SUMMARY OF THE PhD THESIS

PHASE MODULATION OF LASER PULSES IN TWO-PASS SECOND-HARMONIC GENERATION AND BY PROPAGATION IN A RESONANT MEDIUM

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I. PRELIMINARIES AND GOALS

The development of the first lasers also triggered the development of pulsed lasers. Since then, the research on and with pulsed lasers has been growing continuously, and it is hard to overestimate its impact on other fields of sciences. Due to the development of mode-locked solid-state lasers in the last decade, it is now routine to generate pulses as short as few femtoseconds. Another important direction of laser research is the development of widely tunable cw lasers.

Femtosecond and widely tunable lasers radiate mainly in the long-wavelength range of the visible spectrum and in the near infrared. However, for an ever increasing variety of applications, the availability of shorter-wavelength ranges is essential. Frequency conversion in nonlinear optical processes is the common method for accessing other spectral ranges; one of the most often used is second harmonic generation (SHG) in nonlinear crystals. The most important practical criterion for short pulse SHG is the preservation of pulse duration in the conversion process. A neccesary condition for this is a sufficiently broad SHG bandwidth, which includes the entire pulse spectrum. The frequency doubling bandwidth — being crucial for the SHG of tunable lasers, too — is inversely proportional to the crystal length, while the conversion efficiency is proportional to its square. Hence, the bandwidth requirement limits the crystal length, and in turn, drastically lowers the conversion efficiency

One solution is to use intracavity or external resonant-cavity techniques, where the fundamental peak power inside the resonator is enhanced typically by a factor of ~10 as compared to the extracavity output. The disadvantage of these methods is that the conversion efficiency is significantly reduced by changing the resonator length by a fraction of the wavelength. In case of monochromatic lasers, the two-pass arrangement can increase the conversion efficiency without the use of a resonator. The double-pass configuration has the advantage that no interferometric accuracy is required for the adjustment; the optical elements can be displaced by several millimeters without seriously degrading the conversion efficiency. However, the simple two-pass arrangement can not be used for broadband or short-pulse SHG, since it reduces significantly the doubling bandwidth of the crystal.

Another solution is to use dispersion compensation for increasing the efficiency of broadband SHG. By compensating for the frequency dependent phase mismatch between the fundamental and its second harmonic (SH), the effective doubling bandwidth of the crystal can be increased considerably. Hence, in case of ultrashort pulses, the whole pulse spectrum can be frequency doubled with a longer crystal, with increased efficiency. However, in addition to achromatic phase matching, in a dispersion-compensated scheme care has to be taken to the preservation of the fundamental pulse duration, too.

One goal of the present work is the investigation of a new dispersion-compensated SHG scheme, capable of frequency doubling femtosecond pulses as well as widely tunable monochromatic laser sources with an increased efficiency. The proposed setup is based on the double-pass configuration improved by a proper dispersive element.

One of the most fundamental processes with femtosecond pulses is their resonant interaction with atoms. Short pulses, propagating in a resonant medium, can develope an amplitude and phase structure of considerable complexity. The pulse shaping depends on many parameters including atomic density and population, input pulse structure and intensity, and has been thoroughly investigated both theoretically and experimentally in the past. Over many years, the experimental efforts concentrated on spectral and pulse intensity-correlation measurements. Although these investigations were carried out with high precision, and good agreement between theory and experiment was found, they allow no direct access to the electric field of the pulses. For a more direct comparison between theory and experiment, a full (i.e. both amplitude and phase) characterization of the electric field of the pulses is desired. Due to the tremendous progress during the last decade in ultrashort-pulse diagnostic techniques, it is now routine to completely characterize the time dependence of the electric field of a short pulse using, for example, frequency resolved optical gating (FROG). Although full pulse characterization is essential in understanding complex pulse shaping phenomena, these methods have found application in studying resonant pulse propagation only recently.

The other goal of the present work is the experimental study of resonant laser-atom interaction by measuring both the amplitude and the phase of femtosecond pulses interacting with a resonant atomic medium, using the FROG technique. In addition to the pulses after resonant interaction, the incoming pulses are included in the electric field measurements as well, enabling a detailed study of the influence of initial phase modulation on the resonant reshaping of ultrashort laser pulses.

II. METHODS OF INVESTIGATION

The formalism introduced by Martinez was used to calculate the phase shift of the prism pair, which considerably simplified the calculations. The second harmonic field was calculated in the fixed-field approximation. A computer program was written to carry out the numerical calculations.

As the laser source for the resonant pulse propagation experiments at low intensity, a Vitesse type femtosecond oscillator (Coherent Inc.) was used, delivering 96 fs pulses with 803 nm central wavelength and 32 nm spectral width. In some measurements, the pulses were compressed by a prism compressor to 38 fs, having only third-order (and small higher-order) spectral phase. Nearly transform-limited, sufficiently attenuated amplified pulses from a tunable Mira type femtosecond oscillator (Coherent Inc.) were used as well, with 24 nm spectral width at 793 nm. High-intensity pulses were generated by amplifying the oscillator pulses with an Odin type multipass Ti:sapphire amplifier (Quantronix Corp.), pumped by an intracavity-frequency-doubled Nd:YLF laser at 527 nm (Quantronix Corp.). For an easy variation of the pulse intensity over four orders of magnitude, an arrangement with reflective attenuators and three possible cell positions was built.

For the FROG measurements, a background-free SH intensity-autocorrelator was built, using a 100 μm thin BBO crystal. The data acquisition with the FROG apparatus was fully computer-controlled, including the readout of the spectra and the control of the autocorrelator delay by driving an Owis step motor. A Bestec POC4M type spectrometer was used for dispersing the autocorrelator signal. A FROG retrieval computer program from Femtosoft Technologies was used to reconstruct the electric field of the laser pulses from the measured FROG traces.

In the regime of linear laser-atom interaction classical dispersion theory (linear electron oscillator model) was applied, while in the nonlinear regime the Maxwell-Bloch equations (semiclassical model) were numerically solved.

III. RESULTS

1. The two-pass SHG scheme, with a dispersive element inserted between the nonlinear crystal and the mirror, has been investigated with respect to broadband SHG [1,4]. It has been shown that in case of perfect dispersion-compensation — when the relative phase shift of the dispersive element compensates for that of the nonlinear crystal over its entire SHG bandwidth — the bandwidth of the two-pass arrangement is twice as broad as that of the simple (i.e. with no compensating element) two-pass arrangement, and equals to the one-pass bandwidth of the crystal. The SHG efficiency of the dispersion-compensated two-pass scheme is four times higher than the one-pass efficiency.

2. With numerical calculations it has been shown that the two-pass SHG arrangement, with a prism pair inserted as the dispersive element, can provide four times higher SHG efficiency than that of the one-pass scheme, while preserving the one-pass bandwidth. Thus, the two-pass scheme with the inserted prism pair can be used for broadband frequency doubling of tunable monochromatic laser sources with an increased efficiency [3].

According to the calculations, in the two-pass arrangement, up to ~70 nm one-pass bandwidth, first-order compensation of the relative phase shift ΔkL of the nonlinear crystal can be achieved with the inserted prism pair. An explanation of this effect has been given, based on the angular dispersion of the prism pair [3]. The prism-pair compensated two-pass bandwidth reaches at least 90% of that of the one-pass below ~30 nm one-pass SHG bandwidth. The increasing bandwidth-narrowing in the two-pass scheme above ~30 nm one-pass bandwidth has been shown to be the result of the second-order relative phase shift of the prism pair, which fails to match the compensation condition.

3. With numerical calculations it has been shown that the dispersion-compensated two-pass arrangement, with a prism pair as the dispersive element, can be used for frequency doubling of femtosecond pulses, with four times higher efficiency than that of the one-pass scheme [1]. The limiting effects for the arrangement have been analysed with respect to pulse duration and intensity [3].

According to the calculations, the width of pulses longer than ~40 fs is essentially preserved in the doubling process. Pulses shorter than this experience an increase in SH pulse duration by more than 20%, due to the decreased SHG bandwith caused by the second-order relative dispersion of the prism pair. The increasing nonlinear phase limits the two-pass dispersion compensation to lower intensities. The dispersion-compensated two-pass scheme with a prism pair may be used for SHG of unamplified pulses from femtosecond oscillators.

4. Using the FROG technique, the temporal amplitude and phase structure of resonant femtosecond pulses interacting with Rb vapor was experimentally determined for various initial pulse structures and intensities [2]. In addition to the pulses after resonant interaction, the incoming pulses were included in the electric field measurements as well, which turned out to be essential for the study of the dependence of the resonant interaction on the initial pulse structure.

5. The dependence of the resonant interaction on the initial pulse structure has been experimentally demonstrated by measuring the electric field of both the incoming and the transmitted pulses. It has been shown that for a detailed study of the resonant reshaping of femtosecond pulses an exact knowledge of both the amplitude and the phase structure of the incoming pulses is necessary. Only the calculations starting from the measured initial electric field were able to reproduce the resonant pulse shaping with sufficient accuracy in the linear as well as in the nonlinear interaction regime.

The pulse shaping is more pronounced in the linear interaction regime. In short time intervals, extending over a few pulse durations following the main pulse, the phase modulation of the incident pulse strongly influences the reshaping process, whereas on longer time scales it is governed mainly solely by the phase shift of the resonant atomic system [2].

IV. PUBLICATIONS

Journal Papers:

1. J. A. Fülöp, A. P. Kovács and Zs. Bor:
   Dispersion-Compensated Two-Pass Arrangement for Second Harmonic Generation of Femtosecond Pulses
   Laser Physics **10** (2000) 437

2. R. Netz, T. Feurer and J. A. Fülöp:
   Influence of phase modulation on the reshaping of ultrashort laser pulses in resonant three-level systems
   Phys. Rev. A **64** (2001) 043808

3. József A. Fülöp, Attila P. Kovács and Zsolt Bor:
   Broadband dispersion-compensated two-pass second harmonic generation of femtosecond pulses
   Opt. Commun. **188** (2001) 365

Conference Papers:

4. J. A. Fülöp, A. P. Kovács and Z. Bor:
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   SPIE **3573** (1998) 59

5. J. A. Fülöp, A. P. Kovács and Z. Bor:
   Two-pass Second Harmonic Generation of Ultrashort Laser Pulses
   5th Congress on Modern Optics, 1998, Budapest, Hungary

6. J. A. Fülöp, A. P. Kovács and Z.Bor:
   Improved Two-pass Second Harmonic Generation of Femtosecond Pulses
   8th International Laser Physics Workshop, 1999, Budapest, Hungary

V. FURTHER PUBLICATIONS

7. Fülöp J. A.:
   Rövid impulzusok kétutas frekvenciakétszerezése
   XXIV. Országos Tudományos Diákköri Konferencia, 1999, Debrecen

8. R. Netz, T. Feurer and J. A. Fülöp:
   Influence of phase modulation on the reshaping of ultrashort laser pulses in resonant three-level systems
   Annual Report, 2001, Institut für Optik und Quantenelektronik, Friedrich-Schiller-Universität Jena