Doctoral Thesis

Leveraging Novel Nonlinear Devices for Compact Gigahertz Frequency Combs

Author(s):
Mayer, Aline S.

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LEVERAGING NOVEL NONLINEAR DEVICES FOR COMPACT GIGAHERTZ FREQUENCY COMBS

A thesis submitted to attain the degree of
DOCTOR OF SCIENCES of ETH ZURICH
(Dr. sc. ETH Zurich)

presented by
ALINE SOPHIE MAYER

MSc ETH Physics, ETH Zurich

born on 14.04.1990

citizen of La Chaux-de-Fonds (Neuchâtel)

accepted on the recommendation of
Prof. Dr. Ursula Keller, examiner
Prof. Dr. Derryck T. Reid, co-examiner

2018
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Symbols and Abbreviations

Symbols

- $A$: amplitude of fast absorber recovery
- $A_{abs}$: mode size on the absorber [m$^2$]
- $A_{gain}$: mode size in the gain [m$^2$]
- $c$: speed of light in free space [m·s$^{-1}$]
- $E_p$: pulse energy [J]
- $E_{crit}$: critical pulse energy [J]
- $f$: frequency [Hz]
- $f_n$: n-th optical comb mode
- $\Delta f$: frequency deviation [Hz]
- $f_{CEO}$: CEO-frequency [Hz]
- $f_{opt}$: optical frequency [Hz]
- $f_{rep}$: repetition rate [Hz]
- $F$: fluence [J·m$^{-2}$]
- $F_2$: inverse absorption coefficient [J·m$^{-2}$]
- $F_{Kerr}$: Kerr-lens focal power [m$^{-1}$]
- $F_{sat}$: saturation fluence [J·m$^{-2}$]
- $F_{sat,abs}$: absorber saturation fluence [J·m$^{-2}$]
- $F_{sat,gain}$: gain saturation fluence [J·m$^{-2}$]
- $g^{(1)}$: first-order temporal coherence
- $h$: Planck’s constant [J·s]
- $I$: intensity [W]/electrical current [A]
- $L$: loss
- $L_{dbc}(f)$: single-side band noise [dBc·Hz$^{-1}$]
- $M^2$: beam quality factor
- $n$: refractive index
- $n_2$: nonlinear index of refraction [m$^2$·W$^{-1}$]
SYMBOLS AND ABBREVIATIONS

\( N \) \quad \text{soliton order}

\( PID \) \quad \text{proportional-integral-derivative amplifier}

\( P_{pk} \) \quad \text{peak power [W]}

\( Q \) \quad \text{cavity Q-factor}

\( R \) \quad \text{reflectivity}

\( \Delta R \) \quad \text{modulation depth}

\( R_{\text{lin}} \) \quad \text{linear reflectivity}

\( R_{ns} \) \quad \text{non-saturable reflectivity}

\( \Delta R_{ns} \) \quad \text{non-saturable losses}

\( S_{\phi}(f) \) \quad \text{power spectral density of the phase noise [rad}^2\text{-Hz}^{-1}]\]

\( S_{f}(f) \) \quad \text{power spectral density of the frequency noise [Hz}^2\text{-Hz}^{-1}]\]

\( t \) \quad \text{time [s]}

\( T_R \) \quad \text{cavity round-trip time [s]}

\( V \) \quad \text{voltage [V]}

\( \chi \) \quad \text{electric susceptibility}

\( \beta_2 \) \quad \text{GVD coefficient [s}^2\text{-m}^{-1}]\]

\( \gamma \) \quad \text{nonlinear coefficient [W}\cdot\text{m}^{-1}]\]

\( \phi_{\text{CEO}} \) \quad \text{CEO-phase [rad]}

\( \lambda \) \quad \text{wavelength [m]}

\( \nu \) \quad \text{optical frequency [Hz]}

\( \nu_{\text{opt}} \) \quad \text{optical center-frequency [Hz]}

\( \sigma_{\text{abs}} \) \quad \text{cross-section for absorption [m}^2]\]

\( \sigma_{\text{em}} \) \quad \text{cross-section for emission [m}^2]\]

\( \sigma(\tau) \) \quad \text{frequency stability [Hz]}

\( \tau_{\text{FWHM}} \) \quad \text{pulse duration (full width at half maximum) [s]}

\( \tau_{\text{slow}} \) \quad \text{slow absorber recovery time [s]}

\( \tau_{\text{fast}} \) \quad \text{fast absorber recovery time [s]}

\( \tau_p \) \quad \text{pulse duration [s]}

\( \omega \) \quad \text{angular frequency [s}^{-1}]\]

\( \eta \) \quad \text{efficiency, power in coherent peak}
Abbreviations

AC  autocorrelator
AOM  acousto-optical modulator
AM  amplitude modulation
AR  antireflection
CALGO  Calcium Gadolinium Aluminate CaGdAlO$_4$
CEO  carrier-envelope offset
CW  continuous wave
DBR  distributed Bragg reflector
DFG  difference frequency generation
DPSSL  diode-pumped solid state laser
DUT  device under test
Er  Erbium
FM  frequency modulation
FFT  fast Fourier transformation
FSR  free spectral range
FWHM  full-width half-maximum
GDD  group delay dispersion
GTI  Gires-Tournois interferometer
GVD  group velocity dispersion
KLM  Kerr lens modelocking
KYW  Potassium Yttrium Tungstate KY[WO$_4$]$_2$
KGW  Potassium Gadolinium Tungstate KGd[WO$_4$]$_2$
LiSAF  Lithium Strontium Aluminium Fluoride
MIXSEL  modelocked integrated external-cavity surface emitting laser
MSA  microwave spectrum analyzer
NA  numerical aperture
Nd  Neodymium
OC  output coupler
OPA  optical parametric amplifier
OPO  optical parametric oscillators
OSA  optical spectrum analyzer
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>PBS</td>
<td>polarizing beam splitter</td>
</tr>
<tr>
<td>PCF</td>
<td>photonic crystal fiber</td>
</tr>
<tr>
<td>PLL</td>
<td>phase-locked-loop</td>
</tr>
<tr>
<td>PPLN</td>
<td>periodically poled lithium niobate</td>
</tr>
<tr>
<td>PSD</td>
<td>power spectral density</td>
</tr>
<tr>
<td>QCL</td>
<td>quantum cascade laser</td>
</tr>
<tr>
<td>QML</td>
<td>Q-switched modelocking</td>
</tr>
<tr>
<td>RBW</td>
<td>resolution bandwidth</td>
</tr>
<tr>
<td>RF</td>
<td>radio-frequency</td>
</tr>
<tr>
<td>RIN</td>
<td>relative intensity noise</td>
</tr>
<tr>
<td>ROC</td>
<td>radius of curvature</td>
</tr>
<tr>
<td>SC</td>
<td>supercontinuum</td>
</tr>
<tr>
<td>SCG</td>
<td>supercontinuum generation</td>
</tr>
<tr>
<td>SESAM</td>
<td>semiconductor saturable absorber mirror</td>
</tr>
<tr>
<td>SHG</td>
<td>second harmonic generation</td>
</tr>
<tr>
<td>SNR</td>
<td>signal-to-noise ratio</td>
</tr>
<tr>
<td>SPM</td>
<td>self-phase modulation</td>
</tr>
<tr>
<td>SSA</td>
<td>signal source analyzer</td>
</tr>
<tr>
<td>TDL</td>
<td>thin disk laser</td>
</tr>
<tr>
<td>TBP</td>
<td>time–bandwidth product</td>
</tr>
<tr>
<td>Ti</td>
<td>Titanium</td>
</tr>
<tr>
<td>VCO</td>
<td>voltage controllable oscillator</td>
</tr>
<tr>
<td>VECSEL</td>
<td>vertical external-cavity surface-emitting laser</td>
</tr>
<tr>
<td>XPM</td>
<td>cross-phase modulation</td>
</tr>
<tr>
<td>Yb</td>
<td>Ytterbium</td>
</tr>
<tr>
<td>ZDW</td>
<td>zero dispersion wavelength</td>
</tr>
</tbody>
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A major part of the work presented in this thesis has been published in the journal and conference papers listed below. The original manuscripts of the publications highlighted in bold are re-printed with permission in this cumulative thesis. The visual format of the manuscripts has been adapted to fit the one-column-style of the thesis, but the individual reference lists as well as the citation and spelling/grammar style of the different journals have not been modified.

Journal papers


Conference papers


nonlinearities" in Laser Congress 2017 (ASSL, LAC), (Optical Society of America, Nagoya, 2017), paper ATh1A.3., talk


Frequency Comb in the Mid-Infrared," in *Conference on Lasers and Electro-Optics* (Optical Society of America, San Jose, California, 2016), p. SF2O.1, talk


European Quantum Electronics Conference (Optical Society of America, Munich, 2015), p. CD_2_5, talk


Abstract

From the first demonstration of the laser to novel chip-scale devices - research on optical frequency combs has undergone a considerable development over the last decades. Thanks to the efforts of many groups around the world to understand, engineer and control this versatile type of light source, frequency combs of all kinds are being developed to meet the demands of diverse applications. New experiments involving frequency combs are continuously emerging across many disciplines of science, ranging from spectroscopic measurements in chemistry and biology to the calibration of telescope spectrographs in astronomy.

This thesis summarizes results that are part of this ongoing effort, with a focus on one particular aspect: frequency combs based on compact modelocked solid-state lasers with gigahertz pulse repetition rates.

Modelocked laser sources with such high pulse repetition rates are for instance interesting for experiments that require high sampling rates in the time domain. Due to the Fourier transform linking the time and frequency domain, the high repetition rate translates into a large separation between the evenly distributed sharp frequency lines in the spectral domain, i.e. the frequency comb. The larger the spacing between the comb lines, the easier it becomes to resolve the lines individually, which means they can be used as a “ruler” for very precise frequency measurements. Nonlinear optical processes can be used as tools to shape the frequency comb for the desired application, i.e. for instance to convert the comb light to a different frequency or broaden its spectral coverage. These nonlinear processes inherently scale with the light intensity and hence the pulse energy. For high-repetition rate pulse trains based on modelocked laser, this energy requirement represents one of the main challenges: for a fixed average power, the higher the pulse repetition rate, the lower the energy carried by each pulse. Scaling the pulse energy by using a series of amplifiers is one way to tackle the issue, but this is usually paired with adding complexity and noise to the system.
In this thesis, novel approaches will be presented that address the challenge from a different angle: by leveraging new types of nonlinear optical devices, we demonstrate efficient frequency conversion processes that can be achieved at low pulse energies (i.e. picojoules), hence reducing the complexity and cost of high repetition rate frequency comb systems.

The directly diode-pumped laser sources that will be discussed in this thesis are based on Ytterbium-doped crystals emitting at a center wavelength around 1 µm. They rely on semiconductor saturable absorber mirror (SESAM) to start and stabilize the pulsed operation, i.e. modelocking. In the introductory chapter of this thesis, we will have a closer look at the operating principle of this type of lasers and review the state-of-the-art performance of laser-based GHz frequency combs. Each chapter thereafter consists of results that have been published in peer-reviewed journals. The articles have been integrated into the thesis by adapting their visual format, but keeping their original content. The four papers are presented following the chronological order in which they were published.

Paper 1 presents the first demonstration of octave-spanning spectral broadening, so-called supercontinuum generation (SCG), in silicon nitride waveguides. These novel devices are transparent from the visible to the mid-infrared spectral region and enabled us to generate a coherent octave-spanning supercontinuum with only ~36 pJ of pulse energy, which is an order of magnitude less than using traditional silica fibers.

In Paper 2, we show that the supercontinuum obtained from the silicon nitride waveguides is suitable to enable an offset-stabilized comb from a 1 GHz diode-pumped solid-state laser. The comb offset, i.e. the offset of the comb lines with respect to the absolute zero frequency was detected in a so-called self-referenced manner using $f$-to-$2f$ interferometry and stabilized by providing feedback to the pump diode of the laser. Careful engineering of the laser system and the feedback loop allowed us to reach record-low noise performance for GHz solid-state lasers.

Diode-pumped solid-state lasers based on Ytterbium-doped crystals emitting light at 1 µm have proven to be reliable and power-scalable sources that can be scaled into the GHz pulse repetition rate regime while emitting short (<100 fs) pulses. However, many applications of frequency combs, in particular in the field of
molecular spectroscopy for environmental gas monitoring, require comb lines in the mid-infrared spectral region, ideally from 2 - 5 \( \mu \text{m} \) or even beyond. In Paper 3, we have transferred our stable 1-GHz frequency comb into this mid-infrared region by using a frequency conversion technique called optical parametric amplification (OPA), which was carried out in waveguides made of periodically poled lithium niobate (PPLN). We discuss how to take advantage of the waveguide properties to obtain high (> 35 dB) parametric amplification and efficient energy transfer from the pump (output of the 1-GHz laser at 1 \( \mu \text{m} \)) to the signal (spectral portion around 1.5 \( \mu \text{m} \) obtained via SCG using the same 1-GHz laser) and the idler beam. The idler is an inherently offset-free frequency comb that can be tuned from 2.5-4.2 \( \mu \text{m} \).

Finally, we present a novel type of modelocked laser cavity based on self-defocusing nonlinearities in Paper 4, which allowed us to develop a Watt-level femtosecond 10-GHz laser in a compact straight cavity design. Pushing SESAM-modelocked solid-state lasers to high repetition rate is generally associated with issues caused by the build-up of strong pulse energy fluctuations, so-called Q-switching instabilities, which can damage the laser components before stable operation is achieved. We show that by engineering a dynamic self-defocusing lens in a two-dimensionally patterned PPLN crystal, we can create a mechanism that protects the cavity elements from damage. This mechanism is based on cascading of second-order nonlinearities, an effect that provides a way to generate tunable intracavity self-phase modulation (SPM). The latter can be negative in sign, which then allows for the formation of femtosecond soliton pulses using positive material dispersion.

Each of the papers mentioned above is embedded in this thesis with some introductory remarks and further discussion where it is appropriate. The results presented here show that, by combining robust SESAM-modelocked laser designs with the countless facets offered by low-energy nonlinear optics, we can create versatile and compact frequency comb sources in the GHz regime to meet the needs of a diverse range of applications.
Kurzfassung


Diese Arbeit befasst sich mit Ergebnissen, die Teil dieser fortlaufenden Bemühungen sind und legt dabei den Schwerpunkt auf einen bestimmten Aspekt: Frequenzkämme basierend auf kompakten modengekoppelten Festkörperlasern mit Gigahertz-Pulswiederholraten.


In dieser Arbeit werden neue Ansätze vorgestellt, die diese Herausforderung aus einem anderen Blickwinkel betrachten: Durch die Nutzung neuer nichtlinearen optischen Bauelemente demonstrieren wir effiziente Frequenzkonversionsprozesse, die bei niedrigen Pulsenergien (Picojoule) funktionieren und somit die Komplexität und Kosten von Frequenzkammssystemen mit hoher Repetitionsrate niedrig halten. Die in dieser Arbeit verwendeten direkt diodengepumpten Laserquellen basieren auf Ytterbium-dotierten Kristallen, die bei einer Zentralwellenlänge von 1 \( \mu \)m emittieren. Sie beruhen auf einem sättigbaren Halbleiterspiegel (SESAM), welcher den gepulsten Betrieb, d.h. die Modenkopplung in Gang setzt und zu stabilisiert. Im Einführungskapitel dieser Arbeit werden wir die Funktionsweise dieser Laserart genauer ansehen und den neuesten Stand der Technik bzgl. der Leistungsfähigkeit laserbasierter GHz-Frequenzkämme vorstellen. Die darauffolgenden Kapitel bestehen jeweils aus Ergebnissen, die in Fachzeitschriften veröffentlicht wurden. Die Artikel wurden in die Arbeit integriert, indem ihr visuelles Format angepasst, ihr ursprünglicher aber Inhalt beibehalten wurde. Die vier Beiträge werden in chronologischer Reihenfolge präsentiert.

In Paper 1 wird erstmals die oktavenüberspannende spektrale Verbreiterung, sogenannte Superkontinuum-Erzeugung (SCG), in Siliziumnitrid-Wellenleitern demonstriert. Diese neuartigen Strukturen sind vom Sichtbaren bis zum mittleren Infrarotbereich transparent und ermöglichen es uns, ein kohärentes oktavenübergreifendes Superkontinuum mit nur \( \sim 36 \) pJ Pulsenergie zu erzeugen, d.h. mit einer Größenordnung weniger Energie als mit herkömmlichen Quarzglasfasern.

In Paper 2 zeigen wir, dass sich das aus den Siliziumnitrid-Wellenleitern erhaltene Superkontinuum dazu eignet, einen Offset-stabilisierten Kamm aus einem 1-GHz diodengepumpten Festkörperlaser zu generieren. Der Kammversatz, d.h. der Offset der Kammlinien in Bezug auf die absolute Nullfrequenz, wurde in einer sogenannten selbst-referenzierten Weise unter Verwendung von \( f \)-zu-\( 2f \)-
Interferometrie detektiert und stabilisiert, indem ein Rückkopplungssystem die Pumpdiode des Lasers steuert. Sorgfältiges Engineering des Lasersystems und der Rückkopplungsschleife ermöglichte es uns, eine für GHz-Festkörperlasern besonders rauscharme Rauschleistung zu erreichen.


der eine Möglichkeit zur Erzeugung einer durchstimmbaren Eigenphasenmodulation (SPM) bietet. Letztere kann ein negatives Vorzeichen besitzen, was die Bildung von Femtosekunden-Soliton-Pulsen unter Verwendung positiver Materialdispersion ermöglicht.

Chapter 1

Introduction

In order to use light as a tool – whether for scientific experiments or in our daily life – it usually has to be controlled and manipulated in space, time or frequency. Early on already, mankind realized that light could be somehow guided spatially using items such as lenses and mirrors. Descriptions of geometrical optics date back to the ancient islamic and greek civilizations. The search for mechanism to generate short bursts of light in time became popular in the 19th century with the invention of flash photography. Although the description of light as an electromagnetic wave with a certain wavelength/frequency was developed in the same century, altering the frequency of light however, i.e. changing its color, only became a practicable thing after the invention of the laser (short for “light amplification by stimulated emission of radiation”) in the 1960’s [1]. Altering the frequency of an electromagnetic wave requires a nonlinear process and is hence strongly depending on the intensity of the light source. With the invention of the laser, this type of light source became available and nonlinear optics has since then become an integral part of optical technology development. In this introductory chapter, we will briefly summarize the nonlinear optics concepts that are essential to the understanding of modelocked lasers as well as the various frequency conversion processes that will be further addressed in the following chapters of this thesis. We will also introduce the concept of soliton modelocking with semiconductor saturable absorber mirrors (SESAMs) and provide an overview of state-of-the art modelocked lasers at GHz pulse repetition rates.
1.1 Nonlinear Optics

Maxwell’s equations provide the basis for the interaction of electromagnetic waves with matter. In absence of external charges and currents, the equations for the electric field \( E \) and the magnetic field \( H \) read as follows:

\[
\nabla \times E = -\frac{\partial B}{\partial t} \\
\nabla \times H = \frac{\partial D}{\partial t} \\
\n\nabla \cdot D = 0 \\
\n\nabla \cdot B = 0
\]

with the electric displacement field \( D = \varepsilon_0 E + P \) and the magnetic flux density \( B = \mu_0 (H + M) \). The magnetization \( M \) and the polarization \( P \) denote the response of the material. From these equations, one can derive the wave equation that governs the propagation of light through a dielectric medium,

\[
\nabla^2 E - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} E = \mu_0 \frac{\partial^2}{\partial t^2} P
\]

where \( c = \sqrt{\mu_0 \varepsilon_0}^{-1} \) is the velocity of light in vacuum. The polarization \( P \) is a tensor quantity and can be expanded in a Taylor series using the corresponding electric susceptibilities \( \chi^{(m)} \):

\[
P^{(1)} = \varepsilon_0 \sum_i \chi^{(1)}_{ij} E_j + \varepsilon_0 \sum_{jk} \chi^{(2)}_{ijk} E_j E_k + \varepsilon_0 \sum_{jkl} \chi^{(3)}_{ijkl} E_j E_k E_l + ... \quad (1.3)
\]

The linear response of the medium is represented by the first term, i.e. \( m=1 \), and is commonly expressed via the complex-valued refractive index \( n \) of the material, where \( n^2 = 1 + \chi^{(1)} \). The higher-order terms constitute the nonlinear response and give rise to effects that are nowadays widely exploited in the field of laser optics.

Table 1 summarizes some of the most important effects and how they relate to the work presented in this thesis. To simplify the notation in this qualitative overview, we assume that the electric field is a monochromatic plane wave propagating along the \( z \)-direction, i.e.
\[ E(r,t) = Ae^{i(kz-\omega t)} + A^*e^{-i(kz-\omega t)} , \tag{1.4} \]

and that multi-wave interactions take place collinearly. The intensity in SI-units can then be expressed as \( I = 2\varepsilon_0 nc|A|^2 \). The various effects will be discussed in more detail within the corresponding chapters.

**Note:** Depending on the context, the electric field of a plane wave is often also defined as

\[ E(z,t) = \frac{1}{2}(Ae^{i(\omega t-kz)} + A^*e^{-i(\omega t-kz)}) \tag{1.5} \]

with the factor 1/2 and a different convention for the sign of \((\omega t-kz)\). In this case, the intensity is \( I = (1/2)\varepsilon_0 nc|A|^2 \). This notation will be used for instance in Chapter 4.


### 1.1.1 Nonlinear effects: overview

<table>
<thead>
<tr>
<th>Order</th>
<th>Name</th>
<th>Useful expressions</th>
<th>In this thesis</th>
</tr>
</thead>
<tbody>
<tr>
<td>2nd</td>
<td>Second-harmonic generation (SHG)</td>
<td>( P_{\text{SH}}^{(2)} = \epsilon_0 \chi^{(2)} A_0^2 e^{i\Delta k z} e^{i(2k_0 z - 2\omega_0 t)} + \text{c.c.} )</td>
<td>Chapter 2, f-to-2f interferometry</td>
</tr>
<tr>
<td></td>
<td></td>
<td>phase-matching: ( \omega_{\text{SH}} = 2\omega_0, \Delta k = 2k_0 - k_{\text{SH}} = 0 )</td>
<td></td>
</tr>
<tr>
<td>2nd</td>
<td>Optical Rectification (OR)</td>
<td>( P_{\text{OR}}^{(2)} = 2\epsilon_0 \chi^{(2)} A_0 A_0^\ast )</td>
<td>----</td>
</tr>
<tr>
<td>2nd</td>
<td>Sum frequency generation</td>
<td>( P_{\text{SG}}^{(2)} = 2\epsilon_0 \chi^{(2)} A_1 A_2^\ast e^{i(k_1 + k_2 z - (\omega_1 + \omega_2)t)} + \text{c.c.} )</td>
<td>Chapter 2, Supercontinuum generation</td>
</tr>
<tr>
<td></td>
<td></td>
<td>phase-matching: ( \omega_3 = \omega_1 + \omega_2, \Delta k = k_1 + k_2 - k_3 = 0 )</td>
<td></td>
</tr>
<tr>
<td>2nd</td>
<td>Difference frequency generation (DFG)</td>
<td>( P_{\text{DFG}}^{(2)} = 2\epsilon_0 \chi^{(2)} A_1^\ast A_2 e^{i(k_1 - k_2 - k_3) z - (\omega_1 - \omega_2)t} + \text{c.c.} )</td>
<td>Chapter 3, Mid-infrared frequency comb</td>
</tr>
<tr>
<td></td>
<td></td>
<td>phase-matching: ( \omega_3 = \omega_1 - \omega_2, \Delta k = k_1 - k_2 - k_3 = 0 )</td>
<td></td>
</tr>
<tr>
<td>2nd</td>
<td>Optical parametric amplification/oscillation (OPA/OPO)</td>
<td>DFG with amplification of ( \omega_2 )-component ( \omega_1 ): &quot;pump&quot;, ( \omega_2 ): &quot;signal&quot;, ( \omega_3 ): &quot;idler&quot;</td>
<td>Chapter 3, Mid-infrared frequency comb</td>
</tr>
<tr>
<td>2nd</td>
<td>Cascading</td>
<td>( \Delta k &gt; 0 ) and no energy transfer to second-harmonic wave intensity-dependent refractive index: ( n(I) = n + n_2^\ast \omega ) with ( n_2^\ast = \frac{1}{\Delta k} \frac{\omega_0 \big( \chi^{(2)}(\omega_0) \big)^2}{2\epsilon_0 c^2 n_{\omega_0}^2 n_{2\omega_0}} )</td>
<td>Chapter 4, Self-defocusing 10-GHz laser</td>
</tr>
<tr>
<td>3rd</td>
<td>Third-harmonic generation (THG)</td>
<td>( \chi^{(3)} = \chi^{(3),\text{electronic}} + \chi^{(3),\text{Raman}} )</td>
<td>----</td>
</tr>
<tr>
<td></td>
<td></td>
<td>( P_{\text{TH}}^{(3)} = \epsilon_0 \text{Re} \left( \chi^{(3),\text{electronic}} \right) A_0^3 e^{i\Delta k z} e^{i(3k_0 - 3\omega_0)t} + \text{c.c.} )</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>phase-matching: ( \omega_{\text{TH}} = 3\omega_0, \Delta k = 3k_0 - k_{\text{TH}} = 0 )</td>
<td></td>
</tr>
</tbody>
</table>
### 3rd Kerr effect

\[ P_{\text{Kerr}}^{(3)} = 3\varepsilon_0 \text{Re}\left(\chi_{\text{3, electronic}}^{(3)}\right) |A_0|^2 E_0(r,t) \]

intensity-dependent refractive index caused by instantaneous electronic response:

\[ n(I) = n + n_2 I \]

with

\[ n_2 = \frac{3}{4\varepsilon_0 c n_m^2} \text{Re}\left(\chi_{\text{3, electronic}}^{(3)}\right) \]

Chapter 2-4

Self-phase modulation,

Soliton modelocking,

Self-focusing

### Cross-phase modulation (XPM)

Two-beam interaction: nonlinear refractive index change of for beam (2) caused by intensity of beam (1)

\[ n(I^{(2)}) = n^{(2)} + 2n_2 I^{(1)} \]

Chapter 2

Supercontinuum generation

### Two-photon absorption (TPA)

intensity-dependent complex change in refractive index:

\[ n(I) = n + \frac{\beta_{\text{TPA}}}{2k_0} I \]

\[ \beta_{\text{TPA}} = -i \frac{3\omega_0}{2\varepsilon_0 c^2 n_m^2} \text{Im}\left(\chi_{\text{3, electronic}}^{(3)}\right) \]

Chapter 4

SESAM roll-over

### Four-wave mixing

\[ \omega_1 + \omega_2 = \omega_3 + \omega_4 \]

\[ \omega_1, \omega_2 : "\text{pump}" \]

\[ \omega_3 = \omega_1 - (\omega_2 - \omega_1) : "\text{Stokes}" \]

\[ \omega_4 = \omega_2 + (\omega_2 - \omega_1) : "\text{anti-Stokes}" \]

Chapter 2

Supercontinuum generation

### Raman scattering

scattering with optical phonons

\[ P_{\text{Raman}}^{(3)} = 6\varepsilon_0 \chi_{\text{3, Raman}}^{(3)} |A_0|^2 E_{\text{Raman}}(r,t) \]

Chapter 2

Supercontinuum generation

### Brillouin scattering

scattering with acoustic phonons

----

Table 1: Overview of important nonlinear effects and the context in which they play a role for the work presented in this thesis.


1.2 Optical pulses and frequency combs

In the previous paragraph, we have provided an overview of different nonlinear effects using the formalism of monochromatic plane electromagnetic waves, which corresponds well to the output of a single-frequency continuous wave (cw) laser. However, the high peak intensities that are usually required to efficiently drive these processes are difficult to reach in practice for lasers in cw operation. We can nevertheless reach these intensities without the need for more average power by concentrating the energy in time, i.e. by generating an optical pulse train.

Mathematically, such a pulse train can be obtained by the discrete superposition of plane waves with different frequencies, but identical phases. In the time domain, the real-valued electric field \( E(t) \) of a pulse train can be written as

\[
E(t) = \left( A(t)e^{i\omega_0 t} \right) \ast \left[ \sum_n \delta(t - nT_r) e^{i2\pi f_{CEO} n} \right] + \text{c.c.,}
\]

(1.6)

where \( A(t) \) represents the temporal pulse envelope (e.g. Gaussian or hyperbolic secant shape), \( \omega_0 \) is the carrier angular frequency, \( T_r = \frac{1}{f_{\text{rep}}} = \frac{2\pi}{\Delta\omega_{\text{rep}}} \) represents the time between subsequent pulses and \( f_{\text{CEO}} \) is the carrier-envelope offset (CEO) frequency (see Figure 1.1). The amplitude \( A(t) \) and hence also \( E(t) \) can be obtained via the Fourier-Transform

\[
A(t) = \frac{1}{2\pi} \int \tilde{A}(\Delta\omega) e^{i\Delta\omega t} d\Delta\omega
\]

\[
E(t) = \frac{1}{2\pi} \int \tilde{E}(\omega) e^{i\omega t} d\omega
\]

(1.7)

where \( \Delta\omega = \omega - \omega_0 \) and the pulse spectrum is expressed as

\[
\tilde{E}(\omega) = \tilde{A}(\Delta\omega) \sum_n \delta(\omega - n\Delta\omega_{\text{rep}} - 2\pi f_{\text{CEO}}) + \text{c.c.}
\]

(1.8)

This discrete superposition of equally spaced spectral lines is commonly called a frequency comb (Figure 1.1(b)). The comb lines are centered around the angular
carrier frequency $\omega_0$ and are $2\pi f_{\text{rep}}$ apart. The position of the comb with respect to the absolute zero frequency is determined by $2\pi f_{\text{CEO}}$.

![Figure 1.1](image)

Figure 1.1: Time and frequency domain. (a) Representation of a pulse train in the time domain. After the time $T_{\text{CEO}}$, the peak of the envelope coincides again with the peak of the underlying oscillations of the electric field. (b) Representation of the pulses in the frequency domain. The pulse spectrum consists of sharp lines that are equally spaced by a distance corresponding to the pulse repetition rate.

### 1.3 Modelocking

In the mathematical description of a pulse train above, we have assumed a superposition of waves with a fixed phase relation. In a laser cavity, these waves correspond to the different longitudinal modes that are supported by the resonator. While the optical length of the resonator determines the spacing between the different modes ($\Delta\omega_{\text{rep}} \propto (L_{\text{opt}})^{-1}$), their phases are initially random. In order for a pulse to coherently build up, these phases need to be locked, hence the term “modelocking”. In practice, this means that a mechanism has to be in place in the laser cavity, which favors pulsed over continuous wave operation. We can generally differentiate between two methods:

**Active modelocking:** An externally driven element, such as for example an acousto- or electro-optic modulator, is used to modulate the losses in the laser cavity. The frequency of this loss modulation thus determines pulse repetition rate.

**Passive modelocking:** Using passive (i.e. not externally driven) mechanisms, it becomes possible to achieve pulse durations well below the switch time of an active modulator. As an example, losses can be introduced to the cw-beam by clipping it at
an aperture in the cavity or by inducing a change of the beam radius in the gain medium. If a pulse with high intensities happens to be triggered by a perturbation of the cavity, the beam will undergo self-focusing as a consequence of the optical Kerr effect. Since the reduced beam diameter fits through the aperture or leads to a better overlap between the pump and the laser beam, the laser will experience less losses in pulsed operation than when operating in continuous wave. Methods that are based on drastic changes of the cavity mode sizes due to the nonlinear Kerr effect are generally referred to as Kerr-lens modelocking.

Another major method consists of using a saturable absorber, i.e. an optical component that absorbs light at lower intensities, but saturates (becomes transparent) for high intensities. This saturation effect will thus favor modelocking operation, since the pulses are less subjected to losses due to their high peak intensity. The lasers developed within the framework of this thesis all relied on this second method, more specifically on semiconductor devices called SESAMs [2] (more details in section 1.4).

1.3.1 Soliton modelocking

Although the modelocking trigger mechanism for the lasers presented here is not based on the Kerr-lens principle, the Kerr effect still plays an important role for the pulse shaping in time and frequency, since we chose to develop lasers that operate in the so-called soliton regime. Solitons are pulses that are characterized by a particularly stable propagation in both the time and frequency domain. The stable propagation is a result of the balance between the phase change induced by group velocity dispersion and the nonlinear phase change induced by the Kerr effect (called self-phase modulation (SPM)). The temporal profile of a soliton pulse follows a squared hyperbolic secant (sech) function and the pulse duration is usually defined as the full-width-at-half-maximum (FWHM) of the pulse intensity:

$$I(t) \propto A_0^2 \text{sech}^2 \left( \frac{t}{\tau} \right), \quad \tau_{\text{FWHM}} \approx 1.7627 \cdot \tau$$

In a soliton modelocked laser consisting of a Kerr-medium of length $L_{\text{Kerr}}$ and dispersive elements yielding a group delay dispersion $\text{GDD}_{\text{tot}}$ per roundtrip, the pulse duration (assuming two passes in the Kerr medium) scales according to
\[ \tau_{\text{FWHM}} = 1.7627 \cdot \frac{\text{GDD}_{\text{tel}} \lambda_0^2 w_{\text{Kerr}}^2}{4 L_{\text{Kerr}} n_2^2 |U_p|} \]  

(1.10)

where \( n_2 \) is the nonlinear index, \( w_{\text{Kerr}} \) is the beam waist in the Kerr medium, \( \lambda_0 \) is the center wavelength, \( U_p \) is the pulse energy.

For soliton pulses, the peak power \( P_{pk} \) can be expressed using the following relations

\[ U_p = P_{av} f_{\text{rep}}, \quad P_{pk} \approx 0.88 \cdot \frac{U_p}{\tau_{\text{FHWM}}} \]  

(1.11)

where \( P_{av} \) denotes the average power. Most commonly, the nonlinear index has a positive value as a result of the intrinsic positive third-order susceptibility of the material and the SPM needs to be compensated with negative GDD. In Chapter 4 however, we will show that soliton modelocking can also be achieved with the sign of those two quantities reversed.

### 1.4 Semiconductor saturable absorber mirror (SESAM)

SESAMs are semiconductor devices that enable stable and self-starting modelocking. They consist of a highly reflective distributed Bragg reflector (DBR) and an absorber layer (e.g. quantum well or quantum dots) located close to the surface Figure 1.2 (a)). The absorbing quantum well/dot layer is designed in order to saturate at the intracavity pulse energy expected to be delivered by the laser. For lasers where very high powers are targeted, the absorber section can contain several absorber layers in order to increase the saturation fluence [3-5]. For laser operating in the GHz-regime at Watt-level output powers, SESAMs with only one absorber layer are usually sufficient.

A SESAM is commonly characterized by the following parameters:

**Modulation depth** \( \Delta R = R_{ns} - R_{\text{lin}} \): difference between the reflectivity of the fully saturated SESAM and the reflectivity at low fluences

**Saturation fluence** \( F_{\text{sat}} \): fluence at which the reflectivity has increased by 1/e of the modulation depth
**Nonsaturable losses** $\Delta R_{ns} = 1 - R_{ns}$: unwanted losses that remain in the saturated state and can lead to heating of the SESAM

**$F_2$-parameter**: quantifies induced absorption effects such as two-photon absorption, which lead to a roll-over of the reflectivity curve at high fluences

**Recovery time**: time constant over which the absorber recovers after having been saturated by a pulse. Usually, the recovery can be described using a bi-exponential function; the short time scale ($\tau_{\text{fast}} \sim 100$ fs) corresponds to thermalization within the valence and the conduction band, while the longer time scale ($\tau_{\text{slow}} \sim \text{ps-ns}$) describes the process of electron-hole recombination across the band gap.

Using the parameters described above, we can express a model function for the reflectivity of a SESAM as a function of the incident fluence $F = \frac{U_p}{\pi w^2}$ (i.e. pulse energy per beam area) [7]:

$$R(F) = R_{ns} \left[ \frac{1 + R_{lin} / R_{ns} \left( e^{F/F_{sat}} - 1 \right)}{F / F_{sat}} \right] e^{-F/F_2} \quad (1.12)$$
This equation is valid for a flattop beam profile. Inside a laser cavity however, the beam will usually have a Gaussian shape. Hence, to obtain the actual reflectivity for a Gaussian beam, we have to integrate over the spatial energy distribution, leading to

$$R^{\text{Gaussia}}(F) = \frac{1}{2F} \int_0^{2F} R(z) \, dz$$

with the substitution $z = 2F \exp(-2r^2 / w^2)$.

The SESAM is considered to be a "slow" saturable absorber due to its finite recovery time. In contrast, artificial saturable absorbers based on e.g. nonlinear phase shifts (such as Kerr lens modelocking) are called "fast", since the modulation follows the optical power in a quasi-instantaneous fashion. In order to generate short pulses (i.e. much shorter then the recovery time of the SESAM) it is advantageous to combine the benefits of the SESAM with the principle of soliton modelocking described above [8]. By introducing an appropriate amount of intracavity dispersion to balance the nonlinear self-phase modulation, a stable pulse train of short pulses can be initiated and maintained: any potential continuous wave background that may still experience gain during the recovery of the SESAM will not be able to acquire enough nonlinear phase and will thus just be dispersed in time. Hence, the growth of such background instabilities will be suppressed.
1.5 State-of-the-art GHz modelocked lasers

The work presented in this thesis is based on the successful development of high-power SESAM-modelocked solid-state lasers based on Ytterbium (Yb)-doped gain materials. In this section, we would like to briefly discuss the key advantages of this technology and place it into the general landscape of state-of-the-art modelocked lasers in the GHz pulse repetition rate regime. Further comparisons with other frequency comb technologies and additional references can also be found in the introduction paragraphs of the Papers 1-4 integrated in this thesis.

1.5.1 General overview

Modelocked lasers can be classified under many aspects. An obvious categorization consists of distinguishing between different laser gain materials (crystals, glasses, ceramics, semiconductors etc.) and geometries (e.g. bulk, fiber, thin disk). The choice of the gain material will determine e.g. the wavelength, the achievable spectral bandwidth as well as aspects such as costs and the possibility of mass production. The geometry will be linked to factors such as power scalability, heat management or ruggedness. The goal of the laser research community not only consists of developing novel materials as well as geometries, but also consists of pushing the performance of the different configurations in order to exploit the full potential of each technology. The performance goals are of course closely linked to the targeted applications.

The research results presented in this thesis are driven by the need for robust and compact sources to generate frequency combs with large comb line spacings in the GHz range. Since nonlinear optical processes are used to broaden the spectral coverage of a frequency comb as well as for the comb stabilization itself, it is worth comparing potential laser sources with respect to their ability to drive such processes. Since the efficiency of most processes can be increased by a combination of high power and short pulses, the peak power (i.e. average power/(repetition rate x pulse duration)) can be used as useful criterion to compare the performance. In Figure 1.3, we show an overview of modelocked laser results with a pulse repetition rate > 1 GHz and peak powers above 10 W in fundamental modelocking. The graphs only contain results that were obtained directly out of the laser oscillator, i.e. without external amplification of compression of the pulses. Until the year 2012, Kerr-lens modelocked lasers based on titanium sapphire (Ti:sapphire) were clearly leading the...
field of high peak power GHz sources. Over the last five years however, significant advances were demonstrated with another class of modelocked laser: ytterbium (Yb)-doped crystals. Furthermore, the performance of semiconductor disk laser such as vertical external cavity surface emitting lasers (VECSELs) modelocked integrated external cavity surface emitting lasers (MIXSELs) experienced a significant push [38, 55].

Figure 1.3: Overview of modelocked laser sources with pulse repetition rates > 1 GHz in the year 2012 vs. 2017 [9-54]. Only results that were achieved directly out of the oscillator (i.e. without external amplification and/or compression are represented in this graphs. The circled result correspond to the 1-GHz laser used for the experiments presented in chapter 2 and 3, while our newest result marked with the yellow start will be the topic of chapter 4.
1.5.2 SESAM modelocked diode-pumped Yb:CALGO lasers

The development of high-power lasers based on Yb:CALGO ($\text{Yb}^{3+}:\text{CaGdAlO}_4$) in the 1-5 GHz range was carried out in our research group prior or in parallel to the work presented here and is discussed in [56]. The 10-GHz Yb:CALGO laser marked with a star in Figure 1.3 will be presented in chapter 4 of this thesis.

Yb:CALGO [57] is a uniaxial crystal with two molecules per unit cell, the latter having a volume of $V=160.8240 \text{ Å}$ [58]. In the host material CaGdAlO$_4$, both the Ca$^{2+}$ and Gd$^{3+}$ ions can be substituted by Yb$^{3+}$. The material is characterized by a rather high thermal conductivity of $6.3 \text{ Wm}^{-1}\text{K}^{-1}$ along the c-axis and $6.9 \text{ Wm}^{-1}\text{K}^{-1}$ along the a-axis at a doping concentration of 2 at. % [59]. In contrast to Kerr lens modelocked Ti:sapphire lasers, Yb:CALGO lasers offer several key advantages:

- The high thermal conductivity as well as a low quantum defect (emission wavelength around 1050 nm, pump 980 nm) help to reduce unwanted effects such as thermal lensing and hence allow for power scaling.

- Low-cost spatially multimode pump diodes at 980 nm can be used for direct pumping, eliminating the need for cumbersome high-brightness green pumps based on frequency-doubled solid-state lasers.

- SESAM can be used for self-starting and stable modelocking that does not require initiation of the modelocking process at the edge of the laser cavity stability zone.

Compared to other Yb-doped materials such as the tungstates KGW or KYW (which already provide larger spectral gain bandwidths than e.g. the classic high-power laser material Yb:YAG) CALGO offers an even smoother and broader emission bandwidth. Pulses with bandwidths that support pulses as short as 32 fs have been demonstrated in a 100-MHz laser based on an Yb:CALGO gain crystal [60].

The goal of this thesis was to demonstrate that SESAM-modelocked Yb:CALGO lasers are ideal sources to create compact low-noise frequency combs in the GHz regime and that their operating range can be extended to repetition rates of $>10$ GHz. In the following chapter, we will discuss the challenges that had to be overcome in order to achieve these goals and will present the successful outcomes in form of the original peer-reviewed publications.
Chapter 2

Frequency comb offset detection and stabilization

In this chapter, we discuss the challenge of obtaining a stabilized frequency comb with GHz line spacing. As described in section 1.2, the absolute position of the comb lines on the frequency axis is determined by the comb offset $f_{CEO}$ and the spacing between the lines, which corresponds to the repetition rate $f_{rep}$. In order to control and stabilize those two degrees of freedom, they first have to be detected. While the pulse repetition rate can directly be measured using a photodiode with a bandwidth $> f_{rep}$, the comb offset is trickier to detect. This is due to the fact that the offset frequency $f_{CEO}$ does not represent an actual line of the comb carrying some fraction of the light, but is merely an extrapolation from the few 100-THz-range (in which the actual comb light is situated on the frequency axis) down to absolute zero. The frequency $f_{CEO}$ is thus situated in the microwave range and its detection requires an additional effort. In the following sections, we will briefly describe the so-called $f$-to-$2f$ method for detection $f_{CEO}$ as well as the principle of supercontinuum generation, which allows for the generation of the octave-spanning spectra necessary for this technique.

2.1 Frequency comb offset detection

In the late 1990’s, several techniques were suggested to detect the carrier-envelope offset frequency [61-63]. Among those techniques, $f$-to-$2f$ interferometry became one of the most common ways of detecting the CEO of a modelocked laser comb. Its principle is sketched in Figure 2.1. Light from the low-frequency end of the comb is frequency-doubled using SHG in a nonlinear crystal. This frequency-doubled light is then recombined in time and space with the corresponding high-frequency part of
the comb in a heterodyne fashion. The resulting beating signal, corresponding to $f_{CEO}$, can then be detected using a sufficiently sensitive photodetector. Since the $f$-to-$2f$-method does not require any additional laser to detect $f_{CEO}$, but is only based on the beating of different parts of the same comb, it is also often referred to as “self-referencing”.

![Figure 2.1: Principle of f-to-2f interferometry to detect the carrier-envelope offset frequency $f_{CEO}$.](image)

The pre-requisite for this technique is that the comb lines span at least one octave. For the output spectrum of most modellocked laser, this is however not the case. As an example, a transform-limited 100-fs soliton pulse with a center wavelength of 1050 nm has a bandwidth of 11.8 nm, while 500 nm or more (depending on the exact center wavelength) would be required for an octave. Before being sent into the $f$-to-$2f$ interferometer, the spectrum thus needs to be broadened by an interplay of different nonlinear processes, a technique generally referred to as supercontinuum generation (SCG).

### 2.2 Supercontinuum generation (SCG)

The first demonstration of supercontinuum generation was reported in bulk glass in 1970 [64]. Since then, SCG has become a field of research on its own due to the high number of possibilities offered by combining various platforms and a large range of input light parameters. Different physical mechanisms are governing the SCG process depending on the platform properties such as

- the nonlinearity of the material
- the geometry and length (i.e. bulk/fiber/waveguides)
• the dispersion properties (combination of material properties and dispersion effects caused by the geometrical confinement of the guided light)

as well as the input pulse parameters, in particular
• the center wavelength
• the input bandwidth/pulse duration including potential chirp
• the pulse energy/peak power

The propagation of the pulse within the SCG platform can be described using the generalized nonlinear Schrödinger equation (GNLSE), where $A(z,T)$ represents the spatio-temporal pulse envelope of the electric field $E(z,T) = A(z,T)\exp(-i\omega_0 T)$ and $\alpha$ denotes the linear power attenuation:

$$\frac{\partial A(z,T)}{\partial z} + \frac{\alpha}{2} A(z,T) - \sum_{k=0}^{i+1} \frac{i^k}{k!} \beta_k \frac{\partial^k A(z,T)}{\partial T^k} = i\gamma \left( 1 + \frac{i}{\omega_0} \frac{\partial}{\partial T} \right) \left( A(z,T) \int_{-\infty}^{\infty} R(T')|A(z,T-T')|^2\,dT' \right)$$ (2.1)

This equation assumes a time frame that is moving along with the pulse, i.e. $T = t - \beta_1 z$, where $\beta_1$ is the inverse of the group velocity. The propagation constant $\beta(\omega)$ contains the information about the dispersion properties of the platform and can be expanded in a Taylor series:

$$\beta(\omega) = n(\omega) \frac{\omega}{c} = \sum_{k=0}^{i} \frac{1}{k!} \beta_k (\omega - \omega_0)^k, \text{ with } \beta_k = \frac{d^k \beta}{d\omega^k} \bigg|_{\omega=\omega_0}$$ (2.2)

The group velocity dispersion represents an important property of the SCG platform and is given by the coefficient $\beta_2$ with $[\beta_2] = \text{s}^2\text{m}^{-1}$. For fibers, the dispersion is often indicated using the parameter $D = -2\pi c / \lambda^2 \cdot \beta_2$ with $[D] = \text{ps (nm⋅km)}^{-1}$. The dispersion regime is called normal (or positive) if $\beta_2 > 0$ (i.e. $D < 0$) and anomalous (or negative) for $\beta_2 < 0$ (i.e. $D > 0$). The nonlinear coefficient $\gamma$ contains information about the material nonlinearity of the SCG platform (via the nonlinear index $n_2$) as well as the geometry (via the effective mode area $A_{\text{eff}}$):

$$\gamma = \frac{\omega_0 n_2}{c A_{\text{eff}}}$$ (2.3)
The nonlinear response function \( R(t) \) includes both the quasi-instantaneous electronic response \( h_e(t) \) and the delayed Raman response \( h_R(t) \):

\[
R(t) = (1 - f_R) h_e(t) + f_R h_R(t)
\]  

(2.4)

The coefficient \( f_R \) determines the fractional contribution of the delayed Raman response to the nonlinear polarization and is thus material-dependent (e.g. \( f_R = 0.18 \) for silica fibers). Note that the response time-domain functions \( h_e(t) \) and \( h_R(t) \) are linked to the nonlinear susceptibility \( \chi^{(3)} \) (section 1.1) via the Fourier transform.

### 2.2.1 Supercontinuum simulations including noise

Numerical simulations of the SCG process were performed within the framework of this thesis. The MATLAB code used for the purpose of simulating SCG in silica fibers was originally developed by the group of Prof. John M. Dudley. The code solves the GNLSE using the split-step Fourier method: for each propagation step along the length of the fiber, the linear effects are implemented by multiplications in the frequency domain, while the nonlinear steps are treated in the time domain [65]. In order to predict the influence of noise on the spectra and their coherence (see section 2.2.2 for definition), the code takes into account two types of noise: quantum-limited shot noise on the input pulse (added in the time domain) and spontaneous Raman noise. The latter is implemented by adding an additional stochastic variable \( \Gamma_R \) (originating from the quantum description of SCG [66-69]) to the right-hand side of Eq. (2.1):

\[
\ldots = i\gamma \left( 1 + \frac{i}{\omega_0} \frac{\partial}{\partial T} \right) A(z,T) \int_{-\infty}^{\infty} R(T') A(z,T-T') dT' + i\Gamma_R(z,T) \]  

(2.5)

The following relation holds for the frequency domain correlations of \( \Gamma_R \):

\[
\langle \tilde{\Gamma}_R(z,\Omega) \tilde{\Gamma}_R'(z',\Omega') \rangle = \frac{2f_R \hbar \omega_0}{\gamma} \text{Im} h_R(\Omega) \left( n_{th}(\Omega) + \Theta(-\Omega) \right) \delta(z-z') \delta(\Omega-\Omega')
\]  

(2.6)

where \( n_{th}(\Omega) = \left[ \exp(h\Omega / k_B\vartheta) - 1 \right]^{-1} \) is the Bose distributions of the phonons with frequency \( \Omega \) in the fiber at a finite temperature \( \vartheta \) and \( \Theta \) is the Heaviside step function. In a qualitative picture, this term can be understood as the effect of spontaneous interactions between the incoming photons and the phonons in the
fiber. At a temperature $\vartheta$, those phonons occupy the energy level $\hbar\Omega$ with a probability determined by the Bose distribution. In contrast to stimulated Raman scattering, these spontaneous effects add noise to the generated SC and can affect its coherence. In Paper 2, we published numerical investigation of the influence of spontaneous Raman noise on the coherence in the context of comparing SCG platforms with significant Raman action (such as silica fibers) and without (e.g. silicon nitride).

2.2.2 Coherence properties

In the context of SCG, the term coherence refers to the correlation between spectra generated by different pulses [70]. The so-called first-order coherence $g_{12}^{(1)}$ is defined as

$$g_{12}^{(1)}(\lambda) = \frac{\langle |\hat{E}_1(\lambda)|^2 \rangle}{\langle |\hat{E}_1(\lambda)|^2 \rangle^{1/2}}$$

where $\hat{E}_1(\lambda)$ and $\hat{E}_2(\lambda)$ correspond to the spectra generated by pulse 1 and 2 respectively. Using the simulation code described above, independent SC pairs $[\hat{E}_1(\lambda),\hat{E}_2(\lambda)]$ with different random noise can be generated to calculate $g_{12}^{(1)}$ as an ensemble average.

Experimentally, the first-order coherence can be measured by spectrally interfering the SC generated by one pulse with the SC obtained from the adjacent pulse. This interference can be created in a Michelson-interferometer, where one arm provides a delay corresponding to the pulse repetition period. By recording the fringes (i.e. the mean spectral density $I(\lambda) = \langle |\hat{E}(\lambda)|^2 \rangle$) using an optical spectrum analyzer and extracting the fringe visibility

$$V(\lambda) = \frac{I_{\text{max}}(\lambda) - I_{\text{min}}(\lambda)}{I_{\text{max}}(\lambda) + I_{\text{min}}(\lambda)}$$

the first-order coherence may be determined according to [68]

$$g_{12}^{(1)}(\lambda) = V(\lambda) \frac{I_1(\lambda) + I_2(\lambda)}{2I_1(\lambda)I_2(\lambda)}^{1/2} C(\lambda)$$
where the calibration factor \( C(\lambda) \) accounts for the fact that the power levels may be slightly different for the two individual arms of the interferometer.

The degree of coherence obtained in SCG experiment depends on numerous factors and has been the topic of extensive studies [71-73]. A rather obvious reason for the loss of coherence is noisy input pulses with strong amplitude fluctuations. But also the type of nonlinear interactions happening inside the SCG plays a significant role. The more the generation of new spectral components relies on the amplification of noise rather than stimulated processes, the lower the coherence. Degenerate four-wave-mixing (FWM), where two pump photons with the same optical frequency split into a lower-energy (Stokes) and higher-energy photon (anti-Stokes), can for instance lead to decoherence if the Stokes and anti-Stokes photon frequencies do not overlap with the spectrum of the input pulse, but are seeded from noise. This tends to be the case for long pulses (i.e. in the picosecond regime) where the input spectrum is very narrow.

### 2.2.3 Soliton order and soliton fission

The pulse train generated by the modelocked lasers used in this thesis consists of fundamental soliton, as described in section 1.3.1. To estimate the coherence of supercontinua seeded with soliton pulses, Dudley et al. [71] have introduced a useful metric, called the soliton order \( N \). It depends both on the soliton pulse duration \( \tau_{\text{FWHM}} \) and pulse energy \( U_p \) as well as the nonlinear- and dispersion properties of the SCG platform:

\[
N = \frac{\tau_{\text{FWHM}} U_p}{\sqrt{2 \cdot 1.7627 \beta_2}} \sqrt{\gamma}
\]  

(2.10)

In contrast to fundamental solitons, higher order solitons \( (N \geq 2) \) periodically change in the spectral and temporal domain during propagation. This effect can be used to generate efficient spectral broadening: By launching the higher-order soliton into a fiber that provides anomalous dispersion and positive SPM, its spectrum will broaden and the pulse will get compressed in time. At the point of maximum compression, so-called soliton fission may occur. In that case the soliton pulse breaks up into multiple (up to \( N \)) fundamental soliton pulses, which are ejected consecutively in time. The characteristic length after which fission occurs inversely scales with the soliton order, i.e.
\[ L_{\text{fiss}} = \frac{L_D}{N}, \text{ where } L_D = \frac{\tau_{\text{FWHM}}^2}{(1.7627)^2 |\beta_2|}. \] (2.11)

The fission process is susceptible to noise perturbations; hence to maintain good coherence, simulations and experiments have shown that the soliton order should be kept < 10 [74].
2.3 Introductory remarks to Paper 1

The supercontinuum generation experiments performed within the framework of this thesis were motivated by the need of an octave-spanning spectrum for self-referenced stabilization of GHz frequency combs. For a fixed average power, increasing the pulse repetition rate leads to a reduction in pulse energy and peak power. This reduction could be compensated by externally amplifying the pulses after they exit the modelocked laser cavity. This process is however known to add noise and complexity to the system. As discussed in the section above, noise on the input pulse can severely degrade the coherence of the supercontinuum. Good coherence however is a pre-requisite for the detection of the CEO frequency. A drop in coherence of the SCG will lead to background noise in the microwave domain, which in turn will reduce the signal-to-noise ratio (SNR) of the CEO beat signal or even completely destroy it.

We thus set ourselves the challenge to find a SCG platform that is capable to efficiently broaden pulses with peak powers well below 1 kW. Thanks to a collaboration with the groups of Prof. Alexander Gaeta and Prof. Michal Lipson, we got access to a new type of platform: silicon nitride (Si$_3$N$_4$) waveguides. Paper 1 presents our first results using this platform for the detection of the CEO frequency in the case of a 100 MHz and a 1 GHz laser. In an additional joint publication first-authored by A.R. Johnson [75], we used the fringe-method described in section 2.2.2 to provide an alternative proof that our octave-spanning supercontinuum indeed has a high coherence (0.90 on average from 675-725 nm and 0.99 from 1350-1450 nm, i.e. the sections used for $f$-to-$2f$ CEO detection).
Paper 1:

Frequency comb offset detection using supercontinuum generation in silicon nitride waveguides

Authors: Aline S. Mayer (100 MHz laser design, experimental setup and measurements)
Alexander Klenner (1 GHz laser design)
Adrea R. Johnson (waveguide design)
Kevin Luke (waveguide fabrication)
Michael R. E. Lamont (waveguide simulation code)
Yoshitomo Okawachi (supervision)
Michal Lipson (supervision)
Alexander L. Gaeta (supervision)
Ursula Keller (supervision)

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Frequency comb offset detection using supercontinuum generation in silicon nitride waveguides

A. S. Mayer,1,A. Klenner,1 A. R. Johnson,2 K. Luke,3 M. R. E. Lamont,2,3,4
Y. Okawachi,2 M. Lipson,3,4 A. L. Gaeta,2,4 and U. Keller1

1Department of Physics, Institute of Quantum Electronics, ETH Zurich, 8093 Zurich, Switzerland
2School of Applied and Engineering Physics, Cornell University, Ithaca, New York 14853, USA
3School of Electrical and Computer Engineering, Cornell University, Ithaca, New York 14853, USA
4Kavli Institute at Cornell for Nanoscale Science, Cornell University, Ithaca, New York 14853, USA

Abstract: We present the first direct carrier-envelope-offset (CEO) frequency detection of a mode-locked laser based on supercontinuum generation (SCG) in a CMOS-compatible silicon nitride (Si$_3$N$_4$) waveguide. With a coherent supercontinuum spanning more than 1.5 octaves from visible to beyond telecommunication wavelengths, we achieve self-referencing of SESAM modelocked diode-pumped Yb:CALGO lasers using standard f-to-2f interferometry. We directly obtain without amplification strong CEO beat signals for both a 100-MHz and 1-GHz pulse repetition rate laser. High signal-to-noise ratios (SNR) of > 25 dB and even > 30 dB have been generated with only 30 pJ and 36 pJ of coupled pulse energy from the megahertz and gigahertz laser respectively. We compare these results to self-referencing using a commercial photonic crystal fiber and find that the required peak power for CEO beat detection with a comparable SNR is lowered by more than an order of magnitude when using a Si$_3$N$_4$ waveguide.

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OCIS codes: (320.7090) Ultrafast lasers; (320.6629) Supercontinuum generation; (230.7370) Waveguides; (130.4310) Nonlinear (190.4390) Nonlinear optics, integrated optics.

References and links


1. Introduction

Stabilized frequency combs based on ultrafast lasers [1-4] have been a significant breakthrough with many important applications in various fields within chemistry, biology and physics. The frequency comb offset (i.e. the carrier envelope offset, CEO) is directly related to the exact position of the electric field underneath the pulse envelope [1], which has become important for highly nonlinear systems and attosecond science [5]. The broad coherent optical spectra obtained by supercontinuum generation (SCG) are the basis for many biomedical imaging techniques [6, 7]. Stabilized optical frequency combs enable the measurement of optical frequencies with a very high precision, which is essential for many spectroscopic applications ranging from the detection of molecular transitions to the calibration of astronomical spectrographs [8-10]. In the context of optical frequency metrology, frequency combs provide a direct link between optical and microwave frequencies, a property that has been exploited to strongly reduce the complexity of the optical atomic clocks, leading to a new generation of systems for the primary definition of time [11]. Currently, the comb line spacing obtained from commercially available sources is usually not exceeding a few hundred megahertz. Gigahertz line spacings and an increased power per mode would however be beneficial for important applications such as the generation of ultra-low noise microwave signals [12] or to enable the resolution of individual lines in calibration procedures for astronomical spectrographs [10].

Great efforts have been made to obtain a stabilized 10-GHz frequency comb from a Ti:sapphire laser [13]. However, Ti:sapphire systems still rely on an expensive and complex single-mode pumping scheme and Kerr lens modelocking [14] requires operation at the edge of the cavity stability range [15]. The cost-efficient and more compact fiber-based frequency comb sources on the other hand work in a regime with comparably high cavity losses, which reduces the cavity quality factor (Q) and results in deteriorated noise properties [16-18]. The limited power of gigahertz fiber laser oscillators has not allowed for comb stabilization without additional amplification.

Low noise properties and reliable operation can be obtained using diode-pumped solid-state lasers (DPSSL’s) modelocked with a semiconductor saturable absorber mirror (SESAM) [19]. Recently, the successful stabilization of the frequency comb offset from a gigahertz SESAM-modelocked DPSSL has been demonstrated [20]. The short pulses (64 fs) combined with an average output power of more than 1 W provides sufficient peak power for coherent octave-spanning SC which is typically obtained by additionally broadening the pulse spectrum in an external nonlinear device. To date silica-based optical fibers have been the most common SCG platform to self-reference modelocked lasers. The most efficient spectral broadening is
generally obtained if soliton effects dominate the SCG process, i.e. if the fiber is pumped within its anomalous group velocity dispersion (GVD). Standard silica fibers provide anomalous GVD for wavelengths above $\approx 1.3 \, \text{µm}$. For lasers operating in the 1-µm regime however, the design of fibers with anomalous GVD for the pump wavelength is more complex, involving for instance a microstructured cross-section. Only a limited choice of such fibers is commercially available and the required peak power remains a challenge particularly for lasers at high repetition rate and/or with pulse durations longer than 100 fs.

Recent investigations have focused on reducing the requirements for coherent SCG by using novel nonlinear devices. The research on chip-based supercontinuum generation in materials such as periodically poled lithium niobate (PPLN) [22-25], chalcogenide [26-28], silicon [29, 30], amorphous silicon [31], Hydex [32] and silicon nitride ($\text{Si}_3\text{N}_4$) [33, 34] has opened up interesting new possibilities in terms of compactness, nonlinearity and dispersion engineering. Chip-based self-referencing of modelocked lasers has so far only been demonstrated with PPLN waveguides using pulse energies of 600 pJ from an erbium-doped fiber laser [25], and 7 nJ from of a thulium-doped fiber laser system [23].

Silicon waveguides have the advantage to provide very high nonlinearities and thus reduce the pulse energy requirements. SCG is however only possible with pump wavelengths longer than $\approx 1.1 \, \text{µm}$ (absorption edge). Furthermore, silicon-based devices allow for low-cost large-scale production using established complementary metal-oxide semiconductor (CMOS) fabrication infrastructure. Most recently, a mid-infrared comb (1.5 µm to 3.3 µm) has been achieved from a silicon nanophotonic wire with only 16 pJ of coupled pulse energy and its phase coherence was verified by beat note measurements with several narrow line-width sources [35].

Here we present the first $f_\text{CEO}$ detection of a modelocked laser based on a CMOS-compatible platform using $\text{Si}_3\text{N}_4$ waveguides for highly efficient broadband coherent SCG pumped in the 1-µm regime. We show CEO-beat signals from both a 100-MHz DPSSL with a high signal-to-noise ratio of $> 25$ dB and a 1-GHz DPSSL with $> 30$ dB. Advances in the growth of thick low-loss $\text{Si}_3\text{N}_4$ layers (> 500 nm) of optical quality have enabled the fabrication of dispersion-engineered, low-loss waveguides [33, 34, 36], establishing $\text{Si}_3\text{N}_4$ as a promising platform for SCG. Due to its large energy bandgap, $\text{Si}_3\text{N}_4$ does not suffer from high nonlinear two-photon absorption (TPA) at 1-µm. The high refractive index contrast between $\text{Si}_3\text{N}_4$ (core) and silicon dioxide ($\text{SiO}_2$, cladding) allows for tight optical confinement within a waveguide structure. The nonlinear processes necessary for supercontinuum generation are supported by the high nonlinear index $n_2$ of $\text{Si}_3\text{N}_4$ (10 times higher than silica) [37, 38]. The coupled peak power required for octave-spanning SCG is lowered by more than an order of magnitude as compared to previously reported results based on commercially available PCF [20]. Besides the compactness of such waveguide devices and the reduced peak power requirement, we expect additional benefits from the adjustable waveguide structure to control dispersion and nonlinearity during pulse propagation. All this makes the approach very attractive for future frequency comb sources based on even more compact ultrafast semiconductor lasers in the multi-gigahertz regime [39].
2. Experimental setup

2.1 100-MHz and 1-GHz SESAM modelocked diode-pumped solid-state lasers (DPSSLs)

The performance of the Si$_3$N$_4$ waveguides is demonstrated here using two ultrafast SESAM modelocked DPSSLs. The lasers are based on Yb:CaGdAlO$_4$ (Yb:CALGO) laser gain materials pumped with a multi-transverse-mode laser diodes at a wavelength of 980 nm with emission around 1060 nm. A first series of measurements was done using a 100-MHz pulse repetition rate in order to determine the peak power levels required for octave-spanning SCG while staying at moderate average powers. We then successfully continued with a 1-GHz pulse repetition rate, consequently increasing the average power sent into the waveguides by a factor of 10. By comparing the SC generated in both cases and observing the stability over several hours of continuous operation, we rule out any significant thermal or other average-power related degradation of the waveguide devices. The detailed properties of both lasers are listed in Table 1.

Table 1. Properties of the diode-pumped solid-state lasers (DPSSLs) used for SCG in a Si$_3$N$_4$ waveguide

<table>
<thead>
<tr>
<th>Laser</th>
<th>Megahertz DPSSL</th>
<th>Gigahertz DPSSL</th>
</tr>
</thead>
<tbody>
<tr>
<td>Repetition rate</td>
<td>99.1 MHz</td>
<td>1.025 GHz</td>
</tr>
<tr>
<td>Gain medium</td>
<td>3-mm long Yb:CALGO</td>
<td>2-mm long Yb:CALGO</td>
</tr>
<tr>
<td></td>
<td>BMU25A-975-01-R03,</td>
<td>LIMO 60-F200-DL980-LM,</td>
</tr>
<tr>
<td></td>
<td>Oclaro</td>
<td>Lissotschenko Mikrooptik GmbH</td>
</tr>
<tr>
<td>Pump</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pump wavelength</td>
<td>980 nm</td>
<td>980 nm</td>
</tr>
<tr>
<td>Laser output power</td>
<td>&lt; 1.2 W</td>
<td>&lt; 1.7 W</td>
</tr>
<tr>
<td>Total output coupling rate</td>
<td>1.4%</td>
<td>2%</td>
</tr>
<tr>
<td>Pulse durations</td>
<td>65 – 96 fs</td>
<td>&gt; 63 fs (92 fs after isolator)</td>
</tr>
<tr>
<td>Center wavelength</td>
<td>1062 nm</td>
<td>1055 nm</td>
</tr>
<tr>
<td>SESAM</td>
<td>Single AlAs–embedded InGaAs</td>
<td>Single AlAs–embedded InGaAs</td>
</tr>
<tr>
<td></td>
<td>quantum well + SiO$_2$ coating</td>
<td>quantum well (uncoated)</td>
</tr>
<tr>
<td>saturation fluence</td>
<td>$\approx 5.8 \mu$J/cm$^2$</td>
<td>$\approx 11 \mu$J/cm$^2$</td>
</tr>
<tr>
<td>modulation depth</td>
<td>$\approx 2.8%$</td>
<td>$\approx 1.4%$</td>
</tr>
</tbody>
</table>

2.2 The Si$_3$N$_4$ waveguides for supercontinuum generation (SCG)

Our SCG platform consists of Si$_3$N$_4$ waveguides embedded in SiO$_2$. The Si$_3$N$_4$ is first deposited as a film onto an oxide-clad silicon wafer. The waveguide structure is patterned using electron beam lithography and etched with reactive ion etching. A second layer of SiO$_2$ is then deposited as a top cladding. The waveguide used for the $f_{CEO}$-detection is 7.5 mm long, with a cross section of 690 × 900 nm, and is spiraled within a 1 mm by 1 mm square that corresponds to the maximum beam lithography tool field. The radius of the bends is kept greater than 100 $\mu$m to minimize additional dispersion effects. The waveguide is tapered to facilitate coupling into the fundamental spatial mode and is designed to end 3 $\mu$m before the facet of the SiO$_2$-clad chip. Light is thus first coupled into the lower-index SiO$_2$ cladding before entering the Si$_3$N$_4$ waveguide, which reduces the coupling losses caused by Fresnel
The propagation losses were measured to be 0.7 dB/cm at a wavelength of 1.55 µm. The dispersion profile is engineered to have two “zero group velocity dispersion wavelengths” (ZDWs), thereby providing a 200 nm window of anomalous group velocity dispersion (GVD) around the pump wavelength (see Fig.1). The effective nonlinearity $\gamma$ is 3.25 $W^{-1} m^{-1}$ at 1055 nm.

Fig. 1. Simulated dispersion profile of the 690 nm by 900 nm waveguide. The waveguide provides anomalous group velocity dispersion (GVD) around the 1060-nm pump wavelength and has two “zero group velocity dispersion wavelengths” (ZDWs).

2.3 Experimental setup

A complete overview of the experimental setup is shown in Fig. 2. In both laser cavities the output coupler is used in a folding-mirror configuration with two output beams. While one beam was coupled into the waveguide for SCG, the other one was used for further diagnostics.

In case of the 100-MHz laser, we used a dispersion-free variable power attenuation stage consisting of a half-wave plate and a silicon window used in reflection under Brewster angle, enabling tuning of the waveguide input power without altering any other pulse parameters. Before focusing into the waveguide, the size of the collimated beam is adjusted using a telescope and the polarization is rotated with a half-wave plate to match the TE-mode of the waveguide. No optical isolator was needed for the MHz laser, since the back reflections from the waveguide facet did not cause any perturbation.

For the 1-GHz laser the average power incident on the waveguide is a factor of 10 higher to achieve comparable pulse energies. Thus a Faraday isolator was required to block the back reflections from the waveguide. Combined with a half-wave plate, the isolator also acts as a power attenuation stage. The positive group delay dispersion from the isolator is compensated with a transmission grating pair in a double-pass configuration.

In order to detect the CEO frequency of our lasers, the waveguide output is collimated and sent into a quasi-common path f-to-2f interferometer. The spectral portion around 1360 nm is frequency doubled in a periodically poled lithium niobate (PPLN) crystal and then collinearly recombined in space and time with the 680-nm components of the supercontinuum. After passing through a bandpass filter, the beat signals are detected using a gigahertz avalanche photodiode (APD) and displayed on a microwave spectrum analyzer (MSA).
Fig. 2. Overview of the setup: The output of the 100-MHz pulse repetition rate laser (top left) is attenuated to the desired level using a half-wave plate and a Si window under Brewster angle. When using the 1-GHz laser (bottom left), the power is adjusted by rotating the half-wave plate in front of the Faraday isolator. We use a grating pair to compensate for the group delay dispersion in the isolator. After expansion in a telescope and polarization adjustment, the beam is focused into the Si$_3$N$_4$ waveguide (top right). The microscope image shows the chip containing several waveguides. The spiraled structure of one waveguide is re-drawn on top for better visibility. The CEO frequency is detected using an f-to-2f interferometer (bottom right). The dichroic mirror splits the 680-nm and 1360-nm to allow for a variable time delay. Frequency doubling at 1360 nm is then achieved in the PPLN crystal and the SHG signal is collinearly overlapped with the 680-parts of the supercontinuum. After a narrowband optical filter, the beat signals are detected using an avalanche photodiode (APD) and displayed on an microwave spectrum analyzer (MSA).

The $f_{\text{CEO}}$ detection of the 100-MHz laser is performed with 79-fs pulses at a coupled average power of 3 mW (24 mW incident power on the waveguide) and with 92-fs pulses at 37 mW (237 mW incident) for the 1-GHz laser. The characterization of the sech$^2$-shaped input pulses including a second-harmonic intensity autocorrelation, the optical spectrum and a microwave spectrum for both lasers are shown in Fig. 3 and Fig. 4, respectively.
3. Supercontinuum generation (SCG)

The experimentally observed evolution of the SC with increasing pulse energy is depicted in Fig. 5(a). The coupled pulse energies range from 0.6 pJ to 36 pJ, corresponding to average laser powers from 4 mW to 237 mW of our 1-GHz laser taking into account a coupling efficiency of 15%. The pulses launched into the waveguide at a repetition rate of 1.025 GHz have a duration of 92 fs (after isolator and gratings) and a spectral full-width-half-maximum (FWHM) of 17 nm (see Fig. 4). The supercontinuum extends over almost the full range of the optical spectrum analyzer (OSA) Ando AQ-6315A (specified for 350 – 1750 nm), spanning more than 1.5 octaves (600 nm – 1750 nm).

Figure 5(b) shows the simulated spectra obtained using a split-step Fourier method to solve the generalized nonlinear Schrödinger (GNLS) equation including contributions from third-order nonlinearity, higher-order dispersion, and self-steepening. The dispersion profile (Fig. 1) used for the SC simulations was calculated with a finite element mode solver. The broadening of the SC is in good agreement with the simulations and the spectral parts used for f-to-2f interferometry are correctly predicted. The best agreement is obtained by re-calibrating the absolute simulated input powers by a multiplication factor of 0.85 compared to the power levels in the experimental case. The slight discrepancy between actual and simulated coupled pulse energies to achieve the same spectral broadening may be due to small amounts of power being coupled to higher-order waveguide modes during propagation.
Fig. 5. Supercontinuum (SC): (a) Experimentally observed spectra at different coupled pulse energies, plotted with 20 dB-offsets. The grey shaded area marks the wavelength region where the optical spectrum analyzer (OSA) ANDO AQ-6315A records second-order diffracted light (first-order for > 600 nm) which can generate additional spurious signals not really present in the actual SC. (b) Simulated spectra with the absolute energies re-scaled by a factor of 0.85 compared to the experimental values.

Fig. 6. “Spurious” and “real” frequency components: (a) Spectrum of the gigahertz laser output (Fig. 4b) without spectral broadening in the waveguide recorded with the OSA ANDO AQ-6315A at a RBW of 5nm. Internal filtering imperfections of the OSA can lead to first-order diffracted light of longer wavelengths being recorded again in the short wavelength region (<600 nm), giving rise to a spurious signal peak at half the input wavelength. (b) Photo of the supercontinuum obtained with 36 pJ of coupled pulse energy using a diffraction grating with 1250 lines/mm. Frequency components ranging from green to red are clearly visible by eye, indicating that a “real” spectral content is also present between 500-600 nm.

Additionally, we recorded features on our spectrum analyzer in the visible range from 350 nm to 600 nm that are not reproduced by our theoretical model. The OSA Ando AQ-6315A uses second-order diffracted light to detect wavelengths below 600 nm and first-order diffraction for > 600 nm. Spurious signals can thus occur if first-order diffracted long wavelength components are internally not perfectly filtered for the short-wavelength scan (or vice versa). The peak visible already at low pulse energies at exactly half (529 nm) the pump wavelength peak (1058 nm) is such a spurious signal (Fig. 5(a)), further confirmed with the
spurious signal shown in Fig. 6(a). We however verified the existence of real spectral components in the 500–600 nm range of the supercontinuum obtained with 36 pJ using the angular dispersion of a simple diffraction grating alone (Fig. 6(b)). Components extending from green to red were clearly visible by eye. Even though the bulk $\chi^{(2)}$ vanishes due to the centrosymmetry of Si$_3$N$_4$, a second-order response can arise at the interface of the Si$_3$N$_4$ core with the SiO$_2$ cladding, as demonstrated previously in the case of Si$_3$N$_4$ ring resonators [41]. As a consequence, phase matching between the fundamental and higher order waveguide modes can lead to the generation of second or even higher order harmonic components and thus lead to features not predicted by the purely Kerr-effect-based simulations. Coupling to higher order waveguide modes can also lead to slight changes in the effective dispersion profile experienced by the propagating pulse and thus induces a spectral shift of dispersive-wave peaks, which has been shown using algorithms based on pulse propagation in a waveguide with full space- and time-resolution [42]. The higher-order mode effects are not included in our model, as such simulations are several orders of magnitude more computationally expensive compared to solving the GNLS for the fundamental mode, which in most cases already provides very good predictions.

4. Frequency comb offset detection

Using the setup described above, we were able to detect the frequency comb offset, given by the carrier envelope offset (CEO) frequency, of both SESAM-modelocked diode-pumped Yb:CALGO lasers and thus prove that the SC generated in the Si$_3$N$_4$ waveguide is highly coherent. In Fig. 7 and 8, we show the detected $f_{\text{CEO}}$ beat signals and the corresponding SC spectra that were used for $f$-to-$2f$ interferometry. The depicted data was obtained with 30 pJ (36 pJ) of coupled pulse energy, corresponding to 337 W (345 W) of peak power for the 100-MHz (1-GHz) laser.

![Fig. 7. 100-MHz result: (a) Supercontinuum (SC) obtained with 3.3 mW of coupled average power at 99.1 MHz. The spectral components used for the $f$-to-$2f$ interferometry are marked in red. (b) Microwave spectrum showing the pulse repetition rate $f_{\text{rep}}$ and the CEO beat frequencies $f_{\text{CEO,1}}$ and $f_{\text{CEO,2}}$ at 14 MHz and 86 MHz, respectively.](image-url)
Although the average power was increased by an order of magnitude in the case of the 1-GHz laser to obtain the required peak power, the performance of the waveguide was not affected in any noticeable way, as can be concluded from the similarity of the spectra for 100-MHz and 1-GHz case and the fact that no thermal or material degradation could be observed over several days. A signal-to-noise ratio (SNR) of >25 dB with a FWHM of ≈0.2 MHz was obtained for the CEO beat of the 100-MHz laser and >30 dB with a FWHM of ≈2 MHz for the 1-GHz laser. By tuning the laser pump current of the 100-MHz laser to sweep the pulse duration over the whole single-pulse modelocking range from 65 fs (shortest pulses before onset of double-pulsing) to 96 fs (modelocking threshold), we were able to shift the position of CEO beats over a range of 20 MHz, i.e. \( f_{\text{rep}}/5 \). The SNR of the CEO beats is sufficient for additional microwave signal processing, i.e. it will allow us to perform further characterization of the CEO noise properties in the near future. Furthermore, the fact that the CEO beat center frequency can be shifted over a substantial range when adjusting the laser pump power is an essential requirement for ultimately stabilizing the CEO frequency to an external radio-frequency reference by providing feedback to the current of the pump diode.

Table 2. Parameters required for CEO beat detection with a SNR of 30 dB for the same 1-GHz laser using either a 7.5-mm long Si\(_3\)N\(_4\) waveguide or 1 m of the commercial PCF NL-3.2-945 from NKT Photonics.

<table>
<thead>
<tr>
<th>SCG platform</th>
<th>Input</th>
<th>Output</th>
<th>Coupling efficiency</th>
<th>Pulse duration</th>
<th>Coupled peak power</th>
<th>Coupled pulse energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si(_3)N(_4) waveguide</td>
<td>237 mW</td>
<td>37 mW</td>
<td>15 %</td>
<td>92 fs</td>
<td>0.34 kW</td>
<td>36 pJ</td>
</tr>
<tr>
<td></td>
<td>500 mW</td>
<td>400 mW</td>
<td>80 %</td>
<td>63 fs</td>
<td>5.45 kW</td>
<td>390 pJ</td>
</tr>
</tbody>
</table>

In Table 2, we compare the CEO beat detection of our 1-GHz laser using SCG in the Si\(_3\)N\(_4\) waveguide to SCG in a 1-m-long commercial PCF (NL-3.2-945, NKT Photonics). Using the same f-to-2f interferometer as in [20], 16 times less coupled peak power is needed to obtain a comparable SNR (>25 dB). This important peak power reduction is a consequence of the more than 140 times higher nonlinear coefficient provided in our Si\(_3\)N\(_4\) waveguide (3.25 W\(^{-1}\)m\(^{-1}\)) compared to the PCF (0.023 W\(^{-1}\)m\(^{-1}\)). A similar FWHM of the CEO beat (≈2 MHz) is obtained in both cases, indicating that the linewidth of the CEO frequency is not primarily affected by the choice of the supercontinuum platform. The coupling efficiency is
defined herein as the ratio of the average power measured free-space directly before and after the waveguide or PCF, respectively. The efficiency of the SCG process in Si$_3$N$_4$ can be further increased, as the coupling losses of the waveguides are not fundamental and can be improved by optimization of the taper design and coupling procedure.

5. Conclusion

We have presented the first CEO detection of a modelocked laser based on SCG in a Si$_3$N$_4$ waveguide. Strong CEO beat signals (> 25 dB) were obtained for both a 100-MHz and a 1-GHz repetition rate SESAM modelocked diode-pumped Yb:CALGO laser using a standard $f$-to-$2f$ interferometer. More than an order of magnitude less coupled peak power is needed to generate an octave-spanning coherent SC from a 1-GHz laser by using a Si$_3$N$_4$ waveguide instead of a commercial PCF to achieve a comparable SNR for the same 1-GHz laser.

CMOS-compatibility, compactness and tailored dispersion engineering are convincing assets that Si$_3$N$_4$ offers as a platform for self-referencing of modelocked lasers in the 1-µm range. In particular, the low peak power requirement represents an important advantage when going to longer pulses or multi-gigahertz repetition rates and may enable all-semiconductor frequency comb generation directly from compact semiconductor disk lasers such as VECSELs [43, 44] or MIXSELs [45] in the near future.

Acknowledgments

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2.4 Frequency comb offset stabilization

Modelocked lasers represent a highly dynamical system, where the system parameters are strongly coupled to each other via different physical mechanisms. The CEO offset frequency is one of the parameters with the highest sensitivity to these couplings. Hence, measuring the properties of the CEO signal generally reveals a lot about the underlying processes happening in the laser cavity. Furthermore, detailed knowledge about the behavior of the CEO frequency is a pre-requisite for its control and thus ultimately the stabilization of the frequency comb.

2.4.1 Noise in modelocked lasers

“Noise” is a very generic term describing a plethora of perturbations that can affect a laser system. We can classify noise either by its origin, i.e. mechanical/electrical/spontaneous quantum noise etc., or by the impact it has on the system, i.e. how a modelocked pulse train is affected by it.

![Diagram of interactions between noise sources and laser parameters](image)

Figure 2.2: Interactions between potential noise sources (red) and the laser parameters. While the parameters in blue can usually not directly be accessed, the parameters in green represent measurable quantities. Figure inspired from [76] and [77].
In Figure 2.2, we provide an exemplary overview of how the different modelocked laser parameters can be linked and where potential noise sources may enter the system.

The topic of noise in modelocked lasers has been thoroughly discussed in the literature and we refer to those publications for rigorous mathematical derivations [77-79]. Here, we aim to provide a short summary of the types of noise that can be measured and usually belong to the full characterization of a modelocked laser/frequency comb.

2.4.2 The ideal pulse train

In the time domain, we can describe a pulse train as the convolution of the pulse envelope and a train of delta functions (see section 1.2). In the case of the output of a femtosecond modelocked laser, the duration of the pulses is much shorter than the time span between the pulses, i.e. the cavity roundtrip time \(T_R\). Hence, we can approximate an ideal noise-free pulse train \(I(t)\) using a sum of delta functions, where \(P_{av}\) represents the average optical power of each “infinitely short” pulse (Figure 2.3):

\[
I(t) = P_{av} \cdot T_R \cdot \sum_{n=-\infty}^{\infty} \delta(t - nT_R)
\]  

(2.12)

In the frequency domain, we can derive a power spectral density (PSD) that reads

\[
S(f) = P_{av}^2 \cdot \sum_{n=-\infty}^{\infty} \delta(f - nf_{rep}),
\]  

(2.13)

where \(f_{rep} = 1/T_R\) corresponds to the pulse repetition rate.

Figure 2.3: Schematic representation of an ideal pulse train in the time and frequency domain.
2.4.3 Timing jitter/Phase noise of the repetition frequency

Due to the various noise mechanisms depicted above, the arrival time of the pulses can suffer from random delays (timing jitter) $\Delta T_R(t)$ with respect to their ideal arrival time $nT_R$. Adding these fluctuations to Eq. (2.12), we obtain the following expression for a pulse train perturbed by timing jitter:

$$I_{\Delta T_R}(t) = P_{av} T_R \cdot \sum_{n=-\infty}^{\infty} \delta(t - nT_R - \Delta T_R(t))$$

(2.14)

Assuming that those arrival fluctuations $\Delta T_R(t)$ are still small compared to the time $T_R$ between the pulses and using the Fourier shifting theorem, the phase noise PSD can be obtained as

$$S_{\Delta T_R}(f) = P_{av}^2 \sum_{n=-\infty}^{\infty} \left[ \delta(f - nf_{rep}) + (n \cdot 2\pi f_{rep} \cdot \Delta \tilde{T}(f - nf_{rep}))^2 \right],$$

(2.15)

where $\Delta T(f)$ denotes the Fourier transform of $\Delta T_R(t)$. Figure 2.4 depicts schematically how the time- and frequency signals are affected.

Figure 2.4: Schematic representation of a pulse train with timing jitter: in the time domain, the pulses fluctuate randomly around the ideal arrival time. In the frequency domain, the timing jitter leads to sidebands that increase proportionally to $n^2$.

2.4.4 Amplitude noise

Not only the arrival time of the pulses can fluctuate, but also their amplitude. Assuming random normalized power fluctuation $N(t)$, Eq. (2.12) extends to

$$I_{N(t)}(t) = P_{av} T_R (1 + N(t)) \cdot \sum_{n=-\infty}^{\infty} \delta(t - nT_R).$$

(2.16)
Similar than for the timing jitter, we can determine a PSD according to the following expression

\[ S_{N(t)}(f) = P_{av}^2 \cdot \sum_{n=-\infty}^{\infty} \left[ \delta(f - nf_{rep}) + (\tilde{N}(f - nf_{rep}))^2 \right], \tag{2.17} \]

where \( \tilde{N}(f) \) represents the Fourier transform of \( N(t) \).

![Figure 2.5: Schematic representation of a pulse train with amplitude noise: in the time domain, the pulses fluctuate randomly around the ideal optical power level. In the frequency domain, this amplitude noise leads to sidebands that are identical for all the harmonics of the repetition frequency.](image)

Both amplitude and timing jitter lead to side-bands on the spectral power signal in the frequency domain. Note however that the amplitude noise is independent of the harmonic number \( n \), whereas the phase noise scales as \( n^2 \). Any realistic modelocked laser will always exhibit a mixture of both noise types.

### 2.4.5 Measuring amplitude and phase noise

For the noise measurements carried out within the framework of this thesis, we employed a signal source analyzer (SSA) (Agilent E5052B) capable of performing phase- and amplitude noise measurements separately.

The SSA measures the phase and amplitude noise of an input carrier frequency \( f_c \) (e.g. \( f_{rep} \) or \( f_{CEO} \) in our case) using the phase lock loop (PLL) method. In this method, a reference source (internal low-noise oscillator of the SSA) is locked onto the input signal, i.e. the internal oscillator follows the frequency \( f_c \). Both the reference signal and the input signal are fed into a double balanced mixer (also referred to as the phase detector). The output of the mixer then consists of the sum and difference frequency components. The sum frequency \( 2f_c \) is filtered out by a low-pass filter. The difference frequency is nominally around DC (i.e. 0 Hz) with an average voltage output of 0 V, but the voltage output contains AC fluctuations proportional to the noise of the two mixer input signal. Assuming that the noise contribution of the
reference oscillator signal signal is negligible, the phase and amplitude noise of the input signal $f_c$ can determined in the following way:

**Amplitude noise:** To extract the amplitude noise, the reference signal follows the input signal in phase (or out of phase by 180°). In that case, the phase fluctuations are suppressed and the mixer output signal is dominated by the amplitude fluctuations.

**Phase noise:** In order to obtain the phase fluctuations, the reference signal is set to be in quadrature (i.e. 90° out of phase) with respect to the input signal and the signal levels are chosen in order to saturate the mixer. In this way, the voltage output of the saturated mixer is a signal that is directly proportional to the phase fluctuations, whereas the amplitude fluctuations are suppressed:

$$V_{out}(t) = K \cdot (\Delta \varphi_{\text{input}}(t) - \Delta \varphi_{\text{ref}}(t)) = K \cdot \Delta \varphi_{\text{input}}(t) \quad (2.18)$$

Here, $K$ is the phase detector constant in V/rad and we assumed a low-noise reference oscillator, i.e. $\Delta \varphi_{\text{input}}(t) \gg \Delta \varphi_{\text{ref}}(t)$. The PSD of these voltage fluctuations can be determined as $S_V(f) = |\mathcal{F}\{V_{out}(t)\}|^2$. Normalization by $K^2$ then leads to the power PSD of the phase noise $S_\varphi(f)$ in units of rad$^2$/Hz. Note that the Agilent SSA instrument (like many other common analyzers) outputs the data in form of the single sideband (SSB) phase noise $L_{\text{dBc}}(f)$ in dBC/Hz. The following relations hold:

$$L_\varphi(f) = 10 \frac{L_{\text{dBc}}(f)}{10}$$

$$S_\varphi(f) = 2 \cdot L_\varphi(f) \quad (2.19)$$

By integrating the PSD, we can calculate the integrated phase noise according to

$$\sigma_{f_{\text{low}}, f_{\text{high}}} = \sqrt{\int_{f_{\text{low}}}^{f_{\text{high}}} S_\varphi(f) df}. \quad (2.20)$$

When measuring the phase noise of the pulse repetition frequency $f_{\text{rep}}$, it is common to normalize this value by the frequency of the harmonic $n$ at which the power spectral was determined, which then leads to the so-called root-mean-square (rms) timing jitter $\sigma_T$:
\[ \sigma_T[f_{\text{low}}, f_{\text{high}}] = \frac{1}{2\pi n f_{\text{rep}}} \sqrt{\int_{f_{\text{low}}}^{f_{\text{high}}} S_\phi(f) df} \] (2.21)

### 2.4.6 Frequency noise and the \( \beta \)-separation line

When characterizing the noise of frequency combs, it is also often common to calculate the frequency noise PSD (units: Hz\(^2\)/Hz), which is related to the phase noise PSD described above by

\[ S_f(f) = f^2 S_\phi(f) \] (2.22)

where \( f \) denotes the noise frequency.

Di Domenico et al. [80] have introduced the useful concept of the \( \beta \)-separation line, defined as

\[ S_{f,\beta}(f) = 8 \ln(2) f / \pi^2. \] (2.23)

This line separates the frequency noise spectrum into two regions, which represent different contributions to lineshape of the carrier frequency \( f_c \): only the spectral components for which the frequency noise is higher than the \( \beta \)-separation line contribute to the linewidth. Frequency noise in the area below the line only affects the wings of the lineshape. This fact can be nicely illustrated when measuring the noise of stabilized signals: if the stabilization loop manages to push the residual frequency noise entirely below the \( \beta \)-separation line, the microwave spectrum analyzer trace will reveal a coherent peak. This situation is then often referred to as a “tight lock”. The linewidth of this delta-function-like coherent peak is only limited by the resolution bandwidth of the analyzer. Examples of such measurements are presented in Paper 2, where we achieved a tight lock of the CEO frequency of a 1 GHz frequency comb.

### 2.4.7 Phase noise of the CEO frequency

In section 2.1, we have discussed the detection of the carrier-envelope offset (CEO) frequency. Before entering the details of how to stabilize this essential comb parameter, it is worth remembering its physical origin: inside the laser cavity, the dispersion \( n(\omega, x) \) (material dispersion, mirror coatings etc.) leads to a discrepancy between the phase velocity \( v_p \) of the center frequency \( \omega_0 \) and the group velocity \( v_g \).
of the pulse envelope. By integrating over the cavity round-trip length $L$, we obtain the accumulated group-phase offset

$$\Delta \phi_{GPO} = \omega_0 \int_0^L \left( v_g^{-1} - v_p^{-1} \right) dx = \int_0^L \frac{\omega_0^2}{c} \frac{dn(\omega, x)}{d\omega} dx. \quad (2.24)$$

The carrier-envelope offset phase is then defined as

$$\Delta \phi_{CEO} = \Delta \phi_{GPO} \mod(2\pi). \quad (2.25)$$

Taking the time derivative leads us to the following expression for the CEO frequency:

$$f_{CEO} = \frac{1}{2\pi f_{rep}} \Delta \phi_{CEO} \quad (2.26)$$

Any fluctuation of the intracavity dispersion will thus have an impact on the noise PSD of the CEO frequency. Since these fluctuations are being multiplied by $f_{rep}$, the CEO frequency of a high-repetition rate laser will exhibit broader sidebands for the same amount phase fluctuation than a laser operating at a lower pulse repetition rate.
2.4.8 Signal stabilization – basic considerations

Before designing a stabilization procedure for any frequency signal, it is useful to examine the following points:

**Noise of the signal:** How “noisy” is the signal, i.e. what type of fluctuations (amplitude, frequency) occur and how strong are they? Does the noise show any characteristic frequencies that need to be taken care of?

**Origin of the noise:** What are the possible sources of noise and at which level do the perturbations enter the system?

**Passive stabilization:** Can any of the noise sources be eliminated or can their impact at least be mitigated without the need for an active feedback loop?

**Active stabilization, choice of the actuator:** Which actuator(s) can be used to provide feedback to the system?

**Active stabilization, performance of the actuator:** How “strong” is the feedback mechanism (i.e. what is the maximum modulation that can be achieved)? How “fast” can feedback be provided (i.e. what is the maximum bandwidth)?

**Active stabilization, response of the system:** How does the system respond to this actuator (transfer function)?

**Active stabilization, choice of the reference:** Which source can provide a suitable reference signal? Does the residual noise of the reference itself represent a limiting factor for the intended level of stabilization?

Following this reasoning scheme, we have established a procedure to obtain a tight CEO frequency lock of our 1 GHz Yb:CALGO laser. The results are presented in Paper 2.
2.5 Introductory remarks to Paper 2

One of the main goals of this thesis was not only to detect the CEO frequency using Si₃N₄ waveguides, but also to show that the CEO frequency could be tightly locked. Since we were the first ones to use Si₃N₄ as a platform for self-referencing a frequency comb, we wanted to first investigate the potential influence of this new type of supercontinuum platform on the CEO detection compared to using commercially available photonic crystal fibers. These two platforms mainly differ with respect to their nonlinearity (Si₃N₄ offers a nonlinear parameter $\gamma$ that is >100 times higher than the most nonlinear fibers available at 1 $\mu$m), and the significance of the Raman effect for spectral broadening. While one often relies on Raman scattering to shift the spectral components towards longer wavelengths in silica fibers, the influence of Raman for SCG in Si₃N₄ is negligible. In the first part of Paper 2, we discuss numerical simulations that aimed at investigating whether this may have an influence on the coherence of the supercontinuum and hence the CEO detection.

The second part of Paper 2 is dedicated to the stabilization of the CEO frequency. In the following, we will briefly discuss some of the technical details related to the results presented in the article.

2.5.1 Passive mechanical stability

The 1-GHz Yb:CALGO laser cavity used for the results presented here has been optimized for high passive mechanical stability. To reduce mechanical vibrations and resonances, all mirror are mounted on short rigid posts and fixed in holders without any translational or tip/tilt degree of freedom. The only exception is the SESAM, which acts as the alignment end mirror and thus is kept in place by a mount providing tip/tilt (Figure 2.6). The Yb:CALGO crystal is temperature-stabilized to 17°C using a Peltier-element, which is connected to a water-cooled copper heat sink. In order to avoid turbulence-induced vibrations, we reduced the water flow to the minimum necessary for proper operation of the Peltier element. The original cavity design and laser performance was published by Klenner et al. [81] and represents the groundwork for the experiments presented in this thesis. The passive stability of the cavity was evaluated by measuring the phase noise of the repetition rate signal. The extracted timing jitter for the free-running laser amounted to 16 ps for an integration range of 1 Hz to 100 kHz (198 fs for 1 Hz to 100 kHz) [81].
2.5.2 Spatially multimode pump with/without VHG

The publication by Klenner et al. [81] also contains the first demonstration of the stabilization of the CEO from a diode-pumped GHz solid-state laser. The CEO detection was performed using SCG in a PCF and an integrated residual phase noise of the stabilized CEO signal of 744 mrad [1 Hz, 5 MHz] was achieved.

For the work published by Klenner et al. as well as for Paper 1 in this thesis, the Yb:CALGO crystal was pumped using a spatially multimode fiber-coupled pump diode (LIMO 60-F200-DL980-LM, Lissotschenko Mikrooptik, fiber core diameter 100 µm, NA=0.22) that did not feature internal wavelength stabilization. In the meantime, the same pump diode became available with an integrated volume holographic grating (VHG). The latter stabilizes and narrows the optical output spectrum as shown in Figure 2.7(a). Within the framework of our CEO stabilization experiments, we thus also evaluate the effect of this VHG wavelength stabilization on the performance of the laser and in particular the properties of the CEO signal. The relative intensity noise (RIN) of both the VHG and non-VHG diode is shown in Figure 2.7(b) and reveals a 20 dB-drop of the noise level for the VHG stabilized version. To ensure a meaningful comparison, we thus repeated the CEO detection experiment using a PCF in parallel to the detection experiment using the Si₃N₄ waveguide and found the following: while the choice of the platform did not influence the CEO-linewidth in a measurable way, the pump wavelength stabilization reduced the linewidth by an order of magnitude (Figure 2.7(c)). This
drastic improvement may be a result of several different factors. For one thing, any noise of the CEO that originates from the transfer of pump RIN to the CEO will be reduced when lowering the RIN. Additionally, it is worth noting that the main absorption peak around 980 nm in Yb:CALGO is rather sharp, i.e. the absorption cross-section drops by a factor of ~2 when deviating from the peak by ±3 nm. Hence, a fluctuation of the pump wavelength can easily be translated into a fluctuation of the absorbed pump power. Lastly, although the VHG did not lead to an increase of the pump brightness in terms of the $M^2$-value, it may help to stabilize the freckle-pattern in the beam profile and hence reduce noise related to spatially varying pump intensities in Yb:CALGO crystal.

Figure 2.7: Effect of a wavelength-stabilized pump diode. (a) Optical spectrum of the VHG-stabilized diode. (b) Relative intensity noise measurements revealing a considerable drop of the noise level in the 100 Hz to 1 MHz range.
2.5.3 Feedback electronics

In this section, we will describe the phase-locked loop (PLL) used for the CEO stabilization results presented in Paper 2. A PLL generally consists of 4 main components: the voltage controllable oscillator (VCO), the reference signal, the phase detector and the loop filter. These components are highlighted in the setup overview (Figure 2.8) and described in more detail below.

**Voltage controllable oscillator (VCO):** In our case, the CEO frequency represents the quantity that can be controlled by applying a correction signal to the current of the pump diode. Since the signal processing is done on a voltage signal, the correction signal is converted in a voltage-to-current (V-I) converter before being directly applied to the +/- poles of the pump diode.
**Reference signal:** An analog signal generator (E8257D Agilent PSG) provides the low-noise reference signal to which the CEO frequency is phase-locked.

**Phase detector:** The phase detector is the element that compares the CEO signal to the reference. In this setup, we use a digital phase detector (Menlo DXD200), which can handle a maximum phase error of $\pm 32 \times 2\pi$. Analog phase detectors such as double balanced mixers (DBM, see section 2.4.5) have a maximum phase error of $\pm \pi/2$ (but become nonlinear at much smaller values already). The PLL will unlock when the maximum phase error is reached. Hence, for strongly fluctuating signals such as the CEO frequency of a GHz laser, the ability to capture the signal over a larger phase error range is a strong requirement.

**Loop filter:** The loop filter treats the error signal generated by the phase detector in order to convert it into an appropriate correction signal for the VCO. Most commonly, a proportional-integral-derivative (PID) controller is used to transform the error signal $e(t)$ into a correction signal $u(t)$ according to

$$u(t) = K_p e(t) + K_i \int_0^t e(t')dt' + K_d \frac{de(t)}{dt}.$$  \hfill (2.27)

Using the Laplace transform defined as $F(s) = \int_0^\infty f(t)e^{-st}dt$, we obtain the transfer function $U(s)$:

$$U(s) = G(s)E(s) = K_p \left(1 + \frac{1}{s \omega_i} + \frac{1}{s \omega_D}\right)E(s),$$  \hfill (2.28)

where $\omega_i = K_i / K_p$ and $\omega_D = K_p / K_D$ represent the integral and derivative corner frequencies. Figure 2.9 qualitatively shows the function $G(s)$ for controller used in our experiments (VESCENT Laser Servo D2-125). As a particularity, this controller offers a second integrator. Thus, two different integral corner frequencies can be chosen ($\omega_i$ and $\omega_{pi}$), which turned out to be useful in order to refine the feedback signal and improve the noise suppression in particular in the 10 Hz - 1 kHz region. For the stabilization results presented in Paper 2, we used $\omega_i = 500$ Hz, $\omega_{pi} = 1$ kHz, $\omega_D = 100$ kHz.
In addition to the 4 main components of the PLL, we used an additional RC-filter to damp the noise of the voltage line entering the system through the pump diode power supply. The filter consist of a 0.22-Ohm-resistor and 24 capacitors with a capacitance of $C = 4.7 \, \mu F$ each.

Figure 2.9: Gain curve of the VESCENT D2-125 PFD controller featuring a second integrator (adapted from [82]).
Paper 2:

Gigahertz frequency comb offset stabilization based on supercontinuum generation in silicon nitride waveguides

Authors: Aline S. Mayer* (setup and measurements, simulations)  
Alexander Klenner* (1 GHz laser design, simulations)  
Adrea R. Johnson (waveguide design)  
Kevin Luke (waveguide fabrication)  
Michael R. E. Lamont (waveguide simulation code)  
Yoshitomo Okawachi (supervision)  
Michal Lipson (supervision)  
Alexander L. Gaeta (supervision)  
Ursula Keller (supervision)

*equally contributing first authors

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Note: The Si₃N₄ waveguide used in Paper 1 and 2 is the same. In Paper 1, we published the dispersion curve as originally designed. The fabrication tolerances can easily lead to discrepancies between the designed and actual dispersion of the waveguide. In Paper 2, we used the experimentally obtained SC spectrum in order to retrieve a more accurate fit for the actual dispersion curve.
Gigahertz frequency comb offset stabilization based on supercontinuum generation in silicon nitride waveguides

Alexander Klenner,1,2,† Aline S. Mayer,1† Adrea R. Johnson,2,3 Kevin Luke,4 Michael R. E. Lamont,3 Yoshitomo Okawachi,2 Michal Lipson,5 Alexander L. Gaeta,2 and Ursula Keller1

1 Department of Physics, Institute of Quantum Electronics, ETH Zurich, 8093 Zurich, Switzerland
2 Department of Applied Physics and Applied Mathematics, Columbia University, New York, NY 10027, USA
3 School of Applied and Engineering Physics, Cornell University, Ithaca, NY 14853, USA
4 School of Electrical and Computer Engineering, Cornell University, Ithaca, NY 14853, USA
5 Department of Electrical Engineering, Columbia University, New York, NY 10027, USA

† These authors contributed equally to this work.

Abstract: Silicon nitride (Si3N4) waveguides represent a novel photonic platform that is ideally suited for energy efficient and ultrabroadband nonlinear interactions from the visible to the mid-infrared. Chip-based supercontinuum generation in Si3N4 offers a path towards a fully-integrated and highly compact comb source for sensing and time-and-frequency metrology applications. We demonstrate the first successful frequency comb offset stabilization that uses a Si3N4 waveguide for octave-spanning supercontinuum generation, and achieve the lowest integrated residual phase noise of any diode-pumped gigahertz laser comb to date. In addition, we perform a direct comparison to a standard silica photonic crystal fiber (PCF) using the same ultrafast solid-state laser oscillator operating at 1 µm. We identify the minimal role of Raman scattering in Si3N4 as a key benefit that allows to overcome the fundamental limitations of silica fibers set by Raman-induced self-frequency shift.

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References and links


### 1. Introduction

Optical frequency combs consist of ultrafine optical lines that are equally spaced in the frequency domain with an uncertainty down to the $10^{-19}$ level [1]. Over the past 15 years, the generation and stabilization of such combs [2-4] has enabled control of the cycles of light, which represents a major achievement for laser research and high precision optical metrology [5, 6]. The generated comb can be fully stabilized by phase-locking both the spacing of the comb lines and the comb offset, which is also known as the carrier envelope offset (CEO). While stabilizing the comb line spacing of a frequency comb generated by a modelocked laser requires the stabilization of the pulse repetition rate, i.e. by simply acting on the position of a cavity mirror, the detection and stabilization of the CEO is more challenging.
Self-referenced comb offset stabilization is one of the preferred schemes, as it allows for phase locking without a highly-stable external optical reference. For the most common self-referencing technique based on \( f\)-to-\( 2f \) interferometry, optical comb spectra spanning at least one octave are required [2]. Such ultrabroadband spectra are typically generated through the process of supercontinuum generation (SCG). Traditionally, SCG is done at MHz pulse repetition rates by propagating a modelocked femtosecond laser pulse through a highly nonlinear silica fiber. Such systems are designed to be embedded in specialized laboratories for experiments that require operation at medium to high pulse energies. However, the size and cost of these setups currently prevents wide-spread comb applications outside the laboratories. For many applications in spectroscopy and time-and-frequency metrology, low-energy frequency combs in the 1 to 10 GHz regime are highly desirable [7-10]. Such gigahertz combs typically generate lower peak intensities and therefore would greatly benefit from more efficient SCG. The strength of the nonlinear interactions leading to spectral broadening of a pulse propagating inside a waveguide is determined by the nonlinear phase shift \( \gamma P L \), where \( P \) is the power and \( L \) is the waveguide length. The nonlinear parameter \( \gamma \) is defined as

\[
\gamma = \frac{2\pi n_2}{A_{\text{eff}} \lambda}
\]

where \( n_2 \) is the material nonlinear index, \( \lambda \) is the vacuum wavelength, and \( A_{\text{eff}} \) is the effective area of the mode propagating inside the waveguide. For a given length \( L \), i.e. fixed accumulated dispersion, lowering the pulse energy required to obtain an octave-spanning coherent spectrum is thus directly related to increasing the value of \( \gamma \). An obvious first approach to obtaining a high value of \( \gamma \) is thus to choose a waveguide core material with a high \( n_2 \). Fig. 1(a) gives an overview of the material platforms that have been used to demonstrate octave-spanning SCG. \( \text{Si}_3\text{N}_4 \) sticks out as a highly promising material for nonlinear optics and frequency comb generation since it offers a high nonlinear index of \( 2.4 \times 10^{-15} \text{ cm}^2/\text{W} \) and a broad transparency range from visible wavelengths to the mid-IR [11].

Ultrafast diode-pumped solid-state laser oscillators [12] have most recently produced an ultrabroadband supercontinuum (SC) using chip-based silicon nitride (\( \text{Si}_3\text{N}_4 \)) photonic waveguides [13, 14]. Multi-octave-spanning coherent spectra have been obtained with an order of magnitude less pulse energy as a direct result of the higher nonlinearity provided by these silicon-based photonics platform [15] compared to silica fibers. In comparison to materials with similar nonlinear indices, such as lithium niobate, lead-bismuth-silicate and tellurite glasses, \( \text{Si}_3\text{N}_4 \) has the additional benefit of being compatible with the complementary metal-oxide-semiconductor (CMOS) fabrication techniques, which are widely used in industry. Compact and robust \( \text{Si}_3\text{N}_4 \) waveguides can be produced at low costs on wafer scale using standard deposition and lithography methods. Simple dispersion engineering by adjusting the geometrical dimensions of the waveguide allows for designing dispersion profiles with more than one zero-dispersion-wavelengths (ZDW’s) to achieve highly coherent octave-spanning spectra.
Fig. 1. (a) Overview of materials and their nonlinear indices $n_2$ which have been used to generate octave-spanning spectra [11, 17-32], (b) Exemplary representation of the influence of mode-confinement on the nonlinear parameter. The optimal physical core area that yields the maximal $\gamma$ as a function of wavelength was determined using the relation described in Foster et al. [33] for 3 different platforms. The $\gamma$ values divided by the respective material nonlinearity $n_2$ of the actual oxide-clad $\text{Si}_3\text{N}_4$ and the silica PCF used for the experimental results presented in this paper are marked with stars.

Foster et al. [33] provides design guidelines for a maximum value of $\gamma$ as a function of the wavelength and the linear refractive indices of core and cladding. The best result is achieved for the smallest possible core that still enables confinement of the fundamental mode without a significant amount of power being guided in the low-nonlinearity cladding. Fig. 1(b) shows the influence of linear core/cladding index contrast on the nonlinear parameter $\gamma$ as a function of wavelength for 3 exemplary cases: The extreme case of a silica PCF, i.e. corresponding to a silica nanotaper core fully surrounded by air, a $\text{Si}_3\text{N}_4$ with a silica cladding, and an air-clad $\text{Si}_3\text{N}_4$-nanotaper. The optimal core size and the resulting maximum $\gamma' = \gamma/n_2$ value are depicted assuming a Gaussian-like fundamental beam profile, for which the effective area is $A_{\text{eff}} = \pi w^2$ with $w$ denoting the mode radius. The relation between the physical core radius and the radius of the mode is estimated using the Marcuse formula [34]. For comparison, the normalized values $\gamma'$ of the $\text{Si}_3\text{N}_4$/silica waveguide presented in this paper and the photonic crystal fiber (PCF) NL-3.2-945 (NKT Photonics) are included. It can be seen that for the $\text{Si}_3\text{N}_4$ waveguide, the value is close to the theoretically optimum, whereas for the commercial PCF it lies an order of magnitude below a silica nanotaper. This comparatively low nonlinearity is a result of an air-hole patterned cladding which is far from providing the index contrast one would obtain in the case of a fragile, rod-type silica nanotaper. Thus the 140-times higher nonlinear parameter $\gamma$ of the $\text{Si}_3\text{N}_4$-waveguide presented in this paper compared to the PCF (3.25 W$^{-1}$m$^{-1}$ vs. 0.023 W$^{-1}$m$^{-1}$) originates from an order of magnitude difference in both the nonlinear material index $n_2$ and the mode confinement.

In this paper we demonstrate the first self-referenced frequency comb offset stabilization based on SCG in $\text{Si}_3\text{N}_4$ waveguides and show that this platform is particularly suitable for low-energy and low-noise stabilized frequency comb generation. We use a SESAM-modelocked [35] diode-pumped Yb:CALGO [36] solid-state laser with a center wavelength of $\sim 1$ $\mu$m [37, 38] and achieve the lowest integrated residual phase noise of any diode-pumped gigahertz laser comb to date.
Moreover, we numerically investigate the coherence of supercontinua generated in Si$_3$N$_4$ compared to silica fibers. Standard photonic crystal fibers (PCF’s) for wavelengths > 1 µm often rely on the Raman effect to provide sufficient spectral broadening. We show that the coherence of the Raman self-frequency shifted spectral components rapidly degrades with increasing nonlinearity $\gamma$, which prevents coherent SCG and thus CEO beat note detection at low pulse energies. The Raman effect in Si$_3$N$_4$ waveguides is regarded to be negligible (see supplementary part of [39]). The lack of significant Raman gain in single-pass Si$_3$N$_4$ waveguides represents a clear advantage over silica fibers for highly efficient frequency comb generation, since it allows for generating coherent supercontinua with higher nonlinearity and lower energies.

2. Supercontinuum generation in Si$_3$N$_4$ waveguides versus silica PCF

The Si$_3$N$_4$ waveguide used in our experiments is designed to provide anomalous group velocity dispersion surrounding the 1-µm pump wavelength (Figs. 2(a)-(e)). The dispersion of the Si$_3$N$_4$ waveguide is customized by engineering the contribution of the waveguide dispersion using a waveguide cross section of 690 × 880 nm. For our short pump pulses (<100 fs), coherent SCG occurs through higher-order soliton compression and the emission of short- and long-wavelength dispersive waves (DW’s) [40, 41]. The waveguide length is chosen to be 7.5 mm to cut off propagation after DW emission to allow for the broadest spectral bandwidth and to avoid any degradation of coherence. The spectral evolution with propagation distance for this waveguide is shown in Fig. 3(a) for a coupled pulse energy of 26 pJ, with an input pulse centered at 1055 nm, FWHM bandwidth of 18 nm, and a slight positive chirp (800 fs$^2$), corresponding to the experimental parameters provided by our gigahertz diode-pumped solid-state laser oscillator.

For comparison, an example of SCG in a commercially available PCF (NKT NL3.2-945, one ZDW at 945 nm) is shown in Fig. 3(b), using the same pulse duration and chirp as mentioned above, however with 377 pJ of coupled pulse energy. In contrast to the SC generated in the Si$_3$N$_4$ waveguide, the spectrum at the soliton fission point is not yet suitable for f-to-2f interferometry. A fiber length of 1 m is necessary until the ejected soliton has shifted to sufficiently long wavelengths via the Raman self-frequency shift and shorter wavelengths are accessed via cross phase modulation (XPM) of the shifted soliton and the DW. For the experimental input parameters mentioned so far, the first-order coherence is close to unity over the whole spectral bandwidth for both platforms (Figs. 5(c) and (d)). However, in order to obtain the same spectral span with as little energy as in the case of the Si$_3$N$_4$ waveguide, either the fiber length or the nonlinear parameter of the silica fiber would have to be increased accordingly (i.e. via tapering of the cross section).

In the following, we will show that increasing the nonlinear phase shift in a silica fiber results in a deterioration of the coherence due to the Raman effect. For this purpose, we numerically solve the general nonlinear Schrödinger equation (GNLSE) [40, 42] for a scan of nonlinear parameters $\gamma' = K \times \gamma$ and pulse energies $E_p' = E_p / K$, where $K$ is increased from 1-40 in steps of 4. We keep the dispersion profile unaltered and maintain a constant soliton order (i.e. $N = 3$) by decreasing the input power accordingly, so that the spectral width and shape of the supercontinuum remains the same.
Fig. 2. Si$_3$N$_4$ photonic waveguide chip. (a) A Si$_3$N$_4$ waveguide with a rectangular cross-section is sandwiched in a SiO$_2$ layer on the buried oxide of a Si substrate. Optimized light coupling with comparably low losses of < 8 dB is achieved using inverse tapers of 100-µm length and SiO$_2$ gaps of 3 µm between taper and facet at the input and output.[43] (b) The photograph shows a typical Si$_3$N$_4$ chip that incorporates 7 waveguides of various lengths. An SEM image of the rectangular cross-section is shown in (c). (d) The fundamental guided mode which is guided in the waveguide was simulated with a finite element solver. The waveguide bending radii are kept >100 µm to avoid additional dispersion effects. The resulting dispersion curve featuring two zero-dispersion wavelengths (ZDW’s) is shown in (e).

The GNLSE is solved with a split-step Fourier-transform algorithm [42]. The nonlinear step is done with a second-order Runge-Kutta method as published in Dudley et al. [40]. Both stimulated Raman as well as spontaneous Raman effects are included. The latter are thermally induced and depend on the Bose-Einstein distribution of vibrational modes in the material at a given temperature. The effect is independent of the input power, but scales with the nonlinearity as $\sqrt{\gamma}$.

We observe a clear degradation of the coherence of the generated SC, which is particularly pronounced for the spectral components around the Raman-shifted soliton and even stronger for its XPM wave at short wavelength (Fig. 3(c)). The noise processes that may deteriorate the coherence can be divided into three main components; technical laser noise, input shot noise and spontaneous Raman scattering. While the first one is discarded in these simulations, the input shot noise is accounted for with one photon per mode. The spontaneous Raman process describes photon-phonon scattering at the silica crystal. In Fig. 3(d) the coherence values at the soliton (1360 nm) and the XPM wave (680 nm) is depicted for different scaling factors $K$. A scaling factor of $K = 40$ leads to a drop in coherence for the PCF below 80% at 1360 nm (green area) and 60% at 680 nm (purple area). Essentially the same coherence degradation is observed when the shot noise is turned off and only spontaneous Raman effects are taken into account (green and purple circles). In the case of pure input shot
noise (green and purple crosses), i.e. spontaneous Raman scattering switched off, only a slight influence is noticeable.

This coherence degradation becomes even more pronounced for higher soliton orders (i.e. \( N = 7 \)). It becomes dominant whenever the Raman self-frequency shift has a large contribution to the SCG process in a highly nonlinear silica fiber. Especially in the wavelength range from 1-1.5 \( \mu \)m, most silica PCF’s provide a single ZDW and the Raman self-frequency shift is needed to extend the spectrum to longer wavelengths. A platform that does not exhibit significant Raman gain, as it is the case for \( \text{Si}_3\text{N}_4 \), represents a clear advantage for, achieving highly coherent supercontinua even at very low energies. In general, the highest coherence is usually achieved close to the soliton fission point. Designing the platform such as to provide an octave-spanning spectrum at the fission point thus ultimately optimizes the quality of the CEO beat note detected by \( f\)-to-\(2f\) interferometry.

![Fig. 3. Spectral evolution of the SC and the loss of coherence due to the Raman effect. (a) The SC in \( \text{Si}_3\text{N}_4 \) is generated within 7.5 mm by dispersive wave (DW) emission at long and short wavelengths. (b) The photonic crystal fiber (PCF) initially generates a DW at short wavelengths. During the 1-m propagation length a Raman-induced soliton self-frequency shift and cross-phase modulation (XPM) between soliton and DW generates the required bandwidth. (c) A potentially high nonlinearity in the PCF (high scaling factor \( K \)) causes a significant degradation of the coherence of the frequency-shifted soliton and XPM wave. (d) The coherence for the PCF drops below 80% at 1360 nm (green area) and 60% at 680 nm (purple area) for \( K = 40 \). The coherence loss is mainly caused by the spontaneous Raman response in the PCF. Simulations with Raman but without shot noise are shown as purple and green circles. Simulations with shot noise only (green crosses: 1360 nm; purple crosses: 680 nm) show much less degradation.](image-url)
3. Self-referenced frequency comb offset detection

The ultrafast laser used for comparing the performance of both SCG platforms is a SESAM-modelocked diode-pumped Yb:CALGO [36] laser that generates pulses with 64-fs duration at a pulse repetition rate of 1.025 GHz and can provide an average output power as high as 1.8 W at a center wavelength of 1055 nm described in more details in [37]. Without any additional pulse amplification or nonlinear compression, the beam is either coupled into the silica PCF or the Si$_3$N$_4$ waveguide described in the previous section (Fig. 4).

In order to detect the carrier-envelope-offset frequency ($f_{\text{CEO}}$) of our laser, we use a Michelson-type $f$-to-$2f$ interferometer, where light around 1360 nm is doubled in a periodically poled lithium niobate (PPLN) crystal and overlapped in space and time with the spectral components at 680 nm. The generated beat signal is recorded by an avalanche photodiode (APD). The CEO beat notes and corresponding optical spectra obtained with 36 pJ (377 pJ) of coupled pulse energy in the Si$_3$N$_4$ waveguide (silica PCF), are shown in Figs. 5(a)-(d). Our numerical simulations are in very good agreement with the experimental spectra and show that the SC of both platforms are highly coherent. We note that the Si$_3$N$_4$-based spectrum spans about 500 nm more bandwidth and features less amplitude modulation especially in the spectral parts used for the CEO beat note detection. The beat note detection for a self-referenced stabilization requires a high degree of coherence but also sufficient temporal overlap of both spectral components.

While the absolute signal-to-noise ratio (SNR) of the CEO beats also depends on factors such as the amount of light sent onto the APD and the resolution bandwidth of the microwave spectrum analyzer, the difference in signal-to-signal ratio between the CEO beat signals and the repetition rate signal mainly originates from the spectral shape and coherence of the SC spectra, i.e. the distribution of optical power and noise over the wavelength range of interest, and thus is an inherent property of the SCG platform. Balanced optical power levels in both interferometer arms result in the highest signal-to-signal ratio. Although the pulse repetition rate signal is filtered out in the CEO-stabilization feedback loop, maximizing this signal-to-signal ratio also has an implication on the achievable SNR of the CEO signal as it allows for
reaching the same SNR (of the CEO signal) with less optical power on the APD and thus avoiding a higher noise floor and saturation effects.

An important improvement of the free-running CEO signal was achieved by pumping the modelocked laser with a laser-diode that has an integrated volume holographic grating (VHG). The grating stabilizes and reduces the spectral bandwidth of the spatially multimode pump diode (Lissotschenko Mikrooptik GmbH). The passive pump wavelength stabilization reduces the linewidth of the free-running $f_{\text{CEO}}$ by one order of magnitude (Fig. 6).

![Graphs showing frequency comb offset detection and stabilization](image)

**Fig. 5.** Detection of the comb offset frequency ($f_{\text{CEO}}$) of a 1.025-GHz SESAM modelocked diode-pumped solid-state laser using either (a) a Si$_3$N$_4$ waveguide or (b) a photonic crystal fiber (PCF) under the same laser operating conditions (input pulse spectrum centered at 1055 nm, FWHM 18 nm) and using the identical $f$-to-$2f$ interferometer with coupled pulse energies of 36 pJ and 377 pJ, respectively. SC spectrum generated in (c) Si$_3$N$_4$ and (d) the PCF. Both Si$_3$N$_4$ and PCF platforms provide fully coherent octave-spanning SC, however the Si$_3$N$_4$ waveguide provides 500 nm more bandwidth than the PCF. The SC generated in Si$_3$N$_4$ features much less amplitude modulation, which is highly desirable for applications. Simulations are shown in black, while colors denote experimental results. The indicated spectral portions at 1360 nm and 680 nm are used for $f$-to-$2f$ detection. Although both platforms provide similar CEO signal-to-noise ratios, the signal contrast to the repetition rate at 1.025 GHz is improved by 15 dB for Si$_3$N$_4$. RBW: resolution bandwidth, OSA: optical spectrum analyzer, FTIR: Fourier transform infrared spectrometer.
4. Carrier envelope offset stabilization of a compact low-noise frequency comb

We use the generated strong CEO beat signal to perform the first frequency comb offset stabilization based on SCG in Si$_3$N$_4$. The linewidth of the free-running CEO scales with the pulse repetition rate: Any jitter of the phase slip per cavity roundtrip, $\Delta \varphi_{\text{CEO}}(t)$, is multiplied by the repetition rate to yield the CEO frequency according to the relation $f_{\text{CEO}}(t) = (\Delta \varphi_{\text{CEO}}(t) / 2\pi) \times f_{\text{rep}}$ [2, 45]. As a result, the stabilization of combs at gigahertz repetition rate is more challenging. The VHG-stabilized pump laser significantly lowers the required bandwidth of the feedback electronics compared to previous results [37]. The comb-offset of the gigahertz source laser was phase-locked to an RF reference source (Agilent PSG Analog Signal Generator E8257D) using an analogue feedback loop acting on the current of the pump diode. The feedback electronics consist of a phase detector (Menlo DXD200), whose output is filtered by a PID-controller (Vescent D2-125) and subsequently converted into a current signal using a homebuilt Voltage-to-Current (V-I) converter. The freerunning well as the locked CEO beat note at 73.9 MHz are shown in Fig. 7(a). In Fig. 7(b), the 200-Hz-span zoom at 1-Hz resolution bandwith (microwave spectrum analyzer limit) shows the strong coherent peak with minor power line noise at 50 Hz.
Fig. 7. Stabilized comb offset frequency ($f_{CEO}$) based on SC generated in Si$_3$N$_4$ waveguide. (a) Free-running (blue) and stabilized $f_{CEO}$ showing a strong coherent peak at the reference frequency of 73.9 MHz (red) recorded with a microwave spectrum analyzer (MSA). Stabilization feedback loop resonances are visible at around 150 kHz. (b) Zoom into coherent peak at 1 Hz resolution bandwidth shows only minor line-noise at 50 Hz. (c) The frequency noise power spectral density (PSD) of the stabilized comb offset frequency measured with a signal source analyzer (Agilent E5052B) drops integrally below the $\beta$-separation line which is a necessary requirement for a tight phase lock [46]. (d) The integrated residual phase noise (PN) is 304 mrad [1 Hz, 5 MHz].

The noise analysis of the phase-locked comb-offset frequency reveals a drastic drop of the in-loop CEO frequency noise upon onset of the feedback loop. The residual frequency noise power spectral density (PSD) lies integrally below the $\beta$-separation line (Fig. 7(c)), which is a necessary requirement for a tight phase-lock [46]. The integrated phase noise amounts to 304 mrad [1 Hz, 5 MHz] (Fig. 7(d)). To our knowledge, this is the lowest reported value from any gigahertz diode-pumped laser comb to date. This low value is achieved also thanks to the use of pump wavelength stabilization. We have repeated the locking experiment with the CEO beat note based on the PCF without a noticeable difference in the noise performance. As reported in Klenner et al. [37], a more sophisticated high-bandwidth analogue electronic feedback loop, which acts on the current of the multimode pump diode, is also capable of stabilizing $f_{CEO}$-beat notes with a larger FWHM and a lower SNR (i.e. as obtained without pump wavelength stabilization).

5. Conclusion

We have shown the first self-referenced comb offset stabilization of a modelocked laser based on SCG in Si$_3$N$_4$ waveguides. The stabilized SESAM modelocked laser provides, to the best of our knowledge, the lowest integrated residual phase noise of any diode-pumped gigahertz laser comb to date. The combination of highly coherent SCG in the Si$_3$N$_4$ waveguide and passive wavelength stabilization of the diode-laser pump leads to this superior performance.
The energy requirement for octave-spanning SCG is reduced by more than an order of magnitude compared to a commercial silica PCF. Moreover, the core/cladding composition of Si$_3$N$_4$ and SiO$_2$ allows for high confinement while preserving the possibility to engineer the nonlinear parameter $\gamma$ as well as the dispersion profile for various laser sources without the need for fragile suspended nanotapers.

We show that the absence of a significant Raman gain is a key benefit for low-noise SCG at low energies with a spectrum of the SC which is also exhibits much less amplitude modulations. The compatibility of Si$_3$N$_4$ waveguides with CMOS technology opens new avenues for stabilized compact frequency combs. We can envision an ultra-compact chip-device for integrating all three necessary stages for self-referencing: SCG, $f$-to-$2f$ interferometer with detection and control electronics. Such a chip-device would greatly reduce the cost and complexity and increase the robustness of stabilized optical frequency combs and further promote their use outside of the laboratory environment.

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Chapter 3

Extending the wavelength range

In the previous chapters, we have presented the advantages of Yb:CALGO as a gain material to obtain decent power and short pulses from a directly diode pumped laser. However, the center wavelength of Yb-doped materials (around ~1 µm) limits the application of this laser in the field of atmospheric gas spectroscopy. With the exception of a few molecules, such as e.g. acetylene [83, 84], most molecules that are interesting for their impact on the environment have their characteristic absorption lines at wavelengths > 3 µm, i.e. the so-called mid-infrared (mid-IR) region [85, 86]. We thus set ourselves the challenge to transfer our 1-GHz comb at 1 µm into the mid-infrared by making use of waveguide nonlinear optics to avoid the need for additional pulse amplification stages. In the following, we provide a brief introduction to the principle of optical parametric amplification (OPA), which was the technique we used to extend the wavelength range of our 1-GHz frequency comb into the mid-IR.

3.1 Optical parametric amplification (OPA)

Optical parametric amplification is a nonlinear process that originates from the second-order material response (see section 1.1). The process involves the interaction of three waves: the pump with frequency ω_p, the signal at ω_s and the idler at ω_i, which fulfill the photon energy conservation law

\[ \hbar \omega_p = \hbar \omega_s + \hbar \omega_i. \]  \hspace{1cm} (3.1)

The phase mismatch between the wave vectors of these waves can be expressed as

\[ \Delta \vec{k} = \vec{k}_p - \vec{k}_s - \vec{k}_i. \]  \hspace{1cm} (3.2)
In case the interaction takes place in a quasi-phase matched (QPM) medium (e.g. periodically poled lithium niobate (PPLN) in Paper 3), the grating vector $K_g$ term has to be taken into account, which for all-collinear propagation leads to the scalar expression

$$\Delta k = k_p - k_s - k_i - K_g,$$

where $K_g = \frac{2\pi}{\Lambda_g}$ ($\Lambda_g$: poling period). 

$$\Delta k = k_p - k_s - k_i - K_g,$$ \hspace{1cm} (3.3)

Assuming cw plane waves, we can write down coupled wave equations for the pump, signal and idler fields $E_p, E_s$ and $E_i$ propagating in $z$-direction

$$\frac{dE_p}{dz} = -i\kappa_p E_s e^{i\Delta k z},$$
$$\frac{dE_s}{dz} = -i\kappa_s E_p^* e^{-i\Delta k z},$$
$$\frac{dE_i}{dz} = -i\kappa_i E_p E_s^* e^{-i\Delta k z},$$ \hspace{1cm} (3.4)

where $\kappa_j = 2\pi d_{eff}/(n_j \lambda_j)$ and $d_{eff} = \frac{1}{2} \chi^{(2)}$. Usually, the pump is much stronger than the signal and the idler fields. Hence, in particular at the beginning of the interaction process, an undepleted pump can be assumed, i.e. $\frac{dE_p}{dz} = 0$. If the idler field is initially zero and the interaction is phase-matched (i.e. $\Delta k = 0$), we obtain the following expressions for the signal and idler fields after a propagation distance $L$,
Extending the wavelength range

\[ E_i(L) = -i E_i^*(0) \frac{\kappa E_s}{\Gamma} \sinh(\Gamma L) \]
\[ E_s(L) = E_s(0) \cosh(\Gamma L) \]

where \( \Gamma = \sqrt{\kappa \kappa_s |E_p|^2} \) denotes the gain coefficient. The single-pass gain \( G_s(L) \) for the signal field can be calculated as

\[ G_s(L) = \frac{I_s(L) - I_s(0)}{I_s(0)} = \sinh^2(\Gamma L), \]

where with the intensities being defined as \( I_j = \frac{1}{2} n_j c \epsilon_0 |E_j|^2 \).
3.2 Introductory remarks to Paper 3

In Paper 3, we show how starting from a laser frequency comb centered around 1 µm with a comb line spacing of 1 GHz, we can obtain a mid-IR frequency comb in an energy-efficient way by leveraging the tailorable nonlinear properties of chip-scale Si$_3$N$_4$ and PPLN waveguides. The Si$_3$N$_4$ waveguide is the same as used for the results described in Paper 1 and 2.

3.2.1 PPLN chip

The PPLN waveguide chip was fabricated by C. Langrock at Standford University using reverse proton exchange (RPE) [87, 88]. The chip is 25 mm long, 6 mm wide, 0.5 mm high and contains 90 waveguides. On the input side, the waveguides have a tapered region to facilitate coupling into a single spatial mode. The waveguides are periodically poled over a length of 18.5 mm. A section of the chip is shown in Figure 3.2. The 90 waveguides have poling periods ranging from 17 to 30 µm. For the OPA experiments presented in Paper 3 the ones with poling periods between 24.60 and 26.49 µm were used.

![Microscope picture of a section of the PPLN waveguide chip](image)

Figure 3.2: Microscope picture of a section of the PPLN waveguide chip. The actual waveguides are not visible, but their location is indicated by the black/white stripes representing the periodically poled areas.

3.2.2 Waveguide dispersion properties for simulations

In order to simulate the propagation of the pump, signal and idler pulses inside the waveguide, we need to know its dispersion properties. When dealing with pulses
instead of continuous waves, the group index $n_g$ has to be derived from the refractive index $n$ according to

$$n_g(\lambda) = \frac{c}{v_g} = c \frac{\partial k_n}{\partial \omega} = \frac{\partial}{\partial \omega} \left( \omega n(\omega) \right) = n(\omega) + \omega \frac{\partial n}{\partial \omega} = \frac{\partial \left( \frac{2\pi n(\lambda)}{\lambda} \right)}{\partial \left( \frac{2\pi}{\lambda} \omega \right)}.$$ (3.7)

When propagating inside a waveguide instead of bulk, the actual dispersion is expressed by the effective quantities $n_{\text{eff}}$ and $n_{\text{g,eff}}$, which are not purely material properties anymore, but also contain the effect of the spatial mode confinement, i.e. the waveguide dispersion. As will be described in Paper 3, the change in refractive index as a function of the wavelength and transverse position within the waveguide was obtained from proton diffusion simulations as reported by Roussev [89]. While this leads to reasonable values for IR wavelengths, the mid-IR properties are less known. Hence, we used our experimental data set to extrapolate the effective index for the mid-IR part of the spectrum (wavelengths $> 2 \mu m$).

**Fixed propagation simulation inputs**

- Waveguide grating vector $K_{\text{wg}}$ for each waveguide (from poling period design)
- Pump wavelength $\lambda_p$ (fixed by laser center wavelength)
- Signal wavelengths $\lambda_s$ (experimentally determined for each waveguide)

**Calculated quantities**

- Idler wavelengths $\lambda_i = \left( \lambda_p^{-1} - \lambda_s^{-1} \right)^{-1}$ (assures phase-matching)
- Effective waveguide index for the idler wavelengths

$$n_{\text{i,eff}} = \lambda_i \left( \frac{n(\lambda_p)}{\lambda_p} - \frac{n(\lambda_i)}{\lambda_i} - \frac{1}{K_{\text{wg}}} \right)$$

- Effective waveguide group index for the idler wavelengths

$$n_{\text{g,eff}} = c \cdot \frac{\partial \left( \frac{2\pi}{\lambda} n_{\text{i,eff}} \right)}{\partial \left( \frac{2\pi}{\lambda} \right)}$$
Paper 3:

Offset-Free Gigahertz Midinfrared Frequency Comb Based on Optical Parametric Amplification in a Periodically Poled Lithium Niobate Waveguide

Authors: Aline S. Mayer (experimental setup, measurements and simulations)
Christopher R. Phillips (simulations)
Carsten Langrock (PPLN waveguide fabrication)
Alexander Klenner (1 GHz laser design)
Adrea R. Johnson (Si$_3$N$_4$ waveguide design)
Kevin Luke (Si$_3$N$_4$ waveguide fabrication)
Yoshitomo Okawachi (supervision)
Michal Lipson (supervision)
Alexander L. Gaeta (supervision)
Martin M. Fejer (supervision)
Ursula Keller (supervision)

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We report the generation of an optical-frequency comb in the midinfrared region with 1-GHz comb-line spacing and no offset with respect to absolute-zero frequency. This comb is tunable from 2.5 to 4.2 µm and covers a critical spectral region for important environmental and industrial applications, such as molecular spectroscopy of trace gases. We obtain such a comb using a highly efficient frequency conversion of a near-infrared frequency comb. The latter is based on a compact diode-pumped semiconductor saturable absorber mirror–mode-locked ytterbium-doped calcium-aluminum gadolynate (Yb:CALGO) laser operating at 1 µm. The frequency-conversion process is based on optical parametric amplification (OPA) in a periodically poled lithium niobate (PPLN) chip containing buried waveguides fabricated by reverse proton exchange. The laser with a repetition rate of 1 GHz is the only active element of the system. It provides the pump pulses for the OPA process as well as seed photons in the range of 1.4–1.8 µm via supercontinuum generation in a silicon-nitride (Si$_3$N$_4$) waveguide. Both the PPLN and Si$_3$N$_4$ waveguides represent particularly suitable platforms for low-energy nonlinear interactions; they allow for mid-IR comb powers per comb line at the microwatt level and signal amplification levels up to 35 dB, with 2 orders of magnitude less pulse energy than reported in OPA systems using bulk devices. Based on numerical simulations, we explain how high amplification can be achieved at low energy using the interplay between mode confinement and a favorable group-velocity mismatch configuration where the mid-IR pulse moves at the same velocity as the pump.

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I. INTRODUCTION

The midinfrared spectral region between 2 and 20 µm covers the strong vibrational transitions of a variety of molecules that play an important role in environmental, medical, and industrial diagnostics. The ability to detect and quantify the presence of such molecules or to investigate their properties on a more fundamental level is thus directly linked to the availability of a light source capable of probing these transitions. Laser-frequency combs - i.e., lasers whose spectra consist of a series of equally spaced discrete optical lines - combine three essential assets: the high brightness of the light leads to a high detection sensitivity, the narrow linewidth of the individual comb lines allows for high-resolution measurements, while the large spectral bandwidth enables fast simultaneous detection of multiple species.

The success of optical-frequency combs in the near-infrared region has been strongly tied to the advancement of mode-locked lasers in that wavelength range [1–5]. Well-established gain media include Ti:sapphire [6] emitting around 800 nm, and various host crystals doped with ytterbium (Yb) or erbium (Er) emitting in the 1- and 1.5-µm regions, respectively [7–9]. Various approaches have recently been pursued to extend the spectral coverage of frequency combs into the midinfrared region [10]. Direct approaches include alternative laser gain materials for mode-locked solid-state and fiber lasers [11–13] or semiconductor devices such as quantum-cascade lasers [14,15]. Another approach relies on exploiting different aspects of nonlinear optics, such as supercontinuum generation (SCG) in fibers [16–18] and waveguides [19–21], or Kerr-comb generation in microresonators [22,23].

The challenge these approaches have in common is the difficulty to detect and control the comb offset frequency [24–26], i.e., the parameter that defines the exact position of the evenly spaced frequency-comb lines on the absolute-frequency axis. This problem can be circumvented by difference-frequency generation (DFG): in this nonlinear process, the low-frequency part of a comb (termed the “signal”) is mixed with the high-frequency components (the “pump”) of the same comb in a medium exhibiting a second-order (χ(2)) nonlinearity, resulting in a difference-frequency comb (the “idler”) which will be offset-free [27].

A configuration where the signal gets significantly amplified during this mixing process is known as an optical parametric amplifier (OPA). DFG- and OPA-based mid-IR frequency combs have already been demonstrated using bulk devices of various materials, such as periodically poled lithium niobate (PPLN) [28–32], GaSe [33], AgGaSe2 [34], CdSiP2 [35], and orientation-patterned GaAs [36]. Because of the limited interaction length caused by diffraction and material dispersion, single-pass bulk OPAs typically require watt-level pump beams and several hundreds of milliwatts of initial signal power to achieve powers per comb line >1 µW in the mid-IR region. Schemes based on high-power oscillators [37], laser preamplification of a pump and/or signal beam [38], or an intracavity OPA [39] have been demonstrated. Higher efficiencies in converting a near-IR frequency comb to the mid-IR region can be obtained in a parametric oscillator [40–45]. However, the passive comb-offset stability will be lost and the implementation of an active stabilization [46] is instead required to eliminate the offset. The development of stabilized mid-IR frequency combs therefore benefits from a robust and compact configuration that allows for efficient frequency conversion at low energies with passive comb-offset stabilization, using a single mode-locked laser oscillator as the only active medium. Here, we demonstrate chip-scale waveguide technology as a compact low-energy platform for generating widely tunable, offset-free mid-
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IR frequency combs. A diode-pumped solid-state laser operating at 1 µm with a repetition rate of 1 GHz serves as a single active source with two output beams. While one beam is directly used to pump an OPA process in a PPLN waveguide, the other is spectrally broadened in a silicon-nitride (Si$_3$N$_4$) waveguide to generate signal photons in the wavelength range of 1.4 – 1.8 µm. The idler can be tuned from 2.5 to 4.2 µm by laterally translating the PPLN chip across waveguides with varying quasi-phase-matching (QPM) periods [47]. With just 300 pJ of pump-pulse energy and initial seed powers of <20 nW per comb line, we achieve microwatt-level comb-line powers in the mid-IR region. Compared to systems based on nonlinear fibers for SCG and bulk PPLN for the OPA process [38], the required pulse energy for obtaining the same power per comb line is lowered by nearly 2 orders of magnitude. In the following, we provide the details of the experimental setup, present the mid-IR comb results, and explain via numerical simulations how the approach leverages favorable aspects of the waveguide dispersion in order to maximize the achievable OPA gain. Moreover, our simulations, which are in excellent agreement with our experimental results, show that waveguides offer the possibility to enter high-gain OPA regimes that are inaccessible to bulk devices at low energies. The ability to perform efficient supercontinuum-seeded frequency conversion at low pulse energies, as demonstrated and explained here, could enable multi-gigahertz comb-resolved sources based on chip-scale nonlinear-optical devices, directly driven by highly compact laser oscillators, thereby removing the need for laser amplifiers and bulk frequency converters in such systems.

II. EXPERIMENTAL SETUP

The passively mode-locked laser oscillator shown in Fig. 1(a) consists of a 2-mm-long ytterbium-doped calcium-aluminum gadolynate (Yb:CALGO) [48] emitting at 1053 nm and pumped at 980 nm using a spatially multi-mode pump diode. The laser is mode locked with a semiconductor saturable absorber mirror (SESAM) [49] and can produce pulses as short as 63 fs at a repetition rate of 1.025 GHz, with an output power of up to 1.7 W (when both output beams are combined) [50]. One of the output beams is coupled to a 7.5-mm-long Si$_3$N$_4$ waveguide (spiraled onto a square of 1×1 mm) with a cross section of 690 × 900 nm [Fig. 1(b)] [51–53]. A coupled pulse energy of 40 pJ (coupling efficiency 15%) is sufficient to obtain a supercontinuum spanning from 650 to 1800 nm, as shown in Fig. 2(a). Using a long-pass filter, the spectrum is cut at 1400 nm and sent into the PPLN waveguide as a seed for the OPA process.
FIG. 1. (a) Experimental setup showing the two output beams of the 1-GHz laser cavity. The negative second-order intracavity dispersion necessary to achieve soliton mode locking is provided by a Gires-Tournois-interferometer-(GTI-) type mirror. Isolators prevent potential back reflections from the waveguide facets into the laser. Grating pairs are used to compensate for the isolator dispersion and additionally stretch the pulse in the OPA pump arm. (b) Sketch of the (1 × 1)-mm chip with the 7.5-mm-long Si₃N₄ waveguide embedded in silicon dioxide (SiO₂). (c) Excerpt of the PPLN chip containing buried RPE waveguides in regions with different poling periods. The first 6.5 mm of the 2.5-cm-long chip are unpoled, and the waveguides are tapered to facilitate single-mode coupling.

The PPLN-waveguide chip with a dimension of 25 × 6 × 0.5 mm contains 90 waveguides fabricated by reverse proton exchange (RPE). The RPE method exhibits a first step of exchanging lithium ions with protons, using a diffusion process to create a region with a higher refractive index capable of guiding light. In order to obtain buried waveguides that support Gaussian modes and efficient nonlinear mixing, the protons near the surface are subsequently removed in a reverse-proton-exchange step. The RPE waveguides used here are fabricated with a 12 µm width and an exchange depth of 2.3 µm. This depth, which is larger than in typical PPLN waveguides designed for telecom applications [54], is chosen in order to guide the mid-IR wavelengths. At the input side of the waveguide, the width of the lithography-mask pattern is adiabatically tapered to 2 µm to allow for efficient and single-mode coupling of the input near-IR beams. The different waveguides are periodically poled, with poling periods ranging from 17 to 30 µm to achieve QPM.

Coupling into both waveguides as well as beam collimation at the output is performed in free space using antireflection-coated lenses. A Faraday isolator protects the laser cavity from potential back reflections from the waveguide facets. Grating pairs are used to compensate for the dispersion introduced by the isolators. Angled waveguide facets could be used in the future to eliminate the isolators. While the pulse at the input of the Si₃N₄ waveguide is
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FIG. 2. (a) Supercontinuum obtained with 40 pJ of coupled pulse energy in the Si$_3$N$_4$ waveguide. The shaded part is used as a signal input for the OPA. (b) Scan of the amplified signal spectra obtained by laterally scanning the chip across the different QPM periods, recorded with a grating-based optical spectrum analyzer (OSA). (c) Normalized mid-IR spectra recorded with a Fourier-transform infrared spectrometer (FTIR) ranging from 2.5 to 4.2 µm and corresponding to the amplified signal spectra shown in (b). The right axis displays the absolute power levels, with a maximum of 10 mW at 3.55 µm. The power drop near 3.45 µm is due to a slight defect in this particular waveguide, resulting in less overall transmitted power. (d) Enlargement of an FTIR trace recorded with instrument-limited resolution of 3.6 GHz using the free-space port after approximately 1 m of propagation in air (orange) and absorption cross sections of water (dark blue) and carbon dioxide (light blue) taken from the HITRAN database.

recompressed to a nearly-transform-limited 85 fs, the pulses in the pump arm are purposely stretched to nearly 800 fs to maximize the pump-signal interaction in the PPLN waveguide. The general advantage of pump-pulse stretching in a waveguide configuration is discussed in Sec. IV.

III. RESULTS

A. Amplification and mid-IR spectra

Amplified spectra, obtained by scanning through the waveguides with QPM periods from 24.60 to 26.49 µm, are shown in Fig. 2(b). With a maximum pump-average power of 310 mW coupled into the PPLN waveguides, we are able to amplify the spectral region from 1.4 to 1.8 µm obtained by SCG in the Si$_3$N$_4$ waveguide by up to 35 dB. The corresponding mid-IR idler spectra range from 2.5 to 4.2 µm, with an average power reaching 10 mW at 3.5 µm [Fig. 2(c)]. Given the comb-line spacing of 1.025 GHz set by the laser, this value corresponds to an average power per comb line of 4 µW. The DFG process leads to passive cancellation of the laser-comb offset; therefore, the stability of the mid-IR comb lines depends only on the stability of the laser repetition rate - and thus the laser-cavity length. Here, sufficient stability is achieved with low-drift mirror mounts and by boxing the setup. By mounting the SESAM on a piezoelectric actuator as described in Ref. [55], such ultrafast laser combs can be fully
stabilized with a long-term stabilization loop, and the comb lines can also be shifted by a desired amount.

The amplified signal spectra are recorded with a grating-based optical spectrum analyzer [(OSA), Ando AQ-6315A]. A Fourier-transform infrared spectrometer [(FTIR), Thorlabs OSA2015] is used for the idler spectra. The path length of approximately 1 m between the output of the PPLN waveguide and the free-space input of the FTIR analyzer is sufficient to observe distinctive absorption features in the ambient air. By magnifying the mid-IR comb generated in the waveguide with the QPM period of 26.35 µm [Fig. 2(d)], we can clearly identify the presence of water (H₂O) and carbon-dioxide (CO₂) absorption lines by comparing the spectrum recorded by the FTIR with the corresponding absorption cross sections provided by the high-resolution transmission molecular absorption (HITRAN) database.

B. Noise analysis

The relative intensity noise (RIN) of a frequency comb is an important parameter, as it can limit the achievable signal-to-noise ratio in spectroscopic applications such as dual-comb spectroscopy [56]. In an OPA-based system, the RIN can increase during pre-amplification of the pump and/or signal, nonlinear broadening steps, and the OPA process itself. RIN characterization at each stage of the setup thus helps us to identify the bottlenecks and, ultimately, to design low-noise systems. Figure 3 shows the RIN in our setup measured at base band using appropriate photodiodes (Silicon Thorlabs PDA100-EC for 980 nm, InGaAs Thorlabs PDA10CS-EC for 1–1.8 µm, HgCdTe VIGO PVI-4TE-6 + MIPDC-5 for 3.5 µm) and a signal-source analyzer (Agilent E5052B). The noise performance of the gigahertz-laser oscillator is set by its multimode pump diode. Since no preamplifier is used, this noise level also corresponds to the RIN of the OPA pump.

![Figure 3](image-url)

**FIG. 3.** Relative-intensity-noise (RIN) measurements showing the influence of supercontinuum (SC) generation, supercontinuum filtering, and optical parametric amplification for the waveguide where highest amplification is achieved. The root-mean-square (rms) RIN noise integrated over the interval (1 Hz, 3.5 MHz) is indicated in parentheses for each measurement. The overall noise limit is set by the pump diode of the 1-GHz laser.

To investigate the impact of the OPA process itself, noise measurements are recorded using the waveguide that provides highest gain and absolute idler power (QPM period 25.47 µm, signal wavelength 1.50 µm). The RIN of the idler is, as expected, very similar to the RIN of
we observed, however, a noise increase of approximately 30 dB with respect to the pump noise level (Fig. 3). In order to determine the origin of this noise increase, further measurements are performed. We verify that the shot-noise levels, which depend on the wavelength and the optical power of the photodiode, are well below each of the respective RIN measurement results. The measured RIN of the supercontinuum over the full wavelength range accessible by the InGaAs photodiode (1–1.8 μm) is comparable to the gigahertz-laser output, with the exception of white-noise contributions above 100 kHz and technical noise around 100 Hz [Fig. 3, full supercontinuum (SC)]. However, the RIN of the supercontinuum after a 15-nm bandpass filter centered at 1.5 μm is similar to the OPA output (Fig. 3, filtered SC, before OPA). This filter bandwidth is chosen to correspond to the bandwidth of the amplified signal. It is well known that the interplay of the various mechanisms responsible for spectral broadening during the SCG process can lead to strongly-wavelength-dependent RIN [57], which becomes apparent when using narrow-band filters.

We can thus conclude from these observations that, despite the high gain, the OPA process itself is not adding a significant amount of noise, but that the noise increase stems rather from the SCG process in the Si3N4 waveguide. In the experiment presented here, the supercontinuum is optimized, above all, for broad bandwidth and spectral coherence [52], but the RIN may be minimized further by numerically analyzing the wavelength dependence of various noise types [58] and adapting the waveguide design accordingly.

IV. DISCUSSION

The experimental OPA results presented above exploit several advantageous properties that waveguides offer in comparison to bulk devices. Simulations in agreement with our experiments will be shown in this section, along with a general discussion on how to take advantage of those waveguide properties to achieve high gain - and thus high conversion efficiency - of a near-IR into a mid-IR comb.

A. Energy-dependent gain

For a phase-matched interaction assuming an undepleted, plane-wave pump field \( E_p \) and no initial idler field \( (E_i(0) = 0) \), the signal field at the output of an OPA device with length \( L \) can be written as [59]

\[
E_s(L) = E_s(0) \cosh(\Gamma L),
\]

where \( \Gamma \) is the gain parameter defined as

\[
\Gamma = \sqrt{\kappa_i \kappa_s |E_p|}
\]

with \( \kappa_j = 2\pi d_{ej}/(n_j \lambda_j), j = i, s \) (idler/signal) and where \( d_{ej} \) denotes the material-dependent effective nonlinear coefficient. Assuming sufficiently long pump pulses to provide constant pump intensity for the signal pulse during their interaction, we can approximate the magnitude of the pump field \( |E_p| \) as a function of the peak power \( P_p \) 

\[
P_p \sim \frac{U_p}{\tau_p}.
\]
\[ |E_p| - \sqrt{\frac{2}{\pi^2 n_p \epsilon_0 c}} \sqrt{\frac{2U_p}{\pi w_0^2 \tau_p}} \]  

where \( U_p \) is the pulse energy, \( \tau_p \) the pulse duration, \( n_p \) the refractive index, and \( w_0 \) the beam waist. To maximize the interaction in a bulk device, the diffraction length of the beam (and thus the beam radius) is often set to match the distance \( L_{\text{GVM}} \) over which pump and signal pulses walk off each other due to group velocity mismatch (GVM)

\[ w_0^2 k_p = L_{\text{GVM}} = \tau_p \left( \frac{1}{v_p} - \frac{1}{v_s} \right)^{-1}, \]

where \( k_p = n_p \frac{2\pi}{\lambda_p} \) denotes the pump wavenumber. For a given set of phase-matched pump/signal/idler frequencies, the achievable gain will be independent of the pump pulse duration and can only be scaled via the pulse energy,

\[ \Gamma L_{\text{GVM}} \approx C_{p,s,i} U_p (\text{bulk}), \]

with a proportionality factor \( C_{p,s,i} \) containing the wavelength-dependent material properties. If the pump pulse is too short, then confocal focusing according to Eq. (4) may yield an intensity above the material damage threshold. In this case, the pump pulse can be stretched to avoid damage. However, the diffraction still limits the achievable gain according to Eq. (5).

In a waveguide device however, the interaction is not limited by diffraction anymore, thus eliminating the relation imposed in Eq. (4) for the mode size as a function of GVM. The gain can now additionally be scaled via the pump pulse duration and the effective mode area \( A_{\text{eff}} \), which takes into account the modal overlap inside the waveguide,

\[ \Gamma L_{\text{GVM}} \approx C'_{p,s,i} \sqrt{\frac{\tau_p}{A_{\text{eff}}}} (\text{waveguide}), \]

where \( C'_{p,s,i} = C_{p,s,i} \sqrt{\frac{\pi^2}{2k_p^2(v_p^{-1} - v_s^{-1})}} \). Thus high gain can be maintained by stretching the pump pulse duration despite lowering the pulse energy. The waveguide cross-section is then chosen such as to optimize the overlap of the guided pump, signal and idler modes (see Sec. III B). Stretching our pump pulses to \( \sim 800 \text{ fs} \), as described in the experimental section, and taking advantage of the tight mode confinement provided by the PPLN waveguide thus allow us to achieve high gain with nearly one order of magnitude less pulse energy than a best-case estimate of a bulk interaction.

B. Pump versus idler group-velocity mismatch

In the presence of not only GVM between pump and signal but also idler walk-off and group-velocity dispersion of all of the waves, a more-general description of the OPA process is required [60]. In order to explain the gain variations observed experimentally across the broad signal spectral range, we perform numerical simulations based on a general model of the dynamics inside the PPLN waveguides. The model describes the propagation of the pump,
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signal, and idler pulses through the waveguide, accounting for the wavelength-dependent effective index and modal-overlap coefficients in the waveguide, and it includes both second- and third-order nonlinear properties of the PPLN waveguides [61,62]. In order to determine the dispersion profile of the waveguides, we proceed as follows. First, we simulate the proton diffusion inside the waveguide during the waveguide fabrication process, to obtain a proton concentration profile over the cross section of the waveguide [63]. Following Ref. [63], we obtain the change in refractive index as a function of the wavelength and the transverse position, then calculate the corresponding properties of the fundamental waveguide mode versus the wavelength. These properties are reasonably accurate for IR wavelengths, but less is known about the mid-IR properties. To account for this uncertainty, we apply an additional fixed offset to the effective index for the mid-IR part of the spectrum (wavelengths > 2 µm). This offset is chosen such that the numerically predicted set of the phase-matched signal wavelength versus the QPM period is in good agreement with the experimentally measured dependence. As can be seen in Fig. 4(a), we also include the change in refractive index induced by OH absorption in the material around 2.85 µm [61]. Having calculated the spatial profile of the fundamental mode, an effective area for the OPA process can be defined,

\[ A_{\text{eff}}(\omega_s, \omega_p) = \left( \int_{-\infty}^{0} \int_{-\infty}^{0} | \mathcal{F}(x, y) \times B(x, y, \omega_p) B(x, y, \omega_s) B(x, y, \omega_p - \omega_s) |^2 \quad \text{d}x \text{d}y \right)^{1/2} \quad \text{(7)} \]

where \( \mathcal{F} \) is a normalized nonlinear coefficient accounting for the so-called dead layer (layer at the top of the waveguide, where the second-order susceptibility is erased during fabrication) [54], and \( B(x, y, \omega) \) is the spatial profile of the fundamental waveguide mode with frequency \( \omega \), normalized according to \( \int_{0}^{0} \int_{0}^{0} B(x, y, \omega)^2 \text{d}x \text{d}y = 1 \). An effective pump intensity, \( P_{\text{on}} / A_{\text{eff}} \), can be introduced, which leads to a normalized OPA gain rate \( \gamma = \Gamma / (\sqrt{P_{\text{on}} \text{ power}}) \).

Figure 4(b) shows how the modal overlap integral in Eq. (7) affects the normalized gain coefficient \( \gamma \), over the range of signal wavelengths used in this experiment.

![FIG. 4. (a) Effective group index as a function of wavelength over the range used in the experiment. (b) Gain coefficient normalized with respect to power and propagation length. The gain decreases for shorter signal wavelengths as the mode size of the corresponding idler (the gray wavelength scale on top) increases, reducing the modal overlap.](image-url)
In order to directly visualize the spectrally dependent effect of modal overlap and GVM on the achievable gain, the pulse-propagation simulations assume a flat-top initial signal spectrum with a flat spectral phase. The following input parameters corresponding to the experimental values are used: 280-mW pump power, 1-mW signal power over the whole flat-top spectrum (1300–1850 nm) and 70-fs pump pulses with a negative chirp of $-25,000 \text{ fs}^2$. A general propagation loss of 0.1 dB/cm is included. We assume a nonlinear coefficient of $d_{33} = 19.5 \text{ pm/V}$ [64] and, to obtain improved agreement with the experimentally measured gain, $A_{\text{eff}}$ was scaled by a small factor of 1.17 compared to the directly calculated value from Eq. (7). Without any further adjustments, the simulations (Fig. 5) are able to reproduce remarkably well the features observed in the experiment [Fig. 2(b)].

Looking at the gain curve displayed in Fig. 4(b), one may expect the amplification to monotonically increase with increasing signal wavelength. However, the simulated spectra shown in Fig. 5 are in good agreement with the experimental data presented above: instead of a monotonic increase, a maximal amplification of 35–40 dB is reached in the range of 1450–1500 nm and then a decrease can be observed until the effect of the OH absorption becomes visible for signal wavelengths of around 1650–1700 nm. This trend can be explained as follows: while the gain coefficient $\gamma$ contains information about the spatial overlap of the idler with the pump and the signal mode inside the waveguide, it does not take into account the temporal behavior of the idler pulses. As can be inferred from Fig. 4(a), the effective group velocity of the idler, $v_{\text{eff}}(\lambda) = c / n_{\text{group, id}}(\lambda)$, crosses the velocity of the pump (intersection with the red dashed line) when scanning the QPM periods. It is for the signal wavelengths corresponding to this intersection—i.e., where the idler moves at nearly the same velocity as the pump—that we observe maximum signal amplification. Figure 6 illustrates and describes the three regimes that we encounter in the scan:

1. At a signal wavelength of between 1500 and 1650 nm, both the signal and the idler propagate with a higher velocity than the pump. Although a high spatial overlap is given, the short temporal overlap inhibits further amplification [Fig. 6(a)].
2. For wavelengths of around 1450–1500 nm, the corresponding idler temporally stays with the pump, leading to the buildup of a strong idler pulse and maximal signal amplification [Fig. 6(b)].
(3) In the third regime (<1450 nm), the signal and the idler have opposite group velocities with respect to the pump [Fig. 6(c)]. This configuration acts as a “trap” for the signal and idler pulses, as they are pulled towards each other, therefore ensuring a long interaction length and, potentially, high amplification. However, the amplification becomes highly suppressed in this wavelength range due to the increasing mode size of the idler and the resulting poor spatial overlap.

From these experimental and numerical observations, we conclude that the highest gain - and thus the most efficient mid-IR idler generation - is achieved by designing a waveguide where the idler group velocity is as close as possible to that of the pump while maintaining a high spatial overlap.

FIG. 6. Illustrative sketch and magnitude of the interacting fields with the center of the pump pulse setting the reference frame for all figures. The dashed white lines indicate the expected temporal lead or lag of signal and idler pulses with respect to the pump due to group-velocity mismatch (GVM). (a) Idler faster than pump. The three pulses overlap over only a short distance: the energy transfer from pump to signal and idler is limited. (b) Idler follows pump. As the signal is passing through the pump pulse, the generated idler photons stay overlapped with the pump and coherently add up to form a strong pulse. (c) Idler slower than pump, i.e., opposite signs for signal and idler. Signal and idler “drag” each other along, leading to a longer interaction with the pump (the “trapped” state). Note the change in magnitude of the electric field (the color bar) compared to (a) and (b). The lower field magnitudes are a consequence of the reduced spatial-mode overlap, which considerably lowers the gain [as shown in Fig. 4(b)].
V. CONCLUSION

In this paper, we address the challenge of nonlinear-optical frequency conversion at low pulse energies with the aim of transferring a 1-GHz frequency comb at 1 µm into the application-relevant mid-IR spectral region. Using a SESAM-mode-locked laser at 1 µm, we have achieved tunable offset-free combs from 2.5 to 4.2 µm with up to 4 µW of power per comb line around 3.5 µm. The comb spectra are generated in a PPLN RPE waveguide by optical parametric amplification. The signal photons for the OPA process are obtained by supercontinuum generation in a silicon-nitride waveguide with only 40 pJ of coupled pulse energy. During the OPA stage, this signal beam is amplified by up to 35 dB using 300 pJ of pump energy. We show that, in contrast to bulk devices, signal amplification in a waveguide OPA can be increased by stretching the pump pulse and exploiting the waveguide dispersion to obtain a similar effective group velocity for pump and idler pulses. Those degrees of freedom provide interesting design opportunities for low-energy frequency conversion of a variety of compact laser sources, including semiconductor lasers [65], without the need for additional laser power amplifiers.

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Extending the wavelength range


Extending the wavelength range


Chapter 4

Pushing to multi-gigahertz repetition rates

The results presented in this thesis so far were obtained with an Yb:CALGO laser operating at a repetition rate of 1 GHz. As can be seen from the overview provided in section 1.5, the development of diode-pumped solid-state lasers at repetition rates above 1 GHz with peak powers of several 100 W has only become successful in recent years. One of the main reasons was the lack for a solution to overcome the challenges set by Q-switching instabilities. In this chapter, we will discuss how we applied the concept of self-defocusing induced by cascading of second-order nonlinearities to solve the Q-switching damage problem and reach Watt-level, femtosecond operation of an Yb:CALGO laser at 10 GHz repetition rate.

4.1 Q-switched modelocking/Q-switching instabilities

The results discussed in this thesis so far were all implicitly based on continuous wave modelocked (cw-ML) pulse trains, i.e. where the pulse energy hardly changes from one pulse to the next (Figure 4.1(a)). However, a pulse train emitted by a modelocked laser may under certain conditions exhibit strong amplitude modulations. If the modulation is created on purpose, e.g. by periodically modulating the cavity losses (i.e. “switching the cavity quality factor (Q-factor)”), the laser operation state is usually referred to as Q-switched modelocking. The modulations can however also arise unintentionally (i.e. without an external driving mechanism) when designing lasers aiming for cw-ML using saturable absorbers. In this case, the term Q-switching instabilities is more appropriate. These instabilities can be qualitatively explained by the fact that for high intracavity peak powers, the cavity losses are reduced due to the temporary saturation of the absorber. Hence, the
relaxation oscillations (i.e. small oscillations of the pulse energy and gain around their steady-state value) may be amplified instead of damped. These fluctuating pulses easily reach peak powers that can damage the optical elements inside a laser cavity.

![Continuous wave (cw) vs. Q-switched modelocking and Q-switching instabilities.](image)

If the intracavity nonlinearities and dispersion have been designed to support the formation of stable cw-modelocked pulses, this cw modelocking state will become energetically favorable above a certain threshold. The intracavity pulse energy necessary to reach this threshold depends on several factors. The threshold increases in particular for laser materials with a low gain cross section, when choosing a SESAM with a large modulation and high saturation fluence, or when the modesizes in the gain and on the SESAM are large [90, 91]. Q-switching instabilities represent a major concern in particular for two categories of modelocked lasers: high-power laser and lasers with very high pulse repetition rates (i.e. in the GHz regime). In the case of high-power lasers, the modesizes need to be increased when scaling up the power in order to stay below the critical damage intensities of the optical components. This however increases the threshold above which cw-ML becomes favorable. For lasers with very high pulse repetition rates (i.e. in the GHz regime) the challenge arises from the fact that the intracavity pulse energy is intrinsically very low. Thus, more pump power is needed to reach the threshold for stable cw-ML. The task when designing lasers that belong to either of these categories thus consists of two parts: lowering the Q-switching/cw-ML threshold on one hand and avoiding damage during inevitable instabilities on the other hand.
In paper 4, we will discuss a novel approach for multi-GHz lasers that tackles both these issues. The 10-GHz laser cavity we will present in Paper 4 contains a phase-mismatched PPLN device that generates a self-defocusing lens to protect the optical elements from Q-switching induced damage. At the same time, this PPLN device provides strong negative self-phase modulation, which allows for soliton modelocking with positive GDD (recall section 1.3.1 on soliton modelocking). The generation of such a self-defocusing effect is based on the principle of cascading of second-order nonlinearities, which we will introduce in the following section.

4.2 Cascading of second-order nonlinearities

Cascading of second-order nonlinearities refers to the process of using a phase-mismatch second-order nonlinear interaction to generate an effective third-order response [92-94]. In order to describe the process, let’s assume a total real-valued electric field that consists of two waves; the fundamental wave \( F \) and its second harmonic \( \text{SH} \), oscillating at double the frequency. This total field can thus be expressed as

\[
E(z,t) = \frac{1}{2} \left( A^1 e^{i(\omega_1 t - k_1 z)} + A^1_\ast e^{-i(\omega_1 t - k_1 z)} \right) + \frac{1}{2} \left( A^\text{SH} e^{i(\omega_\text{SH} t - k_\text{SH} z)} + A^\text{SH}_\ast e^{-i(\omega_\text{SH} t - k_\text{SH} z)} \right)
\]  

(4.1)

Inserting the electric field \( E(z,t) \) into the wave equation (Eq. (1.2) in section 1.1) and considering the material response \( P \) up to third order yields the following coupled wave equations for the complex amplitudes of the fundamental and second harmonic wave:

\[
\begin{align*}
\left( \frac{d}{dz} + \frac{1}{v_gF} \frac{\partial}{\partial t} - i \frac{k_n}{2} \frac{\partial^2}{\partial t^2} \right) A_F &= -i \frac{\omega_F}{2n_F c} \chi^{(2)} A_F^* A_F e^{-i\Delta kz} - \frac{3\omega_F}{8n_F c} \chi^{(3)} \left[ |A_F|^2 + 2|A^\text{SH}|^2 \right] A_F \\
\left( \frac{d}{dz} + \frac{1}{v_g\text{SH}} \frac{\partial}{\partial t} - i \frac{k_n}{2} \frac{\partial^2}{\partial t^2} \right) A^\text{SH} &= -i \frac{\omega_F}{2n_{\text{SH}} c} \chi^{(2)} A^2_F e^{i\Delta kz} - \frac{3\omega_{\text{SH}}}{8n_{\text{SH}} c} \chi^{(3)} \left[ |A^\text{SH}|^2 + 2|A_F|^2 \right] A^\text{SH}
\end{align*}
\]  

(4.2)

The derivation of the equations above contains the slowly-varying-envelope approximation (i.e. \( \frac{d^2}{dz^2} A = 0 \)) as well as dispersion up to second order. The group velocity is denoted by \( v_g = \left( \frac{dk}{d\omega} \right)^{-1} \) and \( k_n = \frac{dk}{d\omega} \) represents the group velocity dispersion, where \( k_n = (\omega / c)n(\omega) \). Note that the simplified notation “\( \chi^{(3)} \)” here
denotes the fast (i.e. electronic) material response only. Furthermore, the tensor structure of the susceptibility has to be taken into account when implementing these equations for a specific configuration (see for example Bache et al. [95]). We can simplify these equations by assuming plane waves (i.e. $\frac{\partial}{\partial t}A = 0$) and considering cases where there is no third-order nonlinearity:

$$\frac{d}{dz}A_f = -i \frac{\omega_F}{2n_{fc}} \chi^{(2)} A_{SH} A_f^* e^{-i\Delta k z}$$

$$\frac{d}{dz}A_{SH} = -i \frac{\omega_F}{2n_{fc}} \chi^{(2)} A_f^2 e^{i\Delta k z}$$

These simplified equations are usually used to describe the process of second harmonic generation (SHG). In the following, we will distinguish between two different regimes: phase-matched SHG on one hand and the regime far off phase-matching on the other hand, which we call the “cascading” regime.

### 4.2.1 Classical phase-matched SHG regime

In the perfectly phase-matched case, the phase mismatch $\Delta k = k_{SH} - 2k_f$ is zero. By taking into account energy conservation, i.e. $|A_f(0)|^2 = |A_{SH}(z)|^2 + |A_f(z)|^2$ and assuming that the SH amplitude is zero at the input we obtain the following expressions for the amplitude of the fundamental and second-harmonic wave after a propagation distance $z$ inside the SHG medium [96]:

$$A_f(z) = |A_f(0)| \text{sech}(\gamma z) e^{i\phi}$$

$$A_{SH}(z) = -i |A_f(0)| \text{tanh}(\gamma z) e^{2i\phi},$$

where $\gamma = \frac{\omega_F}{2n_{fc}} \chi^{(2)} |A_f(0)|$ and $\phi$ represents the arbitrary phase of the fundamental wave at the input $z = 0$. Figure 4.2(a) shows how qualitatively how energy is transferred from the fundamental to the second harmonic as a function of the propagation distance in the SHG medium.
Figure 4.2: SHG vs. cascading. (a) Phase-matched second harmonic generation vs. (b) the cascading regime, where there is no net energy transfer from the fundamental to the second harmonic wave.

### 4.2.2 Cascading regime

Here, we are interested in the regime where the phase mismatch is very large and the transfer of energy from the fundamental to the second harmonic is very weak, i.e. we assume:

- $|\Delta k| \gg 0$
- $|A_f(z)| \approx |A_f(0)|$

Using these approximations allows us to integrate Eq. (4.3) and we obtain [94]

$$A_{sH}(z) = -\frac{1}{\Delta k} \frac{\omega_f}{2n_{sH}c} \chi^{(2)} A_f^2 e^{-i\Delta z}.$$  \hspace{1cm} (4.5)

This second harmonic component can now be treated as a perturbation to the fundamental wave. By substituting Eq. (4.5) into Eq. (4.3), we get the following propagation equation

$$\frac{d}{dz} A_f = i \frac{1}{\Delta k n_f n_{sH} c} \left( \frac{\omega_f}{2c} \chi^{(2)} \right)^2 |A_f|^2 A_f$$  \hspace{1cm} (4.6)

Let us recall the general coupled wave equations, which contained dispersion, second- and third-order nonlinearities and highlight the third-order contribution in red:

$$\left( \frac{d}{dz} + \frac{1}{v_{g,f}} \frac{\partial}{\partial t} - i \frac{k''}{2} \frac{\partial^2}{\partial t^2} \right) A_f = -i \frac{\omega_f}{2n_c} \chi^{(2)} A_{sH} A_f e^{-i\Delta z} - i \frac{3\omega_f}{8n_c} \chi^{(3)} \left[ |A_f|^2 + 2|A_{sH}|^2 \right] A_f$$  \hspace{1cm} (4.7)
By plugging in Eq. (4.5) for the second harmonic amplitude $A_{SH}$, we obtain

$$\left( \frac{d}{dz} + \frac{1}{v_{g,F}} \frac{\partial}{\partial t} - i \frac{k''_{n,F}}{2} \frac{\partial^2}{\partial t^2} \right) A_r = -i \frac{\omega_F}{2n_c} \chi^{(2)}(\frac{1}{\Delta k} \frac{\omega_F}{2n_{SH}c} \chi^{(2)}(2) A_F^2 e^{j\Delta z}) A_r e^{-j\Delta z}$$

$$= -i \frac{3 \omega_F \chi^{(3)}}{8n_c} \left[ |A_r|^2 + 2 |A_{SH}| \right] A_r$$

(4.8)

Re-arranging the terms leads to:

$$\left( \frac{d}{dz} + \frac{1}{v_{g,F}} \frac{\partial}{\partial t} - i \frac{k''_{n,F}}{2} \frac{\partial^2}{\partial t^2} \right) A_r = -i \frac{3 \omega_F}{8n_c} \left[ \frac{2}{3\Delta k n_{SH}c} (\chi^{(2)})^2 \right] |A_r|^2 A_r$$

$$= -i \frac{3 \omega_F \chi^{(3)}}{8n_c} \left[ |A_r|^2 + 2 |A_{SH}| \right] A_r$$

(4.9)

Treating the strongly phase-mismatched second-harmonic wave as a perturbation to the fundamental thus yields a term of the same form as the third-order response marked in red, i.e. $\propto |A_F|^2 A_r$. We can thus view this term as an effective third-order response with a susceptibility $\chi^{(3)}_{\text{casc}}$ as defined in the table below.

<table>
<thead>
<tr>
<th>Intrinsic material - $\chi^{(3)}$</th>
<th>Cascading - $\chi^{(3)}_{\text{casc}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Third-order response term</td>
<td>$\frac{d}{dz} A_r = -i \frac{3 \omega_F \chi^{(3)}}{8n_c}</td>
</tr>
<tr>
<td>Susceptibility</td>
<td>$\chi^{(3)}$</td>
</tr>
<tr>
<td>Nonlinear refractive index</td>
<td>$n_2 = \frac{3}{4} \frac{\chi^{(3)}}{\epsilon_0 c n_F^2}$</td>
</tr>
</tbody>
</table>

Table 2: Analogy between the intrinsic third-order response of the material and the response caused by the cascading regime.

The effect of the third-order material response can be expressed by an intensity-dependent change of the refractive index, i.e.

$$n(I) = n + n_2 I$$

(4.10)
where the nonlinear index is proportional to $\chi^{(3)}$. Accordingly, we can also define a nonlinear index $n_2^{\text{casc}} \propto \chi^{(3)}_{\text{casc}}$ (see Table 2). The latter is inversely proportional to the phase mismatch $\Delta k$ and can thus be tuned in magnitude and sign. As a consequence, operating a $\chi^{(2)}$-material in the cascading regime provides a way to generate tunable Kerr-type response. Hence, both the sign of self-phase modulation (SPM) and Kerr-lensing can be flipped. While negative SPM allows for soliton modelocking with positive GDD, the negative Kerr lens can also be used as a self-defocusing mechanism for high-peak power pulses. How this self-defocusing effect was used to prevent Q-switching damage is detailed in Paper 4.

### 4.2.3 Cascading regime involving pulses

The derivations presented above were based on single-frequency plane waves for simplicity. When the cascading regime is used as a mechanism to obtain soliton pulses, temporal/spectral effects need to be considered as well. Operation in the cascading regime assumes a large phase mismatch $\Delta k$. Especially when aiming for short pulses, i.e. large spectral bandwidths, this requirement may start to break down for frequencies in the wings of the spectral envelope, which will start to approach phase matching due to dispersion. Assuming some frequency component located at an interval $\Delta \omega$ from the pulse center frequency $\omega_F$, i.e. $\omega^* = \omega_F + \Delta \omega$, we can express phase mismatch as

$$\Delta k(\omega^*) = k_n(\omega_{SH} + 2\Delta \omega) - 2k_n(\omega_F + \Delta \omega).$$

(4.11)

Taylor-expanding $\Delta k(\omega^*)$ to first order leads to

$$\Delta k(\omega^*) = k_n(\omega_{SH}) + 2k'_n(\omega_{SH})\Delta \omega - 2k_n(\omega_F) - 2k'_n(\omega_F)\Delta \omega = \Delta k_0 + \text{GVM} \cdot 2\Delta \omega$$

(4.12)

where $\Delta k_0$ denotes the phase mismatch between the pulse center frequency and its second harmonic and GVM refers to the group velocity mismatch, i.e.:

$$\text{GVM} = \frac{1}{g_{SH}} - \frac{1}{g_F} = k'_n(\omega_{SH}) - k'_n(\omega_F)$$

(4.13)

The effect of GVM/broad spectral bandwidth becomes critical when the frequency component $\omega^*$ approaches phase matching, i.e.

$$\Delta k(\omega^*) = \Delta k_0 + \text{GVM} \cdot 2\Delta \omega \approx 0$$

(4.14)
We can associate a pulse duration $\tau_{\text{crit}}$ to this critical bandwidth $2\Delta \omega$ and obtain a criterion that should be fulfilled in order to fully operate in the cascading regime:

$$|\Delta k_0| \gg \frac{\text{GVM}}{\tau_{\text{crit}}} \tag{4.15}$$

Note that the length of the cascading medium does not matter, i.e. using a shorter crystal will not increase the bandwidth over which the cascading approximations hold.

Besides the bandwidth considerations, it is also worth noting that when operating in the cascading regime with pulses, the residual second harmonic field actually consists of two components. When describing pulses, the time derivatives in (4.2) cannot be neglected anymore. Upon integration of the coupled wave equations, we obtain the following expression for the SH field after a propagation distance $L$ [93, 97]:

$$A_{\text{SH}}(L,t) = -\frac{1}{\Delta k_0} \frac{\omega_F}{2n_{\text{SH}} c} \chi^{(2)} \left[ A_F^2(0,t + v_g^{-1} L) e^{i\Delta k_0 L} - A_F^2(0,t + v_g^{-1} L) \right] \tag{4.16}$$

The first term corresponds to Eq. (4.5) and describes the component of the SH field that propagates at the same velocity than the fundamental pulse and leads to self-phase modulation of the latter. The second term denotes an SH component that is generated at the input of the cascading medium and then propagates at its own velocity.

### 4.2.4 Nonlinear losses

The two SH components described above constitute the so-called nonlinear losses, i.e. the energy that is lost to residual SHG despite the large phase mismatch. By taking the ratio of the energy in those two pulse components to the energy in the fundamental pulse, we can calculate the nonlinear losses $\alpha_{\text{NL}}$ per single pass, which amount to [97]

$$\alpha_{\text{NL}} = \frac{1}{3} \frac{1}{(\Delta k_0)^2} \frac{(\omega_F \chi^{(2)})^2}{n_F^2 n_{\text{SH}}^2 c^3} I_{r,\text{peak}} \tag{4.17}$$
where $I_{F,\text{peak}}$ denotes the peak intensity of the fundamental pulse. Note that also here, the length of the cascading medium does not come into play.

**Trade-off: Nonlinear phase vs. nonlinear losses**

For practical modelocking applications, it is useful to bear in mind the relation between the achievable SPM and the nonlinear losses,

$$\alpha_{NL} = \frac{4}{3\Delta k_0 L} \phi_{\text{SPM,casc}}, \quad (4.18)$$

where $\phi_{\text{SPM,casc}} = \frac{2\pi}{\lambda_F} n_2^\text{casc} I_F L$ represents the nonlinear phase acquired by propagating through a cascading medium of length $L$. In order to obtain stable soliton modelocking, this nonlinear phase has to be compensated by an appropriate amount of GDD (see section 1.3.1). A larger nonlinear phase will lead to shorter pulses according to Eq. (1.10), but also implies higher nonlinear losses. The length of the cascading medium can be used as an optimization parameter, i.e. the longer the crystal, the more favorable the ratio. A long crystal (i.e. several centimeters) however constitutes a drawback for certain cavity configurations, e.g. short high-repetition rate laser cavities. In Paper 4 and its introductory remarks, we will discuss a method to suppress the losses by using a specially engineered quasi-phase matching (QPM) pattern in a PPLN device while keeping the crystal length short enough to fit within a 10 GHz cavity.
4.3 Introductory remarks to Paper 4

4.3.1 Preceding experimental results

Cascading using LBO

Several experiments based on cascading of second-order nonlinearities laid the groundwork for the results presented in Paper 4. In [97], we describe an Yb:CALGO laser that was modelocked in the positive-GDD regime by using negative SPM generated in a 20-mm-long lithium triborate (LBO) crystal. The latter was cut at \( \theta = 90^\circ \), which allows for non-critical phase matching (i.e. not relying on precise angular alignment). Both the fundamental and the second harmonic propagate collinearly, but with their polarizations perpendicular to each other (referred to as type I phase matching). The laser operated at a repetition rate of 113 MHz and delivered pulses as short as 114 fs with a maximum average output power of 1.1 W. The phase mismatch was adjusted by tuning the temperature of the LBO crystal between 60-90°C (phase matching for the center wavelength of 1050 nm occurs at 167°C). This tuning range corresponds to an effective nonlinear index \( n_2^{\text{asc,eff}} \) (intrinsic material \( \chi^{(3)} \) plus the cascading \( \chi^{(3)} \)-contribution) of \(-4 \text{ to } -6 \times 10^{-20} \text{ m}^2\text{W}^{-1}\), which is on the same order of magnitude as the Yb:CALGO crystal \((\sim 8 \times 10^{-20} \text{ m}^2\text{W}^{-1})\), but with the sign flipped. The negative SPM achieved in the 20-mm-long LBO was sufficient to overcompensate the positive SPM generated in the 3-mm-long Yb:CALGO crystal, so that soliton modelocked could be achieved with a total cavity roundtrip GDD of +1577 fs\(^2\) corresponding to the material dispersion of the CALGO, the LBO and the 1.5-mm-long YAG Brewster plate used to enforce linear polarization. By scanning the pump power at different phase mismatch settings and measuring the laser parameters including the residual green light exiting the cavity, we were able to confirm the \((\Delta k_0)^2\)-dependence of the nonlinear losses. Decreasing the phase mismatch (i.e. operating around 90°C as opposed to 60°C) yielded a larger nonlinear phase and thus shortened the pulses (e.g. from 145 fs to 114 fs at a power of 1.1 W). However, the pulse duration was ultimately limited by the increasing nonlinear losses, which in combination with the SESAM led to an increased roll-over of the net cavity reflectance. A SESAM with a modulation of 2.8% was used and the nonlinear losses reached up to 0.55 % before destabilizing fundamental modelocking. Once this roll-over was reached, the laser
Pushing to multi-gigahertz repetition rates

switched to double-pulsing mode to reduce the fluence and minimize the losses. At this stage, a further decrease of the pulse duration was not possible anymore.

**Cascading using 2-dimensionally patterned QPM PPLN devices**

LBO has the advantage of being a commercially available off-the-shelf crystal, and using it in the cascading regime works well for Watt-level lasers in the 100-MHz regime. For lasers in the GHz regime however with their reduced intracavity pulse energy, sufficient SPM could only be obtained with a longer crystal or a tighter beam focus. Considering that a linear 10-GHz laser cavity for instance has a free-space length of 1.5 cm, the geometrical implementation becomes very challenging, if not impossible. Furthermore, the SESAM modulation depth often has to be reduced to mitigate Q-switching instabilities. Hence, the nonlinear losses become non-negligible compared to the SESAM modulation and may prevent the laser from modelocking.

Switching to a cascading medium that can provide a larger $n_2^{\text{casc,eff}}$ and taking care of the nonlinear losses thus becomes a necessity. The solution comes in the form of quasi-phase matched (QPM) structures. An additional degree of freedom to tune the phase mismatch is introduced via the poling periods and thus the grating vector $K_g$:

$$\Delta k = k_{SH} - 2k_f - K_g$$

Equation (4.19)

In [98], we presented modelocking in the cascading regime using a custom-designed 2-mm-long PPLN QPM device. The device consists of 3 different types of QPM gratings:

- Gratings with a simple periodic QPM structure: “Control gratings”
- Gratings with longitudinal apodization: The QPM periods are long at the input and output sections of the device and become shorter in the middle. The middle section has the same poling period than the control gratings.
- Gratings with longitudinal apodization and a transversal “fanout” structure: In addition to the longitudinal change, the period also varies smoothly in the transverse direction with respect to the beam.

The purpose of the longitudinal apodization is the reduction of the nonlinear losses. In the input section, the phase mismatch is very large and the losses to the
residual SH very weak. However, the generated SPM is also weak, since the nonlinear phase scales with $\Delta k^{-1}$. To obtain sufficient SPM, the phase mismatch is adiabatically decreased towards the middle section of the device to allow for a strong cascading interaction between the fundamental and the SH wave. This SH is then again turned off by increasing the phase mismatch again towards the output facet (see Paper 4 for visualization), which suppresses the energy lost to residual SHG. In this way, the nonlinear losses can be reduced to $\approx 0.01$ % compared to $\approx 0.25$ % in the non-apodized case. In the middle section, the effective index $n_{2,\text{casc,eff}}$ reaches values that are up to 2 orders of magnitude larger than in LBO. Hence, a substantial amount of SPM can be generated in a short (i.e. 2 mm) crystal already. The goal of the fanout design is to provide the possibility of tuning the amount of SPM by translating the device perpendicularly to the beam.

The different gratings were tested in a 540 MHz Yb:CALGO cavity modelocked with the same SESAM that was already used in the LBO cavity mentioned above. As expected, modelocking could not be obtained with the non-apodized control gratings due to the large nonlinear losses associated with the high $n_{2,\text{casc,eff}}$. Modelocking could be obtained with the apodized, non-fanout gratings, but with more difficulties to trigger the modelocked operation. On the other hand, the apodized gratings with the fanout structure led to reliable self-starting modelocking. We explain this by the fact that an unfavorable nonlinear-etalon type response can arise when the poling periods are perfectly parallel to each other and perpendicular to the beam [98]. We also made similar observations in the case of the birefringent LBO crystal: if the crystal was slightly tilted such that the Poyting vectors (i.e. direction of the energy flux) of the fundamental and SH component propagating at its own velocity were not perfectly parallel anymore, self-starting modelocking was easier to achieve.

In this first proof-of-principle demonstration of an apodized and fanout QPM PPLN device operated in the cascading regime, we obtained 149 fs-pulses at 1.45 W of average output power with $n_{2,\text{casc,eff}} \approx -1.5 \times 10^{-18} \text{m}^2\text{W}^{-1}$ and a net intracavity dispersion of +1237 fs$^2$. By reducing the net amount of GDD to +273 fs$^2$ using a Gires-Tournois-type dispersion compensating mirror, the pulses could be shortened down to 100 fs with an average output power of 760 mW.
The self-defocusing aspect of the cascading regime

Both the proof-of-principle experiments in LBO as well as using the new 2D-QPM structures enabled us to explore the parameter space of SESAM-modelocking in the cascading regime in a way that had not been done before. However, both the 113 MHz and the 540 MHz laser at 1 µm can be considered nowadays as fairly “standard” bulk solid-state laser cavities with geometrical dimensions and pulse energy levels that are ideally suited for SESAM modelocking. In this repetition rate range, femtosecond Watt-level pulses trains can be achieved with more common techniques, e.g. SESAM or Kerr lens modelocking in the conventional soliton regime (i.e. with positive SPM provided by the gain crystal and negative GDD provided by dispersion-compensating mirrors).

The situation looks very different at multi-GHz repetition rates. The small cavities impose strong constraints on the number and choice of components and the intracavity pulse energy is intrinsically so low even for Watt-level output powers, that generating enough SPM becomes difficult using conventional techniques. Furthermore, the enhanced Q-switching-instabilities have a tendency to damage the optical components before cw modelocking is reached. Preventing damage using a self-defocusing cavity design was introduced by Klenner et al. in [19], where a 5-GHz Yb:CALGO laser with ~100 fs and 4 W of average output power is described. The self-defocusing damage prevention mechanism consists of designing the cavity in such a way, that the laser mode size instantaneously increases on the critical intracavity components for high peak powers. The mode sizes on the different elements are primarily defined by the curvatures of the mirrors used to form the cavity. However, the Kerr effect in the gain material can lead to self-focusing of the beam if the intensity is high enough, as it is usually the case during Q-switching instabilities. Most often, the flat SESAM is used as one of the two cavity end mirrors and will be the first component to suffer damage. By placing an appropriately curved mirror after the gain material, this focusing behavior can be turned into defocusing of the mode on the SESAM. Hence, even if the pulse energy temporarily reaches very high values, the fluence (energy per area) stays clamped below the damage threshold of the SESAM. To implement this scheme, the cavity needs at least one curved folding mirror between the gain crystal and the SESAM. Such a V-shaped cavity was still possible to realize in order to reach a repetition rate of 5 GHz [19]. Scaling this
approach beyond 5 GHz however becomes practically impossible due to the strong mirror curvature involved, which leads to a significant beam astigmatism if the angle of incidence is too large. Given the short distances between the elements, the angle however cannot be made sufficiently small without creating space and alignment issues.

In order to scale the repetition rate to 10 GHz, we decided to replace the defocusing functionality of the curved mirror by the self-defocusing Kerr effect obtained in the PPLN cascading device described above as a result of the negative $n_2$. Since this element can be used in transmission, no folding of the cavity is necessary anymore and the simplest possible geometry, a straight cavity, becomes possible. Paper 4 describes the details of this innovative solution and presents the record modelocking results that were achieved for a diode-pumped solid-state laser at such high repetition rates.

### 4.3.2 Notes on Kerr lens calculations

The complex electric field of a Gaussian beam propagating in free space along the z-axis assuming the paraxial approximation (i.e. small divergence), can be expressed as

$$E(r,z) = E_0 \frac{w_0}{w(z)} \exp \left( - \frac{r^2}{w^2(z)} \right) \exp \left( -ikz - ikr^2 \frac{2}{R(z)} + i\vartheta(z) \right)$$

(4.20)

with the following parameters:

- Beam radius: $w(z) = w_0 \sqrt{1 + \left( \frac{z}{z_0} \right)^2}$
- Beam waist (minimal beam radius, defining the position $z=0$): $w_0 = \sqrt{\frac{\lambda w_0^2}{\pi}}$
- Curvature of the wave fronts: $R(z) = z \left( 1 + \left( \frac{z_0}{z} \right)^2 \right)$
- Rayleigh-range (range over which the beam radius increases by $\sqrt{2}$): $z_0 = \frac{\pi w_0^2}{\lambda}$
- Gouy-phase: $\vartheta(z) = \arctan \left( \frac{z}{z_0} \right)$

The effect of an ideal thin lens on a Gaussian beam $E(r,z)$ can be expressed by a phase transformation $t_{\text{lens}}$ according to

$$E_{\text{out}}(r,z) = t_{\text{lens}} E_{\text{in}}(r,z)$$

(4.21)
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\[ t_{\text{lens}} = \exp \left( ik \frac{r^2}{2f} \right) \]  

(4.22)

Thus, the curvature of the wave front after the lens changes according to

\[ \frac{1}{R_{\text{out}}(z)} = \frac{1}{R_{\text{in}}(z)} - \frac{1}{f} \]  

(4.23)

The phase transformation obtained by propagating through a Kerr-medium of length \( L \) can be expressed as

\[ t_{\text{kerr}} = \exp \left( -ikn_2 I(r)L \right) \]  

(4.24)

with the intensity \( I(r) = I_{\text{pk}} \exp \left( -\frac{r^2}{w^2(z)} \right) \). By approximating the intensity profile by a parabola, i.e. expanding the exponential function as \( \exp(x) \approx 1 + x^2 \), and comparing the resulting expression to the one obtained for a thin lens, we find that:

\[ F_{\text{Kerr}} \approx \frac{1}{f} = \frac{4n_2L}{w_{\text{in}}^2} I_{\text{pk}} \]  

(parabolic approximation)  

(4.25)

This parabolic approximation is widely used in literature. However, it tends to overestimate the lens power \( F_{\text{Kerr}} \), since aberrations due to the non-parabolic beam profile wings are neglected.

A more realistic value for the lens power of a Kerr-type medium can be obtained by taking an approach based on modal overlap optimization. The approach can be framed as the following question: Which value for the focal length \( f \) (and thus \( F_{\text{Kerr}} \)) will lead to an optimal modal overlap of a field \( E_{\text{out,lens}} \) experiencing a phase shift \( t_{\text{lens}} \) due to this lens with a field \( E_{\text{out,Kerr}} \) experiencing a Kerr-phase shift \( t_{\text{kerr}} \)?

The modal overlap integral can be defined as

\[ m(E_{\text{out,lens}}, E_{\text{out,Kerr}}) = \int_0^\infty E_{\text{out,lens}}^* (r) E_{\text{out,Kerr}} (r) 2\pi rdr . \]  

(4.26)

The normalized integral is then given by

\[ m_{\text{norm}} \left( E_{\text{out,lens}}, E_{\text{out,Kerr}} \right) = \frac{\left| m \left( E_{\text{out,lens}}, E_{\text{out,Kerr}} \right) \right|}{\left( \left| m \left( E_{\text{out,lens}}, E_{\text{out,lens}} \right) \right| \left| m \left( E_{\text{out,Kerr}}, E_{\text{out,Kerr}} \right) \right| \right)^{1/2}} \]  

(4.27)
i.e. for a perfect overlap, $m_{\text{norm}}(E_{\text{out,lens}}, E_{\text{out,Kerr}}) = 1$.

Using this approach, we numerically found that the overlap converges to an optimum for a focal power of

$$F_{\text{Kerr}} \approx \frac{n_L}{w_{\text{in}}^2} I_{\text{pk}} \quad \text{(modal overlap optimization)} \quad (4.28)$$

i.e. a factor 4 weaker than when using the parabolic approximation. This result is in agreement with calculations determining the Kerr lens using a variational approach reported by Karlsson et al. [99]. For the Kerr lens calculations performed in Paper 4, we thus used Eq. (4.28) with the peak intensity $I_{\text{pk}} = \frac{0.88 E_p}{\pi w_{\text{in}}^2 \tau_{\text{FWHM}}}$ for soliton pulses with pulse energy $E_p$. 
Paper 4:

Watt-level 10-gigahertz solid-state laser enabled by self-defocusing nonlinearities in an aperiodically poled crystal

Authors: Aline S. Mayer (laser design, measurements and simulations)
Christopher R. Phillips (PPLN design and simulations)
Ursula Keller (supervision)

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Chapter 4

Watt-level 10-gigahertz solid-state laser enabled by self-defocusing nonlinearities in an aperiodically poled crystal


Department of Physics, Institute for Quantum Electronics, ETH Zurich, CH-8093 Zurich, Switzerland

Abstract

Femtosecond modelocked lasers with multi-gigahertz pulse repetition rates are attractive sources for all applications that require individually resolvable frequency comb lines or high sampling rates. However, the modelocked laser architectures demonstrated so far have several issues, including the need for single-mode pump lasers, limited output power, Q-switching instabilities, and challenging cavity geometries. Here, we introduce a technique that solves these issues. In a two-dimensionally patterned quasi-phase-matching (QPM) device, we create a large, low-loss self-defocusing nonlinearity, which simultaneously provides SESAM-assisted soliton modelocking in the normal-dispersion regime and suppresses Q-switching induced damage. We demonstrate femtosecond passive modelocking at 10-GHz pulse repetition rates from a simple straight laser cavity, directly pumped by a low-cost highly spatially-multimode pump diode. The 10.6-GHz Yb:CaGdAlO$_4$ (Yb:CALGO) laser delivers 166-fs pulses at 1.2 W of average output power. This enables a new class of femtosecond modelocked diode-pumped solid-state lasers with repetition rates at 10 GHz and beyond.

Introduction

The advancement of passively modelocked solid-state lasers has provided the basis for the development of reliable low-noise frequency combs, which have nowadays become important tools for a large variety of metrology and spectroscopy applications. Frequency combs with line spacings above 10 GHz are powerful tools for spectroscopy with resolved comb lines$^1$, the calibration of astronomical spectrographs$^2$-$^4$, arbitrary waveform generation$^5$, terabit optical transmission systems$^6$ and other applications that require easy access to the individual comb lines. Dual-combs in the gigahertz-regime for spectroscopic applications with fast acquisition times have recently attracted the attention of the scientific community$^7$-$^{10}$. Furthermore, the ultrastable pulse train in the time domain can be used for low-noise microwave photonics$^{11}$ and emerging optical computation systems based on coupled degenerate optical parametric oscillators$^{12}$-$^{14}$. Recently, pulse trains with multi-gigahertz repetition rates have also been used to generate dense pulse bursts that allowed micro-machining experiments to be performed in the ablation cooled regime$^{15}$.

To date, there are several approaches for generating frequency combs with large comb line spacings such as electro-optic (EO) combs$^{16}$-$^{18}$, microresonator combs$^{19,20}$, Kerr lens modelocked (KLM) solid-state lasers$^{21,22}$ or comb-line filtering of e.g. modelocked fibre lasers in the few-hundred MHz-regime. There are different benefits and trade-offs for these techniques. Parametric comb generation using electro-optical modulators or microresonators
Pushing to multi-gigahertz repetition rates offers a way to easily produce combs with > 10 GHz line spacings. However, the formation of stable high-power femtosecond pulses is less straightforward than compared with modelocked lasers. EO combs for example require a series of nonlinear broadening and laser amplification stages, while microresonator combs require locking of a high-coherence parametric pump laser (single spatial mode and single optical frequency) to a high-Q resonator. On the other hand, in modelocked lasers the pulse train in the time domain and the spectral comb structure are intrinsically linked via Fourier transformation, and the intense output pulses are well suited to subsequent nonlinear frequency conversion. KLM modelocked solid-state lasers at 10- or 15-GHz pulse repetition rates provide high-quality frequency combs, but the relatively low intracavity pulse energy at these high pulse repetition rates makes it more difficult to support KLM. The very tight focusing of the pump and laser beam required to initiate KLM via the gain medium requires an excellent pump beam quality (typically $M^2 \approx 1$).

Figure 1 | Modelocking regime and laser setup. a, Conventional soliton modelocking, where positive self-phase modulation (SPM) with $n_2 > 0$ is balanced by negative group-delay dispersion (GDD). In a straight-cavity configuration with a flat SESAM as an end-mirror, high peak powers during Q-switching instabilities cause self-focusing of the laser on the cavity elements, inducing damage and thereby preventing continuous wave modelocking. b, Soliton modelocking using a self-defocusing nonlinearity to provide negative SPM that is balanced by positive GDD. In addition the strong negative effective Kerr nonlinearity creates a dynamic defocusing lens that leads to an increase of the mode size in the event of Q-switching instabilities, hence protecting the elements from damage. c, Schematic and photo of the 10 GHz Yb:CALGO laser cavity, which is pumped by a spatially multimode pump diode (see Methods).
Moreover, modelocking has to be initiated at the edge of the cavity stability region. As an alternative to GHz KLM lasers, a robust fibre-based oscillator with a repetition rate of a few 100 MHz and filtering the comb lines using Fabry-Pérot cavities has also been successfully demonstrated as a way to reach comb line spacings in the multi-gigahertz regime. The complexity of such a system can however rapidly increase depending on the number of external cavities that need to be locked to achieve sufficient suppression of the unwanted intermediate comb lines.

Semiconductor saturable absorber mirrors (SESAMs) enable robust stable passive soliton modelocking without any critical cavity stability criteria. However, at multi-gigahertz repetition rates, Q-switching instabilities are a principal concern. The high peak powers that occur during Q-switching instabilities damage the intracavity components before achieving sufficient power to exceed the threshold for stable continuous wave (cw) modelocking. To avoid these issues, V- and Z-shaped cavities have been designed with properly placed curved mirrors to avoid focusing on critical cavity elements. However, scaling these methods to the 10-GHz-regime has been elusive, since the small cavity size demands multi-functional optical components and severely limits their spatial arrangement.

A straight-cavity design with the SESAM as one of the end mirrors strongly relaxes those alignment constraints, but self-focusing of the beam and hence damage will occur in the cavity when operating in the conventional soliton modelocking regime. In this article, we introduce a new class of ultrafast lasers based on a simple straight cavity which solves the problems described above.

**Results**

**Self-defocusing straight-cavity design**

Instead of relying on the intrinsic (weak) nonlinearity of the gain medium and negative dispersion compensating elements (conventional soliton modelocking regime, Fig. 1a), we engineer a device with a nonlinear refractive index \( n_{2,\text{eff}} \) that is large and negative in sign, thus enabling femtosecond soliton modelocking at very low intracavity pulse energies and with net positive material dispersion (Fig. 1b).

Simultaneously, the negative \( n_{2,\text{eff}} \) implies a dynamic self-defocusing lens which suppresses damage: if an energetic pulse builds up due to Q-switching instabilities, the beam experiences strong self-defocusing, thus keeping the intensity below the damage threshold of the intracavity components.

A compelling approach to obtain such a tuneable self-defocusing nonlinearity is via the cascading of quadratic nonlinearities (CQN). In CQN, a second-harmonic generation (SHG) crystal is used to generate a second-harmonic wave, which subsequently acts back upon the fundamental wave, thereby simulating a Kerr-like nonlinearity with an effective nonlinear refractive index \( n_{2,\text{eff}} \). Varying the phase-mismatch of the SHG process allows the sign and magnitude of \( n_{2,\text{eff}} \) to be engineered, which has enabled soliton modelocking in the normal dispersion regime. A potential drawback of the CQN technique are the losses associated to residual SHG. The losses one obtains using conventional CQN, e.g. in birefringent crystals,
Pushing to multi-gigahertz repetition rates can easily exceed 0.5%\textsuperscript{34} which is intolerable for a SESAM-modelocked laser in the multi-gigahertz regime. The solution comes in the form of quasi-phase matching (QPM) materials: lithographic patterning of such materials now allows us to engineer a large, self-defocusing nonlinearity that is adiabatically excited via a non-uniform QPM structure in order to suppress the SHG losses by more than an order or magnitude (see Methods). The device used here consists of a 2-mm-long periodically poled lithium niobate crystal (PPLN) that is two-dimensionally patterned (Fig. 2). In earlier work\textsuperscript{33}, we have presented the operating principles of this type of device, whose functionality has now for the first time been leveraged to enable a new type of multi-gigahertz all-self-defocusing straight cavity.

Values for the effective nonlinear index $n_{2,\text{eff}}$ ranging from $-1 \times 10^{-18}$ m$^2$W$^{-1}$ to $-7 \times 10^{-18}$ m$^2$W$^{-1}$ can be achieved in this PPLN device, which is approximately two orders of magnitude higher than the intrinsic third-order material nonlinearities of conventional wide band-gap materials. For example, the nonlinear index of the gain material Yb:CALGO\textsuperscript{37} amounts only to $n_{2,\text{intrinsic}}^{\text{CALGO}} \approx +8 \times 10^{-20}$ m$^2$W$^{-1}$. The best modelocking results (presented in Fig. 3 and 4) were obtained at transverse position 2, indicated in Fig. 2a. This position corresponds to a grating vector $K_g=889$ mm$^{-1}$, leading to a phase mismatch of $44.9$ mm$^{-1}$ at 1052 nm. The net total intracavity GDD amounts to $\approx+1280$ fs$^2$ per roundtrip (material dispersion of the 2-mm-long PPLN and the 1.5-mm-long Yb:CALGO crystal, the SESAM was measured to have a reasonably flat GDD profile with a negligible contribution of $-66$ fs$^2$ at 1050 nm).

![Figure 2 | PPLN device. a, Illustration showing a two-dimensional variation of the quasi-phase matching (QPM) domain structure. The grating vector $K_g$ is varied smoothly along the x-direction to enable tuning of the nominal phase-mismatch and hence the effective nonlinear index $n_{2,\text{eff}}$. The variation in $K_g$ along the beam propagation direction has been designed in order to adiabatically turn the SHG interaction on/off: in the input region, the phase mismatch is adiabatically decreased to reach the value desired to obtain strong self-phase modulation (interaction region) and is then increased again in the output region to minimize the nonlinear losses. b, Calculated effective nonlinear index in the device as a function of wavelength for three different values of the phase mismatch, corresponding to the transverse positions (1) – (3) with respect to the incoming beam. The calculations are based on material data for MgO-doped lithium niobate reported by Gayer et al\textsuperscript{36}.](https://example.com/figure2.png)
Modelocking results

The lasing operation can be divided into two regions: at low pump power, mode-beating and Q-switching instabilities are observed. When increasing the pump power further, the laser reliably transitions into self-starting soliton modelocking at around 734 mW average output power (Fig. 4a) initiated and stabilized by the SESAM. Stable cw modelocking with a TEM$_{00}$ beam profile and an $M^2 = 1.01$ (measured with the Thorlabs BP104-IR beam profiler) is maintained up to an average power of 1.2 W. At this power, the laser can be operated for several hours a day without noticeable degradation of the modelocking parameters and repeatable performance over several months has been observed. A frequency-dependent amplitude noise measurement is shown in Supplementary Fig. 1. If the pump power is further increased, higher order spatial modes start to lase, which become visible in the laser beam profile and appear as sidepeaks in the radio-frequency (RF) trace. The onset of higher order modes can be explained by the strong beam divergence of the multi-mode pump within the length of the gain medium, which allows for non-TEM$_{00}$-modes to experience sufficient gain above a certain pump power. Using a pump with a lower $M^2$-value and/or a shorter gain crystal would mitigate this effect. Over this range, the duration of the transform-limited pulses decreases from 227 fs to 166 fs, consistent with the prediction of soliton modelocking, and the centre wavelength shifts from 1052 nm to 1050 nm. Shorter pulse durations could be achieved by decreasing the net positive intracavity dispersion, i.e. by using a Gires-Tournois-Interferometer (GTI)-type coating on the output coupler.

Figure 3 | Modelocking characterization of the 10-GHz Yb:CALGO laser at 1.2 W of average output power. a, intensity autocorrelation of the transform-limited 166 fs-pulses b, optical spectrum centered at 1050 nm with a bandwidth of 7 nm (full width at half maximum, FWHM) measured with a resolution bandwidth (RBW) of 0.08 nm. c, 5-MHz span microwave spectrum recorded with an RBW of 3 kHz showing the repetition rate signal at 10.62 GHz d, 50-GHz span microwave spectrum recorded with an RBW of 100 kHz.
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Laser cavity dynamics

To understand the threshold for cw modelocking, the influence of soliton shaping and gain filtering, as well as the role of the PPLN device on the Q-switching threshold has been quantified using numerical simulations (Fig 4b). These results show that the PPLN device helps to reduce the Q-switching threshold (see Methods). However, a low Q-switching threshold alone is not sufficient to guarantee successful modelocking. Damage may still occur due to Q-switching instabilities that inevitably build up inside the cavity while ramping up the pump power. By exploiting dynamical self-defocusing effects, we are able to suppress damage while transitioning through this regime. In Fig. 5a we show the mode radius on all the cavity elements as a function of the negative Kerr lens, which is induced in the PPLN device. The Kerr lens power $F$ can be expressed as a function of the pulse energy $E_p$, the corresponding soliton pulse duration $\tau_{\text{FWHM}}$, the mode radius in the PPLN $\omega_{\text{PPLN}}$ and the length of the PPLN crystal $L_{\text{PPLN}}$:

$$F = 1.76 \frac{n_{\text{eff}} L_{\text{PPLN}} E_p}{\pi \omega_{\text{PPLN}}^4 \tau_{\text{FWHM}}}$$

(1)
This defocusing lens depends on $n_{2,\text{eff}}$ (Fig. 5b), and thus on the position of the PPLN as described in Fig. 2. As a result of this dynamic defocusing Kerr-lens, the fluence on the SESAM does not increase linearly anymore, but instead shows a clamping behaviour as the mode size increases (Fig. 5c). Moreover, the laser mode size in the gain crystal also increases with stronger Kerr lensing in the PPLN, and hence for a fixed pump power and pump mode size, the gain will eventually drop below unity at a certain pulse energy (Fig. 5d). Based on this energy, we estimate the maximum fluence that may occur during Q-switching (yellow crosses in Fig. 5). For the experimental modelocking parameters (PPLN at position 2) and for a pump power corresponding to the Q-switching threshold, we obtain an upper limit of approximately 40 nJ, beyond which the pulse energy is clamped (the same calculation was performed to obtain the clamping points in Fig. 5c for the other values of $n_{2,\text{eff}}$).

**Figure 5 | Effect of the dynamic defocusing lens in the PPLN on various parameters.**

- **a**, Intracavity mode radius as function of PPLN Kerr lens power.
- **b**, PPLN Kerr lens as function of intracavity pulse energy for the three different PPLN grating vector values $K_g$ of (1) 854 mm$^{-1}$, (2) 889 mm$^{-1}$ and (3) 910 mm$^{-1}$. At a wavelength of 1052 nm, these values correspond to effective nonlinear indices $n_{2,\text{eff}}$ of (1) $-9.99 \times 10^{-18}$ m$^2$W$^{-1}$, (2) $-2.07 \times 10^{-18}$ m$^2$W$^{-1}$ and (3) $-4.24 \times 10^{-18}$ m$^2$W$^{-1}$ respectively.
- **c**, Fluence on the SESAM without and with the effect of the defocusing lens. The grey shaded area corresponds to the region where damage of the SESAM is likely to occur.
- **d**, Gain $g$ (ratio of input intensity to output intensity after two passes through the gain crystal) as function of intracavity pulse energy. Solid line: with lens effect; dashed line: assuming a constant mode size in the Yb:CALGO. Above a certain intracavity pulse energy (grey area), no more gain is experienced by the pulses.
- **e**, Effective intracavity SESAM reflectivity for PPLN position (2): for each pulse energy, the corresponding change in mode size and pulse duration is taken into account. The experimentally observed cw modelocking region from 2.4 to 4 nJ is shaded in grey.
Next we evaluate the response of the SESAM. The AlAs-embedded single-InGaAs-quantum well SESAM used has a saturation fluence of \( F_{\text{sat}} \approx 8 \, \mu \text{J/cm}^2 \), a modulation depth \( \Delta R \approx 1 \% \), non-saturable losses \( \approx 0.12 \% \) and an inverse saturable absorption coefficient \( F_2 \approx 500 \, \text{mJ/cm}^2 \). These parameters were extracted from fitting the SESAM reflectivity curve for a Gaussian beam\(^{42}\) to the reflectivity values measured at a fixed 170-fs pulse duration and constant spot size on the SESAM. To mimic the real response of the SESAM inside the 10-GHz cavity, we need to incorporate the change in mode size due to the PPLN Kerr lens for each energy point. Additionally, since the pulse duration of the soliton decreases with energy, the pulse-duration-dependent \( F_2 \) coefficient\(^{46}\) needs to be scaled for each energy value as well. This leads to the effective intracavity SESAM reflectivity curve shown in Fig. 5e. This curve shows how the lens effect prevents operation too far into the rollover of the reflectivity, which in turn prevents strong absorption in the SESAM and thus potential damage.

**Discussion**

In conclusion, by combining SESAM modelocking with adiabatic excitation of cascaded quadratic nonlinearities, we overcome the issues faced by conventional femtosecond modelocked lasers that have strongly limited access to 10-GHz repetition rates until now. We demonstrated a straight-cavity 10-GHz SESAM-modelocked laser that operates in the positive GDD regime. The intracavity multi-functional 2D-QPM PPLN crystal shapes the pulses in time by providing negative SPM for soliton formation while minimizing the intracavity loss, and simultaneously acts as a dynamic self-defocusing lens, which protects the cavity elements from Q-switching related damage. Combining these effects allowed us to achieve for the first time femtosecond Watt-level fundamental modelocking of a diode-pumped solid-state laser with a repetition rate above 10 GHz. The SESAM as well as the PPLN device are fabricated on a wafer scale, with the custom QPM pattern on the latter defined by a lithographic mask. The pulsed nature of this 1-µm laser source makes it well suited to various frequency conversion techniques with novel nonlinear platforms, such as low-energy supercontinuum generation in silicon nitride waveguides\(^{43}\) or high-gain optical parametric amplification\(^{44}\) to reach the mid-infrared spectral region. The modelocking technique based on self-defocusing nonlinearities can furthermore be deployed at other wavelengths and is expected to be especially favourable at longer wavelengths due to the reduced group velocity mismatch between the fundamental and the second harmonic in the PPLN crystal. The new class of ultrafast lasers enabled by this technique represents an important step towards compact high-power optical frequency combs beyond 10-GHz.
Methods

Laser cavity details

The 1.5-mm-long Yb:CALGO gain crystal is pumped by an internally wavelength-stabilized, spatially multimode diode (Lissotschenko Mikrooptik GmbH, M$^2$≈36) capable of providing up to 60 W at 980 nm. The pump beam is focused through a 12-mm-radius pump-transparent mirror that acts as a 2.8% output coupler for the 1050 nm laser centre wavelength. In order to minimize the thermal load inside the laser cavity, the vertically polarized pump light, which would only be weakly absorbed in the gain crystal, is removed using a polarizing beam splitter. The beam splitter at the same time allows the vertically polarized laser beam to exit.

PPLN device operating principle

The principle behind the self-defocusing nonlinearity is phase-mismatched second-harmonic generation (SHG), often referred to as cascading of quadratic nonlinearities (CQN). Far from phase-matching, the fundamental experiences an effective third-order nonlinear response that can be expressed as a nonlinear index contribution $n_{2,CQN}$ tunable via the phase-mismatch $\Delta k$. Adding it to the intrinsic material nonlinear index $n_{2,intr}$, yields the total effective nonlinear index $n_{2,eff}$:

$$n_{2,eff} = n_{2,intr} + n_{2,CQN} = n_{2,intr} - \frac{4\pi d_{eff}^2}{c n_{F/SH}^2 n_{SH}^2 \lambda_F^2 \Delta k}$$  \hspace{1cm} (2)$$

($\lambda_F$: wavelength of the fundamental, $n_{F/SH}$: linear refractive index at the fundamental/second harmonic, $d_{eff}$: effective nonlinear coefficient).

In our PPLN device, the QPM periods vary transversely across the chip. By translating this 2D-QPM PPLN device with respect to the incident laser beam, a tunable phase mismatch $\Delta k(x) = k_{SH} - 2k_F - k_{F}(x)$ can be obtained, where $k_{F/SH} = 2\pi n_{F/SH} / \lambda_{F/SH}$ denotes the wave vector of the fundamental wave (F) and its second harmonic (SH) respectively.

In order to obtain the largest magnitude of $n_{2,eff}$, the phase mismatch $\Delta k$ has to be made as small possible within the validity of the CQN regime. However, in a normal CQN device, the fraction of power lost to SHG increases according to $\alpha \sim 1/(\Delta k)^2$. Such losses resemble an inverse saturable absorption in the laser, limiting the nonlinearity available before mode-locked operation is destabilized. This issue is especially critical at high repetition rates, where large nonlinearities are required due to the low intracavity pulse energy.

We have developed a new technique to suppress these losses: adiabatic excitation of quadratic solitons\textsuperscript{33}. In this technique, the QPM period is rapidly but smoothly moved far from phase-matching at the input and output sides of the device. In this way, the second-harmonic light, which mediates the large effective nonlinear refractive index, is adiabatically switched “on” via the input segment, is large in the middle segment (leading to a large and negative $n_{2,eff}$ in that segment), and is switched “off” via the output segment, as illustrated in Fig. 2 of the main text. With this technique, the resulting nonlinear losses can be decreased by an order of
Pushing to multi-gigahertz repetition rates

magnitude or more compared to a non-apodized device. The nonlinear chirp QPM profiles used are analogous to apodization in chirped QPM devices, where they efficiently excite adiabatic three-wave mixing processes.

**PPLN operating point**

In our PPLN device, the value of the grating k-vector \( K_g \) in the interaction region (as indicated in Fig. 2a) varies continuously across the transverse position in the device (i.e. perpendicular to the beam propagation direction). This value varies from 854 mm\(^{-1} \) to 934 mm\(^{-1} \) across the transverse profile of the device. The three example operating points depicted in Fig. 2a and the corresponding values of \( n_{2,\text{eff}} \) in Fig. 2b illustrate the trade-offs involved in optimizing modelocking performance. When operating closer to position (1) (large phase-mismatch, \( K_g=854 \) mm\(^{-1} \)), the self-defocusing lens effect becomes too weak and \( Q \)-switching damage can be observed (Fig. 5c). When moving towards a too-small phase mismatch (position (3), \( K_g=910 \) mm\(^{-1} \)), the nonlinear losses increase and are accompanied by a self frequency shift effect. These effects cause the laser to react by shifting to a shorter centre wavelength, which however at the same time moves the pulse away from the spectral region with highest gain in the Yb:CALGO crystal. Optimum modelocking performance was obtained between these extrema, with the PPLN device placed in position (2) (\( K_g=889 \) mm\(^{-1} \)).

**Numerical prediction of the \( Q \)-switching threshold**

In a general sense, stable cw modelocking can be characterized as a state where small deviations around the steady-state gain \( g_s \) and pulse energy \( E_s \) will be damped by the system, instead of being amplified as it is the case for \( Q \)-switching instabilities. Mathematically, these linearized equations for pulse energy \( E \) and gain \( g \) can be expressed in vector form,

\[
T_R \frac{d}{dt} \begin{pmatrix} \Delta E \\ \Delta g \end{pmatrix} = A \begin{pmatrix} \Delta E \\ \Delta g \end{pmatrix}, \quad A = \begin{pmatrix} \frac{\partial G}{\partial E} & \frac{\partial G}{\partial g} \\ -R & \frac{1}{E_{\text{sat},L}} + \frac{T_R}{\tau_L} + \frac{E}{E_{\text{sat},L}} \end{pmatrix}
\]

(3)

where \( T_R \) denotes the cavity round-trip time, \( E_{\text{sat},L} \) the saturation energy of the laser gain medium, \( \tau_L \) is the upper-state lifetime of the gain medium (420 \( \mu \)s for Yb:CALGO\(^{37} \)) and \( G \) corresponds to the total gain (sum of all cavity gain and loss terms), which is zero for the steady state. For stability, the trace of this matrix needs to be negative. The \( Q \)-switching threshold condition above leads to the following (often cited) simplified equation:

\[
E_{\text{th}}^2 = E_{\text{sat},A} \Delta R \left( 1 + \frac{1}{A_A F_2} \right)
\]

(4)

\( E_{\text{sat},A} \): saturation energy of the SESAM, \( A_A \): beam area on SESAM, \( F_2 \): inverse saturable absorption coefficient, \( \Delta R \): modulation depth). Evaluating this expression for the parameters...
of this 10-GHz laser yields a predicted intracavity pulse energy threshold of 9.9 nJ, corresponding to an average output power of 2.9 W (red dashed line in Fig. 4), which is about 4 times higher than we experimentally observe. Although Eq. 4 may be convenient to use, it does not provide the full picture.

We have developed a complete calculation of trace(\(A\)) in the steady-state for quasi-three-level mode-locked lasers. We include soliton shaping, gain filtering using directly measured cross section data for Yb:CALGO\(^{49}\), as well as the exact SESAM and PPLN responses, accounting for the transverse beam profile of the laser and pump beams in each intracavity component (a flat-top pump beam is assumed for simplicity).

Neglecting the PPLN crystal losses, this precise calculation already reduces the predicted threshold to an average power of \(~900\) mW. To further improve the accuracy, the total gain \(G\) needs to contain the residual nonlinear SHG losses in the PPLN crystal, which are analogous to inverse saturable absorption effects. Although the nonlinear losses are minimized by QPM design as described above, we need to assume random duty cycle (RDC) errors due to imperfect fabrication\(^{39}\). Assuming a realistic QPM period jitter of 0.2 \(\mu\)m increases the nonlinear losses from ideally 0.01 % to 0.05%. Including these RDC errors, the predicted \(Q\)-switching threshold output power drops to 724 mW, which is in very good agreement with the experimentally observed threshold.

**Data availability**

The data and simulations codes that support the findings of this study are available from the corresponding authors upon request.

**Acknowledgements**

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**Author contributions**

A.S.M designed the 10 GHz laser cavity and carried out the measurements. C.R.P designed the PPLN device. C.R.P and A.S.M performed the numerical simulations. U.K. led the project. All authors contributed to the final manuscript.

**Competing financial interests**

The authors declare no competing financial interests.
References


Supplementary Note 1

The amplitude noise during clean cw modelocking is very similar to the noise measured in our SESAM modelocked 1-GHz Yb:CALGO laser\(^1\), which operated in the conventional negative-dispersion soliton regime, and did not contain a PPLN crystal. Hence, the cascaded quadratic nonlinearities (CQN) modelocking regime we have accessed during this work does not seem to alter the amplitude noise in any significant way. Both lasers are pumped with the same multimode pump diode, which – as can be seen in Fig. S1 - is the limiting factor for the noise of both lasers. However, by providing feedback to the pump current, we have shown in Klenner et al.\(^1\) that this noise can be handled, leading to a tight lock of the offset frequency of the 1-GHz laser. At 10 GHz, a broader feedback bandwidth is expected to be needed due to the scaling of the phase noise with the repetition rate, but we expect that using suitable electronics, this should still be feasible. Stabilization of the repetition rate frequency could furthermore be implemented by mounting the SESAM on a piezo-element to stabilize the length of the cavity as demonstrated recently by Hakobyan et al.\(^2\).

![Amplitude noise comparison](image)

**Supplementary Figure 1** | Amplitude noise comparison. Amplitude noise of the 10 GHz laser (red) vs. the 1 GHz Yb:CALGO laser (turquoise) and the spatially multimode pump diode (blue). The root-mean-square (rms) noise integrated over the interval (1 Hz, 1 MHz) amounts to: 10 GHz: 0.019 %, 1 GHz: 0.010 %, pump: 0.017 %

Supplementary References

4.4 Prototype development and performance scaling

4.4.1 Compact 10-GHz prototype for full comb stabilization

In paper 4, we presented a 10-GHz Yb:CALGO straight-cavity modelocked laser enabled by a self-defocusing PPLN device. In order to use this laser as a stable frequency comb source, two main challenges have to be addressed: the stabilization of the pulse repetition rate as well as the detection and stabilization of the comb offset. The first requirement is strongly coupled to the mechanical stability of the system. The proof-of-principle version of the laser used for the results of Paper 4 had not been optimized for compactness and stability yet, but rather for flexibility of alignment and interchangeability of components during the development phase. Hence, as a next step, we designed a prototype version of the laser cavity with optimized mechanical properties. In order to enhance the passive stability, it is advantageous to remove as many mechanical stages with translational or rotational degrees of freedom as possible, which however restricts the possibilities for fine-alignment of the laser. We thus evaluated the essential degrees of freedom (see Figure 4.3) and elaborated mechanical mounting solutions that combine compactness and stability with the necessary adjustment precision.

![Figure 4.3: Mechanical degrees of freedom required for the alignment of the 10-GHz laser cavity.](image-url)
The laser housing was manufactured from a single block of aluminum. The bottom of the case has a height profile which was milled according to the height of the different necessary mounts in order to keep all components as close to the monolithic base plate as possible (Figure 4.4). The pump fiber, the water tubes as well as the electrical connections can directly be plugged into the housing.

Figure 4.4: 10-GHz Yb:CALGO prototype laser with active cavity length stabilization mechanism. (a) CAD design. (b) Realization.

As described in the previous sections, the Q-switching behavior is strongly dependent on the SESAM properties, i.e. the modulation depth and saturation fluence. The latter can be tuned via the temperature, since heating/cooling will slightly shift the center wavelength of the absorber layer (~0.3 nm/°C [100]) and thus move the absorption dip with respect to the laser center wavelength. To achieve better control of the Q-switching threshold, we have implemented temperature control of the SESAM. A technical challenge arises from the fact that the SESAM needs to be placed on a Piezo-electric actuator for precise cavity length control. Since the Piezo-ceramics has a poor thermal conductivity, a heat bridge using copper wires was used to connect the SESAM on the Piezo to the Peltier-controlled and water-
cooled heat sink below. This mounting solution allowed us to obtain successful preliminary repetition rate stabilization results.

4.4.2 Pushing the laser performance

In terms of average output power and pulse duration, the laser presented in Paper 4 was limited by two factors: the divergence of the multimode pump diode and the net intracavity dispersion. In this section, we present follow-up work that aimed at testing the effect of a pump diode with lower $M^2$ (i.e. smaller divergence) as well as reducing the intracavity dispersion using custom-made GTI-type mirrors.

**Pump diode comparison**

The specifications of the new pump diode and the one used for the results of Paper 4 are compared in Table 3.

<table>
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<th>Lissotschenko Mikrooptik (LIMO, Paper 4) LIMO60-F100-DL980-EX1931</th>
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<td>Center wavelength [nm]</td>
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</tr>
<tr>
<td>Linewidth [nm]</td>
<td>1</td>
<td>0.6</td>
</tr>
<tr>
<td>Wavelength-stabilized</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>$M^2$</td>
<td>36</td>
<td>17</td>
</tr>
<tr>
<td>Threshold current [A]</td>
<td>6.0</td>
<td>0.6</td>
</tr>
<tr>
<td>Fiber core diameter [µm]</td>
<td>100</td>
<td>105</td>
</tr>
<tr>
<td>Numerical aperture (NA)</td>
<td>0.22</td>
<td>0.15</td>
</tr>
</tbody>
</table>

Table 3: Specifications of two spatially multimode pump diode used to pump the 10 GHz Yb:CALGO laser.

Both feature internal wavelength stabilization and exhibit similar amplitude noise performance. They mainly differ with respect to their beam divergence; while the LIMO diode has an $M^2 \approx 36$, the BWT diode exhibits an $M^2 \approx 17$. In Figure 4.5, we show how the change in $M^2$ affects the mode overlap between the pump beam and
the fundamental Gaussian laser cavity mode ($M^2 = 1$) in the 1.5-mm-long Yb:CALGO gain crystal. While the LIMO diode shows significant divergence within the length of the crystal, the mode delivered by the BWT diode is still decently overlapped with the laser cavity mode. Using the exact same laser cavity configuration, but switching the pump diodes led to an increase in slope efficiency of >8% and an increase of the maximum average output power in cw modelocking of +20%. The latter is a consequence of the improved mode overlap, which shifts the onset of higher order laser modes towards higher pump powers, as predicted in Paper 4.

Figure 4.5: Pump diode comparison.(a) Overlap of the pump beam and the fundamental Gaussian laser mode in the Yb:CALGO gain crystal. (b) Output power slope presented in Paper 4 (pink/red) compared to output power slope using BWT diode (light/dark blue).

**Reducing the intracavity dispersion**

According to the soliton formula (Eq.(1.10)), the pulse duration linearly scales with the amount of net intracavity GDD. The pure material dispersion in the 10-GHz cavity adds up to +1280 fs$^2$. To reduce the GDD, we replaced the input/output coupling mirror by mirrors with the same curvature (12 mm) and transmission properties, but with an additional dispersion-compensating coating. Figure 4.6(a) depicts the effect on the pulse duration and the power slope. With the coating providing a GDD of -400 fs$^2$, the round trip GDD is reduced to 880 fs$^2$ and we obtain a minimum pulse duration of 148 fs at a maximum output power of 1.18 W. Using a coating with a GDD of -800 fs$^2$, we manage to achieve a spectral bandwidth (FWHM) of 11.2 nm, which supports pulses as short as 104 fs. Due to the material dispersion of the beam splitter, through which the pulse train has to exit (see Paper 4), the
measured pulses are chirped to 126 fs. In this configuration however, the maximum average output power was limited to 810 mW. The limitation can be explained by considering the spectral evolution as a function of power (depicted in Figure 4.6(b)). Above the cw modelocking threshold, the spectrum starts to broaden with increasing pulse energy as expected for a soliton. As a result of this process, the long-wavelength edge will get closer to phase-matching in the PPLN crystal (see wavelength-dependence of the nonlinear refractive index shown in Paper 4), which in turn leads to higher nonlinear losses. This will in turn induce a shift of the center wavelength towards shorter wavelengths to avoid these losses. Ultimately, instabilities occur when the wavelength reaches the edge of the gain bandwidth and/or shifts out of the optimum operation range of the SESAM and the output coupler.

![Figure 4.6](image.png)

Figure 4.6: Dispersion and spectral evolution. (a) Comparison of the laser slope for 3 different values of intracavity dispersion: +1280 fs² (no GTI), +880 fs², and +480 fs². (b) Experimentally recorded spectral evolution as a function of the optical output power.
In this thesis, we have discussed the newest advancements in frequency combs based on compact modelocked lasers with GHz repetition rates. In particular, we focused on exploiting the potential of novel nonlinear optical devices to achieve goals such as repetition rate scaling, self-referencing and wavelength conversion.

The lasers presented in this work were based on Yb-doped CALGO, a material that has received increasing attention in recent years due to its advantageous thermal properties as well as its broad and smooth gain bandwidth. This favorable combination allows for the generation of very short pulses (<40 fs) and output powers reaching several Watts. The fact that this material can be pumped at a wavelength of 980 nm using low-cost power-scalable multi-mode diodes represents a significant advantage over e.g. Ti:sapphire laser systems, which have been the state-of-the art high-peak power laser oscillator technology for scientific frequency comb applications so far. Although some progress has recently been achieved in pumping Ti:sapphire laser using green or blue diodes instead of expensive frequency-doubled single mode solid-state lasers [101, 102], the material intrinsically suffers from a large quantum defect, which ultimately limits its power-scalability.

Modelocking was achieved in our lasers using SESAMs, an approach that allows for self-starting and reliable modelocking. The SESAMs were designed, grown and characterized in-house. One of the main challenges when developing SESAM-modelocked lasers at high repetition rates is their inherent susceptibility to Q-switching instabilities. This issue, paired with the tight geometrical constraints and insufficient SPM at low pulse energies, has prevented the scaling of femtosecond Watt-level SESAM-modelocked lasers to repetition rates > 5 GHz for a long time. In this thesis, we introduced a novel approach that overcomes these issues and
demonstrated the first femtosecond Watt-level operation of a DPSSL at 10-GHz repetition rate. Core to the approach is the intracavity use of a 2D-QPM PPLN device that fulfills two functions: providing sufficient SPM for femtosecond modelocking via cascading of quadratic nonlinearities, as well as generating a dynamic defocusing lens that protects the cavity elements from damage in case of Q-switching instabilities. The straight cavity design enabled by this approach is the simplest possible configuration to align and allowed us to implement cavity length control and hence the stabilization of the pulse repetition rate.

In addition to the laser development itself, a significant part of this thesis was devoted to CEO detection and stabilization at GHz repetition rates, in particular with the aim of reducing the peak power required for self-referencing. In collaboration with the group of Prof. A. Gaeta and Prof. M. Lipson at Columbia University, we used our 1-GHz Yb:CALGO laser to demonstrate the first coherent octave-spanning supercontinuum from Si$_3$N$_4$ waveguides. The pulse energy required to obtain this supercontinuum was more than an order of magnitude lower than for the best silica fibers available on the market. Subsequently, this supercontinuum also allowed us to perform an $f$-to-2$f$ based CEO detection. We found that wavelength-stabilization of the diode (not affecting the available pump power) reduced the linewidth of the free-running CEO by an order of magnitude, which significantly relaxes the requirements on the stabilization loop electronics. Hence, we could demonstrate that, contrary to popular belief, low-noise CEO stabilization of a 1-GHz DPSSL is possible even with highly spatially multimode pump diodes.

Last but not least, we also investigated the possibility of transferring our robust 1-GHz Yb:CALGO laser into the mid-IR spectral region while avoiding additional amplification of the pulses. An elegant solution was found in the form of PPLN waveguides, which we used in an OPA scheme with both the pump and signal beam originating from the same laser. This configuration allowed us to generate a tunable offset-free 1-GHz-comb ranging from 2.5 µm to 4.2 µm, with up to 4 µW of power per comb line around 3.5 µm. The experimental results were presented along with guidelines on how to leverage the waveguide properties in order to achieve high (> 35 dB) amplification of the signal wavelength and thus efficiently convert the IR comb into the mid-IR at low energy.
The applications of frequency combs are numerous and several examples were cited in the papers presented in this thesis. For instance, they constitute a promising tool for the spectroscopic detection of gases. A technique that relies on the combination of two combs, called dual-comb spectroscopy [103, 104], has rapidly evolved over the past few years [105]. Its main advantage is that it translates the comb line structure from the optical domain to the more easily measurable radiofrequency domain by beat signal generation of the comb with a second comb of slightly different line spacing (i.e. pulse repetition rate). Recently, results from our research group have shown that it is possible to obtain such a dual-comb setup using only one laser cavity [106] and a proof-of-principle demonstration of water vapor spectroscopy was carried out [107]. These results were obtained using semiconductor disk laser technology, which shows a lot of potential in terms of compactness and wafer-scale producibility. However, the peak power performance of these lasers had not yet reached the level necessary for direct frequency comb self-referencing.

Building upon the work presented in this thesis, we have now achieved preliminary results using a second generation of Si₃N₄ waveguides with adapted dispersion properties to self-reference a 1.6-GHz VECSEL-based frequency comb without any external pulse amplification [108]. This represents a key milestone in the development of compact GHz frequency comb sources. Further analysis of the free-running and stabilized noise performance may soon reveal interesting insights regarding the impact of the gain dynamics on the frequency comb properties, since e.g. the gain lifetime of the semiconductor medium (~ns) considerably differs from the one of rare-earth doped media such as Yb:CALGO (~few 100 µs).

At the same time, the development Yb:CALGO lasers for GHz applications has now reached a state where engineered prototypes such as the one presented at the end of this thesis are being developed and optimized for applications that benefit from robust and transportable frequency comb sources.

In addition, we hope that the work on the different low-energy nonlinear platforms discussed in this thesis will ultimately also benefit the research that is being carried out on developing on-chip compact light sources and optical communication systems, which can certainly be considered one of the hottest technology topics of our decade.
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Curriculum Vitæ

Name: Aline Sophie Mayer
Date of Birth: April 14, 1990 (Basel, Switzerland)
Nationality: Swiss/French

Education

Feb. 2014 – Feb. 2018 Ph.D. studies in the Ultrafast Laser Physics group of Prof. Dr. Ursula Keller, Institute of Quantum Electronics, Department of Physics, ETH Zürich, Switzerland
July 2012 – Nov. 2013 Master studies in Physics, ETH Zürich, Zürich, Switzerland
Jan. – July 2012 Master studies exchange semester, Kungliga Tekniska Högskolan (KTH), Stockholm, Sweden
Sept. 2008 – Aug. 2011 Bachelor studies in Physics, ETH Zürich, Zürich, Switzerland
June 2008 Matura, Gymnasium Kirschgarten, Basel, Switzerland

Research and Professional Experience

Feb. 2014 – Feb. 2018 Ph.D. studies in the Ultrafast Laser Physics group of Prof. Dr. Ursula Keller, Institute of Quantum Electronics, Department of Physics, ETH Zürich, Switzerland

Leveraging Novel Nonlinear Optical Devices for Compact Gigahertz Frequency Combs

Feb. 2014 – Feb. 2018 Teaching assistant at the Department of Physics, ETH Zürich, Zürich, Switzerland

July 2013 – Nov. 2013 Master thesis in the Ultrafast Laser Physics group of Prof. Dr. Ursula Keller, Institute of Quantum Electronics, Department of Physics, ETH Zürich, Switzerland

Soliton Modelocking by Cascading of Second-Order Nonlinearities

Aug. 2012 Semester thesis in the Ultrafast Laser Physics group of Prof. Dr. Ursula Keller, Institute of Quantum Electronics, Department of Physics, ETH Zürich, Switzerland

Second Harmonic Generation FROG

Sept. 2011 - Dec. 2011 Internship in the Macromolecular Crystallography group of Dr. Meitian Wang at the Paul Scherrer Institute (PSI), Villigen, Switzerland

Development of capacitive and optical measurement and calibration procedures for the high-precision positioning systems at the PX synchrotron beamline endstations

Awards

Nov. 2015 Overall best student paper at the Advanced Solid-State Lasers conference (ASSL), Berlin, Germany
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