SUMMARY OF THESIS SUBMITTED FOR A DEGREE OF DOCTOR OF PHILOSOPHY

Modeling of Femtosecond Optical Setups

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I. MOTIVATION AND SCOPE OF THE THESIS

Ultrafast laser technology and spectroscopy applies the pulses of femtosecond light sources for the investigation of material. Due to the short pulse duration, direct investigation of processes that proceed on a very fast timescale is possible. However, short pulses can suffer serious temporal distortions while propagating through an optical system because of the material dispersion or angular dispersion of the optical components. The most important aspect of femtosecond optical system design is to devise arrangements that deliver a short pulse to the sample without significant broadening of the original pulse length.

The process of designing requires effective methods that reliably model the pulse propagation. Numerical results provided by ray-tracing algorithms give accurate information on the spatial properties of the imaging optical systems within the limits of geometrical optics. In case of systems implementing measurements at high temporal resolution, the relative difference between the arrival times of the spectral components should also be taken into consideration. Several papers described the propagation and distortion of femtosecond pulses in optical systems using wave optical theory. These simulations based on a wave optical description of the problem have confirmed earlier results obtained from geometrical optics. However, wave optical models are difficult to be used in practice owing to the complicated numerical calculations they require. Furthermore, several simplifications and approximations must be made to retain a simple model, which can be used in the assessment of practical applications. Thus, attempts at developing methods for reliably modeling femtosecond beam delivery using ray-tracing calculations remain attractive for their simplicity and speed.

One of the objectives of my thesis work was to model and optimize a setup, which is used for the excitation of dynamic gratings by ultrashort light pulses. The calculations concerned impulsive stimulated Raman scattering experiments with phonon-polaritons in GaP.

Formerly it had not been analyzed, which is the most advantageous pumping arrangement for the excitation of transient gratings. Concerning this problem, I have performed a comparative analysis of three earlier proposed setups operating on the same principle, but using different imaging systems.

Another objective was the modeling of optical systems used for the generation and detection of tunable terahertz pulses, in order to determine which is the most suitable for achieving the necessary angular dispersion in the nonlinear material.

The investigation of systems having angular dispersion has drawn my attention to a different problem. In case the optical system has angular dispersion, the commonly used method to determine group delay of the spectral components of a pulse from their geometrical optical path length is only correct for aberration free imaging. I intended to develop a technique that would be also applicable for systems having imaging errors.

II. METHODS OF INVESTIGATION

For the modeling of the optical setups ray-tracing calculations were performed using Beam4 optical design program by Stellar Software. In case of the pump-probe arrangements I defined the surfaces of the optical components, then traced rays form the object space to the image space, thus determining the positions of spots of the pump and probe beams in the sample. For the investigations of the temporal properties, I have determined the optical path length of the different spectral components and calculated the group delay. This way I obtained information about the distortion of the pulse front upon leaving the optical system. I characterized the distortions based on ray-tracing analysis, by determining the difference of the group delays of the short- and long-wavelength components (usually the wavelengths where the spectral intensity of the pulse reaches half its maximum value) along the cross-section of the beam in the image plane. In the course of optimization, I have dislocated the optical elements from their optimal position, thus checking systematically the sensitivity of the setups to alignment errors.

I verified the accuracy of the developed technique for the determination of the group delay by modeling optical systems having angular dispersion, and comparing the obtained simulation results with the ones obtained from the corresponding experiments.

In the course of the experiments of the velocity-matched efficient ultrashort terahertz pulse generation, I used the excitation pulses of a Ti:sapphire regenerative amplifier system. The tilt of the pulse front necessary for velocity-matching was introduced by diffraction off a grating. The pulses were imaged into the electrooptic crystal by different optical systems. For the pump-probe measurement a rapid-scan delay line and efficient signal averaging in a computer were used, while in free space the radiation was detected by a He cooled Si bolometer.

III. NEW RESEARCH RESULTS

1. *I have performed the modeling and optimization of an optical setup for the excitation of dynamic gratings based on ray-tracing calculations* [1].

The holographic grating-reflecting microscope objective setup can be used for the excitation of dynamic gratings by ultrashort light pulses, making measurements with high time resolution possible. Impulsive stimulated Raman scattering experiments demonstrated that the arrangement is capable of creating dynamic gratings with high and variable grating groove density with femtosecond time resolution. Modeling calculations have shown that the setup can be used for experimental techniques that use pulses as short as 25 fs without significant deterioration of the time resolution. The simulation results were confirmed by experimental work.

2. *Based on my modeling calculations, I have investigated and compared the ability of three optical setups to create dynamic gratings* [2].

   Optical techniques involving the generation of dynamic gratings differ in the way in which the interfering beams are created and directed onto the medium. The investigations of pumping arrangements used for pump-probe experiments revealed the limits of the applicable pulse duration, the quality of the spatial overlap of the pulses, the sensitivity of the setups for alignment errors, having a special emphasis on the ability of the setups to create dynamic grating with variable grating groove density.

   It was shown that for imaging with a demagnification ratio other than 1:1 with the telescopic system, the optimal position of the lenses corresponding to the best time resolution can only be found by ray-tracing calculations.

   It was found that a strong correlation exists between the time resolution and the astigmatism of the setup consisting of a microscope objective.

3. *I have developed a method to determine the group delay of the spectral components of a pulse from their geometrical optical length in case of setups having angular dispersion and imaging errors. This technique, which is based on geometrical optics, is a reliable and effective tool for the investigation of the spatial and temporal characteristics of femtosecond optical systems. I have analyzed optical arrangements used for the generation and detection of terahertz pulses to demonstrate the indispensability of the new technique* [3].

   The group delay of the spectral components of a pulse can be determined from their optical path length. The commonly used formula to determine the group delay is only valid for aberration free optical systems. In case the system has imaging errors, the optical paths lengths obtained from ray-tracing calculations should be corrected. It was shown that the negligence of the correction causes significant errors for certain systems having high angular dispersion, such as the pumping arrangement applying ultrashort pulses with titled pulse front.

4. *On the basis of my calculations, I have analyzed optical systems used for the generation and detection of tunable terahertz pulses, thus contributing to the experimental demonstration of the capability of the tilted-pulse-front technique to generate plane waves with tunable frequency in the terahertz range* [4].

   Phonon-polaritons were generated impulsively with a pumping arrangement using pulses with tilted pulse front. The tilt of the pulse front was introduced by diffraction off a reflective grating; however, there are different ways of imaging the grating onto the nonlinear material. Various imaging systems comprised of different optical elements for different nonlinear crystals were modeled and optimized. On the basis of these calculations the experimental systems were built and velocity-matched efficient ultrashort THz pulse generation was achieved in GaP, $LiNbO_3$ and $LiTaO_3$ crystals.

IV. POSSIBLE UTILIZATION OF THE RESULTS

As the majority of my results are related to the modeling and optimization of femtosecond optical setups, primarily they can be made use of by their implementation in the experimental work. Thus the 1st and 4th thesis points have been utilized, since optical systems have been built based on my calculations, and effective experimental work has been conduced for the examination of phonon-polariton dynamics, and terahertz pulse generation.

My research results summarized in the 3rd and the 4th thesis point, which is the extension of the method of determining the group delay of the spectral components of a pulse from their geometrical optical path lengths for systems having angular dispersion, could be used for the analysis of any optical systems that contain optical components with angular dispersion. This embraces a significant group of the femtosecond systems (any setup containing gratings, prisms).

The results of the 2nd thesis point give an unambiguous answer for the lower limits of the applicable pulse duration of setups used for the generation of dynamic gratings, and evaluate the confines of the applicability of each setup. These results can be employed for researches that apply diffractive optical approach to the solution of nonlinear optical measurements with high time resolution.

The research results summarized in the thesis points were mostly done at the *Department of Experimental Physics* of *University of Pécs*, while the experimental results were achieved at the *Max-Panck-Institut für Festkörperforschung*.

V. PUBLICATIONS OF THE RESULTS OF THE Ph.D. WORK

1. J. Hebling, **I. Z. Kozma**, J. Kuhl:
   A compact high-aperture optical setup for excitation of dynamic gratings by ultrashort light pulses
   J. Opt. Soc. Am. B **17**, 1803-1805 (2000)

2. **I. Z. Kozma**, J. Hebling:
   Comparative analysis of optical setups for excitation of dynamic gratings by ultrashort light pulses
   Opt. Commun. **199**, 407-415 (2001)

3. **I. Z. Kozma**, G. Almási, J. Hebling:
   Geometrical optical modeling of femtosecond setups having angular dispersion
   Appl. Phys. B (2002) accepted for publication

4. J. Hebling, G. Almási, **I. Z. Kozma**, J. Kuhl:
   Velocity matching by pulse front tilting for large-area THz-pulse generation
   Opt. Express **10**, 1161-1166 (2002)

Important conference presentations of the results of the Ph.D. work

5. G. Almási, J. Hebling, **I. Z. Kozma**, J. Kuhl:
   Generation and modeling of tunable THz radiation by femtosecond tilted-pulse-front excitation
   19th Congress ICO, Optics for the Quality of Life, Florence, Italy, August 25-30, 2002, **261**

6. J. Hebling, G. Almási, **I. Z. Kozma**, J. Kuhl:
   Generation of tunable THz radiation by femtosecond tilted-pulse-front excitation
   QELS 2002, Long Beach, California, USA, May 19-24, 2002, **QTuD1**

7. J. Hebling, G. Almási, **I. Z. Kozma**, J. Kuhl:
   Generation of tunable THz radiation by femtosecond tilted-pulse-front excitation
   13th International Conference on Ultrafast Phenomena, Vancouver, Canada, May 12–17, 2002, **ME3**

8. **I. Z. Kozma**, J. Hebling, J. Kuhl:
   Comparative analysis of optical setups for excitation of dynamic gratings by ultrashort light pulses
   Diffractive Optics 2001, Budapest, October 9-11, 2001, **P23**
   EOS Topical Meeting Digest Series **30**, 108-109 (2001)

9. J. Hebling, **I. Z. Kozma**, J. Kuhl:
Diffraction grating-reflecting microscope objective set-up for excitation of dynamic grating by ultrashort light pulses
CLEO 2000, San Francisco, California, USA, May 7-12, 2000, **CMK3**

Further publications

10. G. Tomassetti, A. Ritucci, S. V. Kukhlevsky, J. Kaiser, L. Palladino, L. Reale, F. Flora, L. Mezi, **I. Z. Kozma**, T. Limongi, O. Samek, M. Liska.:
Intense XUV emission generated by a capillary discharge based apparatus
Czech J. Phys. **52**, 405-416 (2002)

11. S. V. Kukhlevsky, A. Ritucci, **I. Z. Kozma**, J. Kaiser, G. Tomassetti, A. Reale, O. Samek:
Atomic model calculations of gain in the 46.9 nm transition of Ne-like Ar
Contr. Plasma Phys. **42**, 109-118 (2002)

12. S.V. Kukhlevsky, J. Kaiser, A. Ritucci, G. Tomassetti, A. Reale, L. Palladino, **I. Z. Kozma**, F. Flora, L. Mezi, O. Samek, M. Liska:
Study of plasma evolution in argon-filled capillary Z-pinch devoted to x-ray production
Plasma Source Sci. Technology **10**, 567-572 (2001)

13. S. V. Kukhlevsky, F. Flora, A. Marinai, G. Nyitray, **Z. Kozma**, A. Ritucci, L. Palladino, A. Reale, G. Tomassetti:
Wave-optics treatment of x-rays passing through tapered capillaries
Selected Research Papers on Kumakhov Optics and Application of 1998-2000, M.A. Kumakhov, Ed., SPIE-Russia-Chapter, Vol. **4155**, 61-71 (2000)

14. S. V. Kukhlevsky, F. Flora, A. Marinai, G. Nyitray, **Z. Kozma**, A. Ritucci, L. Palladino, A. Reale, G. Tomassetti:
Wave-optics treatment of x-rays passing through tapered capillary guides
X-Ray Spectr. 29, 354-359 (2000)

15. S. V. Kukhlevsky, J. Kaiser, G. Tomassetti, A. Ritucci, A. Reale, L. Palladino, F. Flora, L. Mezi, **I. Z. Kozma**:
Effect of transverse electrical fields on X-ray amplification in a capillary-discharge Z-pinch
Soft X-Ray Lasers and Applications IV, Ernst E. Fill, Jorge J. Rocca, eds.
Proceedings of SPIE Vol. **4505**, 47-53 (2001)

16. S. V. Kukhlevsky, **I. Z. Kozma**, F. Flora, A. Marinai, L. Palladino, A. Reale, G. Tomassetti, A. Ritucci:
Diffraction of x-rays in capilary optics
Proceeedings of SPIE Vol. **3766**, p. 418-426 (1999)