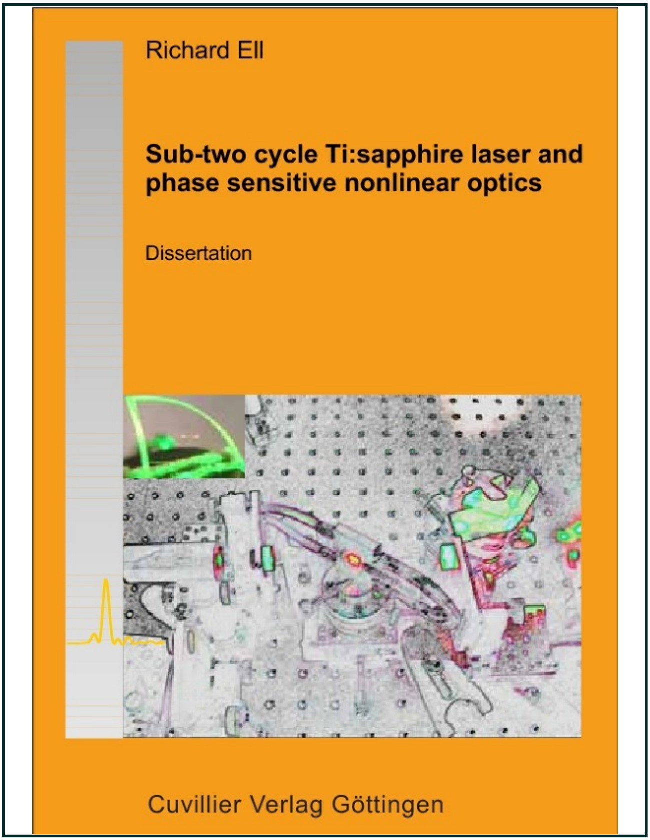
Richard Ell (Autor)

Sub-Two Cycle TI: Sapphire Laser and Phase Sensitive Nonlinear Optics



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Chapter 1

Motivation

Working with ultrashort laser pulses is a very exciting and fascinating field of research in modern optics. From the physicist's point of view, this enthusiasm is caused by the fact that the light pulses generated in our laboratories are actually among the *shortest events controllable by human kind!* It is a challenge to consider and master all physical aspects related to these extremely short light pulses and once they are generated, their complete characterization is another demanding task. From a more technological point of view, research in the field of ultrashort pulse generation is driven by the impressive impact of optical technologies (”Photonics”) and, in particular, lasers onto almost every part of economic and social live today. The laser pulses presented in this work have a time duration of 5 fs (1 fs = 10^{-15} s). Such orders of magnitude are very difficult to imagine for everyone. An amount of 1 million Euros (10^6 Euros), for example, is already beyond the typical order of magnitude people are used to deal with. Hopefully, the reader will keep on reading this dissertation. Interestingly, only ten minutes will have passed after reading the introduction and the outlook of this thesis – this is approximately 10^{-15} of the age of our universe! An ultra–violet laser, e.g. a frequency tripled titanium sapphire laser, with an average power of 1 mW at a wavelength of 200 nm emits 10^{15} photons per second. Considering the speed of light in free space ($\approx 3 \cdot 10^8$ m/s), a 5 fs laser pulse is just a "small slice of photons" with a thickness of less than 3 μm.

The mere generation of ultrashort pulses does not suffice without the proper means of pulse characterization. Apparently, the question is how scientists are able to determine the duration of the incredibly short pulses as discussed above? Unfortunately it is not possible to utilize sampling methods as often performed in signal analysis. The obvious reason is the lack of even shorter reference signals to sample the light pulses. A sophisticated combination of optical self–referencing and standard radiofrequency electronics overcomes this fundamental problem. The resolution achievable with present setups is below 1 fs, whereas the resolution of the involved electronics does not need to be better than 10 ns (corresponding to a bandwidth of about 100 MHz). This is equivalent to the use of a yardstick for a length measurement and obtaining a resolution in the μm range (a rather sophisticated vernier scale would be needed).

Parameters characterizing laser light are very manifold. A laser beam is described by its wavelength, the polarization of the electric field, the divergence and radius of the beam at some reference plane, the optical average power, and finally the amplitude– and phase–noise characteristics. The possible adjustment of a subset or all of these parameters to some specific application determines the lasers versatility. Intentionally, one of the most significant parameters was omitted in the previous list – the temporal regime in which the laser emits its power. Laser sources can be operated either continuously, with constant power over time, or in a pulsed regime. In the latter case, impulses of light are emitted in a periodic manner with ideally no emission in between successive pulses. Consequently, all the light coupled out of the laser is condensed into periodically emitted bunches of photons. This concentration of energy leads to impressive peak powers, which are several orders of magnitude greater than the average power. For better illustration, let us consider the titanium sapphire (TISA) laser, which is subject of this thesis: with an average power of about 100 mW, a pulse period of 10 ns, and a pulse duration of 5 fs, we obtain a peak power of approximately 200 kW – the power of a 270 PS–engine of a sports car. When amplified even further, kHz–repetition rate laser systems reach into the GW regime. And finally, high–power, special low repetition rate lasers generate impulses with TW or even PW peak powers, surpassing the power of nuclear power plants for a very short time [Per99, Kal01]. Focusing the laser beam down to beam waist radii of a few μm leads to huge intensities in the order of 10^{21} W/cm^2. As a result, the electric field strength surpasses the inner atomic Coulomb fields [Bra00]. Hence, the pulsed–laser technology enables the study of new extreme nonlinear regimes never reached before in history of science [Kra98, Bra00, Pau01, Wan03].

Apart from achievable peak powers, intensities, and electric field strengths, an extra motivation of ultrashort pulse generation lies in the potentials of physical, biological, and chemical phenomena as suggested by Table 1.1. It shows various timescales of relevance in physics, biology, and chemistry. One should keep in mind that laser pulses in the order of these timescales are available nowadays. According to the demand, these pulses are furthermore available at different wavelengths, peak powers, and repetition rates. Only ultrafast laser spectroscopy enables us to investigate all the timescales given in Table 1.1. Apparently, lasers in the pulsed operating regime offer the most interesting features for advancing science, and technology beyond established limits. This is exactly the topic of this dissertation: *Pushing the limits of ultrashort pulse generation – the first 5 fs pulses directly from the laser oscillator.*

Applications of lasers are numerous and can be found in every day life, research, industry, and medicine. For example, GaAlAs semiconductor laser diodes are operating in CD–players, and in laser printers. In current DVD–players, the shorter wavelength InGaAlP laser is used. Helium–Neon lasers are found in bar code scanners in supermarkets, and are very dominant in the field of surveying, and ranging[1]. Important industrial lasers for material pro-

[1]During the Apollo 11 and Apollo 14 missions, corner reflectors were located on the surface of the Moon in order to determine the earth-moon distance. A powerful laser pulse was emitted from the MacDonald Observatory in Texas. By the time the laser light hit the moon surface it had spread to about a 3 km radius. The reflection back to the earth is strong enough to be detected. The distance from the moon to Texas was measured within about 15 cm, an accuracy of 10^{-9}.

Timescales (approximate values)	**Physical, biological, and chemical phenomena**
1 ms – 1 μs	– lifetime of upper state levels in solid–state laser materials – relaxation dynamics in phase transition liquid–glass – electron lifetime (T_1) in indirect gap semiconductors
10 ns – 10 ps	– electron lifetime (T_1) in direct gap semiconductors
100 ps – 100 fs	– vibrational dynamics of molecules – photodissociation of molecules
500 fs – 10 fs	– primary charge separation process in photosynthesis – dephasing of phonons, and electrons in semiconductors
$\lesssim$ 1 fs	– nuclear dynamics, e.g. vibrations, fission,... – ionization processes – bound electron dynamics

Table 1.1: Examples for typical timescales in different fields of science, illustrating the potential of pulsed laser sources to probe various dynamics in nature. [Kli97, Sve98, Ult00, Ult02]

cessing are solid–state lasers like Nd:YAG, high power laser diodes, and the classical work horse, the CO_2–gas laser. Microelectronic circuits in chips are unthinkable without using KrF (248 nm) and ArF (193 nm) lasers in Lithography. Various kinds of tunable solid–state lasers (TISA, Nd:glass,...) are useful for LIDAR (Light detection and ranging), an optical radar for the measurement of chemical concentrations or the determination of the speed of moving targets. In medicine, lasers serve as important tools for eye surgery, confocal microscopy, and Optical Coherence Tomography [Way99]. By far the strongest impact of all types of lasers is found in fundamental, and applied research. Namely, in the domains of spectroscopy [Müc01, Sha99, Kra02b, Gad02, Sch02b, Mül02], laser cooling and trapping of atoms or ions [Hän75, Vul00, Bin00, Ket02], laser nuclear fusion [Bal02a, Dit02], optical metrology [Ye00, Zan00, Hol01, Ste01, Jon01, Ram02, Did02], and others [Pov02, Wan03, Phu03]. An overview of laser applications is presented in Ref. [Spe98].

After intensive theoretical investigations on the subject of stimulated emission in the 1950s [Blo56, Sha58], and the first stimulated emission device (MASER: **M**icrowave **A**mplification by **S**timulated **E**mission of **R**adiation) [Gor54] the birth of the optical MASER, or LASER, was initiated by the successful operation of a flashlamp–pumped ruby laser at 694 nm by Theodore H. Mainman at Hughes Research Laboratories in 1960 [Mai60]. Since this early pioneering work, a lot of different kind of lasers have been developed: chemical lasers, free–electron lasers, X–ray lasers, dye lasers; and the most important nowadays: solid–state lasers, semiconductor lasers, and gas lasers [Sve98]. These lasers mainly differ by the active element providing the gain (ions, molecules, electrons, etc.), by the host material (solid–state, gas, semiconductor, etc.), and by the pumping process (optical, chemical, electrical, etc.).

The historical development of ultrashort laser pulses began with the invention of mode locking [DiD64, Har64, Yar65]. Alternative methods for pulse generation in lasers like gain–switching [Cas76], and Q–switching [Soo65] are also used since the early days of laser physics [Sie86]. However, the achievable pulse durations are in the order of the cavity–round–trip time, i.e. in the ns regime. Synchronous pumping allows for the generation of laser pulses $\gtrsim 1\,\mathrm{ps}$. It is preferably used with lasers having short upper state lifetimes, for example in dye lasers, and semiconductor lasers [Yas75, Her82, Yas83, Kaw91, For92]. Ultrashort laser pulses in the ps and fs regime, can only be generated using active and passive mode locking techniques.

Tables 1.2 and 1.3 represent an outline of the evolution of ultrashort pulse history. Emphasis is made on passive mode locking of solid-state lasers, because it allows the generation of the shortest laser pulses among other methods. Important developments concerning fiber lasers, semiconductor lasers, and other types of laser sources are only briefly mentioned. Naturally, the listed evolutionary steps and the cited references can only be a limited extraction of an enormous field of research activity over the last 40 years.

Even though the first mode locked lasers were solid–state lasers[2], dye lasers dominated ultrashort pulse generation until the end of the 1980s, resulting in a long standing pulse duration record of 6 fs. This impressive result was obtained by external compression via combined use of grating and prism pairs, thereby minimizing third order dispersion. In the mid - 1980s, new solid–state host materials were developed and went through extensive spectroscopic characterization. In contrast to active laser dyes, the active ions in these solid–state hosts have comparatively long upper state lifetimes. Mode locking by means of slow saturable absorbers is therefore more difficult and often requires the operation in the soliton regime. As a consequence, the area of mode locking mechanisms that can be described as fast saturable absorbers were developed. Especially Kerr–Lens mode locking, and Additive Pulse mode locking became the methods of choice for ultrashort pulse generation. Triggered by theoretical efforts to understand pulse formation processes in mode locked lasers by Hermann A. Haus in the early 1990s, an avalanche of experimental work generated laser pulses as short as 10 - 15 fs directly from TISA laser oscillators. A major turning point came in 1994, when Robert Szipöcs and Ferenc Krausz invented the principle of dispersion compensation by chirped multilayer coatings, the so–called *chirped mirrors*. These chirped dielectric Bragg stacks are designed in a way that they compensate for higher order dispersion in laser resonators while maintaining high reflectivity over broad bandwidths. In consequence, research groups around the world routinely generated sub–10 fs pulses. Franz X. Kärtner and Ursula Keller advanced the concept of chirped mirrors, leading to so–called *Double–Chirped Mirrors (DCM)*. Adiabatic impedance matching, and an additional anti–reflection coating on top of the mirror reduced residual reflections and lead to reduced oscillations in the dispersion characteristics. Implementation of DCMs rapidly lead to pulse shortenings approaching the two–cycle regime, i.e. only about two oscillations of the electric field within the pulse envelope.

[2] For the readers convenience, references are left out in this paragraph and are listed in the corresponding Tables 1.2 / 1.3

Major inventions and progress	References	Year	Annotations
invention of mode locking (ML)	[DiD64, Har64]	1964/65	DiDomenico,
	[Yar65]		Hargrove,Yariv
active mode locking	[DiD66, Kui70]	1965-70	$\tau_p \lesssim 100$ ps (Nd:gl.)
passive mode locking	[RC65, DeM66]	〃	$\tau_p \lesssim 100$ ps (Ruby)
mode locking of dye lasers	[Ipp72, Kog72]	1972	Rhodamine 6G
effect of gain saturation	[Hau75c, Rud76]	1975/76	$\tau_p \simeq 0.5$ ps
fast nonresonant absorbers	[Dah72, Sal77]	1972/77	optical Kerr effect
colliding pulse mode locking (CPM)	[For81]	1981-85	$\tau_p \simeq 90$ fs
use of prism pairs for negative GDD	[For84]	〃	Fork
soliton–like pulse shaping	[Val85]	〃	$\tau_p \simeq 27$ fs
first sub–10 fs pulse by external	[For87]	1987	$\tau_p \simeq$ **6 fs**
compression of dye laser pulses			
new solid–state host materials	[Mou86, Pet89]	1986/89	sapphire,LiSGaF
	[Smi92]		forsterite, ...
first active ML of Ti:sapphire	[Fre89]	1989	$\tau_p \simeq 80$ ps
first passive ML of Ti:sapphire	[Spe91]	1991	$\tau_p \simeq 60$ fs
additive pulse ML (APM)	[Ipp89, Hau91]		
solitary mode locking	[Bra91]	1990-92	
experimental / theoretical work	[Kel91, Sal91b]	〃	$\tau_p \simeq 70$ fs
on Kerr-Lens mode locking (KLM)	[Hau92]	〃	
semiconductor saturable absorber	[Kel90, Fel91]	〃	$\tau_p \simeq 2$ ps / 1 ps
mirror (SESAM)		〃	
ML with resonant nonlinearities	[Kel92]	〃	review article
ML with nonresonant nonlinearities	[Kra92]	〃	〃
soliton fiber lasers	[Dul91, Hof91]	〃	$\tau_p \simeq 2$ ps / 70 fs
self–starting ML	[Kra91, Riz92]	〃	$\tau_p \simeq 33$ fs
new insight in APM and KLM	[Hau92]	1992	H. Haus
by the master–equation of ML			
progress in pulse shortening by KLM	[Hua92, Pro93]	1992-94	$\tau_p \simeq 17$ fs/13 fs
combined with suitable intra-	[Cur93, Asa93]	〃	$\tau_p \simeq 12$ fs/11 fs
cavity prism materials	[Spi94, Zho94]	〃	$\tau_p \simeq 10$ fs
all solid–state pumped (Nd:YLF)	[Lam94]	〃	$\tau_p \simeq 110$ fs
invention of chirped mirror	[Szi94]	1994	Szipöcs, Krausz
first chirped mirror lasers	[Sti94, Sti95]	1994-98	$\tau_p \simeq 11$ fs / 8 fs
ring lasers with chirped mirrors	[Kas96, Xu96a]	〃	$\tau_p \simeq 10$ fs / 7 fs
theoretical investigations giving more	[Kär95, Kär98]	〃	SESAMS
profound understanding of ML	[Kär96, Che98a]	〃	soliton ML
	[Che99b]	〃	
improvements of the chirped mirror	[Tem98]	〃	refined design
first sub 5 fs pulses by external	[Bal97b]	1997	$\tau_p \simeq$ **4.6 fs**
compression of Ti:sapphire			

Table 1.2: Development of laser mode locking in the past. Milestones are highlighted in bold. τ_p refers to the full width at half maximum (FWHM) pulse duration.

Major inventions and progress	References	Year	Annotations
development of double–chirped mirrors (DCM)	[Kär97, Mat98]	1997/98	Kärtner, Keller
few cycle pulses directly from	[Jun97, Gal99]	1997-99	$\tau_p \backsimeq 6.5$ fs / 6 fs
the laser oscillator	[Sut99, Mor99]	"	$\tau_p \backsimeq 5.8$ fs / 5.4 fs
and by external compression	[Che98b, Shi99]	"	$\tau_p \backsimeq 4$ fs / 4.7 fs
high power / long cavity lasers	[Xu97, Bed99]	"	9 fs/5 nJ , 13 fs/13 nJ
progress in various fields:		2000-03	
infrared short pulse generation	[Tom00, Rip02]		55 fs/20 fs (Cr:YAG)
	[Rob00, Chu01]		71 fs / 14 fs Cr:forsterite
new DCM approach	[Mat00a, Tem01]		backside coated DCMs
double–chirped mirror pairs	[Kär01, Ell01]		$\tau_p \backsimeq 5$ fs
external compression	[ZR01, Bal02b]		5.7 fs / 4 fs (OPA)
	[Xu00b, Xu00a]		liquid cryst. modulator
diode pumping	[Hop02, Uem03]		113 fs/10 fs (Cr:LiSAF)
	[Wag03]		10 fs (Cr:LiCAF)
high repetition rate lasers	[Bar02]		1 GHz broadb. TISA
	[Kra02a]		157 GHz $Nd:YVO_4$
new concepts	[Här02, Bre02]		ML of laser diodes
	[Bru02]		22 W/ 240 fs thin disk

Table 1.3: Development of laser mode locking in the past (continuation). Milestones are highlighted in bold. τ_p refers to the full width at half maximum (FWHM) pulse duration.

The new pulse duration record of 5 fs, one of the main results of this work, was obtained by extending the idea of DCMs to *Double–Chirped Mirror Pairs* (DCMP). In 2001, Franz X. Kärtner and Uwe Morgner added an additional quarter–wave layer in one of the two otherwise identical DCMs. In this setup, they provide a phase–shift of π between the residual dispersion oscillations of the two mirror types. Consequently, these oscillations cancel after successive reflections on the two types of DCMs.

In recent years, research on ultrafast pulse generation has more and more diversified into other fields like new laser materials, diode pumping, thin disk lasers, high repetition rate lasers and mode locking of semiconductor lasers. Namely the new diode pumped materials like chromium doped LiSAF and LiCAF (so–called Colquiriites) offer interesting new perspectives. Without depending on bulky, expensive solid–state pump lasers, they may soon be employed for many applications for example in medical imaging. Pulsed thin disk lasers open up the route to high average power applications in material processing and other domains.

Regardless the mode locking method, the run for further pulse shortening will continue for the next years. Ultrashort pulse generation, either directly from the laser or by external compression in combination with spectral pulse shaping and reliable pulse characterization methods will play an important role in the future. Recently, physics on the timescale of attoseconds became explorable and will continue to be a dominant topic in ultrafast pulse research [Ult02, Kra02b, Mil02b, Mil02a, Ban03].

This thesis is a summary of my research on ultrashort pulse generation and pulse characterization together with new experiments in nonlinear optics concerning the carrier–envelope phase. To better understand the physical challenges of the subject, the following chapter will introduce the fundamental laser principles, focusing on the issue of mode locking. Aspects such as Kerr–lens mode locking, dispersion, and dispersion–managed mode locking are discussed. Chapter 3 presents the design, the setup, and the results of a new 2–foci TISA laser. Theory and measurement of the carrier–envelope phase is discussed in chapter 4. The essential element is the recovery of the temporal evolution of the absolute phase, which demonstrates the potential of nonlinear optics and allows applications in the area of frequency metrology, and phase sensitive nonlinear optics. Afterwards, the important question of pulse characterization is addressed in chapter 5. The implementation of a spectral shearing method for pulse characterization is described. Crucial points such as the role of the nonlinear crystal dispersion, and the calibration procedure are discussed. Two different kinds of ultrafast laser pulses are characterized and also dispersion measurements are demonstrated, using the same setup. The thesis closes with a conclusion and an outlook.