Towards high average power sesam-modelocked thin-disk lasers with short pulse durations

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TOWARDS HIGH AVERAGE POWER
SESAM-MODELOCKED THIN-DISK LASERS WITH SHORT
PULSE DURATIONS

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

presented by
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2015
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Symbols

\( A_A \)  \hspace{1cm} \text{mode area on the absorber} \\
\( A_L \)  \hspace{1cm} \text{mode area on the laser medium} \\
\( D \)  \hspace{1cm} \text{GDD per roundtrip} \\
\( |E_p(z)|^2 \)  \hspace{1cm} \text{field intensity enhancement} \\
\( E_p \)  \hspace{1cm} \text{pulse energy} \\
\( E_{\text{sat},A} \)  \hspace{1cm} \text{saturation energy of the absorber} \\
\( E_{\text{sat},L} \)  \hspace{1cm} \text{saturation energy of the laser medium} \\
\( F_2 \)  \hspace{1cm} \text{induced absorption coefficient} \\
\( F_{\text{dam}} \)  \hspace{1cm} \text{fluence at damage threshold} \\
\( f_{\text{rep}} \)  \hspace{1cm} \text{pulse repetition rate} \\
\( F_{\text{sat},A} \)  \hspace{1cm} \text{saturation fluence of the absorber} \\
\( F_{\text{sat},L} \)  \hspace{1cm} \text{saturation fluence of the laser medium} \\
\( F_t \)  \hspace{1cm} \text{transparency fluence} \\
\( g \)  \hspace{1cm} \text{round-trip power gain} \\
\( I \)  \hspace{1cm} \text{intensity} \\
\( m_L \)  \hspace{1cm} \text{number of passes on the gain per cavity} \\
\hspace{6cm} \text{round-trip} \\
\( n \)  \hspace{1cm} \text{refractive index} \\
\( n_2 \)  \hspace{1cm} \text{nonlinear refractive-index coefficient} \\
\( N_t \)  \hspace{1cm} \text{transparency density}
**Acronyms**

\[ P_{av} \] average power  
\[ P_{peak} \] peak power  
\[ R_{\text{lin}} \] reflectivity of unbleached absorber  
\[ R_{\text{ns}} \] reflectivity of fully bleached absorber  
\[ S \] ratio of pulse- to saturation fluence  
\[ \text{sech} \] hyperbolic secant function  
\[ T_R \] resonator round-trip time  
\[ \beta_{\text{TPA}} \] two-photon absorption coefficient  
\[ \Delta f_g \] FWHM gain bandwidth  
\[ \Delta \nu \] FWHM spectral pulse width in frequency domain  
\[ \Delta R \] modulation depth  
\[ \Delta R_{\text{ns}} \] nonsaturable losses  
\[ \gamma_{\text{SPM, cav}} \] SPM coefficient per roundtrip  
\[ \kappa \] thermal conductivity  
\[ \lambda_c \] central laser wavelength  
\[ \nu_c \] central laser frequency  
\[ \Phi \] soliton phase shift per roundtrip  
\[ \sigma_{\text{abs}, L} \] absorption cross-section at \( \nu_c \)  
\[ \sigma_{\text{em}, L} \] emission cross-section at \( \nu_c \)  
\[ \tau_A \] recovery time of a saturable absorber  
\[ \tau_{1/e} \] 1/\( e \) recovery time of a SESAM  
\[ \tau_{\text{fast}} \] short recovery time of a SESAM  
\[ \tau_{\text{p}} \] FWHM pulse duration  
\[ \tau_{\text{slow}} \] long recovery time of a SESAM  
\[ \xi_{\text{abs}} \] field intensity enhancement at the position of absorber section

**Acronyms**

AIAs aluminium arsenide  
AOM acousto-optic modulator  
AR anti-reflective  
CALGO calcium gadolinium aluminium oxide (CaGdAlO\(_4\))  
CEO carrier-envelope offset  
CPA chirped-pulse amplification  
CTE coefficient of thermal expansion  
cw continuous wave  
Cz Czochralski
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<td>DBR</td>
<td>distributed Bragg reflector</td>
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<tr>
<td>DPSSL</td>
<td>diode-pumped solid-state laser</td>
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<tr>
<td>FWHM</td>
<td>full width at half maximum</td>
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<tr>
<td>GaAs</td>
<td>gallium arsenide</td>
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<tr>
<td>GDD</td>
<td>group-delay dispersion</td>
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<tr>
<td>GTI</td>
<td>Gires-Tournois interferometer</td>
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<tr>
<td>HEM</td>
<td>heat exchanger method</td>
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<td>HHG</td>
<td>high-order harmonic generation</td>
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<td>HR</td>
<td>high-reflective</td>
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<td>IA</td>
<td>induced absorption</td>
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<td>InGaAs</td>
<td>indium gallium arsenide</td>
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<td>KLM</td>
<td>Kerr-lens modelocking</td>
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<td>LT</td>
<td>low temperature</td>
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<td>LuO</td>
<td>lutetium oxide (Lu$_2$O$_3$)</td>
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<tr>
<td>LuScO</td>
<td>lutetium scandium oxide (LuScO$_3$)</td>
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<tr>
<td>MBE</td>
<td>molecular beam epitaxy</td>
</tr>
<tr>
<td>OPCPA</td>
<td>optical parametric chirped-pulse amplification</td>
</tr>
<tr>
<td>PECVD</td>
<td>plasma-enhanced chemical vapor deposition</td>
</tr>
<tr>
<td>Q-factor</td>
<td>quality factor</td>
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<tr>
<td>QD</td>
<td>quantum dot</td>
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<tr>
<td>QML</td>
<td>Q-switched modelocking</td>
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<td>QW</td>
<td>quantum well</td>
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<tr>
<td>RBW</td>
<td>resolution bandwidth</td>
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<tr>
<td>ROC</td>
<td>radius of curvature</td>
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<tr>
<td>RSU</td>
<td>right-side up</td>
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<tr>
<td>ScO</td>
<td>scandium oxide (Sc$_2$O$_3$)</td>
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<tr>
<td>ScYLO</td>
<td>scandium yttrium lutetium oxide ((Sc,Y,Lu)$_2$O$_3$)</td>
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<td>SESAM</td>
<td>semiconductor saturable absorber mirror</td>
</tr>
<tr>
<td>SiC</td>
<td>silicon carbide</td>
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<tr>
<td>SNR</td>
<td>signal-to-noise ratio</td>
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<tr>
<td>SPM</td>
<td>self-phase modulation</td>
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<tr>
<td>STB</td>
<td>substrate-transfer and direct bonding</td>
</tr>
<tr>
<td>Acronym</td>
<td>Description</td>
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<tr>
<td>TDL</td>
<td>thin-disk laser</td>
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<td>TPA</td>
<td>two-photon absorption</td>
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<td>USD</td>
<td>up-side down</td>
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<td>VBG</td>
<td>volume Bragg grating</td>
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<td>YAG</td>
<td>yttrium aluminium garnet ($Y_3Al_5O_{12}$)</td>
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<td>YScO</td>
<td>yttrium scandium oxide ($YScO_3$)</td>
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<td>ZPL</td>
<td>zero-phonon line</td>
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Publications

Parts of this thesis are published in the following journal papers and conference proceedings.

**Journal Papers**


Publications


Conference Papers


Abstract

Many years of research resulted in the dramatic progress made in ultrafast laser technologies. High-brightness, pico- and femtosecond laser systems, which provide average power levels of several 100 W have become an essential part of the high-value manufacturing industry. The combination of high peak intensity and short pulse duration enables unprecedented accuracy in the processing of a variety of materials, including metals, semiconductors, display glass and polymers.

Moreover, the great success story of high-power ultrafast laser systems has provided peak intensities that correspond to the strength of electron binding fields and has opened the door to novel research areas, such as attosecond science. This relatively new sub-field of strong-field physics relies on optical attosecond pulses in the extreme ultraviolet and soft X-ray regime, which are typically produced by means of high-order harmonic generation (HHG). Today, the driving sources of HHG are based on complex amplifier systems and operate typically at low repetition frequencies in the kilohertz regime. As a consequence, experiments suffer from the long acquisition times required to obtain a large signal to noise ratio. Inspired by the growing need for table-top megahertz-driving sources for HHG, there is currently a great research effort focused on the development of cutting-edge ultrafast oscillators.

A key point for efficient HHG is an intense and short driving pulse. Within the scope of this thesis we therefore explore the potential of semi-
Abstract

conductor saturable absorber mirror (SESAM) modelocked thin-disk oscillators with respect to the generation of pulses with short durations (<200fs). The invention of the SESAM nearly 20 years ago was a major milestone for the development of ultrafast laser systems. In order to take full advantage of the SESAM-based modelocking mechanism, we studied the influence of top-mirror structures on nonlinear reflectivity parameters and on the damage threshold of standard antiresonant quantum well (QW) SESAMs, grown at low temperatures. We extended this detailed study to a set of samples with identical absorber sections grown at different temperatures to evaluate the impact of topcoating layers on the recovery dynamics. Furthermore, we performed preliminary experiments with substrate-removed SESAMs to evaluate the potential of such samples with improved heat removal capabilities and flatness, compared to standard-grown SESAMs contacted on copper heat sinks. Our investigations allowed us to establish simple guidelines for the growth and processing of robust SESAMs with high-damage threshold, sustaining high intracavity energy levels.

In a set of experiments, we applied our design guidelines to ultrafast SESAMs grown for the generation of short pulses. The most important parameter in order to achieve short pulses is a broad gain bandwidth of the laser crystal. In this aspect, the group of Yb-doped sesquioxides outperforms the standard thin-disk gain material Yb:YAG. In a first step, we explored the pulse duration limits of the pure sesquioxide Yb:LuO. We were able to achieve 7 W of average power with pulses as short as 142 fs and measured the corresponding carrier-envelope offset (CEO) frequency. In a second step, we scaled the power of this thin-disk oscillator to 25 W and 185 fs. Moreover, we developed SESAM modelocked thin-disk lasers based on the mixed sesquioxides Yb:LuScO and Yb:ScYLO that demonstrated pulse durations of 96 fs, respectively 101 fs, with output powers around 5 W. Currently, the most promising gain material in terms of shortest pulse durations is Yb:CALGO. Experiments based on this gain material enabled the first phase-stabilization of the CEO frequency of a SESAM modelocked thin-disk oscillator and allowed us to obtain sub-50 fs pulses with an average power of 2 W.
A promising alternative approach that extends laser performances to shorter pulse durations combines within one resonator two spatially separated gain media with displaced emission spectra of moderate bandwidth. We successfully implemented, to our knowledge for the first time, this concept into the thin-disk laser technology. The emission spectra of Yb:LuO and Yb:ScO, two gain materials with high thermal conductivity, combined in modelocked operation to a single gain. It supported 103-fs pulses with an average power of 1.4 W and 124-fs pulses at a higher power of 8.6 W.

The ongoing advances in the growth of single crystalline laser materials with high-optical quality are an important key element for the development of future state-of-the-art ultrafast oscillators. A further essential point is the development of substrate-removed, large area SESAMs with outstanding heat removal capabilities and flatness. We believe that the progress in this two areas will enable to push the average power (275 W) and pulse energy (80 µJ) of current leading-edge SESAM-modelocked thin-disk lasers to shorter pulse durations (< 200fs).
Kurzfassung (German)


Kurzfassung (German)


Die bahnbrechende Erfindung sättigbarer Halbleiterabsorberspiegel (SESAMs) vor fast zwanzig Jahren war ein wesentlicher Meilenstein in der Entwicklung ultraschneller Lasersysteme. Um das gesamte Potential dieser Technologie ausschöpfen zu können, analysieren wir die Auswirkung von Spiegelbeschichtungen auf die Sättigungseigenschaften und die Zerstörschwelle antiresonanter Quantentröge (QW) SESAMs. Wir untersuchen des Weiteren den Einfluss der Spiegelbeschichtungen auf die Relaxationszeit einer Reihe von SESAMs, deren identische Absorberstrukturen bei verschiedenen tiefen Temperaturen gewachsen worden sind. Basierend auf richtungsweisenden Experimenten an substrat-entfernten SESAMs, können wir erste Abschätzungen der vielversprechenden Möglichkeiten dieser Absorbereinheiten mit verbesserter Wärmeabfuhr und Oberflächenreiben geben. Die Untersuchungen ermöglichen uns abschliessend, klare Vorgaben zum Wachstum und der Bearbeitung von SESAMs mit hoher Zerstörschwelle zu geben, die bestens geeignet sind für den Einsatz in Laserkavitäten mit hohen Pulspower.


Entscheidend für die Weiterentwicklung ultraschneller Laseroszillatoren ist die Optimierung von SESAM-Kontaktierungsmethoden, die das Wärmeabfuhrvermögen und die Ebenheit der Absorber einheit verbessern. Des Weiteren sind laufend Fortschritte im Wachstum grossflächiger, einkristalliner Scheibenmaterialien mit hoher optischer Qualität zu verzeichnen. Wir sind deshalb zuversichtlich, dass die heutigen Durchschnittsleistungen (275 W) und Pulsenergien (80 µJ) von modengekoppelten Scheibenlasern in Zukunft auch mit kurzen Pulsdauern (< 200 fs) erhältlich sein werden.
Although its invention dates back only a bit more than 50 years, light amplification by stimulated emission of radiation (laser) has become an essential tool in our daily live with a high impact on many different areas [1]. Laser welding, cutting, polishing and hardening allow for metal working with extremely high accuracy and precision down to the micrometer scale [2]. The automotive industry relies on laser cutting and welding of sheets and laser based surface treatments, as for instance the hardening of turbo chargers. Medical implants like stents are high-precision micro-machined with ultrafast lasers and laser surgery precisely cuts and removes tissue [3]. In terms of telecommunication, comprehensive networks of fiber optic communication systems are presently being planned or have already been built [4]. Furthermore, optical data media like CD and DVD use laser diodes to read, write and store large amount of information at a high performance level.

Considering all these different fields of applications it is therefore not an overstatement to say that the laser technology has revolutionized most of our industrial branches no to speak about the possibilities it opened and still opens in fundamental research.

Physical processes involving light-matter interaction with strong laser fields reveal ionization and electron dynamics on the attosecond timescale ($10^{-18}$ s). Studies in the area of strong-field physics rely on intense ultrashort laser pulses with field strengths of the same order of magnitude
as the electron binding fields. In the case of laser wavelengths around 1 µm, this corresponds to an intensity of approximately $10^{14}$ W/cm². The most common way to generate attosecond pulses is based on a form of highly nonlinear frequency conversion called high-order harmonic generation (HHG) [5]. In this process an intense ultrafast source laser (< 100 fs) provides the fundamental driving frequency, typically in the near- or mid-infrared. The ultrashort laser pulse of the driving source is focused in a gaseous target and provokes the electronic motions responsible for the generation of high-order harmonics of the fundamental frequency. The semiclassical three-step model gives an excellent qualitative description of the physical phenomenon responsible for HHG [6, 7, 8].

Currently, passive modelocked Ti³⁺-doped sapphire amplifiers are the workhorse for HHG and attoscience. Ti:sapphire is a tunable gain material with an extremely broad emission bandwidth ranging from 680 nm to 1100 nm and an excellent thermal conductivity of 33 W/Km. Commercially available Ti:sapphire based amplifier systems offer ultrashort pulse durations with high pulse energies in the multi-mJ-range. Without putting in question the remarkable performance of Ti:sapphire amplifier systems it must also be mentioned here, that there exist several drawbacks related to this technology.

Ti:sapphire based oscillators have high demands on their pump lasers. The ideal pump wavelength is in the green spectral range and the relatively short upper-state lifetime of $\approx 3 \mu$s requires extremely high pump intensities with almost diffraction-limited beam quality. Although first progress has been made in frequency-doubled diode and direct-diode pumping, the obtained output performances are far below the standard requirements on driving sources of strong-field processes. Even though pulsed Ti:sapphire amplifier systems have less demanding requirements on their pump laser remains the disadvantage of complexity and, even more crucial, the limitation of the pulse repetition rate to the kHz regime. This constraint on the repetition rate leads to an unwanted low data-acquisition rate in strong-field experiments.
It is for this reason that the development of alternative powerful ultra-fast laser sources at repetition rates in the MHz range is currently a research topic of high interest [9, 10]. The high power levels of such systems result in an increased heat deposition in the gain medium that has to be overcome by choosing a geometry that allows for efficient heat dissipation and cooling. Among the laser technologies fulfilling these requirements we find the slab-, fiber- and thin-disk geometry that are discussed in more detail in chapter 4. All these laser architectures have one thing in common: the shape of their gain material results in a large surface to volume ratio improving the heat removal capabilities. In the last years Innoslab amplifiers [11], fiber systems based on chirped-pulse amplification (CPA) [12] and thin-disk amplifiers [13] have shown the potential of this laser technologies, providing ultrafast pulses with average powers of more than 800 W and repetition rates in the range of 0.8 MHz up to 78 MHz. Furthermore, an optical parametric chirped-pulse amplification (OPCPA) system operating at 0.6 MHz recently enabled the fist demonstration of HHG at high repetition rates [14]. This shows that the above-mentioned novel amplifier technologies are indeed the appropriate tool to drive HHG at unprecedented high repetition rates. However, these systems still consist of a low-power seed oscillator and several amplification stages resulting in undesired overall complexity and the question is now how to solve this remaining drawback.

The closest alternative to an amplifier system is the concept of an ultrafast laser oscillator that directly provides the required peak intensities and pulse durations for HHG. Up-to-date laser oscillators based on thin-disk shaped gain materials hold the record in output powers and pulse energies among all ultrafast laser sources [15, 16]. Since this technology allows for operating all cavity components in reflection, resulting nonlinear effects are reduced to a minimum. In addition, thin-disk lasers (TDLs) support the simple approach of power scaling by increasing mode size on gain material and other cavity components [17, 18]. Femtosecond TDLs are modelocked by means of two different mechanisms: Kerr-lens mode-locking (KLM) [19] and semiconductor saturable absorber mirror (SESAM) modelocking [20]. Recent experiments based on KLM and SESAM mode-locked TDLs outperformed with average powers \( \geq 270 \text{ W} \) and pulse en-
1. **Motivation**

Energies of several tens of µJ\(^{[15, 16, 21]}\). Although the provided pulse durations in the sub-ps range fit to some extent the requirements on a driving source of strong-field experiments, a further pulse compression down to the sub-100 fs range is indispensable to reach the demanded field strengths.

Thus, we present within the scope of this thesis promising gain materials for SESAM mode-locked TDL oscillators with the potential for high output power levels and sub-100 fs pulse durations. The first candidates suitable for short pulses we find among the group of Yb\(^{3+}\)-doped sesquioxides\(^{[22]}\). Crystals as for example Yb:LuScO and Yb:ScYLO are mixed sesquioxides and inhibit a disordered lattice structure providing the large emission bandwidth required for short pulses. Another promising gain material is Yb:CALGO\(^{[23]}\), a novel material with an extremely broad bandwidth, that supported 62 fs-pulses with an average power of 5.1 W in a SESAM modelocked TDL oscillator\(^{[24]}\). Furthermore, we show the implementation of the dual gain concept into the TDL geometry\(^{[25]}\), an innovative concept that combines two gain crystals with shifted emission peaks in a single resonator. In modelocked configuration, the individual emission spectra of two thin disks with good thermal conductivity but moderate emission bandwidth combine to one broad gain spectrum. This approach has a promising future as it relaxes the former stringent condition on the gain material concerning broad emission spectra. We believe that it will allow TDL resonators developed on the basis of high-quality crystals with moderate emission bandwidth the generation of ultrashort pulses with high output performances. The following chapters are structured as follows:

In Chapter 2, we give a brief introduction into different passive modelocking mechanisms with particular emphasis on SESAM modelocked soliton pulses. We present the characterization setups we used to measure nonlinear reflectivity parameters, recovery dynamics and spectral reflectivity of our SESAMs.

In Chapter 3, we present a study on SESAMs with semiconductor and dielectric topcoatings. We investigated the influence of this top-reflectors on nonlinear reflectivity parameters and damage threshold fluence. Our
measurements allowed us to give guidelines on the development of robust SESAMs with optimized parameters for the use in ultrafast high-power oscillators. Furthermore, we started characterizing the next generation of substrate-removed SESAMs. Such SESAMs show high-quality surfaces with remarkably improved flatness, compared to standard indium soldered SESAMs.

Chapter 4 discusses the critical issues that have to be taken into account in order to successfully operate ultrafast lasers at high average power and pulse energy levels. We present the different geometries of high-power solid-state laser technologies that allow to overcome these challenges. These are the fiber-, slab- and thin-disk design. Relevant for our work, were several Yb-doped rare-earth crystals that we present in more detail. Furthermore, we give a short overview of the current-state of the art of femtosecond TDLs.

Chapter 5 presents experimental laser data that we obtained from Yb-doped gain materials with broad emission bandwidths. With these laser crystals, we explored the short pulse duration range (< 200 fs) of SESAM modelocked TDLs. In our experiments, we were able to exploit more than 60 % of the full width at half maximum (FWHM) gain bandwidth of the gain materials that supported the generation of almost transform-limited pulses. By using the sub-100 fs pulses of an Yb:CALGO laser we stabilized for the first time a SESAM modelocked thin-disk oscillator.

In Chapter 6, we present an experiment that successfully implemented the dual-gain concept into the TDL geometry. Two gain crystals with high thermal conductivity but moderate emission bandwidths (Yb:LuO and Yb:ScO) were incorporated in a single resonator. Their emission spectra combined in modelocked operation to a single broad gain spectrum that supported short pulse durations.

Chapter 7 summarizes the reported experimental results and explores possible future developments of high power SESAM modelocked TDL along the groundbreaking direction towards MHz-driving sources of strong field experiments.
Chapter 2

Introduction to SESAM modelocking

The fundamental operation methods of laser oscillators emitting coherent light can be split up in two basic principles: in continuous wave (cw) and pulsed operation. Depending on the type of laser and the technical method applied, typical pulse durations are in the range from nanosecond to few femtosecond time scales. Within this time domain modelocking is the basis for lasers providing ultrashort pulse durations [26]. For many applications in industry and research ultrafast laser pulses have an advantage over cw laser light for a variety of reasons. Material processing driven by ultrafast laser systems in the picosecond-range benefits from a tremendous reduction of deposited heat in the material allowing for much more precise cutting and perforation compared to cw-laser operation [27]. In research, ultrafast lasers allow to explore the dynamics of physical processes at very short timescales [28]. The stabilized frequency comb of a modelocked femtosecond laser, for example, is versatile tool whose implementation ranges from counting optical cycles to enable extremely accurate spectroscopic measurements [29].

2.1 Passive modelocking

The term modelocking originates from the frequency domain. A modelocked pulse consists of a superposition of numerous individual longitudinal cavity modes with a constant phase relation, or in short term “locked” phases. The Fourier transformation of this constellation corresponds to
a train of pulses with durations in the nano- to femtosecond regime: the larger the number of modes involved, the shorter the spatial extent of the pulse in the time domain. The repetition rate of a modelocked pulse train is determined by the cavity length. Either an active or a passive component within the resonator periodically modulates the intracavity losses with the period being an integer multiple of the cavity round trip time. This is in contrast to other pulsing technologies as for example Q-switched modelocking (QML). There, an active device switches independently from the resonator length between low and high intracavity losses represented by a high respectively low quality factor (Q-factor) of the cavity.
Although it is possible to harmonically modelock an oscillator, it usually becomes more difficult to guarantee equidistant pulse spacing and constant pulse energy. Thus, the majority of modelocked lasers operates in its ground mode where only one single pulse is propagating in the cavity. The energy $E_p$ of a fundamental pulse is

$$E_p = P_{av} \cdot T_R = \frac{P_{av}}{f_{rep}},$$

(2.1)
determined by the product of average power $P_{av}$ and round-trip time $T_R$, respectively $P_{av}$ divided by the pulse repetition rate $f_{rep}$. Modelocking generates pulses with a temporal extension that is in general much shorter than the round trip time. For active modelocking the pulse formation is related to the modulation speed of the externally driven switch whereas passive modelocking is based on the intrinsic property of the saturable absorber. The latter one is thus the pulse shaping mechanism that allows for the generation of the shortest pulse durations obtained directly from an oscillator. The following section briefly explains three passive modelocking principles that are illustrated in figure 2.1: KLM [19], SESAM modelocking with dynamic gain saturation and SESAM based soliton modelocking [20]. The main emphasis is on the last of this three pulse shaping mechanisms as the femtosecond oscillators developed within the framework of this thesis rely on this technique.

## 2.1.1 Kerr lens modelocking

KLM relies on a nonlinear optical process of second order that changes the refractive index $n$ of the involved medium. A laser beam of high intensity that propagates through a nonlinear material induces this Kerr effect:

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

(2.2)

Its strength depends on the nonlinear refractive index $n_2$ of the medium and the optical intensity $I$ of the laser beam. If the transverse beam profile is such that the highest intensity is found at the center of the beam, the resulting refractive index change has the same effect as a focusing lens. This lens acts in combination with an intracavity aperture as a loss modulator that provides an energetically more favorable condition in modelocked operation. KLM is therefore based on an artificial absorber with two main
aperture mechanisms [30]. In case of hard aperture KLM, an intracavity iris induces high losses in cw operation. Initial intensity fluctuations in the assigned Kerr medium lead to the formation of a Kerr lens that minimizes the losses of the aperture. In another mechanism for KLM the gain medium itself acts as a soft aperture. In pulsed operation, the intensity-dependent Kerr lens provides a better mode overlap between pump- and laser beam inside the gain crystal. The formation of a Kerr lens is an almost instantaneous process on the timescale of some few femtoseconds. As this response time is much shorter than the temporal expansion of the generated pulses, the pulse shaping mechanism of KLM is based on a fast artificial saturable absorber. KLM is a successful technology that provides pulse durations in the few-cycle regime [31]. However, Kerr lens dynamics require an oscillator operating in cw at an edge of its stability limit. This coupling of the pulse formation process to the laser mode makes the cavity extremely sensitive to misalignment and further lensing effects. Furthermore, KLM is in general not self-starting and relies on a perturbation mechanism. SESAM assisted KLM is thus one possible pulse formation scheme, where the SESAM initiates and stabilizes the KLM [32].

### 2.1.2 SESAM modelocking with dynamic gain saturation

A SESAM is a semiconductor device and thus, in contrast to the KLM mechanism, a real saturable absorber. Its operation principle is, as for all saturable absorbers, based on intracavity loss modulation of the oscillator. The intensity-dependent reflectivity of the SESAM allows to build up a pulse from initial noise fluctuation in cw operation. The relaxation dynamics of the SESAM set an upper limit on the duration of the gain window. That determines in case of fast saturable absorption the demonstrated pulse duration. SESAMs with fast recovery dynamics have typical relaxation times in the few ps range. As the area of our research interest is focused on the generation of pulses with maximal durations of some hundred of femtoseconds, SESAMs act as slow saturable absorbers [33]. One fundamental principle of passive modelocking with slow saturable absorbers relies on dynamic saturation of the gain medium. This pulse formation technology is typically applied on semiconductor lasers. Classical semiconductor materials have upper state life times in the nanosecond
regime. They exhibit therefore high emission cross-sections, as they are inversely proportional to fluorescence lifetime. Moreover, the saturation energy of the gain medium

\[ E_{\text{sat},L} = \frac{h \nu_c A_L}{m_L \cdot (\sigma_{\text{em},L} + \sigma_{\text{abs},L})} \]  

(2.3)

is relatively low and already a single laser pulse passing the gain medium remarkably depletes the inversion level. In formula 2.3 we find the Planck’s constant \( h \), the central laser frequency \( \nu_c \), the laser mode size on the gain medium \( A_L \), the number of passes on the gain per resonator round-trip \( m_L \) and the emission and absorption cross-sections \( \sigma_{\text{em},L} \) and \( \sigma_{\text{abs},L} \) of the laser medium at \( \nu_c \). If the saturation of the SESAM starts at a lower pulse fluence than the dynamics of the gain, stable femtosecond pulse formation can be achieved.

### 2.1.3 SESAM based soliton modelocking

Populated upper laser levels of solid-state materials decay, in contrast to semiconductors, within micro- to milliseconds due to smaller emission cross-sections. Thus, the gain of diode-pumped solid-state lasers (DPSSLs) saturates only for energy levels much higher than usually generated in modelocked operation. As the depletion of the gain is negligible, the net gain window can be 10 to 30 times larger than duration of femtosecond pulses. A cw background propagating within this window experiences in principle the same, or even higher gain than the main pulse and is supposed to disturb the pulsed operation. However, there exists a technology called soliton modelocking that supports stable pulse build up with constant gain and slow absorption modulation [34, 35]. Responsible for the soliton-pulse shaping is a balanced interplay of self-phase modulation (SPM) and group-delay dispersion (GDD) with opposite signs. The generated SPM broadens the spectrum of the pulse that is temporally compressed via GDD. A fundamental soliton preserves its \( \text{sech}^2 \)-shaped envelope during propagation in both, time and frequency domain. The previously mentioned continuum is, however, of lower intensity and thus only disperses and eventually fades away. The SESAM is inserted as an additional element that starts and stabilizes the pulse formation. In a stable
soliton regime with weak destabilizing effects, the FWHM pulse duration \( \tau_p \) satisfies the equation:

\[
\tau_p \approx 1.76 \cdot \frac{2|D|}{|\gamma_{\text{SPM,cav}}|E_p}.
\]  

(2.4)

The parameter \( D \) is the GDD per roundtrip and \( E_p \) the intracavity pulse energy. The parameter \( \gamma_{\text{SPM,cav}} \) represents the SPM coefficient per cavity roundtrip. The SPM coefficient for a laser beam with a Gaussian-shaped intensity profile that propagates through a Kerr medium of length \( L \) is

\[
\gamma_{\text{SPM}} = \frac{4n_2}{\lambda_c} \cdot \int_L \frac{1}{\omega^2(z)} \, dz,
\]  

(2.5)

where \( n_2 \) is the nonlinear refractive index of the material, \( \lambda_c \) the lasing wavelength in vacuum and \( \omega(z) \) the radius of the laser beam. Formula 2.4 neglects certain effects, as for example discreteness of SPM and GDD, finite bandwidth of the gain \( \Delta f_g \) or absorber dynamics. This processes set a lower limit to the achievable pulse duration. By using an analytical model, a rough estimation of this limit was obtained [34]:

\[
\tau_{p,\text{min}} \approx 0.2 \cdot \left( \frac{\Delta f_g}{\Delta R} \right)^{3/4} \cdot \left( \frac{\tau_A}{\Delta R} \right)^{1/4} \cdot \left( \frac{g^{3/8}}{\Phi^{1/8}} \right).
\]  

(2.6)

The modulation depth \( \Delta R \) and recovery time \( \tau_A \) are parameters of the slow saturable absorber that are introduced in more detail in section 2.3. The parameter \( g \) is the round-trip power gain, and \( \Phi \) the soliton phase shift per roundtrip. More detailed information about formula 2.4 and 2.6 can be found in [33]. If the effects of spectral broadening and temporal compression exactly compensate each other, the generated \( \text{sech}^2 \)-pulse is bandwidth-limited. In such a configuration, the product of FWHM pulse duration \( \tau_p \) and FWHM spectral width in frequency domain \( \Delta \nu \) is minimized and yields:

\[
\tau_p \cdot \Delta \nu \approx 0.315.
\]  

(2.7)

The peak power \( P_{\text{peak}} \) of a soliton pulse is given by:

\[
P_{\text{peak}} \approx 0.88 \cdot \frac{E_p}{\tau_p}.
\]  

(2.8)

The technology of SESAM based soliton modelocking completely decouples the mechanism responsible for saturable absorption from spectral
2.2 SESAM structure

A SESAM is, as its name says, a semiconductor mirror of nonlinear reflectivity. A classical SESAM consists of a quantum well (QW) or quantum dot (QD) structure embedded in a spacer layer grown on top of an antiresonant distributed Bragg reflector (DBR). Such a standard DBR design is based on a periodic structure of low and high refractive index materials that ends with a high index layer. The alternating layer pairs have optical thicknesses of a quarter of the lasing wavelength. As a result, the laser light reflects at the interfaces and constructively interferes, forming a standing wave pattern inside the reflector. The antiresonant structure of the DBR is preserved if the thickness of the spacer layer is such that the broadening. This is in contrast to KLM where both, loss modulation and SPM rely on the induced Kerr effect. Therefore, both effects can be optimized independently allowing for more flexibility in the cavity design. In addition, the sensitivity of the cavity to parasitic thermal lensing of intracavity components is reduced.

Figure 2.2: Antiresonant QW SESAM structure consisting of 30 quarter-wave layer pairs of AlAs/GaAs with alternating low- and high refractive indices (left axis) and one InGaAs absorber layer embedded in a GaAs spacer layer. The field enhancement $|E_n(z)|^2$ (right axis) and thus the intensity of the standing wave pattern exhibits a node at the air-semiconductor interface and its value at the position of the absorber layer is $\approx 0.32$. For better visibility the dotted line shows the field enhancement scaled by a factor of 10.
standing wave pattern shows a node at the air-semiconductor interface, as shown in figure 2.2. The intensity of the standing wave pattern inside the SESAM is a function of the penetration depth $z$ [36]:

$$I(z) = n(z) \cdot |E_n(z)|^2 \cdot I_{\text{inc}}.$$  \hspace{1cm} (2.9)

Parameter $n(z)$ is the local refractive index and $E_n(z)$ is the electric field of the standing wave pattern normalized with respect to the electric field of the incoming beam with intensity $I_{\text{inc}}$ [37]. The field intensity enhancement $|E_n(z)|^2$ has a value of 4 outside the semiconductor device and a maximum value of $4/n^2$ inside an antiresonant structure. As the strength of the field intensity $I(z)$ in the absorber layer influences the macroscopic SESAM parameters, we define

$$\xi_{\text{abs}} = |E_n(z = z_{\text{abs}})|^2$$  \hspace{1cm} (2.10)

as the field intensity enhancement at the position of the absorber section $z_{\text{abs}}$. An antiresonant configuration results in an almost constant GDD and $\xi_{\text{abs}}$ over a broad spectral range, beneficial for many applications. Furthermore, it provides a $\xi_{\text{abs}}$ with low values around 0.34 [38]. Hence, antiresonant SESAMs saturate in comparison with resonant devices at a much higher pulse fluences and fulfill the requirements of high-energy level oscillators. The intensity dependent reflectivity of the SESAM arises from the quantum confinement of an incorporated QW or QD structure. A QW is a thin semiconductor layer with a somewhat smaller bandgap energy than the lasing wavelength. Therefore, incident laser light is absorbed and populates the conduction band. The QW structure acts as a one-dimensional trap, which confines the created carriers in direction to the beam axis. It saturates and becomes transparent for high fluences of the laser beam. The transparency fluence $F_t$ of a SESAM is an intrinsic property of the absorber section that remains constant for a specific lasing wavelength and sample temperature. It is is given by [39]:

$$F_t = d \cdot h\nu \cdot N_t,$$  \hspace{1cm} (2.11)

with $d$ the thickness of the absorber section, $h\nu$ the incident photon energy and $N_t$ the carrier transparency density. The geometry of a QW confinement results in a comparatively high $N_t$ which in turn is related to a high
transparency fluence $F_t$ of the SESAM. The combination of an antiresonant structure and one or several QW absorber layers is therefore the key to obtain stable modelocked operation of ultrafast oscillators with high pulse energies. All SESAMs we present within this thesis were grown by molecular beam epitaxy (MBE) in the FIRST cleanroom facility at ETH Zurich. They were antiresonant and consisted of a DBR structure of 30 alternating gallium arsenide (GaAs) and aluminium arsenide (AlAs) quarter-wave layer pairs. The absorber section consisted of one or several indium gallium arsenide (InGaAs) QWs with typical thicknesses of 7 nm up to 10 nm, placed in antinodes of the standing wave intensity pattern. They provided reflection bandwidths of approximately 100 nm with center wavelengths in the range of 1030 nm up to 1050 nm.

2.3 SESAM characterization

We already mentioned in the previous section 2.2, that the SESAM development presented in this thesis focused on antiresonant QW-structures. However, the geometry of the absorber layers and underlying DBR structure is only one among various available methods to optimize the macroscopic properties of SESAMs intended for applications in high-power ultrafast oscillators. The optimization starts with the appropriate choice of the surrounding semiconductor material of the absorber layer and the amount of QWs placed in one or several antinodes of the standing wave pattern. Growth conditions can be adjusted to tweak certain properties of the absorber layer, as for example nonsaturable losses $R_{ns}$ and relaxation time $\tau_A$. Post-processing methods include mirror structures that are placed on top of the absorber layer. Such a reflector at the air interface lowers the field enhancement inside the QW-structure $\xi_{abs}$. This list of methods acts on the optical properties of a SESAM. Hence, a complete characterization of macroscopic SESAM parameters is indispensable for a subsequently successful application in ultrafast oscillators with high power-levels. We describe in the following subsections the measurement setups we used to characterize the corresponding SESAM parameters.
2. Introduction to SESAM modelocking

Figure 2.3: Experimental setup of the nonlinear reflectivity characterization and damage measurements. The laser source was in general an Yb:YAG TDL delivering 1-ps pulses at 1030 nm with 7 W of average power and a repetition rate of 3.9 MHz. The fixed attenuation stage in front of the setup set a maximum pulse fluence level for a given measurements and the variable attenuation stage inside provided a pulse fluence range of more than four orders of magnitude.

Figure 2.4: Typical measurement of the reflectivity of an antiresonant QW SESAM as a function of pulse fluence (discrete points). The graph shows in addition the corresponding fit function with (solid curve) and without induced absorption (dashed curve).
2.3. SESAM characterization

2.3.1 Nonlinear reflectivity

The saturation dynamics of a SESAM result in a fluence-dependent reflectivity. We measured this nonlinear reflectivity with a high-precision setup, shown in figure 2.3, providing a pulse fluence range of more than four orders of magnitude. In general, we used for this setup a Yb:YAG-TDL source that provided 1-ps pulses centered around 1030 nm, with a repetition rate of 3.9 MHz and an output power of 7 W. A detailed description of the concept of this characterization system is presented in [40]. The experimental setup variably attenuated the ultrafast laser beam first and split it then in a reference and a sample beam of equal power. The sample arm contained a lens with a focal length of 20 mm that created a spot diameter of $\approx 20 \, \mu m$ on the sample. We used a high-reflective (HR) mirror with a specified reflectivity of 99.98% to calibrate the measured reflectivity. Figure 2.4 depicts a typical measurement of an antiresonant QW-SESAM that we obtained with this setup. The data points were fitted with a model function that assumes a flat-top intensity profile of the laser beam:

$$R(F_p) = R_{ns} \ln \left( \frac{1 + (R_{lin}/R_{ns}) \cdot \left( e^{F_p/F_{sat,A}} - 1 \right)}{F_p/F_{sat,A}} \right) \cdot \left( e^{-F_p/F_2} \right).$$ (2.12)

Formula 2.12 describes the nonlinear reflectivity of a SESAM as a function of constant pulse fluence $F_p$. This is in contrast to our laser source that provided a Gaussian-shaped transverse intensity profile. In order to obtain a precise match between measured nonlinear reflectivity and fit function, the change in peak fluence has to be taken into account. Reference [41] describes this modified model function that is based on the same fit parameters than formula 2.12. The parameter $R_{lin}$ represents the linear reflectivity of an unbleached SESAM and the parameter $R_{ns}$, in case of absent induced absorption ($F_2 \rightarrow \infty$), the nonlinear reflectivity of a fully saturated SESAM. These two fit parameters determine the modulation depth $\Delta R$ and the nonsaturable losses $\Delta R_{ns}$:

$$\Delta R = R_{ns} - R_{lin}$$ (2.13)

$$\Delta R_{ns} = 1 - R_{ns}.$$ (2.14)

The saturation fluence $F_{sat,A}$ is defined as the fluence required to saturate $1/e$ of the modulation depth $\Delta R$. The transparency fluence $F_t$ of a given
The absorber section defined in equation 2.11 can be estimated by calculating the product of the two fit-parameters $F_{\text{sat,A}}$ and $\Delta R$:

$$F_t \approx \Delta R \cdot F_{\text{sat,A}}.$$  \hspace{1cm} (2.15)

The derivation of formula 2.15 can be found in [39, 42]. An important laser parameter is the ratio of pulse fluence to saturation fluence

$$S = F_p / F_{\text{sat,A}}.$$  \hspace{1cm} (2.16)

Ultrafast oscillators operate typically in an S-parameter range of $\approx 3-10$ [33]. However, stable modelocked operation of high-power thin disk lasers with larger S-parameters (90-100) was reported [43, 16]. A real absorber shows induced absorption at highest pulse fluences. According to formula 2.12, the induced absorption coefficient $F_2$ represents the fluence at which the SESAM reflectivity drops to $\approx 1/e$ of $R_{\text{ns}}$. When the incident pulse fluence on the SESAM is in this "rollover" regime, a single pulse might break up into energetically favored multiple pulses leading to instabilities of the laser operation [44, 45]. One source of the rollover in reflectivity is the nonlinear optical process of two-photon absorption (TPA). If the laser source delivers pulse durations in the femtosecond regime, TPA is even the main effect of induced absorption and $F_2$ can be approximated as [36]:

$$F_{2,\text{TPA}} = \frac{\tau_p}{0.585 \cdot \int \beta_{\text{TPA}}(z) n^2(z) (|E_n(z)|^2)^2 \, dz}.$$  \hspace{1cm} (2.17)

Parameter $\tau_p$ is the pulse duration, $\beta_{\text{TPA}}$ the local TPA coefficient, $n(z)$ the local refractive index and $|E_n(z)|^2$ the field intensity enhancement inside the structure, see formula 2.9. In the case of weak nonlinear TPA, the absorbed intensity within the structure can be calculated as follows:

$$I_{\text{abs}} = \int \beta_{\text{TPA}} I^2(z) \, dz,$$  \hspace{1cm} (2.18)

with $I(z)$ referring to the local intensity of the standing wave pattern. We see that TPA can be reduced with the growth of a few-layer mirror-structure on top of the spacer layer that lowers the field intensity enhancement inside the structure. Another approach replaces the common spacer layer material GaAs by an other appropriate semiconductor material with lower $\beta_{\text{TPA}}$. The $F_2$ derived from TPA is proportional to $\tau_p$ and thus
larger for picosecond laser pulses corresponding to a diminished rollover. However, experimental results with ps-laser sources show significantly stronger induced absorption than theoretically predicted with TPA alone. The origin of the mechanism responsible for the additional absorption is not clearly elucidated yet. So far we discussed possible ways to reduce the effect of induced absorption, in order to prevent instable multipulse operation of ultrafast oscillators with high intracavity energies. However, inverse saturable absorption relaxes in terms of Q-switching instabilities the condition for stable modelocking [46]. At high pulse fluences, a slight increase in energy due relaxation oscillations is damped by the rollover in reflectivity. In such a configuration, stable modelocking is obtained with pulse energies fulfilling the following equation:

$$E_{p,IA}^2 > \frac{E_{sat,A} \cdot \Delta R}{1/F_{sat,L} + 1/A_A \cdot F_2}.$$  

(2.19)

$E_{sat,A}$ is the saturation energy of the absorber, $E_{sat,L}$ the saturation energy of the gain medium and $A_A$ the mode area on the SESAM. If induced absorption ($F_2 \to \infty$) is negligible, formula 2.19 simplifies to:

$$E_{p, no IA}^2 > E_{sat,L} \cdot E_{sat,A} \cdot \Delta R = F_{sat,L} \cdot F_{sat,A} A_A \cdot \Delta R.$$  

(2.20)

with $F_{sat,L}$ the saturation fluence of the gain medium and $A_L$ the mode area on the gain medium. The threshold for stable modelocking is increased, as the SESAM provides lower loss for a pulse of higher energy. This is a destabilizing mechanism of the absorber that is compensated by gain narrowing. A pulse with higher energy experiences more SPM and has therefore a broader spectral bandwidth. This results in a lower effective gain, as the finite gain bandwidth of the laser medium does not support its full bandwidth.

Damage of a SESAM occurs at pulse fluences deep in the rollover (e.g. $S > 150$), where no stable modelocked oscillator is operating. We defined damage of a SESAM as an irreparable change of the structure, resulting in a dramatic drop of the measured reflectivity. The corresponding parameter $F_{dam}$ refers to the minimum pulse fluence where this irreversible drop in reflectivity occurs within less than one second, the precision limit of our measurement setup. We measured the damage-fluence threshold.
2. Introduction to SESAM modelocking

\[ F_{\text{dam}} \]

with the same optical characterization setup required to obtain the nonlinear reflectivity parameters of a SESAM, depicted in figure 2.3. We increased the maximum pulse fluence level of the measurements by adjusting the fix attenuation stage in front of the nonlinear reflectivity setup. In order to find \( F_{\text{dam}} \), we started at a pulse fluence far in the induced absorption regime (\( S \approx 100 \)) and tracked the reflectivity of the sample. We then scanned the pulse fluence in steps of \( \approx 5 \text{ mJ/cm}^2 \). The fluence at which we observed instantaneous damage roughly determined \( F_{\text{dam}} \). To exactly localize \( F_{\text{dam}} \), we performed a fine scan in both, pulse fluence (steps of \( \approx 1 \text{ mJ/cm}^2 \)) and z-position of the sample (in order to find the exact position of the focus). A typical time-to-damage measurement and the resulting damage spots on the SESAM are shown in figure 2.5.

2.3.2 Recovery dynamics

The information about the relaxation times of a SESAM is typically obtained with a pump-probe measurement. A pump-probe setup splits the output of an ultrafast laser at the wavelength of interest in two parts. The pump beam is of relatively high power and saturates the sample to a certain extent. The probe beam is in contrast of such low power that it does

---

**Figure 2.5:** Left: Typical time-to-damage measurement. During (1) the sample arm was blocked and the detected reflectivity was zero. At (2) the sample was exposed to a pulse fluence deep in the rollover regime. Position (3) indicates the exposure time after which the sample irreparably damaged, resulting in a dramatic drop in reflectivity. The sample arm was blocked again (4) and the SESAM laterally moved to perform the next measurement. The damage-threshold fluence \( F_{\text{dam}} \) was defined as the minimum pulse fluence for which (3) occurs within a time frame \( < 1 \text{ s} \). Right: Microscopic image of a tested SESAM where we saw multiple damage spots resulting from the measurements performed to determine \( F_{\text{dam}} \).
2.3. SESAM characterization

Figure 2.6: Noncollinear pump-probe setup for measuring the recovery dynamics of the different SESAMs. The laser source was an Yb:YAG TDL delivering 1-ps pulses at 1030 nm with 150 mW of average power and a repetition rate of 38 MHz. Maximum time delay between pump and probe pulse was 300 ps.

Figure 2.7: Typical pump-probe measurement of the relaxation dynamics of a SESAM as a function of time delay between pump- and probe pulse. The two term exponential function includes the short and the long time constant ($\tau_{\text{fast}}$ and $\tau_{\text{slow}}$) of the recovery process. Under certain conditions it is more convenient to consider the recovery time $\tau_{1/e}$ after which the absorber has recovered to a reflectivity of $1/e$ of the normalized reflectivity at zero time delay.
not affect the nonlinear reflectivity of the device. Short pulses are beneficial as the resolution of the measurement is determined by the pulse duration. Pump and probe pulse are chopped at different frequencies and spatially overlap at the position of the characterized sample. The signal of the reflected pump beam is detected with a lock-in-amplification system at the beat note frequency of the two chopping frequencies. The lock-in procedure filters scattered pump light that accidentally hit the detector. Finally, a variable time delay \( \tau \) between pump and probe pulse allows to measure the recovery dynamics of the sample. The recombination process of a SESAM involves two effects that can be described by a normalized two-term exponential:

\[
R(\tau) = Ae^{-\tau/\tau_{\text{fast}}} + (1-A)e^{-\tau/\tau_{\text{slow}}}. \tag{2.21}
\]

The physical meaning of the fast term \( \tau_{\text{fast}} \) is the intraband thermalization of carriers within the conduction band. The fast recovery of a SESAM is essential for the stabilization and shaping of ultrafast pulses. Typical values of short recovery times are in the range of a few 100 fs. The slow term \( \tau_{\text{slow}} \) describes the interband recombination involving the capture of electrons in mid-gap energy levels. Long recovery times are mainly involved in the self-starting of modelocked oscillators and occur within 1 ps to several 100 ps. Mid-gap energy levels result from growth defects within the crystal structure of the absorber layer \[47\]. The less defects are present, the stronger gets the slow recovery process relative to the fast one. This is described by the increasing weight of the amplitude \( 1-A \) of the slow exponential. The amount of mid-gap traps is reduced when the absorber layer is grown at higher temperatures. This effect is convenient for tuning the relative weight of fast and slow recovery dynamics. Although the exact recombination process of a SESAM includes two recovery parameters, it is under certain conditions beneficial to consider the recovery time for a given level of relaxation. We therefore introduce the parameter \( \tau_{1/e} \) as the time delay after which a SESAM relaxes to a reflectivity of \( 1/e \) of the normalized reflectivity at time zero.

\[
R_n(\tau_{1/e}) = 1/e. \tag{2.22}
\]

A typical pump-probe measurements of a SESAM is depicted in figure 2.7. The absorber layers of the QW-SESAMs introduced in this thesis were
2.3. SESAM characterization

![Graph showing linear spectral reflectivity and wavelength middle vs temperature](image)

Figure 2.8: Left: Linear spectral reflectivity of a SESAM with four 7 nm-thick QW layers measured at different temperatures (steps of 10°). The reflectivity exhibited a broad wavelength dependence as expected from low-temperature MBE grown QW SESAMs. Right: We calculated the rate of the reflectivity shift with temperature by evaluating the wavelength where the curvature of the reflectivity curve changed its sign (circles). We measured the rate of this specific SESAM to be $\approx 0.38$ nm/K.

grown at low temperatures ranging from 250° up to 400°. We measured their recovery times with a noncollinear pump-probe setup based on an Yb:YAG laser delivering 1-ps -pulses at 1030 nm with a repetition frequency of 38 MHz. The pulses were chopped with acousto-optic modulators (AOMs) at several 100 MHz resulting in a beat note frequency of $\approx 70$ kHz.

### 2.3.3 Spectral reflectivity

Our experiments with ultrafast TDLs relied solely on Yb-doped gain crystals and the obtained wavelengths were therefore located in the near infrared. Depending on the host material, the pulses exhibited wavelengths centered around 1030 nm up to 1050 nm. The optical characterization setups presented in the previous sections 2.3.1 and 2.3.2 operated at a fixed wavelength, typically 1030 nm. In addition, we stabilized the SESAM temperature. As a consequence, we measured the macroscopic SESAM parameters for a given wavelength and a given temperature. In some experiments, the central wavelength $\lambda_c$ of our laser was longer than the characterization wavelength. For small differences in wavelength, the shift of the SESAM parameters was negligible. However, an approximation of the absorption properties of our SESAMs at $\lambda_c$ was beneficial for larger shifts.
2. Introduction to SESAM modelocking

in wavelength (> 3nm). We were able to do that by changing the temperature of the SESAM under characterization. Increasing the temperature of a SESAM decreases the energy gap of the QW absorber layer which leads to a red shift of the nonlinear reflectivity parameters. Compared to QD SESAMs, this effect is much more pronounced in QW structures. In order to determine the rate of such a red shift with increasing temperature, we performed temperature-tuned spectral reflectivity measurements with a commercial photospectrometer (Varian Cary 5E). The known red shift of a given SESAM allowed us then to characterize its parameters at the temperature of interest.

To give an example, figure 2.8 shows the spectral reflectivity measurements of a SESAM at different temperatures (steps of 10°) and the calculated rate of the red shift with temperature (≈ 0.38 nm/K). Based on this rate, we estimated the parameters of the SESAM characterized at a wavelength of 1030 nm and a temperature of 10°C to correspond to the SESAM parameters at a longer wavelength of 1034 nm and a sample temperature of 20°C.
Previous studies reported on QD SESAMs optimized for low saturation fluences and short relaxation times [48]. Devices that show such properties are one of the key elements of SESAM modelocked lasers with \( f_{\text{rep}} \) in the GHz-regime. The development of SESAMs with low saturation fluences allowed the demonstration of record-high repetition rates of fundamentally modelocked bulk lasers, achieving 160 GHz in the 1-\( \mu \)m wavelength regime [49] and 100 GHz at 1.5 \( \mu \)m [50]. Moreover, it was a crucial step towards a new class of surface-emitting semiconductor lasers that integrates the saturable absorber section in the gain structure (MIXSEL) [51, 52]. The pulse formation process of MIXSELs is based on dynamic gain saturation and strongly differs from the soliton modelocking mechanism of typical ultrafast TDL, see section 2.1.2. Bulk GHz-lasers provide soliton pulses but they operate in a different repetition rate regime and have usually other requirements on the saturable absorber. Hence, we present in this chapter different studies on QW SESAMs designed and processed for operation in high-power ultrafast TDLs [39]. In a first set of measurements we evaluated in detail the performance of SESAMs sustaining intracavity power levels in the kW-regime and pulse fluences of several mJ/cm\(^2\). Their nonlinear reflectivity parameters were optimized for large saturation fluences, negligible nonsaturable losses, reduced induced absorption and damage thresholds deep in the rollover regime where no stable laser operation is supported. In a second step we extended our study on the specific case of SESAMs operating in TDL oscillators with shortest pulse...
durations (< 200 fs). Formula 2.6 shows that in this range of operation an absorber with relatively large modulation depth and short recovery time reduces the minimum achievable pulse duration. We investigated thus on SESAMs grown at different temperatures in order to find the best compromise between short relaxation dynamics and still negligible nonsaturable losses. We furthermore explored novel contacting methods that demonstrated SESAMs with remarkably improved flatness and heat removal capability. These effects reduce thermal lensing and allow for larger pump spot sizes on the SESAM, beneficial for future high-power TDLs providing output powers reaching the kW-range.

### 3.1 SESAMs with high saturation fluences

An elegant way to obtain SESAMs with large saturation fluences is the growth of a few-layer mirror structure on top of a standard antiresonant SESAM shown in figure 2.2. Such a top reflector increases the finesse of the SESAM that is related to a reduced intensity of the standing wave pattern inside the semiconductor device. It results in a low field intensity enhancement at the position of the absorber layer ($\xi_{\text{abs}}$) that we defined in formula 2.10. As a consequence, the saturation fluence of the SESAM is increased. We remember that the two macroscopic parameters $F_{\text{sat},A}$ and $\Delta R$ are related to each other through the intrinsic transparency fluence $F_t$ of the QW layer, see formula 2.15. Therefore, the increase in saturation fluence results in a reduction of the modulation depth by the same factor. In order to obtain high-finesse SESAMs with present absorption ($R_{\text{lin}} < R_{\text{ns}}$), we placed several QW sections inside the spacer layer of the non-topcoated SESAM resulting in an initially large modulation depth. The antiresonant SESAM structure without top reflector (NTC) consisted of three 10-nm thick QW layers placed in a single antinode of the standing wave pattern. The absorber section was grown at 400°C resulting in a recovery time of $\tau_{1/e} \approx 200$ ps and low nonsaturable losses < 0.1 %. Figure 3.1 shows the original sample and the different mirror structures that were grown on it. The first topcoating consisted of a MBE-grown DBR with four AlAs/GaAs layer pairs. We refer to this semiconductor topcoating as SCTC. We calculated the value of $\xi_{\text{abs}}$ to be reduced by 4
3.1. SESAMs with high saturation fluences

Figure 3.1: SESAMs designed for our study on high-finesse samples. They were optimized for large saturation fluences, negligible nonsaturable losses and damage thresholds deep in the rollover regime. a) Basic SESAM design with 3 QWs in a single antinode and no topcoating (NTC). We calculated the field enhancement at the position of the absorber layer to be \( \xi_{\text{abs}} = 0.32 \). b) Semiconductor topcoating consisting of four quarter-wave AlAs/GaAs layer pairs (SCTC). The parameter \( \xi_{\text{abs}} \) was reduced by a factor of \( \approx 4 \). c) Dielectric topcoating consisting of three quarter-wave SiO\(_2\)/Si\(_3\)N\(_4\) layer pairs (DTC2). This top-reflector reduced \( \xi_{\text{abs}} \) by a factor of \( \approx 5 \).

Figure 3.2: SESAMs designed for our study on high-finesse samples. They were optimized for large saturation fluences, negligible nonsaturable losses and damage thresholds deep in the rollover regime. a) Basic SESAM design with 3 QWs in a single antinode and no topcoating (NTC). We calculated the field enhancement at the position of the absorber layer to be \( \xi_{\text{abs}} = 0.32 \). b) Semiconductor topcoating consisting of four quarter-wave AlAs/GaAs layer pairs (SCTC). The parameter \( \xi_{\text{abs}} \) was reduced by a factor of \( \approx 4 \). c) Dielectric topcoating consisting of three quarter-wave SiO\(_2\)/Si\(_3\)N\(_4\) layer pairs (DTC2). This top-reflector reduced \( \xi_{\text{abs}} \) by a factor of \( \approx 5 \).

and thus we estimated the increase in saturation fluence to be given by the same factor. The other two top reflectors were obtained via plasma-enhanced chemical vapor deposition (PECVD) of two respectively three quarter-wave layer pairs of silicon oxide (SiO\(_2\)) and silicon nitride (Si\(_3\)N\(_4\)). The two dielectric topcoatings (DTC) are in the following called DTC2 and DTC3. In the case of this dielectric topcoatings we expected an increase in saturation fluence by a factor of 3 and 5, respectively. We used the experimental setup presented in section 2.3.1 to measure the nonlinear reflectivity of this set of SESAMs as a function of pulse fluence shown in figure 3.2. The Yb:YAG laser source delivered 1-ps pulses with an average power of 15 W at a repetition rate of 10.7 MHz and the SESAM temperature was set to 20\(^\circ\)C. The measurements confirmed the suitability of our approach,
as all topcoated samples showed saturation fluences $> 150 \, \mu J/cm^2$ and modulation depths between 0.4 % and 0.8 %, see table 3.1. All SESAMs had small nonsaturable losses $< 0.1 \, \%$ resulting in minimal thermal load. We observed a roughly constant product of $F_{\text{sat,A}} \cdot \Delta R$, as the absorber section and thus the transparency fluence $F_t$ was the same for all SESAMs. The increase in saturation fluence given by the ratio $F_{\text{sat,A}} / F_{\text{sat,A,NTC}}$ corresponded to the estimated decrease of the field enhancement factor $\xi_{\text{abs}}$. The successful combination of large saturation fluences with typical modulation depths of SESAMs operating in high-power TDLs is advantageous for oscillators with high energy-levels.

Our investigations additionally showed that all high-finesse SESAMs benefited from a reduced induced absorption, compared to the original sample without top-reflector. Table 3.2 presents the values of the induced absorption coefficient $F_2$ of our test samples, obtained from nonlinear reflectivity measurements. SESAMs with semiconductor topcoating showed an increase of $F_2$ by a factor of $\approx 2$. However, we noticed a much stronger effect on the rollover in case of dielectric topcoatings that reduced $F_2$ by a factor of $\approx 10$ or higher. This observation showed that for SESAMs with identical absorber section, the strength of induced absorption depended on the material composition on top of the spacer layer. In sec-

Figure 3.2: Left: Nonlinear reflectivity measurements of the set of SESAMs we used to study the effect of different topcoatings (SCTC, DTC2, DTC3) on saturation fluence and damage threshold. All topcoated samples had increased saturation fluences and typical modulation depths of SESAMs operating in high-power TDLs. The damage-threshold fluences of the individual samples are indicated by an asterik. The set of SESAMs coated with DTC3 did not damage even at the maximum available pulse fluence of 210 mJ/cm$^2$. Right: Lifetime curves of the basic SESAM (NTC) and the topcoated samples (SCTC and DTC2).
Table 3.1: Saturation properties of the original SESAM without topcoating (NTC) and the corresponding high-finesse structures coated with semiconductor (SCTC) and dielectric (DTC2, DTC3) top-mirrors. The parameters were measured at 1030 nm with a pulse duration of 1 ps. All topcoated SESAMs had saturation fluences > 150 µJ/cm² and modulation depths between 0.4 % and 0.8 %. We observed a roughly constant product of $F_{\text{sat,A}} \cdot \Delta R$ and the increase in saturation fluence corresponded, as we expected, to the decrease of the field enhancement factor $\xi_{\text{abs}}$.

### Table 3.1

<table>
<thead>
<tr>
<th>SESAM</th>
<th>$F_{\text{sat,A}}$ (µJ/cm²)</th>
<th>$\Delta R$ (%)</th>
<th>$F_{\text{sat,A}} \cdot \Delta R$ (µJ/cm²)</th>
<th>$\Delta R_{\text{ns}}$ (%)</th>
<th>$\xi_{\text{abs,NTC}} / \xi_{\text{abs}}$</th>
<th>$F_{\text{sat,A}} / F_{\text{sat,A,NTC}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>NTC</td>
<td>72</td>
<td>2.05</td>
<td>1.4</td>
<td>&lt;0.1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>SCTC</td>
<td>279</td>
<td>0.52</td>
<td>1.5</td>
<td>&lt;0.1</td>
<td>≈ 4</td>
<td>3.9</td>
</tr>
<tr>
<td>DTC2</td>
<td>168</td>
<td>0.71</td>
<td>1.2</td>
<td>&lt;0.1</td>
<td>≈ 3</td>
<td>2.3</td>
</tr>
<tr>
<td>DTC3</td>
<td>247</td>
<td>0.43</td>
<td>1.1</td>
<td>&lt;0.1</td>
<td>≈ 5</td>
<td>3.4</td>
</tr>
</tbody>
</table>

In the previous chapter, we mentioned TPA as one source of induced absorption at high pulse fluences. We therefore used equation 2.17 to calculate the induced absorption coefficient $F_{2,\text{TPA}}$ depending on the TPA-coefficient $\beta_{\text{TPA}}$ inside the structure. As the laser source we used for our experiments provided 1-ps pulses, we calculated $F_{2,\text{TPA}}$ for such a pulse duration. In addition, we assumed the value of $\beta_{\text{TPA}}$ of AlAs, SiO₂ and Si₃N₄ to be negligible compared to that of GaAs, for which we used $\beta_{\text{TPA}} = 20 \text{ cm/GW}$ [53]. As the rollover of the SESAM with three dielectric layer pairs occurred at fluences beyond the maximum available pulse fluence of our measurement setup, we obtained only a rough approximation of the fit-parameter $F_2$. This explains the discrepancy between calculated and measured $F_2$. All other calculations were in good agreement with our measurements, apart from a constant factor of $\approx 2.5$ that indicates the contribution of other absorption mechanisms on top of TPA. This is related to the relatively long pulse duration we used for our study. Previous investigation on induced absorption with 3-ps pulse durations yielded a factor of $\approx 2.6$ between calculated and measured rollover coefficient. [36]. In summary, we found that especially dielectric coatings have a beneficial impact on the induced saturation parameters as they significantly reduce the rollover in reflectivity. This prevents in modelocked operation pulse break-up into multiple pulses at high fluences.

In order to obtain information on the damage behavior of our samples, we carried out a number of experiments deep in the rollover regime. We
measured the damage threshold fluence $F_{\text{dam}}$ of all SESAMs, following the experimental procedure explained in detail in section 2.3.1. It is interesting to note that regardless of the sample, instantaneous damage occurred only at fluences much higher than typical pulse fluences on SESAMs of state-of-the-art SESAM modelocked TDLs [43, 15, 16, 54]. Table 3.2 shows that our high-finesse SESAMs exhibited larger damage thresholds than the basic structure without topcoating. We noticed a significant influence of the dielectric topcoating, shifting $F_{\text{dam}}$ to larger fluences. In the case of two dielectric layer pairs we observed an increase of the damage fluence by a factor of $\approx 4$. Moreover, the sample coated with three dielectric layer pairs sustained the maximum available pulse fluence of $210 \mu\text{J/cm}^2$. This particular SESAM was exposed to the maximum fluence level for several hours without any sign of damage. This is in contrast to the semiconductor top-reflector that caused only a minor increase in $F_{\text{dam}}$. We conclude that the damage threshold fluence strongly depends on the material composition of the specific topcoating layers and consequently on the strength of the TPA coefficient $\beta_{\text{TPA}}$ inside the structure. To obtain a detailed characterization of the damage behavior of our SESAMs we extended our study to pulse fluences lower than the damage threshold fluence. We exposed the test samples to fluences levels ranging from the instantaneous damage fluence to around 80% of this value and recorded the time-to-damage. Our
measurements revealed an exponential dependence of the lifetime on the pulse fluence, as depicted in figure 3.2. We therefore fit the lifetime curves with a single exponential as a function of pulse fluence $F_p$:

$$t(F_p) = t_0 + e^{-(F_p - F_3)/F_4}.$$  \hspace{1cm} (3.1)

The values of the fit parameters $t_0$, $F_3$ and $F_4$ are shown in table 3.2. The parameter $t_0$ corresponds to a small shift of the exponential on the time axis, required to correctly fit the measurements and has no physical meaning. The parameter $F_3$ represents the shift of the exponential on the fluence axis and thus the shift of the damage threshold to higher fluences. The inverse of the parameter $F_4$ represent the decay rate. Hence, a larger value of $F_4$ can be interpreted as a steeper “slope” of the exponential. The increase in $F_3$ of the topcoated test samples was especially pronounced in the case of the dielectric top-reflector. Furthermore, this specific SESAMs had almost a three-fold higher value of $F_4$ than the noncoated sample, suggesting an even larger difference between the lifetimes of this two SESAMs at lower pulse fluences. Our experiments therefore revealed the advantageous effect of top-reflectors, in particular dielectric topcoatings, as they shifted damage threshold fluences and lifetime curves to record-high pulse fluences. Such parameters are beneficial to overcome fluence instabilities below the QML threshold.
It is interesting to note that damage occurred deep in the rollover regime. Furthermore, the increase of the induced absorption coefficient $F_2$ of the topcoated SESAMs was related to a shift of the damage threshold fluence $F_{\text{dam}}$ to higher pulse fluences. This suggests a damage mechanism that is related to the absorbed energy within the structure due to induced absorption. We therefore derived an expression for the amount of absorbed energy per area as a function of pulse fluence $F_p$

$$F_{\text{abs}}(F_p) = F_p \cdot \left(1 - R(F_p)\right), \quad (3.2)$$

taking into account certain approximations. We simplified the expression 2.12 describing the reflectivity $R(F_p)$ of a laser beam with flat-top intensity profile, by assuming a strongly saturated absorber ($F_p \gg F_{\text{sat}}$) with a large induced absorption coefficient ($F_p \ll F_2$) to

$$R(F_p) \approx R_{\text{ns}} \cdot \left(1 - \frac{F_p}{F_2}\right). \quad (3.3)$$

The absorbed energy per area is therefore

$$F_{\text{abs}}(F_p) \approx F_p \cdot \left(1 - R_{\text{ns}} \cdot \left(1 - \frac{F_p}{F_2}\right)\right). \quad (3.4)$$

The SESAMs we used for our study had negligible nonsaturable losses ($\Delta R_{\text{ns}} \to 0$) and therefore ($R_{\text{ns}} \to 1$). Hence, the expression for the absorbed energy per area simplifies to

$$F_{\text{abs}}(F_p) \approx \frac{F_p^2}{F_2}. \quad (3.5)$$

If we associate the damage mechanism of a SESAM to a certain amount of deposited energy within the structure, instantaneous damage occurs once the absorbed energy exceeds a critical level

$$E_{\text{abs}}(F_{\text{dam}}) = E_{\text{crit}} = \text{const.} \quad (3.6)$$

As we used a constant spot diameter of $\approx 20 \ \mu\text{m}$ on our samples, we find

$$F_{\text{abs}}(F_{\text{dam}}) \approx \frac{F_{\text{dam}}^2}{F_2} = \text{const.} \quad (3.7)$$

Our approximations suggests that damage is related to a constant ratio between $\sqrt{F_2}$ and $F_{\text{dam}}$, independently of the specific absorber section.
We therefore had to verify the proportionality between this two parameters for SESAMs with different absorber layers and topcoatings. Figure 3.3 shows the ratio $\sqrt{F_2/F_{\text{dam}}}$ of the basic SESAM with 3 QWs and the corresponding topcoated samples we used for our study and additional data we obtained from samples with 1 QW and 6 QWs and a DBR. We calculated an average ratio of $1.72 \sqrt{\text{cm}^2/\text{mJ}}$ with a standard deviation of $0.24 \sqrt{\text{cm}^2/\text{mJ}}$. The result seems to confirm that the main contribution to damage was given by the deposited energy within the DBR and spacer layer, and that the absorbed energy in the absorber layer was of minor importance. The observed dependence of the damage process on the strength of induced absorption was of great importance, as it gave us the flexibility to tailor the damage threshold of our SESAMs by simply shifting the rollover to higher fluences.

Our study based on SESAMs with semiconductor and dielectric topcoatings revealed important aspects of the influence of top-reflectors on nonlinear reflectivity behavior and damage process. That allows us to provide guidelines on the design and post-processing of SESAM developed for operation in state-of-the-art SESAM modelocked TDL demonstrating highest pulse energy levels:

- Multiple QWs in a single or consecutive antinodes allow to tune the modulation depth of the resulting samples without influencing their saturation fluence. During post-processing, the mirror section placed on top of the spacer layer reduces the initially large modulation depth to typical values of samples operating in SESAM modelocked TDLs.

- If a larger number of QWs is used, the amount of material to add becomes significant. In this case, one could consider replacing the commonly used GaAs barrier layers with another material that exhibits a lower TPA coefficient $\beta_{\text{TPA}}$ and to use, for instance, AlAs spacer layers.

- As the top layers are exposed to high intensities and strongly contribute to induced absorption at high pulse fluences, dielectric material are preferred over semiconductor top-reflectors. SESAM with
dielectric topcoating show significantly reduced induced absorption and consequently, higher damage threshold fluences.

- In order to avoid residual heat deposition in the absorber, the growth temperature of the absorber layers is to be chosen such that the resulting nonsaturable losses are negligible \( \Delta R_{ns} < 0.1\% \).

We investigated in this section on nonlinear reflectivity parameters and damage threshold of SESAMs with QW absorber grown at a temperature of 400°C. This growth temperature provides minimal growth defects within the absorber structure resulting in small nonsaturable losses. However, the relatively long recovery time of \( \tau_{1/e} \approx 200 \text{ ps} \) limits the minimum achievable pulse duration, as shown in formula 2.6. Most of the latest TDL experiments aiming for high output powers were based on the standard thin-disk gain material Yb:YAG. This laser crystal is available with a high optical quality but suffers from a relatively low bandwidth. In this case, the pulse duration is mostly limited by the small FWHM emission bandwidth of the laser crystal and not by the response time of the SESAM. However, in order to explore new pulse duration limits of modelocked TDLs it is beneficial to have SESAMs with short recovery times of only few ps. Hence, we explored the influence of growth temperature on relaxation dynamics, nonsaturable losses and damage fluence of as-grown and dielectric topcoated SESAMs.

3.2 SESAMs with short recovery times

The SESAMs with dielectric top-reflectors that we presented in the previous section showed high damage thresholds, low nonsaturable losses and intermediate recovery times. They successfully initiated and stabilized soliton modelocking of TDLs demonstrating impressive levels of output power and pulse energy [15, 16]. These cutting-edge ultrafast oscillators provided pulse durations of several hundred femtoseconds. In order to pave the way for future SESAM modelocked TDLs that provide these performances with shorter pulse durations, we have to find appropriate gain materials with large emission spectra. In addition, we have to investigate in SESAMs with ultrafast relaxation dynamics as they support shorter pulse duration, see formula 2.6. The response time of a specific absorber
3.2. SESAMs with short recovery times

Figure 3.4: Recovery dynamics of SESAMs with LT-grown QW layers, measured with pulses centered around 1030 nm and a duration of 1 ps. The pump pulse fluence on the sample was \( \approx 250 \, \mu J/cm^2 \). The increased response time of absorbers grown at higher temperature was in agreement with previous studies. The basic SESAM without topcoating (NTC) and the corresponding samples with dielectric top reflectors (DTC1, DTC2) had almost identical relaxation times. For the sake of clarity we do not show the relaxation process of the absorber grown at 350°C.

The main purpose of our investigation was to find the best compromise between short recovery times and nonsaturable losses. Hence, the main purpose of our investigation was to find the best compromise between short recovery times and nonsaturable losses. An additional aspect concerned the damage behavior of SESAMs with LT grown absorber section. According to formula 3.7, the damage process of a SESAM is mainly determined by induced absorption rather then the structure of the specific absorber layer. We therefore expected to measure similar damage threshold fluences for a given SESAM structure, independent of the growth temperature. In addition, we based our study on AlAs spacer layer, as this material exhibits a lower TPA coefficient \( \beta_{TPA} \) than GaAs. For our measurements we used SESAMs with QW layers grown at different low
temperatures. They were additionally topcoated with dielectric reflectors in order to study the influence of the field intensity enhancement inside the semiconductor device on the parameters of interest. We characterized the sample with the nonlinear reflectivity setup shown in figure 2.3 and the standard noncollinear pump-probe setup depicted in figure 2.6 at a pump fluence of \( \approx 250 \, \mu J \). The two optical setups were based on Yb:YAG lasers delivering pulses with a duration of 1 ps, centered around 1030 nm. The representative samples consisted of a set of antiresonant SESAMs with identical QW absorbers grown at temperatures of 270°C, 300°C, 350°C and 400°C. All SESAMs had 4 QW layers of 7 nm thickness, placed two-by-two in consecutive antinodes of the standing wave pattern inside the AlAs spacer layer. During post-processing we coated the samples with one respectively two quarter-wave SiO\(_2\)/Si\(_3\)N\(_3\) layer pairs, referred to as DTC1 and DTC2.

Figure 3.4 shows the normalized saturation of the samples under investigation as a function of time delay between pump- and probe pulse. As expected, we measured increased response times of the absorbers grown at higher temperatures. Our results were therefore consistent with previous studies reporting on relaxation dynamics of LT-grown SESAMs [47, 57]. It is interesting to note that the recovery time \( \tau_{1/e} \) of SESAMs grown at a specific temperature seems to be independent of the additional topcoating layers. Hence, we are able to take advantage of the beneficial effect of di-

\[ \text{Figure 3.5: Nonlinear reflectivity behavior of SESAMs with identical absorber layers grown at } 270^\circ \text{C. The graphs illustrate the effect of dielectric topcoatings (DTC1, DTC2) on nonsaturable losses and induced absorption of the basic structure without topcoating (NTC).} \]
3.2. SESAMs with short recovery times

<table>
<thead>
<tr>
<th>SESAM</th>
<th>$F_{\text{sat,A}}$</th>
<th>$\Delta R$</th>
<th>$\Delta R_{\text{ns}}$</th>
<th>$F_2$</th>
<th>$F_{\text{dam}}$</th>
<th>$\tau_{1/e}$</th>
<th>$\tau_{\text{fast}}$</th>
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Table 3.3: Nonlinear reflectivity parameters and recovery time $\tau_{1/e}$ of SESAMs with QWs grown at 270°C, measured at 1030 nm with a pulse duration of 1 ps. The table shows the parameters of the basic sample (NTC) and the modified SESAMs with one respectively two dielectric quarter-wave layer pairs (DTC1, respectively DTC2).

electric top-reflectors on damage threshold and nonsaturable losses, illustrated in figure 3.5, while the ultrafast response time of the SESAM is preserved. We measured an almost identical damage threshold fluences for a given SESAM structure, as illustrated in figure 3.6. The as-grown SESAMs without topcoating exhibited damage threshold fluences of 25 mJ/cm² at 270°C up to 29 mJ/cm² at 400°C. These values increased by a factor of $\approx 1.6$, respectively $\approx 2.5$ in the case of SESAM with one, respectively two dielectric layer pairs. This observation indicates that the increased lattice defects of absorber sections grown at lower temperatures have only minor influence on the damage threshold fluence of a specific SESAM design. The non-topcoated SESAM with QW layers grown at the lowest temperature of 270°C showed nonsaturable losses of $\Delta R_{\text{ns}} \approx 0.7 \%$, compared to $\Delta R_{\text{ns}} \approx 0.1 \%$ of the other samples. However, the corresponding modified structures with dielectric topcoating had reduced nonsaturable losses see table 3.3.

In summary, our measurements confirmed that the use of dielectric topcoatings is a simple and straightforward method to obtain large damage thresholds and reduced nonsaturable losses, mostly independent of the absorber growth temperature. The obtained results allowed us to extend the guidelines for high-damage threshold SESAMs we derived in the previous section to SESAMs with non-negligible nonsaturable losses and ultrafast response time. These SESAMs were optimized for operation in modelocked TDLs targeting shortest pulse durations. As we expect the strength of induced absorption to increase within the same order of magnitude than the pulse duration decreases, large damage thresholds and
reduced induced absorption are of great importance. This significant aspect in the development of ultrafast SESAMs allowed us to demonstrate first power scaling experiments of SESAM modelocked TDLs with short pulse durations [58, 59].

Our study was based on AlAs barriers, as this material exhibits a lower TPA coefficient $\beta_{\text{TPA}}$ than GaAs. However, the influence of the spacer layer material on induced absorption and recovery dynamics and its interplay with absorber growth temperature is not completely understood yet and requires further investigations. AlAs has in addition a higher thermal conductivity $\kappa$ of $\approx 91 \text{ W/Km}$, than GaAs with $\kappa$ of $\approx 45 \text{ W/Km}$ [55]. Hence, future modifications of the standard SESAM design could involve novel mirror designs with larger amounts of AlAs material, compared to GaAs. Such adapted mirror structures will result in better heat removal.
3.3 Substrate-removed SESAMs

capabilities, beneficial for SESAMs operating in high-power ultrafast oscillators. Another possible improvement for better thermal management of SESAMs is substrate-removed contacting, as we will highlight in the next section.

3.3 Substrate-removed SESAMs

So far, thermal effects in SESAMs have not been the main limitation for power scaling of modelocked TDLs. Deposited heat in SESAM structures is very low and is mostly contributed by the residual nonsaturable losses of the samples. The power-scaling concept of modelocked TDLs relies on increasing the spot sizes both on the disk and on the SESAM. Although we have the design freedom to engineer SESAMs with increased saturation fluences to keep the saturation level constant, spot sizes will most likely also become larger as we move towards several kW of intracavity powers. In future thin-disk oscillators demonstrating such power levels, the laser-induced thermal lensing and surface deformation due to small fractions of absorbed intensity can become significant [60]. In this case, the resulting beam distortion and degradation have to be considered as a limiting factor of the laser performance. Hence, the development of large scale SESAMs with improved thermal management is essential to push the limits of the current thin disk laser technology.

A promising approach to obtain SESAMs with enhanced heat-removal capabilities is based on substrate removal. Standard SESAM structures
shown in figure 2.2, obtained with MBE, are grown on a GaAs substrate with low thermal conductivity of $\approx 45$ W/Km. As the GaAs wafer does not contribute to the optical properties of the SESAM, it only limits the heat dissipation performances of the structure. Hence, substrate removed contacting is a straightforward post-processing method to improve the heat removal of SESAMs.

A possibility to obtain substrate-removed samples, is to grow the SESAM up-side down (USD), first the absorber section embedded in the spacer layer and then the DBR structure. During post-processing, we contact the sample on a heat sink with high thermal conductivity, as for instance copper with a thermal conductivity $\kappa$ of $\approx 400$ W/Km, and remove the substrate. This technique is usually referred to as “flip-chip” bonding. It is a common approach for power scaling of VECSELs, as in such structures the high heat load due to pump light absorption becomes a critical issue [55, 61]. The resulting structure has typical thicknesses of only few $\mu$m and can be very efficiently cooled. The fabrication steps required for contacting USD grown SESAMs are illustrated in figure 3.7. During the time frame of this thesis we developed first test samples, contacted on copper heat sinks. Certain fabrication steps as uniform etching of the etch-stop layer and bonding the samples without pre-stress still appeared challenging. However, we achieved first laser results based on USD grown SESAMs shown in figure 3.8. The SESAM that initiated and stabilized fund-
damental soliton modelocking in single mode operation had the following saturation parameters: $F_{\text{sat,A}} \approx 126 \, \mu J/cm^2$, $\Delta R \approx 1.2\%$ and $\Delta R_{\text{ns}} < 0.1\%$. It exhibited an almost perfect spherical surface deviation with a radius of curvature (ROC) of $\approx 120 \, m$. This value was substantially larger than typical ROC $< 30 \, m$ we measured for SESAMs contacted with the standard indium soldering method. We benefit from such a large ROC as it simplifies the cavity design. Moreover, the negligible astigmatism of this SESAM is a further advantageous property, as it is difficult to compensate for astigmatism with a normal laser setup. The Yb:LuO based thin-disk oscillator demonstrated 26 W of average output power at a repetition rate of 43 MHz with an $M^2 < 1.1$. The pulses were centered around 1034 nm, had a duration of 470 fs and were almost transform-limited with a time-bandwidth product of $\tau_p \cdot \Delta \nu \approx 1.08 \cdot 0.315$. We developed the USD grown SESAM in accordance with the design guidelines presented in section 3.1. Hence, a laser mode diameter on the SESAM of 2 mm and an output coupling rate of 7.8 % resulted in a pulse fluence on the absorber of $\approx 250 \, \mu J/cm^2$. The resulting low saturation parameter ($S \approx 2$) would have allowed us to scale the laser performance to higher power levels. The next step is therefore to optimize certain fabrication steps in order to obtain surfaces with high optical quality. This will allow us to use such USD grown SESAMs with excellent heat removal capabilities in high-power TDL setups with large mode diameters.

**Figure 3.9**: Schematic of the fabrication steps required for contacting a RSU-grown, substrate-removed SESAM.
Another promising contacting method relies on standard right-side up (RSU)-grown SESAMs that in addition contain a 300-nm thick etch stop layer (Al_{0.85}Ga_{0.15}As) between the GaAs substrate and the DBR. The substrate-removed samples were mounted on a super-polished silicon carbide (SiC) heat sink with a thermal conductivity of $\kappa \approx 490$ W/Km. The proprietary crystalline coating process was developed by Crystalline Mirror Solutions (CMS) [62]. The substrate-transfer and direct bonding (STB) method allows for the integration of high-performance single-crystal epitaxial material onto polished and even curved surfaces. A schematic of the fabrication steps required for contacting a RSU-grown, substrate-removed SESAM is shown in figure 3.9. We used a commercial phase shifting Twyman-Green interferometer to compare ROC and surface deformation of a set of samples with identical absorber section, contacted either by means of STB or standard indium soldering [63]. The interferometric measurements revealed significant advantages of STB based SESAMs, see figure 3.10. Their impressively large radii of curvature ($> 1000$ m) exceeded typical values of ROC of traditional SESAMs by more than one order of magnitude. Moreover, standard indium soldered SESAMs tended to show slight astigmatic surface deviations whereas STB based samples were almost perfectly flat, except for some minor defect lines. We measured similar nonlinear reflectivity parameters and ultrafast response times for both
contacting methods. This confirms that macroscopic SESAM parameters remain unaffected during the contacting process. Our preliminary experiments showed that STB based SESAMs have high-quality surfaces with remarkably improved flatness, superior to surface properties of indium soldered SESAMs. In addition, one could consider replacing the copper heat sink with a diamond heat spreader ($\kappa$ of $\approx 2200$ W/Km in order to further improve the heat dissipation capabilities of the structure. We conclude that STB based SESAMs are promising candidates for stable pulse formation in future high-power TDLs with large beam areas, outperforming the current state-of-the-art ultrafast thin-disk oscillators.
Chapter 4

High-power ultrafast thin-disk lasers

The development of ultrafast high-power oscillators and amplifiers over the last years has been impressive. Their continuing success story has revolutionized many material-processing applications. Such high-power laser systems provide pulse durations in the ps-regime and close to diffraction-limited beam quality. In combination with high repetition frequencies, they enable high-speed cutting, remote welding and high-precision micromachining of a wide variety of materials including metals, glasses and semiconductors [27, 64].

In the scientific world, experiments in attoscience [65] and strong-field physics [9] rely on femtosecond driving sources that provide field intensities in the order of electron binding fields. The growing need of these research fields for faster acquisition times and higher yields has pushed the development of ultrafast laser systems with repetition rates in the MHz-range [12, 13, 11]. Such laser technologies, which combine high peak- and average power with nearly diffraction-limited beam quality, are mostly based on rare-earth doped solid-state materials.

Solid-state lasers can be efficiently pumped with diode lasers. However, heating of the laser crystal is inevitable due to the energy difference between pump and laser photons referred to as quantum defect \((1 - \lambda_{\text{pump}}/\lambda_{\text{laser}})\). At high power levels, the heat load becomes significant. The deposited energy evokes a transverse temperature gradient within the gain that has detrimental effects on the laser performance. The
thermo-optic-induced refractive index change leads to thermal lensing. Thermo-mechanical effects cause deformation and stress-induced birefringence [66]. The thermal aberrations can lead to degradation of the beam quality and set a limitation on the power scalability. In order to overcome these critical issues, the geometry of the gain has to be optimized for outstanding heat dissipation capabilities and homogenous transverse temperature profiles. Efficient cooling is achieved when the surface-to-volume ratio of the gain material is maximized. Starting from a typical rod-shaped gain material of solid-state bulk lasers, we can distinguish between three concepts that fulfill this requirement: the fiber-, the slab- and the thin-disk geometry depicted in figure 4.1.

The fiber technology implements the geometry of a long gain medium with small diameter into an optical fiber that confines the laser light in the core due to total internal reflection [67]. Conventional single mode double-cladding fibers have typical core diameters in the range of 6 µm up to 40 µm. The main challenge of high-power and high-brightness fiber lasers are excessive nonlinearities arising from high intensities in the small active area in the core of the fiber. Nonlinear processes as SPM, self-focusing and stimulated inelastic scattering impose limitations on the fundamental-transverse mode performance at high power levels. Ultrafast fiber amplifiers relying on chirped-pulse amplification [68] circumvent the
disadvantageous effect of strong nonlinearities and provide high average powers at MHz-repetition rates [12]. Recent experiments report on first high-repetition-rate HHG close to the MHz regime obtained from a fiber-based OPCPA system [14]. However, the amplifier architecture consists of several optical stages, which implies a rather high system complexity and maintenance effort.

The slab laser configuration reduces the height (y direction in figure 4.1) of the gain medium to typical thicknesses of only few millimeters [69]. In a conventional slab design, the slab-shaped gain crystal is pumped through one of the two polished end-faces (z direction). The laser beam propagates either along a zigzag path through the gain material due to total internal reflection or straight through the slab without any further reflections. The Innoslab concept is based on a longitudinal and partially pumped slab crystal [70]. High-repetition rate amplifier systems based on this technology provide high average power levels in the kW regime with sub-ps pulse durations [71, 11]. The thermally-induced cylindrical lens within the gain material is a challenging engineering task [72]. In addition, the Innoslab technology is based on rather complex amplification stages. We therefore believe that TDLs are the best suited technology to achieve high average output powers directly from a table-top oscillator.

4.1 Thin-disk laser concept

Thin-disk oscillators and amplifiers are based on a geometrical concept that has been proposed first by Giesen et al. in the year 1994 [17]. The sophisticated laser design consists of a thin-disk shaped gain material with typical thicknesses about 100 μm up to 300 μm. In general, pump- and laser radiation propagate along the z-direction (see figure 4.1), perpendicular to the two polished top and bottom surfaces of the disk. The gain crystal is typically used in reflection in form of an “active mirror”. To allow this, the two polished surfaces of the disk have different dielectric coating layers. The front side is coated with an anti-reflective (AR) layer for both laser and pump wavelength, whereas the coating on the backside consists of a HR layer again for the same spectral range. Hence, one reflection of the laser beam at the active disk mirror corresponds to a double pass
through the gain medium. The short optical path length within the gain medium efficiently reduces disadvantageous nonlinearities, as for instance self-focusing, to a minimum. Moreover, the small thickness of the gain material results in an almost constant temperature gradient within the pumped volume. This corresponds to a one-dimensional heat flow along the propagation axis allowing for efficient cooling of the thin disk through the back. The excellent heat dissipation capabilities of the thin-disk geometry successfully mitigate optical aberrations due to thermo-optically induced birefringence by more than an order of magnitude compared to rod-type shaped gain crystals [18].

As the interaction of the pump light with the active medium is restricted to a short optical path length, the fraction of absorbed pump photons within a single pass is limited to few percent. The pump arrangement depicted in figure 4.2 overcomes this limitations. The radiation of a multimode diode laser is collimated by a set of lenses. An optical arrangement usually consisting of four prisms and a parabolic mirror focuses the pump beam under an angle several times on the gain crystal. Depending on the collimation optics, the pump spot diameter on the disk can range from few millimeters up to several centimeters. The TDLs we present in this thesis were all pumped with 24 passes on the gain medium. Considering
4.1. Thin-disk laser concept

Figure 4.3: Left: Schematic of a water-cooled thin disk. The thin gain-crystal is mounted on an appropriate heat sink with high thermal conductivity. Water-cooling of the heat sink through the back allows for efficient removal of the energy deposited during laser operation. Right: In general, the thin disk is used in reflection in form of an active mirror. To allow this, the front side is coated with an AR layer for both laser and pump wavelength, whereas the coating on the backside consists of an HR layer for the same spectral range.

typical disk thicknesses and doping concentrations this configuration resulted in most of the experiments in a pump absorption > 98 %. However, current state-of-the-art pump modules are available with up to 44 passes on the laser crystal. Such an arrangement is beneficial for extremely thin disks optimized for outstanding heat removal capabilities.

4.1.1 Mounting of thin disks

The thin-disk crystal is mounted on a heat sink consisting of a material with high thermal conductivity. During laser operation the heat sink is water-cooled from the back. A schematic of the cooling scheme of a thin-disk crystal is shown in figure 4.3. Heat-sink materials that match the thermal expansion of the gain material are advantageous to reduce stress during the mounting procedure and later on during laser operation. In the case of Yb:YAG, this is achieved with the use of heat sinks made of copper-tungsten alloys that match the coefficient of thermal expansion (CTE) of the gain crystal. There exist different thin-disk contacting methods, as for instance indium-tin soldering or gluing. In order to compare these mounting techniques, we measured for three Yb:CALGO thin-disks with different thicknesses the change in diopter with increasing absorbed pump intensity. The result is shown in figure 4.4. The thinnest disk with 100 µm-thickness was glued on a diamond heat sink. We observed no change in
Figure 4.4: We measured the change in diopter for three Yb:CALGO thin-disks as a function of absorbed pump intensity. We observed no change in diopter for the diamond-glued disk with 135 µm-thickness within the measured pump intensity range (cross). The diamond-glued disk of 300 µm thickness (circle) showed a similar strength of thermal lensing than the thinner 223 µm-thick crystal soldered on copper-tungsten (triangle).

diopter up to the maximum absorbed pump intensity of 2.3 kW/cm². This is in contrast to the diamond-glued disk of 300 µm-thickness. It showed a linear increase in diopter with increasing absorbed pump intensity and a similar strength of thermal lensing as the thinner, 223 µm-thick crystal soldered on copper-tungsten. Our measurements indicate that gluing the disk on diamond reduces the strength of deformation and thermal lensing. However, further investigations on different thin-disk compounds are required in order to provide guidelines on the choice of heat sink materials and the corresponding contacting technique.

Although an appropriate choice of the contacting method and heat-sink material is of great importance, optical, mechanical and thermal properties of the gain medium have an even higher impact on the laser performance. We therefore explain in the next section the requirements on a thin-disk gain material and introduce different gain crystals ideally suited for the use in a TDL configuration.
4.2 Thin-disk laser gain materials

A key point for the outstanding heat removal capabilities of TDLs is the minimized thickness of the gain medium. In order to obtain the geometrical shape of the thin-disk unit several processing steps are required. Mechanical procedures as cutting, polishing and mounting rely on superior mechanical properties, as for instance a relatively high hardness of the gain material, see table 4.1 and 4.2. In order to absorb the pump light efficiently, the short optical distance of a thin-disk crystal has to be compensated for by multiple pump beam passes on the disk and a comparatively high doping concentration of the gain medium. Thus, a gain material with negligible quenching processes like cross-relaxation and upconversion even at high doping levels is required [81]. Furthermore, a small quantum defect is an indispensable property, as the deposited heat within the crystal structure has to be minimized.

Ytterbium (Yb)-doped rare-earth materials are suitable thin-disk gain materials. They have lasing wavelengths in the 1 µm spectral domain similar to the lasing wavelengths of standard titanium (Ti)-doped sapphire around 800 nm. However, Ti:sapphire oscillators have typically low doping concentrations around 0.25 at.% and suffer from large quantum defects higher than 30%. Yb-doped rare-earth crystals, in contrast, have typical doping concentrations around 2 at.% due to the simple energy level scheme of the $\text{Yb}^{3+}$-ion with only two 4$f$ Stark manifolds. Furthermore, they have low quantum defects of only few percent, a result of the quasi-three level laser transition of Yb-doped crystals. The drawback of a quasi-three-level gain medium is its increased laser threshold due to reabsorption. However, the thin-disk geometry allows for efficient cooling resulting in a low population density of the lower laser-level.

Relevant for an efficient pumping is a high absorption cross-section with preferably large bandwidth. Yb-doped host materials exhibit maxima in their absorption cross-sections within the spectral range of 900 nm up to 1000 nm, see figure 4.5. This spectral domain is accessible with the radiation of high-power diode lasers. Nowadays, volume Bragg grating (VBG) systems enable to stabilize the wavelength of high-power diode lasers within spectral bandwidths smaller than 0.5 nm. They allow for
## 4. High-power Ultrafast Thin-Disk Lasers

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<th>Yb:ScO</th>
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**Table 4.1:** Crystal properties of Yb:YAG and the pure sesquioxides Yb:LuO, Yb:ScO and Yb:YO based on values found in literature. The table shows the common pump wavelength and the corresponding absorption cross-section and bandwidth, emission peak wavelength and the corresponding cross-section and FWHM bandwidth and other relevant parameters. The crystals are grown using the heat exchanger method (HEM) or Czochralski (Cz) growth.
4.2. Thin-disk laser gain materials

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</tbody>
</table>

Table 4.2: Crystal properties of Yb:CALGO and the mixed sesquioxides Yb:LuScO and Yb:ScYLO based on values found in literature. The table shows common pump wavelength and the corresponding absorption cross-section and FWHM bandwidth, emission peak wavelength and the corresponding cross-section and FWHM bandwidth and other relevant parameters. The crystals are grown using the heat exchanger method (HEM) or Czochralski (Cz) growth.
Figure 4.5: Left: Absorption cross-section of Yb-doped YAG, LuO, LuScO and CALGO in a spectral range of 910 nm up to 1050 nm. Right: Gain cross-section of Yb-doped YAG, LuO, LuScO and CALGO in a spectral range of 980 nm up to 1100 nm. The inversion level $\beta$ represents the fraction of active Yb-ions in the upper laser level. We estimate that we operate all our experiments at an inversion level lower than 0.15.
pumping at the typically narrow zero-phonon line (ZPL) around 969 nm up to 979 nm, see figure 4.5.

Within the framework of this thesis we were exploring new pulse duration limits of modelocked TDLs. In this aspect, it is important to use laser crystals with flat and broadband gain spectra. The inversion level of the Yb-ions during laser operation determines the gain cross-section of a laser crystal:

\[ g(\lambda_L) = \beta \cdot \sigma_{em}(\lambda_L) - (1 - \beta) \cdot \sigma_{abs}(\lambda_L). \] (4.1)

The parameter \( \beta \) represents the fraction of active Yb-ions that are in the upper laser level. The parameters \( \sigma_{em}(\lambda_L) \) and \( \sigma_{ab}(\lambda_L) \) are the emission and absorption cross-sections at the lasing wavelength \( \lambda_L \). According to formula 2.6, the minimum achievable pulse duration is proportional to the round-trip power gain and therefore influenced by the inversion level. We estimate to operate all our experiments at an inversion level \( \beta \) lower than 0.15. However, TDLs providing record-high average power have usually large output coupling rates to avoid an excess of nonlinearities within the resonator. This corresponds to a stronger saturated gain resulting in a more pronounced gain peak at the center wavelength. Thus, a flat spectral gain profile with broad bandwidth is essential for the generation of ultrashort pulses at high power levels. In a real laser resonator most of the optical components, as for instance dispersive mirrors, output coupler and SESAM, have a spectrally limited bandwidth and shape the gain accordingly. Taking into account these spectrally dependent losses of the cavity elements yields an accurate expression of the gain cross-section \( g(\lambda_L) \).

However, it is a complicated task to estimate the influence of the specific laser configuration on the gain cross-section. It is often more adequate to refer to emission cross-section and bandwidth that are both intrinsic properties of the gain material, rather than the inversion-dependent gain. In order to evaluate the potential of a laser crystal for the generation of short pulse durations we therefore compared the emission bandwidths shown in table 4.1 and 4.2.
In summary, high-power TDL experiments targeting the generation of short pulses require a gain material with the following properties [82]:

- Superior mechanical strength in order to sustain certain processing steps as cutting, polishing and contacting.
- Potential for high doping levels combined with minimal parasitic energy transfer processes within the crystal.
- Low quantum defect to minimize the amount of deposited energy within the crystal.
- High thermal conductivity for efficient heat removal.
- Absorption cross-section maxima in a spectral range that is accessible with the radiation of high-power diode lasers.
- Large emission bandwidth supporting short pulse durations.
- Low thermo-mechanical effects to reduce stress-induced birefringence.
- Low thermo-optic effect $dn/dT$ to reduce thermal lensing.
- Large emission cross-section and long upper-state lifetime to obtain a low laser threshold.

In the following, we present several Yb-doped host crystals relevant for our experimental work on thin-disk oscillators. We compare these gain materials with respect to their power-scaling capabilities and their potential for exploring the limits of minimum achievable pulse durations.

### 4.2.1 Yb-doped YAG

The first demonstration of a TDL [17] was based on Yb$^{3+}$-doped yttrium aluminium garnet (Y$_3$Al$_5$O$_{12}$), usually abbreviated to YAG. This host material has a melting point of 1940°C and can be grown by the crucible-based Czochralski technique or other procedures, resulting in single crystals with high optical quality and dimensions of several centimeters [83, 84]. The gain material Yb:YAG has a good thermal conductivity of 7 W/(m·K) at an Yb-ion concentration density of $8 \cdot 10^{20}$ cm$^{-3}$. Advantageous is its large emission cross-section of $1.9 \cdot 10^{-20}$ cm$^2$, compared to other Yb-doped host materials. Furthermore, its broad absorption band-
width of 12 nm at 940 nm allows for cost-efficient pumping, as wavelength-stabilized pump radiation is not required. Alternatively, Yb:YAG can be pumped with VBG stabilized diode lasers at the zero phonon line of 969 nm, resulting in a smaller quantum defect. In summary, Yb:YAG offers a number of advantageous properties and is at present the standard gain material of commercial TDLs. Moreover, ultrafast thin-disk oscillators delivering more than 200 W of average power are up-to-date always based on Yb:YAG [85]. The drawback of this gain crystal is its small emission bandwidth of 9 nm. However, KLM of an Yb:YAG thin-disk oscillator demonstrating impressive values of average and peak power with a pulse duration in the range of 210 fs up to 330 fs continues with the success story of this gain material [21].

4.2.2 Yb-doped sesquioxides

The first experiment with a modelocked TDL exceeding the 100 Watt level [54] was based on Yb-doped lutetium oxide (Lu$_2$O$_3$), usually abbreviated to LuO. This host crystal belongs to the group of rare-earth sesquioxides with cubic lattice symmetry. The pure rare-earth sesquioxides have a chemical compositions of $RE_2O_3$ ($RE = Lu, Sc, Y$) in which the ratio of metal- to oxygen ions is two to three. In the case of mixed sesquioxides the chemical composition has the same ratio of metal- to oxygen ions and $RE = Lu_aSc_bY_c$ with $a + b + c = 1$ [22]. As the cation density of Yb-doped sesquioxides is about twice that of Yb:YAG, an Yb-ion concentration of 1 at. % in a sesquioxide crystal roughly corresponds to an Yb-ion concentration of 2 at. % in YAG.

Yb-doped sesquioxides have a number of advantageous properties that makes them excellent candidates for thin-disk gain materials. First, their properties are independent of the crystal cut due to the isotropic lattice structure. Second, they have large absorption cross-section at their ZPL around 975 nm and benefit therefore from a low quantum defect, compared to Yb:YAG pumped at 940 nm, see figure 4.5. And third, they have larger emission bandwidths than Yb:YAG, beneficial for the generation of short pulses.
In terms of power scaling, the pure sesquioxide LuO is the most promising alternative to the standard host material YAG. In general, the thermal conductivity of a laser crystal drops significantly with increasing doping concentration. Responsible for this effect are the different atomic weights of the substituted cation and the doping ion. This mass difference disturbs the heat-transport that in insulators like YAG or sesquioxides is mainly based on phonon propagation [86]. As a consequence, the thermal conductivity is lower at higher doping concentrations. Due to the small mass difference between Yb and Lu the phonon scattering is minimal and results in a superior thermal conductivity of Yb:LuO of 12 W/(m·K) at an Yb-ion concentration density of $8 \cdot 10^{20}$ cm$^{-3}$ [74]. Furthermore, Yb:LuO has a lower thermo-optic effect $dn/dT$ than Yb:YAG suggesting a minimal thermal lensing effect even at highest pump intensities [76].

The Yb-doped scandium oxide (Sc$_2$O$_3$) crystal exhibits the largest absorption cross-section and emission cross-section among all Yb-doped sesquioxides. Due to the large mass difference between yttrium and scandium of $\approx$ 93 g/mol, its thermal conductivity drops to a value comparable to that of Yb:YAG. The drawback of Yb-doped sesquioxides is their high melting point exceeding 2400$^\circ$C. It makes the growth of large single-crystalline regions with high optical quality a challenging task. Up to now the most promising results were obtained using the heat exchanger method (HEM) [87]. There is a further complicating factor in the case of yttrium oxide (Y$_2$O$_3$) that undergoes a phase transition from a hexagonal to a cubic structure at a temperature around 2280$^\circ$C. This is in contrast to LuO and ScO that only have cubic lattice structures [88].

The host crystals lutetium scandium oxide (LuScO$_3$) and scandium yttrium lutetium oxide ((Sc,Y,Lu)$_2$O$_3$) are mixed sesquioxides. Their disordered lattice structure results in broad emission bandwidths of 22 nm, respectively 18.5 nm. Hence, these two gain materials have within the group of Yb-doped sesquioxides the greatest potential for the generation of ultrashort pulse durations. Their FWHM gain bandwidths for typical inversion levels of TDLs theoretically support a minimum pulse duration of 51 fs (Yb:LuScO) and 62 fs (Yb:ScYLO) [75]. The increase in spectral bandwidth comes at the expense of a reduced thermal conductivity around 3.5 W/(m·K) at an Yb-ion concentration density of $8 \cdot 10^{20}$ cm$^{-3}$.
4.2. Thin-disk laser gain materials

[74]. The thermal management is therefore the critical issue of Yb:LuScO and Yb:ScYLO gain crystals.

4.2.3 Yb-doped CALGO

At present, the shortest pulses of a modelocked TDL [24, 89] were obtained by using Yb-doped calcium gadolinium aluminium oxide (CaGdAlO$_4$), usually abbreviated to CALGO. This host material belongs to the family of crystals with the chemical formula $A$$BC$$O_4$, where $A$ denotes Ca or Sr, $B$ represents an yttrium or another rare-earth ion and $C$ is Al or Ga [23]. Yb:CALGO has a relatively broad ZPL at 979 nm resulting in a low quantum defect of approximately 6% to 7%, a characteristic feature of Yb-doped rare-earth compounds. It has a similar cation density than Yb:YAG and thus, its doping concentration given in at.% is directly comparable to that of Yb:YAG. The melting temperature of Yb:CALGO is low around 1700°C. It simplifies the growth of laser crystals with high optical quality that can be obtained by the Czochralski (Cz) technique.

The lasing properties depend on the orientation of the crystal, a result of the uniaxial lattice structure. Regarding the geometrical shape of a thin-disk, a cut of the crystal along the optical axis (c-direction) is beneficial as it accounts for isotropic mechanical and thermal properties in the disk plane. In the crystal structure of CALGO, the two cations Ca$^{2+}$ and Gd$^{3+}$ equally share the same lattice site and can be both replaced by Yb$^{3+}$-ions. Depending on whether the surrounding lattice sites are mostly occupied by Ca or Gd cations, the Yb-ion experiences a different electrostatic field that shifts its $4f$ energy levels accordingly. This phenomenon could explain the presence of a plateau in the emission cross-section of Yb:CALGO in between 1000 nm and 1050 nm with a FWHM of $\approx$ 80 nm [90, 91]. The small mass difference between Gd and Yb suggests only a minimal decrease in thermal conductivity with increased doping concentration. However, in the case of Yb:CALGO the thermal conductivity depends not only on the doping level but the ratio of substituted Gd to Ca ions as well [78].

Studies made on this relatively new laser crystal reported of the observation of discrete scattering centers that might affect the optical perfor-
4. High-power ultrafast thin-disk lasers

Figure 4.6: Overview graph of ultrafast TDL experiments that shows average output power versus pulse duration. If not indicated differently, the thin-disk oscillators were SESAM modelocked and based on single crystalline gain materials. The shortest pulse durations that were achieved with a given gain material and pulse formation mechanism are labeled with the name of the corresponding host crystal. The results we present within this thesis are shown in color.

mance of Yb-doped laser oscillators [91]. However, the origin and the impact of this scattering centers on the laser performance are not completely elucidated yet and require further investigations.

Until 2007, only the two gain materials Yb:YAG and Yb:KYW have been used in modelocked thin-disk oscillators [92, 93]. Since then, a variety of novel single crystalline and ceramic gain materials has been developed and tested in the power-scalable TDL configuration. A detailed description of all these materials would go beyond the scope of this thesis. In the next section we give a short overview of the laser performance of SESAM and Kerr-lens modelocked ultrafast TDLs in the femtosecond regime.

4.3 SESAM and Kerr-lens modelocked thin-disk lasers

Since their first demonstration in the year 2000 [92], modelocked TDLs have outperformed other ultrafast oscillator technologies in terms of average power and pulse energy. The overview graph shown in figure 4.6
plots the average output power of femtosecond TDL experiments versus the demonstrated pulse duration. The results presented in this thesis are shown in color.

The first modelocked thin-disk oscillator that reached an average power exceeding the 100 W-level was based on the gain material Yb:LuO. It delivered 141 W of average power at a pulse duration of 738 fs and held the record for the highest average power from a modelocked TDL for many years [54]. Stable soliton modelocking only tolerates a maximum nonlinear phase shift that scales with intracavity peak power. The relatively low output- and intracavity energy of the pulses allowed to operate the Yb:LuO resonator in air. Later TDL experiments aiming for high power- and energy levels had to take appropriate measures to reduce the intracavity peak power of the oscillator. A possible approach to overcome the limitations of excessive nonlinearity is to increase the output coupling coefficient of a given laser setup. In order to operate the laser efficiently, the round-trip gain has to be increased by using an active multipass cell. Such a cavity design allows to reach higher output peak powers at a similar (or even lower) amount of generated SPM [43]. Alternatively, the oscillator can be operated in helium or in a vacuum environment in order to eliminate parasitic nonlinearities caused by the air inside the resonator. A TDL based on the latter approach provided 275 W of average output power and a pulse energy of 17 \( \mu \)J at a repetition rate of 16.3 MHz [15]. The obtained pulse duration was 583 fs. By using the same concept to reduce nonlinearity, energy scaling was successfully achieved and resulted in the recent demonstration of a pulse energy of 80 \( \mu \)J [16]. In this case, the thin-disk oscillator operated at a repetition rate of 3 MHz and an average power of 242 W. The achieved pulse duration was 1.07 ps.

KLM has attracted attention as a pulse formation mechanism that enables to exploit a large fraction of the available gain bandwidth. First experiments using a KLM TDL based on Yb:YAG delivered 17 W of average power at a pulse duration of 200 fs [94]. Recently, an energy scaling experiment of a KLM Yb:YAG laser demonstrated a maximum output power of 270 W at a pulse duration of 300 fs [21]. However, the saturable absorber action for KLM is defined with the laser cavity. The resonator operates therefore in cw mode close to the cavity stability limit [95].
In summary, most of the latest energy and power scaling experiments of modelocked TDLs were based on the standard gain material Yb:YAG. As it exhibits a relatively narrow emission spectrum, the demonstrated pulses had typical durations of several 100 fs. Currently, we still require external pulse compression stages to reduce the pulse durations of these oscillators for the targeted applications in strong-field science[96, 97]. As a consequence, there is a strong research effort in extending the performance levels of current state-of-the-art TDLs to shorter pulse durations. In order to overcome the trade-off between average output power and pulse duration, novel broadband thin-disk gain materials that meet the spectroscopic and thermo-mechanical requirements have been developed during the last years. They provide broad gain bandwidths due to their disordered lattice structure that combines shifted emission spectra of the constituting components.

In the next two chapters, we explore the potential of these promising gain materials with respect to the generation of short pulse durations (<200 fs) in SESAM modelocked TDLs.
Chapter 5

Experimental laser results: single broadband thin-disk gain materials

In this chapter, we present experiments where we explored the pulse duration limits of novel thin-disk gain materials with broader FWHM emission bandwidths than the standard laser crystal Yb:YAG. These gain materials are therefore promising candidates for the development of high-power TDLs that outperform current leading-edge ultrafast thin-disk oscillators based on Yb:YAG in terms of minimum achievable pulse duration. We carried out single-gain TDL experiments based on Yb:LuO, Yb:LuScO, Yb:ScYLO and Yb:CALGO. The results were published in [98, 59] [99] [75] and [24, 89]. The Yb-doped sesquioxide crystals were grown by the HEM at the Institut für Laser-Physik in the Universität Hamburg [100]. All disks were slightly wedged ($\approx 0.1^\circ$) in order to avoid parasitic reflections that can lead to pulse instabilities in modelocked operation. In our experiments, the disks were pumped with fiber-coupled VBG stabilized diode lasers with narrow spectral linewidth $<0.5$ nm, emitting at 976 nm (Yb-doped sesquioxides) or at 979.5 nm (Yb:CALGO). The corresponding thin-disk modules were arranged for 24 pump passes through the disk, enabling in most cases an efficient absorption of the pump radiation. Throughout the experiments, we used the thin-disk as a folding mirror in a single pass configuration and the SESAM as one end mirror. As discussed in section 3.2, the SESAMs were specifically designed to have ultrafast relaxation dynamics, beneficial for the generation of short pulse
5. Experimental laser results: single broadband thin-disk gain materials

5.1 Thin-disk lasers based on Yb:LuO

The gain material Yb:LuO is a TDL crystal with the potential for power scaling at short pulse durations, as it combines an excellent thermal conductivity with a relatively broad emission bandwidth, compared to Yb:YAG. The first TDL based on this laser crystal delivered a maximum output power of 24 W at a pulse duration of 523 fs [101]. This result was then further improved to a shorter pulse duration of 329 fs with an average output power of 40 W [102]. The power scalability of Yb:LuO TDLs was confirmed in an experiment that delivered 140 W of average power with 738 fs pulses [54].

Here, we explore the performance of Yb:LuO in the short pulse duration regime (< 200 fs). We used a Yb:LuO disk with a thickness of 150 µm and a doping concentration of 3 at. %, enabling a pump light absorption...
5.1. Thin-disk lasers based on Yb:LuO

Yb:LuO thin-disk on diamond

GTI-type mirror

YAG plate

OC

Yb:LuO thin-disk on diamond

-3000 mm

25 W

185 fs

Figure 5.2: Schematic of the Yb:LuO laser cavity adapted for power-scaling experiments. We were able to demonstrate single-transverse-mode operation with diffraction-limited beam quality up to 25 W of average power. The pulse duration at the highest output power was 185 fs.

We obtained stable modelocking up to an average power of 7 W. At this average power, pulses as short as 142 fs were obtained with an optical-to-
Figure 5.3: Top: In the first experiment based on a Yb:LuO thin-disk oscillator delivering 7 W of average power, we measured a non-collinear autocorrelation signal (left) that revealed a pulse duration of 142 fs. The optical spectrum of the pulses (right) had a FWHM of 8.5 nm. Bottom: In the second experiment providing a power of 25 W, we measured an autocorrelation signal (left) corresponding to a pulse duration of 185 fs. The FWHM of the modelocked spectrum (right) was 6.7 nm.

optical efficiency of 15%. The two graphs at the top of figure 5.3 show the measured autocorrelation signal and the corresponding optical spectrum of the pulses. The laser operated at a repetition rate of 64 MHz. The pulses were close to the transform-limit of the spectrum with a time-bandwidth product of 0.34. It is worth noticing that the achieved modelocked spectrum of 8.5 nm was more than 70% of the FWHM of the available gain bandwidth. Further relevant laser parameters can be found in table 5.1.

The short pulse durations allowed us to measure, at the time of the experiment for the first time, the carrier-envelope offset (CEO) beat frequency of a modelocked thin-disk oscillator without any external amplification or pulse compression stage. By using only 65 mW of the output of the laser, we were able to generate a coherent octave-spanning supercontinuum in a highly nonlinear photonic crystal fiber. We detected the CEO beat signal using a standard \( f \)-to-\( 2f \) interferometer, achieving a signal-to-noise ratio (SNR) > 25 dB with a resolution bandwidth of 3 kHz. The CEO frequency
was tunable with the pump current at a rate of 33 kHz/mA. The experiment is explained in more detail in [98].

In a second experiment we scaled both the pump spot diameter on the disk from 1.9 mm to 2.6 mm and the mode diameter on the SESAM from 350 µm to 600 µm. In addition, we increased the output coupling coefficient from 4 % to 6 % to relax intracavity conditions, see table 5.1. As we were aiming for higher average power with short pulse duration, we used a topcoated version of the SESAM that successfully started and stabilized the pulse formation in the previous experiment. The topcoating consisted of a quarter-wave layer pair of SiO₂/Si₃N₄ that optimized the SESAM parameters for the use at higher intracavity energy levels. We estimated the SESAM parameters at the laser wavelength of 1035 nm, a sample temperature of ≈18 °C and a pulse duration of 1 ps to be: saturation fluence of ≈110 µJ/cm², modulation depth of ≈0.9 %, nonsaturable losses of ≈0.4 %, recovery time (τ₁/ₑ) of ≈2 ps. Figure 5.2 shows a schematic of the cavity that contained two GTI-type mirrors that accounted for -2200 fs² second-order dispersion per roundtrip. The 7 mm-thick YAG plate provided in stable modelocked operation the necessary SPM to obtain 185 fs-pulses with an average power of 25 W. The two graphs at the bottom of figure 5.3 show the measured autocorrelation signal and the spectral shape of the pulses with a time-bandwidth product of 0.349. The repetition rate was 66 MHz resulting in a pulse energy of 0.38 µJ and a peak power of 1.8 MW. The observed increase in pulse duration can be explained with several contributing effects. The larger output coupling rate of the laser corresponded to a higher gain per roundtrip. As a consequence, the laser operated at a higher inversion level resulting in a smaller gain bandwidth, see figure 4.5. Furthermore, the minimum achievable pulse duration was increased due to the higher gain of the cavity configuration and the smaller modulation depth of the SESAM (see formula 2.6).

5.2 Thin-disk lasers based on Yb:LuScO and Yb:ScYLO

A successful technique to obtain broader gain bandwidths is the stoichiometric combination of similar laser crystals with shifted spectral emission peaks. The growth of Yb-doped mixed sesquioxide materials allows to
Figure 5.4: Schematic of the TDLs based on Yb-doped LuScO, respectively ScYLO, demonstrating a pulse duration of 96 fs with 5.1 W of average power, respectively 101 fs-pulses with an average power of 4.6 W.

Figure 5.5: Top: The autocorrelation signal (left) of the Yb:LuScO TDL revealed a pulse duration of 96 fs at the highest output power of 5.1 W. We measured the modelocked spectrum (right) to have a FWHM of 12.5 nm. Bottom: The autocorrelation signal (left) of a similar experiment based on Yb:ScYLO providing 4.6 W corresponded to a pulse duration of 101 fs. The FWHM of the optical spectrum of the pulses was 12 nm.
obtain broad FWHM gain bandwidths. They support lower transform-
limited pulse durations as the gain spectrum of the pure sesquioxide
crystal Yb:LuO (see section 4.2.2). For example, the gain spectrum of
a SESAM modelocked TDL based on Yb:LuScO theoretically supports a
time-bandwidth limited minimum pulse duration around 50 fs. This es-
timation does not consider self-broadening effects and is explained in
[75]. Previous TDL experiments based on Yb:LuScO provided an aver-
age power of 7.2 W and 227-fs pulses [103] and a higher output power
of 23 W with 235-fs pulses [58]. The first Yb:ScYLO thin-disk oscillator
demonstrated an average power of 3.9 W with a pulse duration of 236 fs
[104].

We explored new pulse duration limits of Yb-doped LuScO and ScYLO
in the thin-disk configuration. In a first experiment, we used a Yb:LuScO
disk mounted on a 1.4 mm-thick diamond heat sink. The diamond was di-
rectly water-cooled at its back surface to ensure an efficient heat removal.
The Yb-ion concentration of 3 at. % and a thickness of 200 µm allowed to
absorb more than 95 % of the pump radiation. To obtain stable mode-
locked operation, we introduced five dispersive mirrors that accounted
for -2800 fs² GDD per roundtrip, balanced during laser operation by SPM
generated within two YAG plates with thicknesses of 5 mm and 7 mm.
We used an output coupling coefficient of 2.6 % that was the best com-
promise between average power and pulse duration. As in the Yb:LuO
based experiments described in the previous paragraph, we used an an-
tiresonant SESAM with four 7 nm-thick QW layers placed two-by-two in
consecutive antinodes of the standing wave pattern created in the AlAs-
spacer layer grown at 270 °C. By using 1-ps-pulses, we approximated
the SESAM parameters at the laser wavelength of 1039 nm and a sample
temperature of ≈18 °C to be: saturation fluence of ≈90 µJ/cm², modulation
depth of ≈1.2 %, nonsaturable losses of ≈0.4 % and recovery time (τ₁/ₑ) of
≈2 ps. A schematic of the laser cavity that provided a single-transverse-
mode beam profile in cw and modelocked operation is shown in figure
5.4.

We obtained clean single-pulse operation up to 5.1 W of average power
with an optical-to-optical efficiency of 11 %. The autocorrelation signal
shown at the top of figure 5.5 revealed a pulse duration of 96 fs at the
maximum power level. With a FWHM of the modelocked spectrum of 12.5 nm, the pulses were nearly transform-limited with a time-bandwidth product of 0.33. In this experiment we were exploiting more than 60% of the FWHM of the provided gain bandwidth. The laser operated at a repetition rate of 77.5 MHz resulting in a pulse energy of 0.07 µJ. Further output- and intracavity parameters are listed in table 5.1.

In a second experiment we used a Yb:ScYLO disk with a thickness of 250 µm and a Yb-doping level of 3 at.%. We built a TDL with a similar cavity layout than the Yb:LuScO-oscillator of the first experiment, see figure 5.4. As we were targeting short pulse durations we used a low output coupling rate of 1.8%. A total of five dispersive mirrors yielded a GDD of -3520 fs per roundtrip. We used two YAG plates with thickness of 6 nm and 7 nm to provide the required SPM. The thin-disk oscillator operated at a central wavelength of 1036.5 nm. The approximated SESAM parameters of our laser setup are listed in table 5.1. The thin-disk oscillator delivered in fundamental modelocked operation a pulse duration of 101 fs at an output power of 4.6 W. The repetition rate of the resonator demonstrating diffraction limited beam quality was 75 MHz. The autocorrelation signal and the optical spectrum of the pulses are shown in the two graphs at the bottom of figure 5.5. In this experiment, we exploited more than 60% of the FWHM of the gain spectrum.

The broad emission spectra of both gain materials allowed us to obtain similar laser performances at short pulse durations. Yb:ScYLO has a slightly narrower emission bandwidth and no further advantages over Yb:LuScO. In principle, the combination of Yb:YO and Yb:ScO should result in a gain material with larger FWHM emission bandwidth than Yb:LuScO. However, the high-quality growth of such Yb-doped yttrium scandium oxide (YScO₃) crystals is extremely challenging (see section 4.2.2). Hence, further laser experiments based on Yb-doped sesquioxides with mixed crystalline structure will most probably be based on Yb:LuScO.
5.3. Thin-disk lasers based on Yb:CALGO

Figure 5.6: Schematic of the TDLs based on Yb:CALGO. They provided 62 fs-pulses with an average power at 5.1 W and with an improved dispersion management pulses as short as 49 fs with an average power of 2 W.

Figure 5.7: Top: In the first experiment, we measured an autocorrelation signal (left) corresponding to a pulse duration of 62 fs. The spectral shape of the pulses (right) had a FWHM of 22.5 nm. Bottom: A better dispersion management in the second experiment, allowed us to measure an autocorrelation signal (left) resulting in a pulse duration of 49 fs. The modelocked spectrum (right) had a FWHM of 24.7 nm.
5.3 Thin-disk lasers based on Yb:CALGO

Another promising candidate for the generation of short pulses with high average power is Yb:CALGO. For more detailed information about the material properties of this laser crystal, see section 4.2.3. Low-power bulk modelocked lasers demonstrated pulse durations as short as 47 fs with 38 mW of average power [105] and 40 fs pulses with 15 mW of average power [106]. In both cases, the oscillator delivered chirped pulses that required extracavity dispersion management to compress the pulse duration to the transform-limit of the spectrum. Furthermore, remarkable performance was achieved with a high-power bulk modelocked laser that provided 12.5 W of average power with 94 fs-pulses [107]. The suitability of Yb:CALGO for ultrafast high-power operation in the thin-disk geometry was also confirmed in recent results that reached an average power of 28 W with a pulse duration of 300 fs [108]. The first Yb:CALGO TDLs that provided sub-200 fs pulse durations demonstrated 135 fs-pulses and an average power of 1.3 W [108].

In our experiments, we continued to exploit the potential of this gain material. We used a commercially available Yb:CALGO disk with a thickness of \( \approx 220 \mu m \) and a dopant concentration of 3.1 at.\%, that resulted in a pump light absorption >90\%. The crystal was c-cut in order to obtain isotropic mechanical and thermal properties in the disk plane. In addition, it allowed us to operate the laser in the optical \( \sigma \) polarization, which exhibits a broader and smoother gain cross-section than the \( \pi \) polarization, an advantage for short pulse generation. The disk was vacuum soldered by using indium at 180°C onto a copper-tungsten heat sink (20\% of copper). This material has a CTE at room temperature of \( 8.3 \cdot 10^{-6} K^{-1} \), which is close to the CTE of Yb:CALGO for the a axis (\( 10.1 \cdot 10^{-6} K^{-1} \) [23]). Nevertheless, the mounted disk showed strong astigmatism due to the not yet optimized mounting technique. We measured a smallest ROC of -4.8 m along one direction, and a highest ROC of infinity perpendicular to this direction. The pump spot diameter on the disk was 2.1 mm. We first tested the disk in cw operation in a linear multimode cavity. It consisted of the disk as the one end mirror and a curved mirror (ROC = -100 mm) with an output coupling rate of 0.9\% as the other end mirror. In this configura-
tion, we obtained up to 45 W of output power with an optical-to-optical efficiency of 45 % and a slope efficiency of 54 %.

Currently, the highest slope efficiency of an Yb:CALGO TDL operating in cw multimode is 70 % [79]. This results was based on a crystal with a high doping concentration of 5.4 at. % and a thickness of 200 µm. In our experiments, we used an Yb:CALGO crystal with a similar thickness but lower doping level. Thus, a larger amount of passes would have allowed to optimize the pump light absorption [109]. Furthermore, investigations made on this relatively new gain material reported on scattering centers that might have influenced the laser performance [91].

The schematic of the cavity supporting single-transverse-mode operation with an $M^2 < 1.1$ is shown in figure 5.6. With an output coupler transmission of 3 %, we obtained in cw operation a maximum average power of 14 W with an optical-to-optical efficiency of 19 %. The output power was not pushed further to avoid damage of our crystal at pump intensities $> 3 \, \text{kW/cm}^2$. In order to obtain stable modelocking, we inserted a 5.3 mm-thick YAG plate into the cavity and replaced three HR-mirrors with GTI-type mirrors. In this configuration, we estimated to have a total GDD per roundtrip of -300 fs$^2$, taking into account the positive second-order dispersion of the Brewster plate. We used a SESAM with a single QW absorber layer and no topcoating. We characterized its nonlinear reflectivity parameters directly with our Yb:CALGO modelocked TDL, which allowed us to measure the SESAM parameters at the center wavelength of our laser setup (1051 nm) and also to measure the rollover in reflectivity with short pulse durations (85 fs). The latter was important as the induced absorption at high fluences is mostly due to TPA and much more pronounced in the case of short pulses, see formula 2.17. In order to avoid laser operation in the rollover it is therefore beneficial to know the strength of induced absorption for the given pulse duration. At a temperature of approximately 18°C, we measured a saturation fluence of $\approx 10 \, \mu\text{J/cm}^2$, a modulation depth of $\approx 1.4 \, \%$, nonsaturable losses of $\approx 0.5 \, \%$ and an induced absorption coefficient $F_2$ of $\approx 275 \, \text{mJ/cm}^2$.

With an output coupling rate of 2.5 %, we were able to achieve fundamentally modelocked pulses with a duration of 62 fs and an average
power of 5.1 W, see figure 5.7. The repetition rate of our oscillator was 65 MHz, corresponding to a pulse energy of 0.08 µJ and a peak power of 1.1 MW. The generated nonlinear phase shift at our maximum intracavity peak power was 600 mrad. The optical-to-optical efficiency was moderate (≈7 %) partly due to the astigmatism and the limited optical quality of the mounted laser crystal. The optical spectrum of the pulses had a FWHM of 23 nm. At an estimated inversion level of approximately 10 %, this corresponded to 66 % of the FWHM of the gain bandwidth. The pulses had a slight chirp most likely originating from uncompensated higher-order dispersion of our laser cavity, resulting in a time-bandwidth product of 0.38.

In order to overcome this chirp, we exchanged the GTI-type mirrors with samples that showed flatter GDD profiles around the center wavelength. We kept the amount of GDD per roundtrip that we had in the previous experiment (-300 fs$^2$). In addition, we reduced the output coupling coefficient to 1.6 %. In this second experiment, we obtained an average output power of 2 W and a pulse duration of 49 fs at a repetition rate of 65 MHz.

This result showed that from an engineering point of view, special attention has to be paid to the flatness and bandwidth of the used GTI-type mirrors. The two graphs at the bottom of figure 5.7 show the corresponding autocorrelation signal and mode locked spectrum. We measured a beam quality of $M^2$<1.1. The improved dispersion management balanced a nonlinear phase shift of 490 mrad for the peak of the pulse and provided nearly transform-limited pulses with a time-bandwidth product of 0.325.

The demonstrated record-short pulse durations enabled us to measure and stabilize the CEO frequency of the Yb:CALGO TDL. We set the laser parameters of the first cavity layout to 2 W of average power and a pulse duration of 90 fs. By launching only 2 % of the output power into a highly nonlinear photonic crystal fiber, we generated a coherent, octave spanning supercontinuum. By using a standard $f$-to-$2f$ interferometer for CEO detection, we measured the CEO beat frequency with 33 dB of SNR in a 100 kHz resolution bandwidth. We achieved a tight lock of the CEO frequency at 26.2 MHz by active feedback to the pump laser. The residual in-loop integrated phase noise was 120 mrad within a range from 1 Hz up to 1 MHz. Our results showed that it was possible to reliable lock the CEO frequency of a TDL despite the strongly spatially multimode pump-
ing scheme of thin-disk oscillators. More detailed information about the experiment are presented in [110].

5.4 Summary

In summary, we showed in this chapter different TDL experiments based on broadband gain materials that reached pulse durations well below 200 fs. They were still restricted to moderate pulse energies (< 1 µJ) and average output powers (< 10 W) due to the challenging requirements for the generation of short pulses. The broad gain bandwidth necessary to support short pulse durations comes, for example, usually at the expense of reduced thermo-mechanical properties of the laser crystal. In the next chapter, we present experiments that were based on the dual-gain concept, which is a promising way to overcome this limitation.
### 5. Experimental Laser Results: Single Broadband Thin-Disk Gain Materials

<table>
<thead>
<tr>
<th>Yb-doped</th>
<th>LuO</th>
<th>LuO</th>
<th>LuScO</th>
<th>ScYLO</th>
<th>CALGO</th>
<th>CALGO</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>output parameters</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>pulse duration ($\tau_p$)</td>
<td>142 fs</td>
<td>185 fs</td>
<td>96 fs</td>
<td>101 fs</td>
<td>62 fs</td>
<td>49 fs</td>
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<tr>
<td>average power</td>
<td>7 W</td>
<td>25 W</td>
<td>6 W</td>
<td>4.6 W</td>
<td>5 W</td>
<td>2 W</td>
</tr>
<tr>
<td>energy</td>
<td>0.11 µJ</td>
<td>0.38 µJ</td>
<td>0.07 µJ</td>
<td>0.06 µJ</td>
<td>0.08 µJ</td>
<td>0.03 µJ</td>
</tr>
<tr>
<td>repetition rate</td>
<td>64 MHz</td>
<td>66 MHz</td>
<td>77.5 MHz</td>
<td>75 MHz</td>
<td>65 MHz</td>
<td>65 MHz</td>
</tr>
<tr>
<td>peak power</td>
<td>0.7 MW</td>
<td>1.8 MW</td>
<td>1.6 MW</td>
<td>0.5 MW</td>
<td>1.1 MW</td>
<td>0.6 MW</td>
</tr>
<tr>
<td>center wavelength ($\lambda_c$)</td>
<td>1035 nm</td>
<td>1035 nm</td>
<td>1039 nm</td>
<td>1036.5 nm</td>
<td>1051 nm</td>
<td>1051 nm</td>
</tr>
<tr>
<td>FWHM optical spectrum</td>
<td>8.5 nm</td>
<td>6.7 nm</td>
<td>12.5 nm</td>
<td>12 nm</td>
<td>22.5 nm</td>
<td>24.7 nm</td>
</tr>
<tr>
<td>Fraction of emission (gain) spectrum</td>
<td>65%</td>
<td>52%</td>
<td>57%</td>
<td>68%</td>
<td>28%</td>
<td>31%</td>
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<tr>
<td>optical-to-optical efficiency</td>
<td>15%</td>
<td>18.5%</td>
<td>11%</td>
<td>-</td>
<td>7%</td>
<td>-</td>
</tr>
<tr>
<td>factor above transform limit</td>
<td>1.08</td>
<td>1.11</td>
<td>1.05</td>
<td>1.07</td>
<td>1.21</td>
<td>1.03</td>
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<tr>
<td>output coupling rate</td>
<td>4%</td>
<td>6%</td>
<td>2.6%</td>
<td>1.8%</td>
<td>2.5%</td>
<td>1.6%</td>
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<tr>
<td>average power</td>
<td>175 W</td>
<td>416 W</td>
<td>196 W</td>
<td>256 W</td>
<td>204 W</td>
<td>125 W</td>
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<tr>
<td>energy</td>
<td>2.7 µJ</td>
<td>6.3 µJ</td>
<td>2.5 µJ</td>
<td>3.4 µJ</td>
<td>3.1 µJ</td>
<td>1.9 µJ</td>
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<tr>
<td>peak power</td>
<td>16.9 MW</td>
<td>30.0 MW</td>
<td>23.2 MW</td>
<td>29.7 MW</td>
<td>44.5 MW</td>
<td>34.5 MW</td>
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<td>GDD per roundtrip</td>
<td>-1970 fs</td>
<td>-1150 fs</td>
<td>-995 fs</td>
<td>-1560 fs</td>
<td>-300 fs</td>
<td>-300 fs</td>
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<tr>
<td>nonlinear phase shift for the peak of the pulse</td>
<td>306 mrad</td>
<td>104 mrad</td>
<td>339 mrad</td>
<td>475 mrad</td>
<td>245 mrad</td>
<td>391 mrad</td>
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<td><strong>SESAM parameters</strong></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>approximated to $\lambda_c$</td>
<td>yes</td>
<td>yes</td>
<td>yes</td>
<td>yes</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>measured with $\tau_p$</td>
<td>no</td>
<td>no</td>
<td>no</td>
<td>no</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>$F_{satA}$</td>
<td>36 $\frac{µJ}{cm^2}$</td>
<td>110 $\frac{µJ}{cm^2}$</td>
<td>90 $\frac{µJ}{cm^2}$</td>
<td>36 $\frac{µJ}{cm^2}$</td>
<td>10 $\frac{µJ}{cm^2}$</td>
<td>10 $\frac{µJ}{cm^2}$</td>
</tr>
<tr>
<td>$\Delta R$</td>
<td>3.3%</td>
<td>0.9%</td>
<td>1.2%</td>
<td>3.3%</td>
<td>1.4%</td>
<td>1.4%</td>
</tr>
<tr>
<td>$\Delta R_{ref}$</td>
<td>0.7%</td>
<td>0.4%</td>
<td>0.4%</td>
<td>0.7%</td>
<td>0.5%</td>
<td>0.5%</td>
</tr>
<tr>
<td>$\tau_{1/e}$</td>
<td>2 ps</td>
<td>2 ps</td>
<td>2 ps</td>
<td>2 ps</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>mode radius</td>
<td>350 µm</td>
<td>600 µm</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
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<tr>
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<td>[59]</td>
<td>[99]</td>
<td>[75]</td>
<td>[24]</td>
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Table 5.1: Laser parameters obtained from experimental laser results based on the broadband gain materials Yb:LuO, Yb:LuScO, Yb:ScYLO and Yb:CALGO and the corresponding SESAM parameters.
Chapter 6

Experimental laser results: first dual-gain thin-disk oscillator

We presented in the previous chapter experimental laser results based on a set of broadband gain materials that pave the way towards the ultimate goal to generate both high average power and sub-100 fs pulses directly from the oscillator. As we discussed in section 4.2.2, the heat transport in Yb-doped rare-earth crystals is mainly based on phonon propagation. As a broadband emission spectrum comes at the expense of a disordered lattice structure, mixed Yb-doped host materials have often a low thermal conductivity (see table 4.2), which hinders average power scaling in the thin-disk geometry. An interesting alternative approach that extends laser performances to shorter pulse durations is to combine two spatially separated gain media with displaced emission spectra within one resonator. In modelocked operation, the two independent gain materials support a single pulse with a broad gain spectrum. Therefore, this combination allows for working with gain materials that exhibit moderate emission bandwidths but high thermal conductivities while reaching out for shorter pulse durations. To date this concept has been successfully implemented in oscillator and amplifier systems. A laser oscillator based on bulk ceramic Yb:ScO and Yb:YO gain media delivered 53 fs pulses with an output power of 1 W [111] and the modelocking of a dual-gain Nd:glass laser yielded 38-fs pulses at 40 mW of output power [112]. Furthermore, the combined gain spectra of a regenerative thin disk amplifier generated sub-
Experimental laser results: first dual-gain thin-disk oscillator

6. Experimental laser results: first dual-gain thin-disk oscillator

fs pulses with an energy of 500 µJ [113]. Here, we present the successful implementation of this method to the modelocked TDL technology that has been published in [25]. Our first proof-of-principle experiments are based on a dual-gain SESAM-modelocked thin-disk oscillator that combines the gain of the two Yb-doped sesquioxides LuO and ScO with high thermal conductivity in the same resonator. These two gain crystals have moderate emission bandwidths of 13 nm and 12 nm, see table 4.1. We have shown in the previous chapter an experiment based on Yb:LuO alone that provided 7 W of average power with pulses as short as 142 fs and a spectral bandwidth at FWHM of 8.5 nm, exploiting therefore more than 60% of the emission bandwidth of the laser crystal [98]. To the best of our knowledge no ultrafast TDL based on a single Yb:ScO crystal has been demonstrated so far, but a bulk SESAM-modelocked Yb:ScO laser delivered 230-fs pulses [114]. However, we do not expect this gain material to outperform Yb:LuO in a single resonator configuration in terms of shortest pulse generation due to the slightly narrower emission bandwidth of Yb:ScO. In the following we present the experimental setup that allowed us to obtain shorter pulse durations than previously achieved with the single-gain Yb:LuO-TDL confirming the potential of this dual-gain approach.

6.1 Dual-gain thin-disk laser setup

The layout of the SESAM-modelocked TDL used for the dual-gain mode-locking experiments is shown in figure 6.1. Both sesquioxide crystals used in the modelocked oscillator were grown by the HEM at the Institut für Laser-Physik in the Universität Hamburg [100]. They have a wedge of 0.1° in order to avoid parasitic reflections that could result in modelocking instabilities and were indium-soldered on copper heat sinks. The Yb:LuO disk with a doping concentration of 2 at. % has a thickness of 250 µm and a diameter of 5.2 mm. The 2.4 at. % doped Yb:ScO disk is 300 µm in thickness and 6 mm in diameter. Two separate fiber-coupled VBG stabilized diodes, emitting at the maximum absorption wavelength of ≈ 976 nm of the crystals, were used to optically pump the gain materials. At the pump wavelength the absorption cross-section for Yb:LuO is $3.1 \cdot 10^{-20}$ cm$^2$ and $4.4 \cdot 10^{-20}$ cm$^2$ for Yb:ScO, see table 4.1. The two multimode pump-fibers
6.1. Dual-gain thin-disk laser setup

Figure 6.1: Cavity design of the dual-gain resonator supporting fundamental transverse mode operation up to 10 W cw. The Yb:LuO- and the Yb:ScO disk were separated by a distance of ≈ 1.15 m. The SESAM and the output coupler were the two end mirrors of the linear cavity. We measured for all experiments an $M^2 < 1.2$.

have an NA of 0.22 and a core diameter of 600 µm. We used two pump modules both arranged for 24 pump passes on the corresponding disk.

Single-transverse-mode operation is crucial for modelocking experiments and in such a dual-gain cavity two unknown thermal lensing elements can make the fundamental resonator design very challenging. Therefore, we carefully characterized the quality of the contacted disks by inspecting them with a microscope and by measuring the deformation of the disks under optical pumping with a Michelson interferometer. Figure 6.2 shows the microscopic pictures and the surface deviation measurements of the two disks. The Yb:LuO disk exhibited a good surface quality without any visible grain boundaries. We measured the ROC of the cold disk and the deformation of the disk under pumping levels used in the experiments. The cold disk shows a spherical curvature with an average ROC of -2.5 m. When the disk was water-cooled at a water pressure of about 1 bar, we observed a small change of the average ROC to -2.6 m. During cw single-mode operation, with a pump power of 44 W on a pump spot diameter of 2.3 mm, no additional changes in the average ROC were observed. The cold Yb:ScO disk showed a slight astigmatism and several grain bound-
6. **Experimental laser results: first dual-gain thin-disk oscillator**

![Yb:LuO - disk](image1.png)

*Figure 6.2: Top: The high-quality Yb:LuO crystal did not show any visible grain boundaries under the microscope (left). An interferometric measurement, performed at 1 bar of water pressure at the backside of the disk, revealed a perfect spherical surface of the Yb:LuO disk (right). Bottom: The microscopic picture of the Yb:ScO crystal showed several visible grain boundaries (left). The interferometric measurement, performed again at 1 bar of water pressure at the backside of the disk, revealed slight astigmatism of the Yb:ScO disk. The pump spot on the corresponding disk is marked with a dashed black circle.*

aries. Therefore, we confined the diameter of the pump spot to 1.9 mm and centered the spot on the disk in order to work on an area that shows only small astigmatism. In pumped operation without any lasing at 1 bar of water pressure and 6 W of pump power the average ROC was measured to be -12 m. The quality of the disk clearly limited the power-scalability of our dual-gain oscillator. However, it was the only disk available at the time of the experiment. We first tested the disks separately in cw operation. In a linear resonator consisting of the Yb:LuO crystal and a 100 mm ROC mirror with 1.2% transmission, we were able to achieve up to 50 W in multimode operation with an optical-to-optical efficiency higher than 60% and a slope efficiency of 72%. With the Yb:ScO crystal we obtained optical-to-optical efficiencies higher than 50% in cw multimode operation.
The dual-gain linear resonator with diffraction-limited beam quality had a length of ≈3.6 m and the two disks were separated by a distance of ≈1.2 m. The cavity was designed to operate at the center of the stability zone and exhibited a small sensitivity to the thermal lenses of both disks but with a stronger consideration of the lower quality of the Yb:ScO disk. This oscillator then supported single fundamental-transverse mode operation up to 10 W in cw. We inserted a wedged undoped YAG plate at Brewster’s angle into the cavity. This ensures a linearly polarized output and provides the required SPM for soliton modelocking. In order to produce enough SPM we placed the YAG plate close to the focus with a spot radius of ≈150 µm between the two curved mirrors with a ROC of 500 mm. The SESAM inserted as one end mirror was the only loss modulator inside the cavity and started and stabilized the soliton modelocking.
6. Experimental laser results: first dual-gain thin-disk oscillator

All experiments were done with an antiresonant SESAM without top-coating, designed and grown in accordance with our guidelines on SESAMs with short recovery times presented in section 3.2. The absorber section consists of four InGaAs QWs with a thickness of 7 nm, placed two-by-two in consecutive antinodes of the standing wave pattern inside the spacer layer. The absorber layers were grown at a low temperature of 270°C resulting in a fast recovery time of $\tau_{1/e} \approx 3$ ps. As explained in section 2.3.3, increasing the temperature of a SESAM decreases its band gap energy, which leads to a red shift of the corresponding absorption properties. For the SESAM used in our experiments we measured this red shift to be 0.38 nm / K, see figure 2.8. During modelocking, we controlled the temperature of the SESAM using a Peltier element. This additional degree of freedom allowed us to find the best SESAM parameters for modelocked operation of the dual-gain configuration with short pulse durations. The temperature-dependent SESAM parameters shown in figure 6.3 were measured at a center wavelength of 1030 nm with 1-ps pulses over a temperature range of -20°C up to 60°C. It is clearly visible that with increasing temperature the modulation depth $\Delta R$ was increased whereas the saturation fluence $F_{\text{sat,A}}$ was initially reduced and then remained approximately constant, as expected from the red shift of the SESAM absorption edge. The nonsaturable losses $\Delta R_{\text{ns}}$ remained approximately constant and became much smaller than the modulation depth at higher temperatures. In addition we did not observe any change of the recovery time.

During our modelocking experiments we confirmed that a small modulation depth of the SESAM at room temperature (20°C) is not strong enough to fully lock the two gain materials. We observed two cw instabilities at 1034 nm and 1041 nm, shown in the graph at the top of figure 6.4. At an increased temperature of 30°C, the modulation depth at the center gain wavelength of Yb:LuO (1034 nm) increased, but remained low for the peak-gain wavelength of Yb:ScO (1040 nm). Therefore, cw breakthrough originating from the gain of the Yb:ScO disk was still observed, see the graph in the middle of figure 6.4. The SESAM parameters at a temperature of 40°C allowed us to achieve stable and smooth $\text{sech}^2$-shaped optical spectra with broad bandwidths that supported short pulse durations. The measurement of such a modelocked spectrum is shown in the graph at

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Figure 6.4: Modelocked pulse spectra for three different SESAM temperatures. At 20°C and 30°C the two separate gain materials with maximum emission cross-section around a wavelength of 1034 nm (Yb:LuO), respectively 1041 nm (Yb:ScO), were not fully locked because the SESAM did not provide a large enough modulation depth. We therefore observed cw instabilities at 1034 nm and 1041 nm (top and middle). In order to obtain a fully modelocked spectrum the modulation strength of the SESAM had to be increased with the higher temperature of 40°C (bottom).
Figure 6.5: Top: We measured in the first experiment a non-collinear autocorrelation signal (left) that revealed a pulse duration of 103 fs. The corresponding optical spectrum of the pulses (right) had a FWHM of 12.8 nm. It was centered at 1037.7 nm in between the two emission cross-section maxima of Yb:LuO (blue) and Yb:ScO (yellow). This clearly demonstrated that both gain materials were contributing to the spectral shape of the pulses. Bottom: In the second experiment, we measured an autocorrelation signal (left) corresponding to a pulse duration of 124 fs. The modelocked spectrum (right) was centered at 1038.5 nm, again in between the two emission peaks of Yb:LuO and Yb:ScO.

the bottom of figure 6.4. At 40°C, the SESAM parameters at the center wavelength of 1038 nm were approximately as follows: \( \Delta R \approx 2.9\% \), \( F_{\text{sat,A}} \approx 39 \mu \text{J/cm} \) and \( \Delta R_{\text{ns}} \approx 0.8 \% \).

In a first experiment we worked with a large amount of SPM in order to find the shortest pulse durations of the dual-gain cavity configuration. We introduced a 6 mm thick YAG plate close to the cavity focus and balanced the generated SPM with a total amount of negative GDD of -1170 fs\(^2\) per roundtrip, taking into account the positive dispersion of the Brewster plate. A low output coupling rate of 4.4% yielded a spectrum centered at a wavelength of 1037.7 nm with a FWHM of 12.8 nm and a pulse duration of 103 fs corresponding to a time-bandwidth product of 0.367. The two graphs at the top of figure 6.5 show the modelocked pulse spectrum...
6.1. Dual-gain thin-disk laser setup

Figure 6.6: Top left: We used a microwave spectrum analyzer set to a RBW of 1 kHz to measure the pulse repetition rate. We did not observe any side peaks around the repetition frequency of 41.7 MHz. Top right: A scan with a RBW of 300 kHz over a larger frequency range did not reveal any unusual features in the pulse train. Bottom: By using a sampling oscilloscope we measured a temporal pulse separation of 24 ns, corresponding to the roundtrip time of our fundamentally modelocked oscillator.

Together with the normalized emission spectra of Yb:LuO and Yb:ScO and the corresponding autocorrelation signal. The output power of 1.4 W, repetition rate of 41.7 MHz and a spot radius of 580 µm on the SESAM resulted in a saturation parameter of \( S \approx 2 \). We did not observe any side peaks around the fundamental pulse repetition frequency and measured a constant intensity of the higher harmonics of the repetition rate, see figure 6.6. The nonlinear phase shift per roundtrip was \( \gamma_{\text{SPM,cav}} \approx 52 \text{ mrad/MW} \).

To confirm fundamental modelocked operation, we increased the autocorrelator time delay up to 70 ps and used a sampling oscilloscope with a fast photodiode (45 GHz). A good beam quality was confirmed by measuring an \( M^2 < 1.2 \).
In a second experiment, we increased the average output power and replaced the Brewster plate by a thinner 4 mm YAG plate. We used an output coupling rate of 7.8% and were able to achieve modelocked operation at 8.6 W of output power and a pulse duration of 124 fs at a center wavelength of 1038.5 nm. The modelocked pulse spectrum together with the normalized emission spectra of Yb:LuO and Yb:ScO and the corresponding autocorrelation signal are depicted at the bottom of figure 6.6. We obtained this result with a pump power of 82 W on the Yb:LuO crystal and a pump power of 55 W on the Yb:ScO crystal. The nonlinear phase shift of $\gamma_{\text{SPM, cav}} \approx 25 \text{ mrad/MW}$ was balanced by a total negative GDD of -2320 fs$^2$ per roundtrip. The saturation parameter of the SESAM was $S \approx 7$. The pulse spectrum had a FWHM of 10.1 nm which resulted in nearly transform-limited pulses with a TBP of 0.348 that is $\approx 1.1$ -times the time-bandwidth limit of 0.315 of a $sech^2$-shaped pulse. We confirmed again single-pulse modelocking and measured a good beam quality with an $M^2 < 1.2$.

We were able to demonstrate, to our knowledge for the first time, a modelocked TDL based on two different gain materials in a resonator. This dual-gain approach combines the shifted gain spectra of doped host materials with moderate emission bandwidth but excellent thermal conductivity to an overall broad gain spectrum in order to obtain pulse durations much shorter than supported by a single gain-crystal. In addition, we took advantage of the temperature-dependence of the bandgap energy of a QW SESAM that gave us the flexibility to tweak absorption parameters during laser operation, compensating for small destabilizing effects. We showed in first proof-of-principle experiments the successful modelocking of a broad spectrum supported by the two separate sesquioxide gain materials Yb:LuO (centered at 1034 nm) and Yb:ScO (centered at 1041 nm). We demonstrated pulses as short as 103 fs with 1.4 W of average power and were able to achieve 124-fs pulses at a higher output power of 8.6 W. The obtained pulse durations were shorter than previously demonstrated using the Yb:LuO gain material alone. The characterization measurements of the Yb:ScO disk suggest that the lower surface quality and thermal lensing of the Yb:ScO disk were mainly limiting the achievable output power. Hence, the current progress in the growth of sesquioxide
materials combined with contacting on diamond heat sinks would result in significantly higher average power of ultrafast pulses with diffraction-limited beam quality. By combining other gain materials with a larger shift in emission cross-section, as for instance Yb:YO and Yb:ScO with a displacement of their emission peaks of 10 nm, the generation of even shorter pulse durations comes within reach. We therefore believe, that the ongoing progress in the growth of crystals with larger shift in their gain peaks together with accordingly adapted SESAM parameters will allows us to exploit the full potential of the dual-gain concept and support higher power levels of modelocked TDLs operating in the sub-100 fs pulse duration regime.
Chapter 7

Conclusion and outlook

The aim of this thesis was to explore new pulse duration limits of modelocked TDLs. We therefore developed SESAM modelocked TDLs based on novel broadband gain materials that provided soliton pulses in the sub-200 fs regime.

The requirements on the SESAM become more stringent with decreasing pulse duration. In particular, induced absorption at high pulse fluences due to TPA is significantly increased for short pulse durations. We therefore investigated the influence of dielectric topcoating layers on optical SESAM parameters. We found that this processing method allows to remarkably increase saturation- and damage-threshold fluence of low-temperature grown QW SESAMs with intermediate response times. For the development of TDLs that produce shortest pulses durations, we used SESAMs with short recovery times and higher modulation depths. We extended our study on dielectric topcoating structures to SESAMs with ultrafast recovery dynamics. In general, a short relaxation time comes at the expense of non-negligible nonsaturable losses. However, we measured for all samples nonsaturable losses that were much lower than the corresponding modulation depth. Our investigation allowed us to successfully grow and process SESAMs with short response times, high saturation fluences and, as a result of reduced induced absorption, damage threshold fluences deep in the rollover regime. SESAMs with such parameters are ideally suited for the use in TDL experiments targeting the generation of short pulses.
These ultrafast SESAMs were therefore an essential point to obtain the results we presented in this thesis. They allowed us to explore the potential of novel thin-disk materials with broad emission spectra in TDL experiments aiming for short pulse durations. We were able to scale the power of a thin-disk oscillator based on Yb:LuO from 7 W to 25 W with a minor increase in pulse duration from 142 fs to 185 fs. Yb-doped LuScO and ScYLO TDLs allowed us to achieve pulse durations of 96 fs and 101 fs with average powers around 5 W. By using Yb:CALGO as thin-disk gain material, we demonstrated 62 fs-pulses with 5.1 W of average power. In a second step, we were able to achieve sub-50 fs pulses with 2 W of average power. The broad gain bandwidth of the laser crystals we used in our experiments was the result of a disordered lattice structure.

The dual-gain concept is a possible alternative to obtain a large gain bandwidth. This approach incorporates two gain materials with similar emission cross-sections but shifted emission peaks in a single resonator. In modelocked operation the two emission spectra combine to a single broad gain spectrum that supports the generation of short pulses. We therefore implemented, to our knowledge for the first time, the dual-gain concept into the thin-disk geometry. In our experiment we inserted two pure sesquioxide gain materials (Yb:LuO and Yb:ScO) with high thermal conductivities in a single TDL resonator. The emission peaks were shifted by 7 nm and combined in modelocked operation to a broad gain bandwidth that supported 103 fs-pulses with an average power of 1.4 W. By using a higher output coupling rate and an accordingly adapted cavity design we successfully scaled the power to 8.6 W with a pulse duration of 124 fs. During the experiments we took advantage of the temperature-dependence of the bandgap energy of our QW SESAM. This effect gave us the flexibility to tweak absorption parameters during laser operation in order to obtain a smooth \( \text{sech}^2 \)-shaped gain profile.

In our proof-of-principle experiments we were still restricted to moderate pulse energies (< 1 µJ) and average output powers (< 10 W) due to the challenging requirements for the generation of short pulses on gain material, laser setup and SESAM design.
The quality of the available broadband materials is not yet comparable with that of the standard thin-disk crystal Yb:YAG. However, it is expected that the quality of these novel materials as well as the corresponding contacting techniques will keep improving. In this aspect, the development of a dual-gain TDL based on Yb:YAG and Yb:LuO is of high interest as both gain materials already showed excellent performance in high-power TDLs. Moreover, a quantitative understanding of the soliton pulse formation process in a dual-gain TDL is an indispensable tool for future experiments. We are confident that a numerical model will give us information on how to fully exploit the potential of the two emission spectra with shifted maxima. In addition, the model will provide details on how to optimize the balance between SPM and GDD in this specific case. Furthermore, it will allow to find basic guidelines for SESAMs specifically designed for the use in dual-gain ultrafast TDLs.

As the spectral shape of the gain is flatter at low inversion levels, TDL experiments aiming for short pulse durations typically use low output coupling rates of only few percent. However, a low output coupling coefficient results in a high intracavity peak power already at moderate output power and pulse energy levels. As nonlinearities scale with peak power, reaching high output performance in a stable modelocked configuration becomes increasingly challenging. In the case of oscillators providing shortest pulse durations this effect is even more critical as the peak power is inversely proportional to the pulse duration. We therefore believe that future high-power TDL experiments with pulse durations lower than 200 fs will require the same measures to avoid excessive nonlinearities as are necessary in current leading-edge TDLs. This implies the operation in a vacuum environment.

Another core element is the development of a new SESAM generation with improved heat removal capabilities and flatness. Promising candidates in this aspect are substrate-removed SESAMs contacted on heat sinks with high thermal conductivity, as for instance SiC or diamond. The combination of an excellent thermal management with superior flatness will be beneficial for the use in future high-power SESAM modelocked TDLs with large mode areas on gain material and absorber.
7. Conclusion and outlook

We believe that considering these key elements will allow to extend current output power and pulse energy levels of leading-edge ultrafast TDLs to shorter pulse durations. This is a crucial milestone, which remains to be solved on the way towards the ultimate goal of HHG at MHz-repetition rates directly driven by a table-top ultrafast oscillator.
Literature


Literature


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