# VILNIUS UNIVERSITY CENTER FOR PHYSICAL SCIENCES AND TECHNOLOGIES

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# THE INFLUENCE OF GROUP DELAY DISPERSION OF THE RESONATOR COMPONENTS ON AN OPTICAL PARAMETRIC OSCILLATOR SYNCHRONOUSLY PUMPED BY FEMTOSECOND PULSES

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# VILNIAUS UNIVERSITETAS FIZINIŲ IR TECHNOLOGIJOS MOKSLŲ CENTRAS

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# REZONATORIAUS KOMPONENTŲ GRUPINIO VĖLINIMO DISPERSIJOS ĮTAKA SINCHRONIŠKAI KAUPINAMAM PARAMETRINIAM ŠVIESOS GENERATORIUI

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# Introduction

The emergence of lasers unlocked a new era in fundamental and experimental sciences. A large number of new phenomena were discovered, many of which are investigated by nonlinear optics. In particular, the understanding of principal laws governing parametric light generation and amplification has boosted laser technology even further. New spectral regions and spectral bandwidths unsustainable by any laser host material can now be reached via different parametric processes. During the past few decades, many different parametric devices have been developed. Which of these should be used depends on various parameters including: pump source characteristics, desired wavelength, spectral bandwidth and available nonlinear materials. In the case of low pump pulse energy, short pulse duration and high repetition rate synchronously pumped optical parametric oscillators (SPOPO) are used. Despite the fact that the first femtosecond SPOPO was demonstrated in 1989 [1], these devices are still continuously under development. Currently three main directions can be distinguished: output power or pulse energy scaling, broadening of tuning range and carrier – envelope phase stabilization [2]. These development directions are indistinguishable from the main areas of applications: nonlinear imaging and microscopy together with frequency comb generation and spectroscopy.

As mentioned, SPOPOs are used when pump pulse energy is as low as a few tens of nJ and the pulse repetition rate is in the order of several tens of MHz. Low pump pulse energy requires numerous passes though the nonlinear crystal, thus an optical resonator must be constructed. On the other hand, short pulse duration and the instantaneous nature of parametric amplification require precise timing. Parametric light generated by the first pump pulse must return to the nonlinear crystal at the exact moment when the next pump pulse arrives. In the femtosecond time scale, group velocity or group delay dispersion (GDD) plays a major role during pulse formation. Moreover, GDD acts as a spectral filter and strongly affects the output wavelength of the femtosecond SPOPO. A complex interplay of various processes like parametric light amplification, self-phase modulation, dispersive pulse broadening, group delay dispersion etc., makes the operation of femtosecond SPOPOs complicated and difficult to interpret from a theoretical point of view. These specific details make the construction of SPOPOs meticulously demanding.

During the past few decades many efforts have been made to develop ytterbium doped laser systems. These lasers emit radiation around 1  $\mu$ m and can generate pulses from ~ 100 fs to a few hundreds of picoseconds. The main attraction point is the versatile power scalability option as these lasers are pumped by high power laser diodes. Thus, ytterbium doped lasers overtook Ti:sapphire systems as a pump sources for SPOPOs due to their

beneficial output power and easier maintenance. Moreover, optical components, and especially optical coatings, evolved together with femtosecond laser systems. Many efforts were devoted to the development of broadband mirrors with desirable and functional dispersion characteristics [3,4]. Firstly, chirped mirrors were used for pulse compression in mode – locked Ti:sapphire laser oscillators [5] and further employed for ultra broadband pulse compression [6]. Such mirrors are essential elements for compact laser systems capable of providing ultrashort, time – bandwidth limited pulses. A significant breakthrough in these two areas was inspirational in developing and nurturing femtosecond synchronously pumped optical parametric oscillators. So the **main task** of this thesis was to investigate the power and temporal characteristics of SPOPOs pumped by the second harmonic of a Yb:KGW laser. Utmost attention was paid to the mirrors group delay dispersion influence on signal wavelength tuning and emergence of pulse to pulse instabilities.

## Novelty

- 1. To our knowledge, this is the first time that a femtosecond synchronously pumped optical parametric oscillator has been built to work in the visible and infrared spectral range while using complementary chirped mirror pairs.
- 2. The study explores how group delay dispersion affects output emission in distinct ways such as synchronous generation of multiple spectra and tuning discontinuity.
- 3. It was shown that a synchronously pumped femtosecond optical parametric oscillator arranged to work in positive dispersion exhibits improved performance regarding output power. Meanwhile, an external prim compressor allowed shorter pulse durations to be achieved than in SPOPO configuration with negative dispersion.
- 4. While investigating GDD influence on resonator performance, it was observed that the emission spectrum changes from pulse to pulse. The variation can range from periodic oscillations to chaotic movement. A method was proposed that allowed the reconstruction of the spectral evolution of the pulse train in the case of the periodic oscillations.

# **Practical Value**

The experiments carried out during the preparation of this Thesis are valuable for the construction of femtosecond synchronously pumped optical parametric oscillators tunable over a wide spectral range. Several aspects can be pointed out:

- It was shown that not only the reflection coefficient but dispersive characteristics of broadband mirrors are important for construction of widely tunable synchronously pumped optical parametric oscillators.
- It was showed that when mirrors with GDD oscillations are used, overall cavity dispersion might get close to zero, thus output wavelength tuning discontinuity can appear.
- It was determined that an SPOPO operating with relatively small positive group delay dispersion would produce higher output power in comparison to the same resonator working in a negative dispersion setup.
- Output pulses of an SPOPO operating with relatively small positive group delay dispersion can be compressed to pulse durations shorter than generated in an SPOPO working in a negative dispersion setup.

## Statements for Defense

- 1. Simultaneous up to N + 1 peaked output spectrum generation might be observed in the spectral range with sufficient gain overcoming cavity round trip losses, if group delay has N extrema in this range.
- 2. Continuous output wavelength tuning and stable operation of an SPOPO is more sensitive to monotonicity of the GDD curve than to an absolute value of GDD. Thus the design of mirrors suitable for a broadly and continuously tunable SPOPO should be chosen in a way that monotonicity of GDD would be immune to deposition errors as much as possible. It is also desirable that the frequency of oscillations and their amplitude would be minimal.
- 3. A femtosecond synchronously pumped optical parametric oscillator operating in a net positive group delay dispersion regime might provide higher output power than an SPOPO operating in a net negative group delay dispersion regime.
- 4. Pulse to pulse instabilities might occur in a synchronously pumped optical parametric oscillator if the spectral wings of the oscillating pulse are in the region of low absolute value of intracavity GDD.

## Approbation

#### Scientific articles in periodical journals with an impact factor which are included in the Web of Science database

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**Other scientific papers,** not directly related to the topic of this dissertation

- I. Pipinytė, R. Grigonis, K. Stankevičiūtė, S. Kičas, R. Drazdys, R. C. Eckardt, V. Sirutkaitis, Laser-induced-damage threshold of periodically poled lithium niobate for 1030 nm femtosecond laser pulses at 100 kHz and 75 MHz, Proc. SPIE 8786, 87861N, 2013.
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#### *Conference presentations, presented by the author*

- K. Stankevičiūtė, I. Pipinytė, J. Vengelis, A. Marcinkevičiūtė, R. Šuminas, R. Grigonis, V. Sirutkaitis, Sinchroniškai Yb:KGW lazerio pirmąja ir antrąja harmonika kaupinami parametriniai šviesos generatoriai, 40-toji Lietuvos nacionalinė fizikos konferencija (LNFK-40), Vilnius, Lietuva (birželio 10–12 d., 2013).
- K. Stankevičiūtė, I. Pipinytė, J. Vengelis, A. Marcinkevičiūtė, R. Šuminas, R. Grigonis, R. C. Eckardt, V. Sirutkaitis, Optical parametric oscillators synchronously pumped by fundamental and second harmonic radiation of femtosecond Yb:KGW laser. SPIE Optics + Photonics, San Diegas, JAV (rugpjūčio 25–29 d., 2013).
- K. Stankevičiūtė, S. Melnikas, S. Kičas, M. Vengris, M. Malinauskas, V. Sirutkaitis, Synchronously pumped femtosecond optical parametric oscillator with broadband chirped mirrors, SPIE Optics + Optoelectronics, Praha, Čekija (balandžio 13-16 d., 2015).

#### Conference presentations, presented by coauther

1. K. Stankevičiūtė, S. Kičas, **I. Pipinytė**, M. Vengris, R. Grigonis, R. Drazdys, V. Sirutkaitis, Investigation of resonators mirrors GDD influence on synchronously pumped femtosecond OPO tuning properties, Europhoton Nešatelis, Šveicarija (rugpjūčio 24–29 d., 2014).

#### Contributions

All experimental work that enabled the writing of this thesis was carried out at the Department of Quantum Electronics at Vilnius University during 2012 – 2017 under the supervision of Prof. Valdas Sirutkaitis. The thesis outlines an assembly of various task of different nature that were performed in order to achieve better scientific knowledge. The indicated effort consists of the construction of multiple SPOPO systems, experimental work, data analysis and interpretation which was performed by the author. Moreover, I am pleased to introduce some of the contributors to whom I am very grateful:

- Prof. V. Sirutkaitis supervised doctoral studies, helped to create conditions for experimental work, consulted on data presentation and its publication;
- Prof. M. Vengris consulted on experimental implementation and data analysis, contributed with publication efforts and helped to automate the experimental setup;

- S. Melnikas and Dr. S Kičas designed and manufactured complementary chirped mirror pairs, consulted on mirror dispersion characteristics;
- PhD student I. Stasevičius performed the numerical simulation of the SPOPO.

## Structure of the Thesis

The doctoral dissertation consists of four chapters:

- In Chapter 1, an introduction to nonlinear optics, review of pump sources, nonlinear crystals, group delay dispersion influence on output wavelength of the SPOPO and dispersion characteristics of chirped mirrors are presented. This chapter is not included in the summary of the doctoral dissertation.
- Chapter 2 describes the investigation of a synchronously pumped optical parametric oscillator with complementary chirped mirror pairs. Main attention is paid to the influence of intracavity group delay dispersion on the wavelength of the output radiation.
- In Chapter 3, the operation of a synchronously pumped optical parametric oscillator operating in the net positive dispersion regime is analyzed.
- In Chapter 4, the investigation of pulse to pulse instabilities in synchronously pumped optical parametric oscillator is presented. The influence of intracavity GDD and other parameters are examined.

# 1 Synchronously pumped optical parametric oscillator with complementary chirped mirror pairs

Expansion of the tuning range is one of the main development trends of femtosecond synchronously pumped optical parametric oscillators. Here, two separate solutions can be implemented: subsidiary stages for other parametric processes are enabled in order to reach higher or lower frequencies; or specially designed mirrors are used so full signal or idler wavelength tuning range could be covered. The main problem here is the lack of broadband mirrors with suitable group delay dispersion. Chirped mirrors are widely used for SPOPOs, but their reflection and optimized dispersion region is often limited to the 100 - 200 nm range. In this chapter, a synchronously pumped optical parametric oscillator with complementary chirped mirror pairs (CMPs) is presented. The main focus is paid on the influence of mirrors group delay dispersion on the signal wavelength tuning range and simultaneous generation of a multi-peaked output spectrum.

#### 1.1 Experimental setup

The experimental setup of the SPOPO based on a 3 mm long BBO crystal is shown in Figure 1.1. The SPOPO was pumped by the second harmonic of a femtosecond Yb:KGW oscillator (*Flint, Light Conversion Ltd.*) operating at a central wavelength of 1026 nm with a repetition rate of 76 MHz and providing an average output power of 4 W. Pump radiation was generated in a nonlinear lithium triborate crystal and separated from fundamental radiation with dichroic mirrors. Maximum power of the second harmonic was 1.9 W, whereas pulse duration was 109 fs. The SPOPO resonator was made out of six mirrors. Two of them were meniscus mirrors with convex side AR-coated for pump radiation at 513 nm, while the other side exhibited high reflection in the signal wavelength range from 630 to 1030 nm and high transmission at the pump wavelength. The other four mirrors were two complementary chirped mirror pairs and had high reflectivity in the signal wavelength range. Three different SPOPO cavity configurations were explored: (1) high dispersion, with 5 mm thick UV fused silica window used as an output coupler; (2) medium dispersion, where the OC was a 2 mm thick Suprasil glass plate; and (3) low dispersion, where the output coupler was removed. Mirror M6 was tilted to provide an additional reflection near 0 deg incidence and a mirror with an average reflection of 93 % was used to couple out signal radiation (see dashed line boxes in Figure 1.1). The group delay and group delay dispersion of every mirror used in the SPOPO cavity was evaluated experimentally. For this, a home-made white



Figure 1.1 Experimental setup. Flint – Yb:KGW oscillator;  $\lambda / 2$  – halfwave plate; L1 - L2 – lenses; LBO – lithium triborate nonlinear crystal; DM – dichroic mirrors; L3 – triplet lens; M1 and M2 – meniscus mirrors; BBO –  $\beta$  – barium borate crystal, M3 - M6 – complementary chirped mirror pairs; GP – glass plate; OC – output coupler.

light interferometer was used. Output pulse duration was measured with a scanning autocorrelator (*Geco, Light Conversion Ltd.*). Mirror M5 was mounted on the translation stage (*Standa Ltd.*) with a step size of 0.312  $\mu$ m. At every step of the translation stage, the output spectrum was recorded with a spectrometer (*AvaSpec-2048, Avantes*), thus giving insight into the output wavelength dependence on cavity length detuning. The cavity length detuning at which the generation stopped was attributed to the reference value of 0. Signal wavelength was determined as the center of mass of every recorded spectrum.

## 1.2 Intracavity dispersion influence on signal wavelength tuning

Two complementary chirped mirror pairs were used in the SPOPO cavity. These mirrors were specially designed so that GDD oscillations of one mirror would minimize the GDD oscillations of the other. The experimentally measured GDD is given in Figure 1.2 (a). The inset shows designed GDD curves. Target value of mirror pair GDD oscillations was  $40 \pm 10$  fs<sup>2</sup>, while the reflection coefficient was higher than 99 %. Considerable discrepancy between measured and designed GDD curves is seen in the infrared spectral range from 900 nm. As previously mentioned, three different cavity arrangements with respect to intracavity dispersion were explored. Corresponding cavity round trip GDD curves are given in Figure 1.2 (b).

Experimentally measured signal wavelength dependences on cavity



Figure 1.2 (a) Experimentally measured GDD of complementary chirped mirrors measured at 0 deg angle and GDD of chirped mirror pair measured at ~ 9 deg angle of incidence. Inset shows designed GDD curves. (b) Cavity round trip GDD for high, medium and low dispersions SPOPO cavity configurations.

detuning in the case of high and low dispersion at selected phase-matching angles are given in Figure 1.3. For clarity, comparison of three different GDD cases is done gradually from high to low dispersion regimes, and from red to infrared spectral regions. It should be noted that the wavelength tuning range is defined as a variation of the central wavelength of the signal pulse with respect to the cavity length detuning.

Continuous tuning can be sustained in the 664 to 916 nm range in the high dispersion regime and in the 623 to 910 nm range in the medium dispersion regime. Here, the idler wavelength covers the 1176 - 2905 nm spectral range. The tuning rate changes from 2.5 nm/ $\mu$ m to 3.8 nm/ $\mu$ m around 700 nm as cavity dispersion is reduced from high to medium regime. The first region of interest is in the spectral range between 735 and 762 nm as the tuning rate changes rapidly at medium dispersion setup and is as high as 26 nm/ $\mu$ m at 744 nm. Such tuning behaviour indicates that the cavity round trip GDD approaches zero. More similar regions are observed in the infrared spectral range. In some cases specific wavelengths are hard to reach and such regions get wider as cavity dispersion is reduced. Analysis of GDD curves indicates that these regions with rapid tuning rate changes are determined by GDD oscillation of certain mirrors. Tuning behaviour is particularly complicated in the low dispersion regime. Here, several tuning gaps were observed. Moreover, cavity GDD changes sign around 725, 836, 888, 922, 945 and 981 nm according to wavelength tuning rate measurements. It should be noted that peaks and troughs of GDD oscillations can shift due to the change in reflection angle. A small variation of angle in combination with a very steep decrease of the GDD curve can lead to drastic change in GDD values at a particular wavelength. Moreover, data obtained during group delay measurements was numerically manipulated in order



Figure 1.3 Relationship between the central wavelength of output spectra and cavity length detuning in (a) high and (b) low dispersion cases.

to calculate GDD and suppress noise. These two reasons can cause errors in evaluated intracavity GDD. Thus, disagreement between wavelengths, where the intracavity GDD curve intersects zero value, and positions, estimated from wavelength tuning rate measurements, is observed. In the infrared spectral region, double peaked output spectra were observed at some cavity length detuning values. Such output spectra are determined by generation while intracavity GDD is close to zero. Two different generation conditions were observed. Peaks of the output spectra moved in different directions as the cavity length was changed. Thus SPOPO generation occurs at GD extremum. Here, GDD values for these two wavelengths have the same value but a different sign. It was also noticed that peaks can move in the same direction during the change of cavity length. This indicates generation between two GD extrema. Here, GDD value and sign are the same for both wavelengths.

Output power characteristics of the SPOPO were investigated in the medium dispersion regime. Maximum output power was 500 mW at 700 nm and 300 mW at 845 nm at 13% and 15% output coupling rates respectively. This corresponds to 27% and 15% pump to signal power conversion efficiency. The generation threshold was below 1 and 1.2 W at different output coupling conditions and increased notably at the marginal phase-matching angles. Pulse duration was measured in low dispersion regime. Here, the SPOPO had a single output beam whereas reflections from the glass plate formed a four output beam. Signal pulse duration at full width half maximum level varied from 102 to 266 fs. Shortest pulses were generated when the SPOPO operated in negative intracavity dispersion regime.

## 1.3 Conclusions

A synchronously pumped optical parametric oscillator continuously tunable from 623 to 910 nm with an average output power of more than 500 mW was demonstrated. To our knowledge, this is first SPOPO with complementary chirped mirror pairs. A nonmonotonous intracavity GDD curve led to a complicated relationship between signal wavelength and cavity length detuning. Rapid tuning rate changes and simultaneous dual–wavelength generation were observed. These results suggest that smooth and continuous output wavelength tuning can be achieved in the positive dispersion regime when intracavity GDD does not exhibit large oscillations shifting intracavity GDD to zero or making it change sign.

# 2 SPOPO operation in positive dispersion regime

The common goal while constructing new SPOPO systems is usually aiming to achieve suitable performance in negative dispersion regime. In this way, a phase mismatch of different spectral components can be compensated. In contrast, a resonator with positive dispersion has a distinctive mechanism that establishes the formation of pulses with stable duration. The present process can be divided into two competing forces – dispersive broadening and spectral narrowing. The latter is caused by limited amplification bandwidth and temporal overlapping of the pump and oscillating pulses. Despite this, experimental research regarding pulse duration dynamics in the SPOPO with positive dispersion regime is very modest. It was shown that GDD oscillations can disrupt continuous tuning of output radiation. This can be solved by offsetting intracavity GDD to values further away from zero i.e. introducing additional positive dispersion. This can be achieved experimentally by adding a glass plate to the resonator cavity. The Brewster angle would allow losses to be minimized, while dispersion control could be managed with two AR coated wedge prism pairs. However, positive GDD has some downsides leading to dispersive broadening and a spectral shape which is not Gaussian. Despite this, a broadly tunable SPOPO with high output power might become advantageous for some applications even with the present downsides. Moreover, output pulses can be compressed with an external compressor which also simplifies construction of the resonator. In this way, output power can be increased as lower losses are accumulated due to fewer reflections from the mirrors and possible removal of the prim pair.

#### 2.1 Experimental setup

Experimental realization is very similar to the one described in Chapter 1.1. Foremost, the Yb:KGW oscillator which acts as pump source had its output power increased to 5 W. Maximum power of second harmonic radiation measured at the front of the focusing lens L3 was as high as 2.7 W, while pump pulse duration was 105 fs. Once SPOPO configuration was modified, signal wave reflection from complementary chirped mirror pairs increased to 28 bounces (see Figure 2.1). Furthermore, a flat mirror was fitted as one of the end mirrors inside the resonator and was fixed on a motorized translation stage with step size of  $0.312 \ \mu$ m. The particular mirror was examined with a white light interferometer and showed a GDD oscillation around -25 fs<sup>2</sup> in the spectral range between 620 and 750 nm, whereas its transmission was measured to be less than 0.3 % in this spectral range. The current set-



Figure 2.1 Experimental setup. L – triplet lens; M1 and M2 – meniscus mirrors; BBO –  $\beta$  – barium borate crystal, M3 – M6 – complementary chirped mirror pairs; M7 – flat broadband mirror, GP – glass plate; OC – output coupler.

up had an output coupling mirror with 8 % transmission. The integrating spectrometer AvaSpec-2048 was synchronized with the translation stage thus recording output spectra at each consecutive step. If no glass plate was inserted in the resonator, negative dispersion reached the lowest value of -900 fs<sup>2</sup> in the spectral range from 695 to 705 nm. The negative values were present in the whole range between 625 and 750 nm only approaching zero near 665 nm. Once different glass plates were introduced, the cavity round trip GDD could be offset by 400, 800, 1600, 2400, 3200 and 4000 fs<sup>2</sup>. For the experiments described below, the optical parametric oscillator was tuned to maximize output power in the 695 – 705 nm spectral range. Crystal orientation was fixed and no more adjustments were performed to its angle during addition of various glass plates.

#### 2.2 Experimental results

Experiments showed that central wavelength tuning becomes very sensitive around GDD regions near zero value. This characteristic is even exaggerated with the insertion of 1 or 2 mm thick glass plates. The broadest tuning range was achieved while no positive dispersion was additionally introduced. This configuration shows sharp tuning rate changes and tuning discontinuities in the 626 - 685 nm region. Introduction of a 1 mm glass plate offsets the GDD curve by 400 fs<sup>2</sup> and also reduces the tuning zone to 688 - 748 nm. Once 2 mm thick glass is inserted, the tuning curve shows a small leap in the 673 - 689 nm range. This part is of additional interest as the resonator GDD curve changes its sign. This is indicated by central wavelength movement and its reverse during monotonous ca-



Figure 2.2 (a) Output power and (b) pulse duration dependence on signal wavelength in case of various SPOPO cavity arrangements. Intracavity dispersion of the SPOPO without glass plate is given at the bottom of the pictures.

vity length adjustment. Once positive dispersion is increased even further with a 4 mm glass plate, the central wavelength returns to steady transition i.e. without any turning points. Thicker glass plates of 6, 8 or 10 mm do not change the tuning curve significantly, only showing a narrowing of the output spectrum. Their tuning ranges are: 666 - 733 nm, 675 - 731 nm and 687 - 713 nm respectively. Output spectrum dependence on cavity length detuning in the resonator without and with a 4 mm thick glass slab is shown in Figure 2.2. Reduction of pump power also has the characteristic of tuning range narrowing. If the pump power is reduced from 2.6 W to 0.9 W, the tuning zone decreases from 107 to 38 nm in the case of a resonator without a glass plate. As a general case, a positive dispersion setup with each additional dispersion increment reduces its output pulse bandwidth while the spectrum keeps the same shape. Here, the spectral form exhibits two maxima on the sides with steep slopes and a minimum in the middle, while the peak on the lower GDD side is more pronounced.

Relationship between central wavelength and output power during various glass plate insertions is depicted in Figure 2.3 (a). Meanwhile Figure 2.3 (b) shows pulse duration at different wavelengths and distinctive dispersion values. Pump power was 2.6 W while measuring data mentioned in this chapter. Firstly, one can observe that the highest output power is reached when a 4 or 6 mm SF10 glass plate is inserted into the resonator. This can be attributed to more favorable amplification conditions. When comparing spectral width, one can see that the addition of a 6 mm glass plate increases bandwidth by 1.6 times. Consistently, a 4 mm plate widens the bandwidth of the output spectrum by 2.4 times (in comparison to the negative dispersion regime). Pulse duration also increases by 2.5 times at maximum output power. In order to explain pulse duration broadening, bandwidth widening and output power increment, one has to look at processes happening inside the SPOPO. The group velocity mismatch length



Figure 2.3 (a) Output power and (b) pulse duration dependence on signal wavelength in the case of various SPOPO cavity arrangements. Intracavity dispersion of the SPOPO without a glass plate is given at the bottom of the pictures.

increases once pulse duration increases due to positive GDD. The delay introduced by cavity length detuning might improve temporal overlapping between pump and signal pulses by delaying the faster one. However, further pulse broadening starts to reduce overlapping between the pump pulse and the wings of the signal pulse, thus lowering amplification efficiency. The results manifest as spectral narrowing and pulse duration broadening. Thus, we can conclude that specific positive dispersion introduction would increase SPOPO generation efficiency.

While positive dispersion is quite low (in the cases of 4 and 6 mm glass plates) the generation threshold is similar to the negative dispersion regime. Further increment of intracavity GDD leads to higher generation thresholds. Moreover, output power emission saturates with an increase of pump power at negative dispersion. Contrary to that, a positive dispersion SPOPO shows a linear dependence between output and pump power in the investigated pump power range. As a result, a parametric oscillator with negative dispersion loses its advantage regarding output power once saturation starts to happen.

The shortest measured output pulse duration was 90 fs at setup without any additional glass plates. The average duration in the spectral range of 687 – 707 nm is 107 fs with standard deviation of 7.5 fs. To enumerate measured pulse duration values obtained with glass plates we get:  $344 \pm$ 40 fs with 4 mm SF10 slab,  $384 \pm 74$  fs with 6 mm SF10 slab,  $414 \pm 69$  fs with 8 mm SF10 slab and  $455 \pm 70$  fs with 10 mm SF10 slab. The longest duration was obtained with 10 mm SF10 glass and estimated 626 fs. One can observe that not only pulse duration but also pulse duration fluctua-



Figure 2.4 (a) Output power and (b) pulse duration dependence on pump power in the case of various SPOPO cavity arrangements.

tion magnitude increases as cavity dispersion increases. Output power and pulse duration dependence on pump power is given in Figure 2.4. When pump power is increased, the resonator with negative dispersion shows a decrease of pulse duration while the positive dispersion setup acts contrarily i.e. pulse duration increases. In both cases the dynamics are determined by spectral broadening. Despite the prevailing configuration of dispersion, when pump power increases more and more spectral components undergo amplification that exceeds ongoing losses. Self–phase modulation also acts on bandwidth broadening. Once the resonator has a negative GDD, a wider spectrum unambiguously influences generation of shorter pulses. Meanwhile in the positive dispersion configuration temporal broadening of different wavelengths is not compensated. Thus generated pulses become longer.

It is important to note that an increase of pump power also reduces the time–bandwidth product which goes from 0.44 to 0.32. Gaussian pulse shape was assumed for pulse duration measurements. However, the duration of such a time–bandwidth limited pulse should be longer than the measured one, thus pulses generated at high pump power are not Gaussian.

#### 2.3 Conclusions

The results of the previously analyzed experiments showed that an SPOPO working in positive dispersion configuration could have higher output power than the same resonator with negative dispersion. Output power increases linearly with pump power at a positive GDD arrangement, but saturates at negative dispersion configuration in the investigated pump power range. In the case of further possibility to increase pump power, an SPOPO with positive dispersion could be a preferable source regarding output power. Moreover, an SPOPO with a positive GDD generated pulses with a non–Gaussian spectral shape. These pulses were not time–bandwidth limited.

# 3 Pulse to pulse instabilities in a femtosecond optical parametric oscillator

Next to the experimental advancement of femtosecond synchronously pumped optical parametric oscillators, new numerical approaches are developed for more precise and detailed description of SPOPOs. Numerical simulation showed that small changes in SPOPO cavity length can transform SPOPO operation from steady state to periodic [7]. Energy repartition among the spectral components of the resonant pulse was observed. Dispersive Fourier technique was applied for pulse to pulse spectrum recording in the buildup stage of a picosecond SPOPO [8]. At high pump powers, pulse to pulse spectra gained a periodic pattern. It is known that a femtosecond SPOPO is highly unstable near zero cavity group delay dispersion [9, 10]. Periodic intensity modulation of the output radiation was observed experimentally in the region near zero group delay dispersion, and was attributed to soliton formation [11]. However, no detailed investigation has been performed.

## 3.1 Experimental setup

The pump radiation generation stage and the SPOPO cavity have the same arrangement as described in Chapter 2.1. However, the pump source was updated to have an average output power of 5.8 W and the maximum power of the second harmonic was as high as 3.1 W. The intracavity GDD was modified by changing the glass plates made out of SF10 glass. Two different output couplers were used with an average transmission of 3%and 8%. No other changes were introduced into the SPOPO cavity. The most important part of the experiment lineup is the detection setup shown in Figure 3.1. The SPOPO output beam passes through two glass plates. One of the reflected beams is focused on the fast photodiode in order to monitor overall amplitude variation. The main output beam is directed to the diffraction grating with 1800 lines/mm groove spacing. The diffracted beam is focused with a large aperture gold plated mirror and directed to three fast photodiodes. Every photodiode registers the amplitude variation of different spectral components of the SPOPO pulse. Spectral resolution is determined by the active area of the photodiode and spectral distance between the monitored wavelengths is restricted by the physical dimensions of the photodiodes. Another beam reflected from a glass plate is focused to the entrance slit of the Shamrock spectrometer. An intensified CCD camera iStar is placed at the output of the spectrometer.

The intensified CCD camera is used for recording a single pulse spectrum in the case of stable periodical oscillation. The oscilloscope sends a synchronization signal to the iStar camera when the signal of the pulse



Figure 3.1 Detection setup. GP1 and GP2 – UVFS glass plates, DG – diffraction grating, L1 and L2 – lenses, M – large aperture mirror, D1 – D4 – fast photodiodes, Shamrock – spectrograph, iStar – intensified CCD camera, OSC – oscilloscope.

train envelope reaches its maximum value. After the synchronization signal, voltage is applied to the multichannel plate for 13 ns, so the CCD camera is exposed to only one pulse. The digital delay generator delays amplification voltage with an additional 13.2 ns for every synchronization signal. Thus a sequence of consecutive spectra is recorded and pulse spectrum dynamics can be reconstructed. Meanwhile, spectral amplitude variation monitoring with photodiodes gives prompt insight into the spectral behaviour of the SPOPO.

#### 3.2 Experimental results

Pulse train sequences are recorded at different pump powers with a 3% output coupler and Suprasil glass plate. Photodiode readings were normalized in the range from 0 to 1. Afterwards, peaks in the data were located, giving the pulse train envelope or amplitude variation of the specific spectral component. A set of such data is given in Figure 3.2. The top row shows amplitude variation at 679 nm, the second row from the top at 695 nm, the third row from the top at 706 nm, whereas the bottom row shows overall energy variation. Based on the shape of the pulse train envelope, four different operation modes can be distinguished: steady state, single peak (Figure 3.2 (a)), double peak (Figure 3.2 (b)) and chaotic operation (Figure 3.2 (c)). Chaotic operation is observed only with a 3% output coupler. In the case of double peak oscillation, the maximums of one spectral component coincide with the minimums of the other, indicating energy repartition between different spectral components. The frequency of oscillations strongly depends on cavity length detuning at the same pump power and can change between different oscillatory regimes as the output wavelength changes. Periodicity



Figure 3.2 Amplitude of different spectral components (top row at 679 nm, second from the top 695 nm, third from the top at 706 nm) and overall energy (bottom row) variation at 1.7 W (a), 2.15 W (b) and 3.05 W (c) pump powers.

in single and double operation regimes strongly depends on pump power too. Spectral components monitoring with photodiodes revealed that oscillation modes change at different internal powers in the case of different output couplers. Such behaviour can be determined by the non-smooth transmission curve of the output coupler, which influences signal spectrum shape and strong oscillation dependence on cavity length detuning.

In order to verify the reliability of the data set obtained, we compared photodiode readings with the amplitude variation of spectral components and the overall energy recorded with the iStar camera. Good agreement was found between these two data sets. At lower pump power, oscillation with a periodicity of 17 cavity round trips (this corresponds to 225 ns) is observed. Whereas, at higher pump power, energy repartition



Figure 3.3 Sequences of SPOPO output spectra recorded with intensified CCD camera at 2.7 W (a) and 2.9 W (b) pump power.

between different spectral components is clearly seen, as the spectrum evolves between two states with clearly distinguishable peaks around 701 and 703.5 nm (see Figure 3.3).

# 3.3 Intracavity group delay dispersion influence on pulse to pulse instabilities

Firstly it should be noted that we never observed oscillating pulse spectra or chaotic operation in the SPOPO operating in the net positive dispersion regime. As the pump power increases, the output spectrum broadens. If the central wavelength of the output radiation is in the negative dispersion regime, but the wings of the spectrum fall in the region of relatively small GDD, oscillation in the output spectrum occurs. In order to understand the influence of intracavity group delay dispersion on the oscillatory behaviour of the SPOPO, the experiment described by T. D. Reid et. al. was reproduced [11]. We kept the folding cavity arrangement, but changed the chirped mirror pairs into quarter wavelength mirrors. The oscillating beam is reflected only five times from these mirrors. At the end of one arm, an 8% output coupler was introduced. Two Brewster angle cut prisms made out of SF11 glass were placed in the path of the oscillating beam in order to compensate the cavity round trip group delay dispersion. Intracavity group delay dispersion was changed gradually. Firstly, the resonator cavity length was shortened and afterwards generation was reestablished by increasing the light path inside the prisms. SPOPO output power, the spectrum amplitude variation of spectral components and, if possible, pulse duration were recorded at every  $\Delta GDD$  point. Experimental and numerical simulation data are presented in Figure 3.4. Overall intracavity dispersion was not evaluated. Experimental and numerical simulation data are presented as a function of the change of the intracavity group delay dispersion ( $\Delta$ GDD). Intracavity dispersion was equal to zero around a value of  $\Delta \text{GDD} \approx 800 \text{ fs}^2$ .

Output radiation had a Gaussian spectrum shape in negative dispersion. The spectrum broadened as intracavity GDD approached zero value. Measured pulse duration also decreased. With an increase of the GDD, the stable regime transformed into single peak oscillations. Here, autocorrelation acquired a pedestal, which became severely pronounced as the operation changed into double peak regime. Operation changed to single peak and to stable regime again, as the intracavity GDD was changed further. Output power and pulse duration increased in the positive dispersion regime, once again showing SPOPO superiority with respect to the negative dispersion regime. Here, output spectra showed a non–Gaussian shape with steep slopes and minimum in the middle. The discrepancy between experimentally obtained and numerically simulated data in the positive dispersion regime is caused by the change of the output wavelength. Cavity length de-



Figure 3.4 (a) SPOPO output power, (b) pulse duration and time bandwidth product dependence on  $\Delta$ GDD. The points depict experimental data, whereas the solid lines represent a cubic spline added to guide the eye.

tuning was not implemented in the model. Thus the central wavelength of the output radiation moved to a shorter wavelength range around 680 nm. Meanwhile, experimental conditions were changed so that the output wavelength would be around 695 nm. Oscillations were observed in the 450  $\rm fs^2$  range experimentally and in the 350  $\rm fs^2$  range numerically.

Oscillations or chaotic operation at the output spectrum were observed in the regions near zero intracavity GDD. Here, the influence of intracavity GDD on the output wavelength decreases, thus no spectral filtering is applied. A chaotic generation regime therefore naturally occurs. However, it is more complicated to understand oscillating bahaviour. First of all, it should be noted that there are two separate aspects determining output wavelength: phase matching conditions and intracavity GDD. If the influence of intracavity GDD decreases, the impact of phase matching conditions increases. Such competition between phase-matching and intracavity GDD might be the cause of the observed oscillations. Nevertheless, other processes like pump regeneration, noncollinear interaction, self–phase modulation, cross-phase modulation and spatial phenomena can also have a non-diminishing effect.

# 3.4 Pulse compression with an external prism compressor

SPOPO output power increases dramatically if the dispersion compensating prisms are removed from the SPOPO cavity. Maximum output power was 690 mW at a pump power of 3.05 W. This corresponds to 22% pump to signal power conversion efficiency. Meanwhile, maximum output power generated in the SPOPO with dispersion compensating prisms was 328 mW

in the negative dispersion regime and 372 mW in the positive dispersion regime. Such a difference in output power is determined by two factors. Firstly, the prisms increase losses in the SPOPO cavity. Secondly, a longer signal pulse leads to longer interaction length in the nonlinear crystal. Non-compressed pulse duration was 352 fs. The external prism compressor was constructed out of two prisms with peak to peak separation of 19.5 cm. The signal beam passes twice through the prisms. Here, beam power decreases by approximately 10% from 640 mW to 576 mW. Output radiation exhibits a specific double peak spectrum with a trough in the center as shown in Figure 3.5 (a). Assuming Gaussian pulse shape measured pulse duration was 55 fs, while the corresponding autocorrelation function is given in Figure 3.5 (b).



Figure 3.5 (a) Output spectrum generated in the SPOPO cavity without dispersion compensating prisms and (b) measured autocorrelation function.

#### 3.5 Conclusions

Experimental investigation of pulse to pulse instabilities in a femtosecond synchronously pumped optical parametric oscillator showed that several operation types can be attributed to SPOPOs: stable, single peak, double peak and chaotic operation. The relationship between pump power, intracavity dispersion and pulse spectrum suggested that pulse to pulse instabilities emerge due to broadening of the pulse spectrum. In the case of broadband pulse spectrum and oscillating intracavity GDD, the wings of the spectrum fall in the low GDD value zone. Thus evoking pulse to pulse instabilities, despite the fact that the GDD value is far from zero for the central wavelength. Monitoring of several components of the pulse spectrum with a set of fast photodiodes gives prompt insight into the nature of SPOPO oscillations. A new method was proposed for reconstruction of pulse spectrum evolution in the case of periodic oscillations. An intensified CCD camera was exposed to a single pulse while its synchronization was based on oscillation patterns.

## Main results and conclusions

- 1. An SPOPO pumped by the second harmonic of a femtosecond Yb:KGW oscillator was demonstrated. Complementary chirped mirror pairs were used for construction of the SPOPO resonator. A nonlinear 3 mm long BBO crystal was used for parametric interaction. Continuous tuning of the output wavelength in the 625 910 nm range was demonstrated via rotation of the nonlinear crystal and cavity length detuning. Maximum output power was as high as 500 mW at 13% output coupling and 1.9 W pump power.
- 2. It was experimentally shown that the wavelength tuning rate increases and tuning accuracy decreases while changing cavity length detuning if the intracavity group delay dispersion approaches the point of zero GDD. The tuning rate changes from 3.8 nm/ $\mu$ m at 700 nm to 26 nm/ $\mu$ m if intracavity GDD decreases from 420 to 225 fs<sup>2</sup>. Gaps in the tuning range appear if the sign of the intracavity GDD changes.
- 3. If the intracavity GDD changes its sign several times in the region of sustainable amplification bandwidth, simultaneous generation of several wavelengths can occur. During the change of GDD sign, intracavity group delay is at the extremum. Therefore, group delay will be equal for two different wavelengths. In the case of such intracavity GDD, a continuous tuning of the cavity length will lead to simultaneous generation of a double peaked spectrum. As the cavity length changes, the existing peaks move in different directions. It was experimentally shown that these peaks can move in the same direction as SPOPO generation occurs near two group delay dispersion has the same sign for both wavelengths. The experimentally observed tuning behaviour was consistent with the evaluated intracavity GDD curve.
- 4. The SPOPO pumped by the second harmonic of the femtosecond Yb:KGW oscillator was optimized for generation in the 695–705 nm spectral region. An increment in output power from 282 to 328 mW was observed as intracavity group delay dispersion was increased from the -900 to 700–1500 fs<sup>2</sup> range and a 3 mm long BBO crystal was used for parametric interaction. Output power decreased as intracavity GDD was increased further to 2100 fs<sup>2</sup>. The increment in output power can be explained by improved interaction conditions. As the pulse duration gets longer in the net positive dispersion regime, a group velocity mismatch length also increases. This leads to a longer nonlinear interaction length in the nonlinear crystal.
- 5. If the central wavelength of the output radiation is in the negative intracavity GDD regime, but the wings of the spectrum fall into re-

gions with relatively small intracavity GDD, the stable operation of the SPOPO can be disturbed. Four different types of operation were observed experimentally. Regimes range from stable, to single peak, double peak and finally to chaotic operation. To reach each consecutive regime, pump power should be increased, which also broadens the output spectrum.

- 6. A new technique suitable for reconstructing the pulse spectrum evolution in synchronously pumped optical parametric oscillators was proposed. This method is based on the periodic behaviour of instabilities. The intensified CCD camera is triggered by a signal generated during the monitoring of spectral components with amplitude modulation. Every time a digital delay is added, the camera is exposed to a different single pulse in the pulse train. Thus, a set of individual pulse spectra is recorded, which shows the pulse spectrum evolution in the synchronously pumped optical parametric oscillator.
- 7. The SPOPO generating 690 mW at 700 nm with  $+600 \text{ fs}^2$  net intracavity dispersion value was demonstrated. This corresponds to 22 % pump to signal power conversion efficiency. Measured pulse duration was as high as 352 fs, while spectrum width at  $1/e^2$  level was 29 nm. Despite a non-Gaussian spectrum shape, pulse compression down to 55 fs was demonstrated with an external prism compressor.

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#### Santrauka

#### REZONATORIAUS KOMPONENTŲ GRUPINIO VĖLINIMO DISPERSIJOS ĮTAKA SINCHRONIŠKAI FEMTOSEKUNDINIAIS IMPULSAIS KAUPINAMAM PARAMETRINIAM ŠVIESOS GENERATORIUI

Femtosekundiniai sinchroniškai kaupinami parametriniai šviesos generatoriai (SKPŠG) itin patrauklūs prietaisai, plačiai taikomi netiesinėje mikroskopijoje bei spektroskopijoje. Nepaisant to, šių prietaisų tobulinimas vis dar labai aktualus, o pastaruoju metu yra nusistovėjusios trys vystymo kryptys: derinimo diapazono plėtimas, išvadinės galios arba impulso energijos didinimas bei stabilios gautinės fazės impulsų generavimas.

Šiame disertaciniame darbe yra pristatomi eksperimentiniai tyrimai, analizuojantys galimybes išplėsti generuojamos spinduliuotės derinimo diapazoną naudojant plačiajuosčius čirpuotus veidrodžius, padidinti generuojamos spinduliuotės galią tinkamai parenkant rezonatoriaus grupinio vėlinimo dispersiją. Daug dėmesio skirta spektrinių nestabilumų stebimų sinchroniškai kaupinamuose parametriniuose šviesos generatoriuose bei dispersijos įtakos šių nestabilumų atsiradimui.

Darbo metu sukonstruotas SKPŠG su suderintų dispersijos osciliacijų veidrodžių poromis. Nors tokie veidrodžiai yra sėkmingai naudojami itin plataus spektro impulsų spūdai, eksperimentiškai parodyta, jog neišvengiamos tokių veidrodžių grupinio vėlinimo dispersijos osciliacijos sutrikdo tolygų generuojamos spinduliuotės bangos ilgio keitimą. Rezonatoriaus grupinio vėlinimo dispersijai, kurios pavidalui didžiausią įtaką turi veidrodžių dispersinės charakteristikos, priartėjus prie nulinės vertės yra prarandamas generuojamos spinduliuotės bangos ilgio keitimo tikslumas, atsiranda trūkiai derinimo srityje. Osciliuojančio pobūdžio rezonatoriaus grupinio vėlinimo dispersijos priklausomybė nuo bangos ilgio lemia itin sudėtingą generuojamos spinduliuotės bangos ilgio priklausomybę nuo rezonatoriaus ilgio išderinimo. Eksperimentiškai nustatyta, jog SKPŠG, veikiančio nedidelės teigiamos rezonatoriaus grupinio vėlinimo dispersijos srityje, generuojamos spinduliuotės išvadinė galia yra didesnė, nei tokios pačios konstrukcijos SKPŠG veikiančio neigiamos dispersijos srityje. Šiuos pasikeitimus lemia palankesnės stiprinimo sąlygos, nes teigiamos dispersijos srityje išauga generuojamų impulsų trukmė ir padidėja atstumas kristale, kuriame vyksta netiesinė sąveika. Srityse, kuriose rezonatoriaus grupinio vėlinimo dispersija priartėja prie nulinės vertės, stebėti nestabilumai generuojamos spinduliuotės spektre. Tokiomis sąlygomis galimas osciliuojančio bei chaotiško pobūdžio impulsų spektrų kitimas. Eksperimentiškai nustatyta, jog generuojamos spinduliuotės centriniam bangos ilgiui esant neigiamoje srityje, stabilus SKPŠG veikimas gali sutrikti, jei impulso spektro sparnai patenka i nedidelės suminės rezonatoriaus grupinio vėlinimo dispersijos sritis.

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