Thesises of the Ph.D dissertation

INVESTIGATIONS IN FEMTOSECOND NONLINEAR OPTICS

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SUMMARY

Ultrashort laser pulse generation, characterization and investigation of processes induced by ultrashort laser pulses are a few of the main topics of today's laser physics. This field of science that produces the shortest time resolution and the highest power density is important not only for physics but also for other fields such as chemistry and biology.

Regarding recent works aiming for obtaining shorter and shorter optical pulses, mirror dispersion compensated solid state lasers offering sub-10 fs pulse durations reached their limits in the second half of the past decade. The 5 fs time limit could be overcome only by more complicated laser systems: the shortest pulse durations can be reached only by generating a broader spectrum in the optical frequency range than that of the shortest pulse Ti:Sapphire lasers with full width at half maximum (FWHM) bandwidths of 80..130 nm around 800 nm. There are two main approaches how to reach the sub-5 fs regime; both of them are based on nonlinear optical processes. In the optical fiber pulse compressor, the required broad spectrum is generated by a third-order nonlinear process called shelf phase modulation (SPM). The second approach, the broadband noncollinearly phase matched optical parametric amplifier (OPA) uses parametric processes. The disadvantage of the above mentioned methods is that they require amplified laser pulses with 30-50 nJ pulse energies for efficient generation of these broad spectra.

There is a possibility to reach the sub-5 fs regime based on unamplified pulses of a Ti:Sapphire laser with pulse energies below 10 nJ: femtosecond optical parametric oscillators (OPO) based on broadband noncollinearly phase matched parametric processes and broadband dispersion compensation by means of chirped mirrors. The basic requirement for building such a system is the high efficiency second harmonic generation (SHG) of the Ti:Sapphire laser pulses; these SHG pulses are used as (synchronous) pump pulses in the broadband phase matched parametric process. Efficient second order frequency conversion based on low pulse energy (1..10 nJ) femtosecond oscillators requires focusing in order to obtain high enough intensities. There are a few models described in the literature that discuss SHG, but none of them are applicable in the case of conversion of strongly focused femtosecond pulses.

In order to reach the above mentioned goals, modelling second-order nonlinear processes turned to be important. In contrast to earlier models described in the literature that are based on computer simulation, I could give an analytical solution of a more general problem that involves focused femtosecond pulses with arbitrary intensity and spectral distribution in the conversion process. The model is based on Fourier-decomposition of the interacting beams. As an advantage of the analytic evaluation of the novel model, it is possibile to derive general statements about the conversion process: exact conditions for optimal conversion efficiency were found and the evolution of the second-harmonic and the fundamental beam in the optimally phase matched case can be given. The model shows an improved beam and spectral quality of the SHG beam relative to that of the fundamental beam. (This observation has been previously reported in the literature only for the quasi-monochromatic and plane wave case respectively.) On the basis of the model, conditions were given under which the above mentioned improved spatial and spectral quality still appeared in the case of focused femtosecond optical pulses exhibiting arbitrary intensity and spectral distribution. An additional important result of the model was that the quality of the fundamental beam was not considerably affected by the conversion process. A few experimental results were presented verifying the main results of the model.

For enhancement of SHG efficiency of short femtosecond laser pulses, a dispersion compensated ring cavity was built. A 400% increase in the conversion efficiency relative to the single pass arrangement was demonstrated with sub-100 fs pulses, however the phase stability of the setup was found to be too poor for practical applications.

Regarding broadband OPO-s, optimal conditions for broadband non-collinear phase matching were theoretically investigated in case of a BBO crystal being pumped by SHG of a 100 fs Ti:Sapphire laser oscillator, which was operated around its gain maximum. It was shown that at the pump wavelength of 430 nm, it is also possible to obtain such a broadband (measured in $\Delta\omega$) phase matching than in the case of the typical pump wavelength of 400 nm. This difference in the pump wavelength could result in lower dispersion and hence easier dispersion compensation in an OPO cavity. In connection with this topic, I also measured the dispersion of chirped mirrors for this wavelength regime by means of spectrally resolved white light interferometry, and optimized a chirped mirror pair compressor in the 550-700 nm signal wavelength range for a BBO based OPA system that was built at the Tokyo University. Using this chirped mirror compressor, 4.7 fs pulses were obtained in the visible for the first time.

Since 1999, there is a new possibility to generate a broad optical spectrum by use of unamplified laser pulses: the development of microstructured optical fibers was a revolutionary step in white light continuum (WLC) generation, but phase properties of the generated WLC have not been reported. The first step in generation of ultrashort pulses with WLC, however, is the characterisation of the spectral phase (chirp) of the WLC. Based on this, compression of the WLC could be possible by the appropriate dispersion compensating elements.

Basic linear and nonlinear optical properties of microstructure optical fiber (MF) samples were investigated. Among others, I found a weak second harmonic signal around 530 nm during our studies. I also experimentally proved optical birefrigence of the MF by investigating polarization properties of the generated WLC.

During experimental studies on the spectral intensity distribution and phase properties of the generated WLC as function of pulse energy and fiber length, temporal pulse splitting near to the zero dispersion wavelength was found. (This observation was predicted by the theoretical model of Seres.) This feature of the pulse evolution limits the minimum pulse duration that can be reached by our current MF samples.

A chirp characterization setup (based on the setup proposed by Baltuska et al) was adapted for our unamplified Ti:Sapphire laser and MF samples: by sum-frequency generation in a relatively thick BBO crystal, chirp of the generated WLC was measured for conditions found to be optimum for pulse compression (the fiber length is a few centimeters, laser operation wavelength is 750 nm). Based on this result, an optical fiber pulse compressor was constructed. The compressed pulses were characterized by a home built second-order interferometric autocorrelator. Two photon absorption in a green LED provided our signal in the autocorrelator. After computer evaluation of the measured interference fringes, a 6-fold pulse shortening was demostrated relative to the 150 fs pulse duration of the input pulse with a central wavelength of 750 nm and pulse energy of 1 nJ.

The microstructure optical fiber can be well suited for femtosecond time-resolved spectroscopy as a broadband source of the synchronous probe beam for transient absorption measurements. Based on my previous results in connection with the spatial quality of the fundamental beam after the SHG conversion process, a cost efficient pump-probe optical setup was built for ultra-broadband femtosecond transient absorption measurements. In this setup, the WLC probe beam is generated by the unconverted fundamental beam after the BBO crystal producing the SHG pump pulses.

We also performed ablation experiments on Au films using the second harmonic pulses and on diamond-like carbon (DLC) films with fundamental pulses of our 100 fs Ti:Sapphire laser source. In these experiments, holes with sub-micrometer diameter were produced, although the actual focal spots (2 microns x 4 microns) were far from the diffraction limit. We found that ablation properties of the metallic and the DLC samples were considerably different. Although both of the samples could be ablated on the sub-micrometer scale, the metal film suffered surface deformation on the micrometer scale. On the contrary, the damaged area was one order of magnitude less in the case of the covalently bounded DLC samples. On the basis of our experiments we can say that DLC films could be important in microfabrication applications.

NEW SCIENTIFIC RESULTS

1. I have given a model based on Fourier-optics, for describing second-order nonlinear processes. With the help of the model, I could give the optimal phase matching conditions in the case of SHG in Type-I phasematched uniaxial crystall placed in the Raileigh-range, using femtosecond pulses with nearly arbitrary spectral and intensity distribution [14].

2. I have given the time evolution of the Fourier-components of the fundamental and the SHG beam near the conditions, described in point 1., in the case of small conversion efficiency and also in the case of optimal phase matching with high conversion efficiency. As a conclusion of the model, at optimal phase matching conditions the intensity and spectral properties of the SHG pulse are improoved related to those of the fundamental beam, which remain unchanged [13][14].

3. Verifying the theoretical conclusion regarding the spatial intensity distribution of the fundamental described in point 2., I have shown experimentally, that the beam-quality of the fundamental beam after the conversion will not be worse than it was before. As a consequence of this the fundamental beam after the conversion process is suitable for coupling into a microstructure optical fiber. As an application I planned and built a cost efficient pump-probe optical setup based on pulses of a Ti:sapphire laser oscillator [13].

4. I have investigated theoretically the possibilities of broadband non-collinear phase matching in case of a BBO crystal being pumped by SHG pulses of a 100 fs Ti:Sapphire laser oscillator, which was operated around its gain maximum. It was shown that at the pump wavelength of 430 nm it is also possible to obtain such a broadband (measured in $\Delta\omega$) phase matching than in the case of the

typical pump wavelength of 400 nm. In connection with this topic I also measured the dispersion of chirped mirrors for this wavelength regime by means of spectrally resolved white light interferometry and optimized a chirped mirror pair compressor in the 550-700 nm signal wavelength range for a BBO based OPA system that was built at the Tokyo University. Using this chirped mirror compressor, 4.7 fs pulses were obtained in the visible for the first time [2].

5. For enhancement of SHG efficiency of short femtosecond laser pulses, I planned and built a dispersion compensated ring cavity. A 400% increase in the conversion efficiency relative to the single pass arrangement was demonstrated with sub-100 fs pulses, however the phase stability of the setup was found to be too poor for practical applications [6][8].

6. I have performed real sub-micrometer ablation on diamond-like carbon sample with 700 ns pulsetrains of a modelocked Ti:sapphire laser at the wavelength of 800 nm [4][5][11].

7. During experimental studies on the spectral intensity distribution and phase properties of the generated WLC, I found temporal pulse splitting near the zero dispersion wavelength as a function of pulse energy and fiber length. (This observation was predicted by the theoretical model of Seres.) This feature of the pulse evolution limits the minimum pulse duration that can be reached by our current MF samples. I also experimentally proved optical birefrigence of the MF by investigating polarization properties of the generated WLC.
I adapted a chirp characterization setup (based on the setup proposed by Baltuska et al) for our unamplified Ti:Sapphire laser and MF samples. Based on this result, I built an optical fiber pulse compressor. I characterized the compressed pulses by a home built second-order interferometric autocorrelator. Two photon absorption in a green LED provided our signal in the autocorrelator. I demostrated a 6-fold pulse shortening relative to the 150 fs pulse duration of the input pulse with a central wavelength of 750 nm and a pulse energy of 1 nJ [1][3][7][9][10][12].

PUBLICATIONS

Papers:

1. P. Apai, S. Lakó, R. Szipőcs, M.B. Danailov: Broad-band photorefractive phase conjugation in a dispersive scheme. Laser Physics, Vol. 10, No. 2, pp. 444-448 (2000).

2. R. Szipőcs, A. Kőházi-Kis, S. Lakó, P. Apai, A. P. Kovács, G. DeBell, L. Mott, A. W. Louderback, A.V. Tikhonravov, M. K. Trubetskov: Negative Dispersion Mirrors for Dispersion Control in Femtosecond Lasers: Chirped Dielectric Mirrors and Multi-cavity Gires-Tournois Interferometers. Appl. Phys. B70, pp. S51-S57 (2000).

3. S. Lakó, J. Seres, P. Apai, R. S. Windeler, R. Szipőcs: Pulse compression of nJ pulses in the visible using microstructure optical fiber and dispersion compensation. (elküldve az Appl. Phys. B-hez)

4. S. Lakó, P. Apai, I. Pócsik, E. Wintner, R. Szipőcs: Sub-Micrometer Pulsetrain Machining of Diamond-Like Amorphous Carbon Films with Femtosecond Pulses of a Ti:sapphire Laser Oscillator (elküldve a J. of Optics A-hoz)

Book issue:

5. S. Lakó, P. Apai, I. Pócsik, E. Wintner, R. Szipőcs: Sub-Micrometer Pulsetrain Machining of Diamond-Like Amorphous Carbon Films with Femtosecond Pulses of a Ti:sapphire Laser Oscillator, in *Ultrafast Phenomena XIII,* in press (Springer), (2002).

Oral and poster presentations:

6. S. Lakó, P. Apai, R. Szipőcs: High Efficiency Second Harmonic Generation of femtosecond Laser Pulses in Dispersion Compensated Ring Cavity, CLEO-Europe '00 Conference, Nice, France, Paper CThE 0013 (2000).

7. R. Szipőcs, S. Lakó, P. Apai, J. Seres: 1O fs-os lézerimpulzusok elállítása kompakt 100 fs-os lézeroszcillátor, optikai szál és diszperzió kompenzálás segítségével. Kvantumelektronika 2000 konferencia

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10. R. Szipőcs, S. Lakó, P. Apai, J. Seres, R. S. Windeler: 150 fs to 25 fs pulse compression of 1 nJ pulses in the visible using microstructure optical fiber for efficient self phase modulation and a prism pair/chirped mirror compressor. In the proceedings of Ultrafast Processes in Spectroscopy, Firenze, Italy, October 28 – November 1, pp. 44 (2001).

11. S. Lakó, P. Apai, I. Pócsik, R. Szipőcs: Sub-Micrometer Pulsetrain Machining of Metallic and Diamond Like Amorphus Carbon Films with Femtosecond Pulses: a Comparison. Ibid. pp. 86 (2001).

12. R. Szipőcs, S. Lakó, P. Apai, J. Seres, R. S. Windeler: 150 fs to 15 fs pulse compression in microstructure optical fibers at 1 nJ pulse energy levels. Winter college on ultrafast nonlinear optics, 18 February- 1 March 2002, Trieste, Italy, Paper SMR.1397-9 (2002)

13. Á. Bányász, S. Lakó, P. Apai, K. Szőcs, R. Szipőcs: Cost efficient pump-probe setup for ultra-broadban femtosecond transient absorption measurements. (Accepted for Laser Physics 2002 conference, July 1-5, Pozsony, SK)

14. S. Lakó: Analytical modelling of second harmonic generation of focused femtosecond laser pulses with arbitrary spectral and spatial distribution (Accepted for Laser Physics 2002 conference, July 1-5, Pozsony, SK)