A.P.Kovács, K.Osvay, "*Dispersion measurement of inert gases and gas mixtures at 800 nm,"* Applied Optics, **47** (2008) 4856-4863.

[T4] Á. Börzsönyi, Z. Heiner, A.P.Kovács, M.P. Kalashnikov, K.Osvay, *"Measurement of pressure dependent nonlinear refractive index of inert gases,"* Optics Express, **18** (2010) 25847-25854.

[T5] K. Osvay, L. Canova, C. Durfee, A. P. Kovács, Á. Börzsönyi, O. Albert, R. Lopez Martens, *"Preservation of the carrier envelope phase during cross-polarized wave generation,"* Optics Express **17** (2009) 22358-22365.

[T6] K. Osvay, M. Mero, Á. Börzsönyi, A.P. Kovács, M.P. Kalashnikov, *"Spectral phase shift and residual angular dispersion of an acousto-optic programmable dispersive filter,"* Applied Physics B (2012) DOI 10.1007/s00340-011-4867-7, in press.

Other publication in a peer-reviewed journal:

[1] P. Jójárt, Á. Börzsönyi, B. Borchers, G. Steinmeyer, K. Osvay, *"Agile linear interferometric method for carrier-envelope phase drift,"* Optics Letters **17** (2012), 836-838.

Patent application:

[2] Á. Börzsönyi, L. Mangin-Thro, K. Osvay, *"Eljárás és berendezés fénnyaláb szögdiszperziójának mérésére,"* Hungarian Patent Application P1100626 (2011.11.14.).

Conference proceedings:

[3] Á. Börzsönyi, K.Osvay, A.P.Kovács, M.Görbe, R.Balogh, M.P.Kalashnikov, *"Measurement of pressure dependent dispersion of femtosecond pulses in air down to 0.01 mbar,"* Ultrafast Phenomena 2006, Pacific Grove, California, USA, paper MH21.

[4] Á. Börzsönyi, K.Osvay, A.P.Kovács, Zs.Heiner, M.P.Kalashnikov, "*Measurement of pressure dependent dispersion of inert gases,"* ECAMP 2007, Th2-27.

[5] Á. Börzsönyi, K.Osvay, A.P.Kovács, Zs.Heiner, M.P.Kalashnikov, "*Pressure dependent dispersion of inert gases at 800 nm,"* CLEO Europe 2007, paper CF-15-MON.

[6] K. Osvay, Á. Börzsönyi, Z. Heiner, A.P. Kovács, M. Kalashnikov, "*Pressure dependent nonlinear refractive index of inert gases,"* XXX ECLIM, 2008, Darmstadt, Germany, WE-0401 (oral).

[7] K. Osvay, Á. Börzsönyi, Z. Heiner, A.P. Kovács, M. Kalashnikov, "*Measurement of Pressure Dependent Nonlinear Refractive Index of Inert Gases,"* CLEO 2009, Baltimore, USA, paper: CMU7 (oral).

[8] K. Osvay, L. Canova, C. Durfee, A.P. Kovács, Á. Börzsönyi, O. Albert, R. Lopez Martens, "*Preservation of the Carrier Envelope Phase in Generation of Cross Polarized Wave,"* CLEO 2009, Baltimore, USA, paper: JTuD39.

[9] K. Osvay, L. Canova, C. Durfee, A.P. Kovács, Á. Börzsönyi, O. Albert, R. Lopez Martens, "*Preservation of CEP upon Generation of XPW Pulses,"* CLEO Europe 2009, Munich, Germany, paper: CG1.5 MON (oral).

[10] Á. Börzsönyi, A.P. Kovács, M. P. Kalashnikov, M. Merő, K. Osvay, *"Measurement of the spectral phase shift and the residual angular dispersion of an AOPDF,"* First International Conference Light at Extreme Intensities LEI' 09, Brasov, Romania, AIP Conference Proceedings 1228 (2010) 138.

[11] K. Osvay, Á. Börzsönyi, A.P. Kovács, M. P. Kalashnikov, M. Merő, "*Spectral phase shift and residual angular dispersion of an acousto-optic programmable dispersive filter,*" Ultrafast Optics 2009 (UFO VII), Arcachon, France.

[12] Á. Börzsönyi, Z. Heiner, M.P. Kalashnikov, A.P. Kovács, K. Osvay, "*Interferometric Measurement of Nonlinear Refractive Index of Inert Gases at Various Pressures,*" Nonlinear Photonics 2010, Karlsruhe, Germany. Paper: 10-C-868-NP.

[13] Á. Börzsönyi, M. Merő, A. P. Kovács, M.P. Kalashnikov, K. Osvay, "*Spectral phase shift and residual angular dispersion of an acousto-optic programmable dispersive filter,*" XXXI ECLIM 2010, Budapest, Hungary, paper: P21.

[14] Á. Börzsönyi, M. Görbe, P. Jójárt, M. Kovács, K. Osvay, "*Independent control of arbitrary high order dispersion of high intensity laser pulses,*" ICUIL 2010, Watkins Glen, USA.

[15] Á. Börzsönyi, P. Jójárt, M. Kovács, M. Görbe, K. Osvay "*Independent Control of Arbitrary Dispersion Order of High Intensity Laser Pulses,*" High-Intensity Lasers and High-Field Phenomena (HILAS 2011), Istanbul, Turkey, Pres. number: HWC9.

[16] P. Jójárt, Á. Börzsönyi, S. Koke, M. Görbe, K. Osvay, "*A simple linear optical measurement of carrier envelope phase shift,*" High-Intensity Lasers and High-Field Phenomena (HILAS 2011), Istanbul, Turkey, Pres. number: HThD5 (oral).

[17] P. Jójárt, Á. Börzsönyi, S. Koke, M. Görbe, K. Osvay, *"A simple linear technique for measurement the carrier envelope offset phase of ultrashort pulses,"*, CLEO 2011, Baltimore, USA, Pres. number: CWI6 (oral).

[18] P. Jójárt, Á.Börzsönyi, B.Borchers, G. Steinmeyer, K. Osvay, "*General Linear Method for Carrier-Envelope Offset Phase Measurements,*" Ultrafast Optics 2011, Monterey, CA, USA, paper Mo12 (oral).

[19] L. Nagy, Á. Börzsönyi, K. Osvay, K. Nagy, Gy. Varo, P. Maroti, M. Terazima, "*Conformation changes after primary charge separation in photosynthetic reaction centers,*" EBSA European Biophysics Congress, Budapest, Hungary, paper P-529.

[20] M. Kovács, Á. Börzsönyi, P. Jójárt, K. Osvay, "*Independent control of arbitrary dispersion order of high power ultrashort pulses,*" Light at Extreme Intensities 2011, Szeged, Hungary

[21] L. Mangin-Thro, Á. Börzsönyi, K. Osvay, "*Real time two-dimensional measurement of angular dispersion of broadband laser pulses,*" Light at Extreme Intensities 2011, Szeged, Hungary