Habilitation Thesis

New approaches to the generation and characterization of few-cycle laser pulses

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NEW APPROACHES TO THE GENERATION AND CHARACTERIZATION OF FEW-CYCLE LASER PULSES

Habilitationsschrift

von

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Zürich, October 2001

(Günter Steinmeyer)
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ABSTRACT

With a gain bandwidth ranging from 650 nm to well above 1000 nm Ti:sapphire lasers offer a vast bandwidth supporting pulses of 5 fs duration. In this work, nearly the entire bandwidth could be utilized for mode-locking with pulse durations of 5.8 fs obtained directly from the laser. Important component in this type of lasers are chirped mirrors for dispersion compensation. Here a new generation of chirped mirrors is introduced. The approach of back-side coating the chirped mirror structure inherently avoids the impedance matching problem of previous chirped mirror generations and holds the potential to achieve dispersion compensation for octave-spanning spectra, which makes this technique particularly interesting for compression of white-light continua. A careful theoretical analysis is presented, followed by experiments with a mode-locked Ti:sapphire laser. Pulses from this laser are currently still the shortest well-characterized laser pulses from an oscillator. Measurement of such short pulses is an extremely important task. In this work several variants of frequency-resolved optical gating and spectral phase interferometry for direct electric-field reconstruction are described. These methods contributed much to the success of the work on the oscillator. With pulses as short as two optical cycles, the relative phase between carrier and envelope starts to become an important parameter in optics. A method to measure this parameter is described, relying on harmonic generation out of different part of the mode-locked laser comb and subsequent heterodyning. The phase noise of this quantity is carefully analyzed, and the laser is finally phase-locked to a reference oscillator with a residual rms phase jitter of only about 20 mrad. In the final part of this thesis, a novel method is described to generate short pulses by frequency doubling them in an aperiodic quasi-phase matching grating structure. An experimental demonstration yielded blue pulses as short as 5.3 fs. This sets a new record for the shortest pulse ever generated with second-harmonic generation and at oscillator repetition rates in the blue spectral range.
1. Introduction

The ability to conduct experiments on dynamical processes in physics is ultimately limited by the shortest possible observation time or by the shortest controllable event that can be created. Experiments in past centuries relied on direct observation by the human eye and were limited to tens of milliseconds. A great step ahead was the use of a pulsed light source for stroboscopic illumination, as first demonstrated in the famous experiments by Muybridge [1, 2]. In these pioneering experiments it was shown that the motion sequence of a galloping horse included a short phase where none of the horse’s hoofs touched the ground. This method was refined to a temporal resolution of a few microseconds, which is sufficient to freeze the dynamics of even the fastest macroscopic mechanical phenomena, e.g., a bullet ripping through a balloon [3]. With the advent of the laser and the discovery of mode-locking of lasers [4, 5, 6], the capabilities of brightly illuminating an experimental scenario greatly improved to picosecond resolution. Still, even on such time scales many phenomena in solid state materials are governed by diffusive processes and undergo rapid dephasing. With continuous improvements to femtosecond pulse duration [7, 8, 9], a new regime became accessible to investigations. In this regime, independent systems excited by photons from a femtosecond source undergo coherent dynamics and will only dephase on longer time scales. Pulse durations of a few fs are also rapid enough to resolve any type of molecular dynamics. For example, the ring opening process of a cyclo-butane molecule yielding two ethylene molecules was investigated by A. Zewail, through the excitation of cyclopentanone molecules with a femtosecond dye laser pulses [10]. Zewail’s experiment clearly showed the reaction to occur via a transition state living a few hundred femtoseconds. This experiment settled an old argument about the existence of an intermediate state, dating back about a century to a postulation of Arrhenius. Zewail’s work was rewarded by 1999’s Nobel prize in chemistry [11]. Recently, the spectral range of femtosecond studies was extended to x-rays, which allows for the investigation of structural changes in solid state materials with extreme temporal resolution [12].

The extreme shortness, however, is only one fascinating aspect of femtosecond laser light sources. Given a pulse duration of a few 10 fs, amplification of such pulses into the mJ range already leads to peak powers of $10^{12}$ W and above. Currently, there are major efforts underway to generate pulses with petawatt peak powers and focus them to an intensity of $10^{20}$ W/cm$^2$ and more [13, 14]. In this intensity regime, field strengths exceed inneratomic binding forces by several orders of magnitude and can even introduce relativistic effects [15, 16]. Amplified femtosecond lasers produce the highest controlled peak power and energy density of any man-made event. A third very important aspect of femtosecond lasers is their spectral coherence. Fourier theory requires that the extreme shortness of a few-femtosecond pulse is accompanied by a spectral width on the order of 100 THz. The modes of a typical few-femtosecond laser form an equidistant comb of a million synchronized phase-coherent oscillators, spaced by the laser’s repetition rate. Recently, it has been shown that the equidistance of the modelocked laser modes is better than one part in $10^{13}$ [17]. This uniformity opens up fascinating new applications in frequency metrology [18]. If one is able to measure one frequency out of the modelocked frequency comb and to control or measure the mode spacing, the frequency comb
can be utilized as a sequence of frequency calibration marks. These marks extend over the entire range of the mode-locked spectrum, and they can be regarded as frequency analogs to distance marks on a ruler. These novel possibilities to bridge the gap between spectrally remote optical frequency standards is considered as one of the major steps towards optical atomic clocks with a potentially much higher precision compared to the current cesium microwave frequency standard. Thus, apart from their shortness, femtosecond laser pulses offer a surprising wealth of applications based on energy concentration, spectral width, and coherence properties.

One of the major driving forces in the femtosecond research during the last 10 years was the Kerr-lens mode-locked Ti:sapphire laser. This laser has become the work horse in ultrafast research as the very first step of femtosecond pulse generation, in amplification of short pulses, compression schemes, and for frequency conversion to other wavelength ranges. In the second chapter of this thesis, the latest refinements of Ti:sapphire lasers for direct generation of sub-6-fs pulses are reviewed. The laser itself as well as most other schemes for the generation of sub-10-fs pulse generation rely on chirped mirrors for the compensation of dispersive broadening effects inside the laser cavity. Double-chirped mirrors have greatly boosted the performance of femtosecond lasers and enhanced the wavelength range accessible to mode-locked operation. But even this type of mirror is limited because of interference effects inside the mirror stack, giving rise to so-called dispersion oscillations. This limiting problem is reviewed in the third chapter. A solution, back-side coated chirped mirrors, is presented and demonstrated in a Ti:sapphire laser. This laser produced the shortest fully characterized pulses from a laser oscillator. The concept of the back-side coated mirror offers virtually unlimited bandwidth of more than one optical octave and is an extremely interesting component for compression of white-light continua and other broadband femtosecond sources.

Accurate measurement and characterization of short laser pulses has experienced great progress in the last few years. In previous years, pulse characterization has nearly exclusively relied on optical autocorrelation. As demonstrated in Chapter 4, this method is particularly unreliable for the complex pulse shapes observed in the sub-10-fs range. Alternative methods, such as frequency resolved optical gating (FROG) and spectral phase interferometry for direct electric-field reconstruction (SPIDER), provide much more information on the pulse shape. These methods set a new standard in pulse characterization. Adaption of these methods to the challenging case of the sub-10-fs oscillator is presented in Chapter 4, including spatially resolved variants and a concise discussion of the pros and cons of the individual methods. The suitability and performance of FROG and SPIDER will be reviewed for the case of the Ti:sapphire laser. It needs to be emphasized that proper characterization of laser pulses is not an end in itself, but provides additional information on detrimental mechanisms that lengthen the pulse or influence the pulse shape. Other than autocorrelation that simply tells that the pulse might be longer than desired, SPIDER and FROG allow the pinpointing of causes that is necessary for a cure.
Given the duration of an optical cycle is only 2.7 fs in a typical Ti:sapphire laser, one finds that within the half width point of the intensity envelope of a sub-6-fs pulse, merely two optical cycles are hosted. With such short pulses, the exact phase of the underlying electric field structure relative to the envelope becomes an interesting parameter for the interaction of such pulses and matter. In Chapter 5, methods will be described and demonstrated to measure the carrier-envelope offset phase and to stabilize it. The original proposal of this method has found widespread use in the meantime and has been used extensively in frequency metrology. Femtosecond lasers may well be the keystone for a new generation of optical clocks based on optical rather than microwave transitions. With the potential of building clocks of unprecedented precision, one hopes to be able to directly monitor minute drifts of fundamental physical constants as suggested by theoretical cosmological studies. Relative drifts of the fine structure constant on the order of a few $10^{15}$ determined from billions of years of look-back time [19, 20] could then be determined within a few days or weeks of observation. At first sight, the mode-locked laser with its broad spectrum is a somewhat surprising tool for ultraprecise measurements of frequencies. In Chapter 5 fluctuation and noise mechanisms of the laser mode comb will be discussed as these may impose a limitation to the use of femtosecond lasers in metrology. Also, the feasibility of experiments in extreme nonlinear optics, exploring a dependence of high-order nonlinear optical processes on carrier envelope offset phase, will be reviewed.

In the early days of ultrafast lasers, researchers sought a broadband laser material and a mode-locking scheme adapted to that material for every wavelength range they explored. A wide variety of dye, color center and other solid state laser materials was explored for picosecond pulse generation. With the availability of a robust and powerful femtosecond laser source at 800 nm, this strategy was traded for wavelength conversion schemes of femtosecond lasers. One important method is optical parametric amplification. Another one, which will be treated in Chapter 6, is direct frequency doubling of short pulses. After a brief review of the traditional approach employing birefringent phase matching, quasi-phase matched pulse compression will be reviewed. This method is very similar to dispersion compensation by chirped mirrors, and it is fully scalable with pulse width. One of the first demonstrations of this method, leading to a world record pulse duration below 6 fs in the blue spectral range, will be presented.

In closing, an outlook on the prospects of ultrafast research will be given. Femtosecond lasers have developed from a specialized laboratory tool that was nearly exclusively used for ultrafast pump-probe spectroscopy studies. Nowadays, a surprising wealth of applications has emerged, including their use as diagnostic tools in medicine, for laser material processing with femtosecond pulses, and for extremely precise frequency measurements.
1.1 References to Chapter 1

2. Sub-6-fs pulse generation from Ti:sapphire laser oscillators

The generation of ultrashort pulses naturally requires optical processes with an extremely wide bandwidth. While the fundamental technique of mode-locking of lasers was discovered in the early days of lasers, it took several decades before mode-locked spectra with several 100 nm bandwidth could be produced. Two key discoveries made this development possible: The Ti:sapphire gain material [1] and the Kerr-lens mode-locking (KLM) technique [2]. We will start giving a brief introduction and then review the latest development necessary to fully exploit the enormous bandwidth of the Ti:sapphire material.

Ti:sapphire provides laser gain from 650 nm to 1100 nm. A simple Fourier transform of the Ti:sapphire gain profile [3] promises a 4.5-fs pulse duration. The mode-locking process requires the introduction of an advantage for short-pulse operation over continuous operation of the laser. In its simplest form, this can be the saturation behavior of a dye or a semiconductor material. A saturable absorber displays a reduced absorption for high intensities compared to low intensities. Unfortunately, the nonlinear saturation behavior is not arbitrarily fast. Many methods have been developed to overcome or reduce response-time limitations of mode-locking (for a recent overview see for example [4]). In the following, we will restrict ourselves to the fundamentals of Kerr-lens mode-locking (KLM) which was discovered in Sibbett’s group in 1991 [2]. Initially the modelocking mechanism was not understood and was somewhat of a mystery. But within a short time after the initial discovery it became clear that the transverse Kerr effect provides an effective fast saturable absorber [5, 6, 7]. KLM is currently the most successful method for sub-10-femtosecond pulse generation directly from a modelocked laser.

The Kerr effect relies on a nonresonant electronic nonlinearity. This effect is extremely fast with response time of less than a femtosecond [8]. The Kerr effect causes an intensity dependent phase-shift

$$\phi_{nl}(x,y,t) = \frac{2\pi}{\lambda} n_2 I(x,y,t) \ell.$$  \hspace{1cm} (2.1)

Here $I$ is the intensity of the laser pulse, $\lambda$ the vacuum wavelength, $n_2$ the nonlinear refractive index, and $\ell$ the length of the nonlinear medium. In the following, we assume a positive sign of $n_2$, as is observed in all dielectrics far away from the band gap. The Kerr effect has implications both transverse to the direction of propagation (dimensions $x$ and $y$) and in the direction of propagation (time $t$). If the central and most intense part of the spatial beam profile is retarded, this corresponds to the focusing action of a lens. Therefore this transverse Kerr effect is called Kerr-lensing. This can be used to translate the phase-nonlinearity of the Kerr effect back into an effective saturable absorber. In KLM the transverse Kerr effect produces a Kerr lens that focuses the high intensity part of the beam more strongly than the low intensity part. Thus, combined with an intracavity aperture the Kerr lens produces less loss for high intensity and forms an effective fast saturable absorber. In addition, one can also obtain an advantage for pulsed
operation of the laser if a laser cavity with an additional Kerr lens has a better overlap of pump and laser mode than the same cavity without the lens.

The nonlinear phase shift caused by the Kerr effect along the beam of propagation is also called self-phase modulation (SPM). This effect is inseparably connected to the Kerr-lensing action and also plays an important role in Ti:sapphire lasers. As SPM delays the central part of an intense laser pulse in respect to the low-intensity wings, it compresses the oscillations of the electric field in the trailing part and stretches them in the leading part of the pulse. This causes a red shift in the pulse front and a blue shift at the end of the pulse. The main point is to see that this phase modulation results in an overall spectral broadening of the pulse. A broader spectrum, however, allows for shorter pulse duration, provided all phase distortions experienced in the nonlinear broadening process are compensated for. In particular, this pulse compression requires sending the spectrally broadened pulse through a delay line that advances the blue and retards the red spectral components of the pulse. Methods to provide exactly the right dispersion compensation for effective pulse compression will be treated in the following Section.

![Fig. 2.1](image_url)

**Fig. 2.1:** The Kerr effect gives rise to an increase of the refractive index with intensity, causing a retardation of the most intense parts of a pulse. In its longitudinal form (A) the Kerr effect causes self-phase modulation; in its transverse form (B) a nonlinear lens is formed in the central part of the beam profile.

In sub-10-fs Ti:sapphire lasers, both effects, the Kerr-lens and SPM go hand in hand. The strong action of pulse compression mechanisms and spectral broadening is illustrated by the appearance of spectral components well outside the gain region of Ti:sapphire [4]. On the other hand, strong SPM action can also lead to specific problems in the sub-10-fs regime. One of these problems is a spectrally-dependent mode size (compare Sect. 4.5). If the different spectral components of a laser pulse experience a different Kerr-lens effect (i.e. if the pulse is chirped) and this will also cause a spectrally dependent spatial mode profile of the laser [9, 10]. Another problem is that SPM together with dispersive effects in the cavity and spectral shaping in the output coupler can create pulses with a complicated spectral structure [11]. Thus, pulses generated in the sub-6-fs regime exhibit strong spectral distortions [12, 13] and are not well described by simple analytical functions (compare Sect. 4). Measurements of such pulses becomes very challenging because no specific pulse shape can be assumed to fit autocorrelation traces. In this
Sub-6-fs pulse generation from a Ti:sapphire laser

regime fits to interferometric autocorrelation measurements tend to strongly underestimate the real pulse duration [14]. Therefore, full characterization in amplitude and phase is necessary for characterization without any further assumptions and uncertainties. The shortest pulses from a KLM Ti:sapphire laser that have been fully characterized in amplitude and phase without any fitting attempts resulted in 5.8 fs FWHM [15].

Previously, pulses in the two-cycle regime were only achievable by extracavity pulse compression [16, 17] or parametric amplification [18]. Both of these techniques rely on a white-light continuum that is produced using high-energy laser pulses. It was recently demonstrated that continuum-generation can be used directly inside an oscillator to extend continuous wave (cw) modelocked laser operation beyond the gain bandwidth leading to sub-6-fs pulses [12]. Morgner et al. reported equivalent pulse durations at a slightly longer center wavelength [13, 19, 20]. In comparison to the previous sources, the laser oscillator offers the advantage of a higher repetition rate at lower system complexity. Some of the history of short-pulse sources has been compiled in Fig. 2.2.

![Fig. 2.2: Development of shortest reported pulse duration over the last three decades.](image)

Circles refer to dye-laser technology, triangles to Ti:sapphire laser systems, squares to optical parametric amplification. Filled symbols indicate results directly achieved from an oscillator, hollow symbols stand for results achieved with additional external pulse compression.

Self phase-modulation (SPM) can broaden the pulse bandwidth, and intracavity continuum generation beyond the gain bandwidth has been observed experimentally [12, 13, 21]. While this suggests that theoretically even shorter pulses are feasible, dispersion compensation supporting the bandwidth needed for a sub-6-fs Ti:sapphire laser pulse is very challenging. After passing through 1 mm of sapphire, the relative group delay between wavelength components at 700 nm and 1 µm is 43 fs. Even for a very thin crystal, a sub-6-fs pulse will temporally stretch, and it will need to be compressed inside the resonator by one order of magnitude. Unfortunately, the chirped mirrors commonly used today typically show deviations from the desired group delay ($T_g$). These deviations tend to oscillate with wavelength and increase with the bandwidth of the mirrors [22].
In the following the key components for obtaining shaped pulse spectra with a transform-limit of $\tau_{\text{FWHM}} < 5$ fs directly from a KLM Ti:sapphire laser will be described. The laser employs broadband double-chirped mirrors (DCMs, [22, 23, 24]) for dispersion compensation and a broadband semiconductor saturable absorber mirror (SESAM, [25, 26]) to assist Kerr-lens modelocking. Spectral shaping is achieved with a custom-designed output coupling (OC) mirror [12, 27]. It is shown how amplitude and/or phase shaping can lead to a significant reduction of $\tau_{\text{FWHM}}$. The laser performance is analyzed with phase sensitive pulse diagnostics, and current limitations are pointed out.

### 2.1 Experimental Results

A sketch of the laser resonator used throughout most of this research is shown in Fig. 2.3. The folded linear cavity has a round-trip cavity length of 3 m. It employs a SESAM at one end of the resonator and a custom designed OC mirror as the second end mirror. All other mirrors are broadband DCMs from a single coating run. The laser crystal has a plane-to-plane thickness of 2.3 mm and a doping level of 0.25 wt.%. The maximum output power attained for the 5.8-fs pulse shown in Fig. 2.4 is 300 mW at an absorbed pump power of 4.7 W from a frequency-doubled, diode-pumped neodymium-vanadate laser (Spectra Physics Millennia-X). Other results presented here were obtained with an argon-ion laser pump laser (Coherent Innova 200), a slightly longer 92-MHz cavity and a similar set of DCMs discussed below, but an otherwise unchanged configuration.

![Fig. 2.3: Ti: Sapphire laser resonator (M1-M4: double-chirped mirrors, OC: output coupling, FS: fused silica prisms with 40 cm apex separation and variable material insertion, SESAM: semiconductor saturable absorber mirror). Extracavity, the spectrally dispersed output beam is recombined with an identical prism sequence, and the dispersion of the substrate of the output coupling mirror is compensated with seven additional reflections off double-chirped mirrors.](image)

Figures 2.4 shows two sets of spectrum and autocorrelation measurements of this laser, achieved with the two different mirror sets. In both examples, pulse shaping via wavelength-dependent OC was employed as illustrated by the comparison of intracavity and extracavity spectra in Fig. 2.4. The intracavity spectra in this Figure both support a
transform-limited pulse duration of $\tau_{\text{FWHM}} = 6.8$ fs, whereas the extracavity transform limit is 5.3 fs in both cases.

**Fig. 2.4:** Sub-6-fs laser pulses. **Left:** (a) Intracavity and extracavity pulse spectrum (spectral power density); (b) measured interferometric autocorrelation (circles) and calculated trace (solid line) that results from an ideal sech$^2$ fit, yielding a 4.5-fs pulse; (c) measured interferometric autocorrelation (circles) and calculated trace (solid line) that results from a fit to the extracavity spectrum and intensity autocorrelation (Section 4.1, [12]). The envelopes of the autocorrelation for the ideal sech$^2$ surround the shaded area. The inset in (c) shows the temporal intensity envelope resulting from the phase retrieval algorithm. Its FWHM is $\tau_{\text{FWHM}} = 5.8$ fs. The extracavity Fourier-transform (FT) limit of $\tau_{\text{FWHM}} = 5.3$ fs is shorter than the intracavity value of $\tau_{\text{FWHM}} = 6.7$ fs, due to spectral shaping via wavelength-dependent output coupling. **Right:** same as left, but for slightly different operating conditions of the laser. In (b) a sinc$^2$ fit yields a 5.9-fs pulse duration; in (c) the solid line was reconstructed from a SPIDER measurement (comp. Section 4.3). A sech$^2$ assumption yields 4.7 fs (not shown). The inset in (c) shows the temporal intensity envelope retrieved with SPIDER. Its FWHM is $\tau_{\text{FWHM}} = 5.9$ fs. The extracavity Fourier-transform (FT) limit of $\tau_{\text{FWHM}} = 5.3$ fs is shorter than the intracavity value of $\tau_{\text{FWHM}} = 6.8$ fs, due wavelength-dependent output coupling.
In the left part of Fig. 2.4, the spectral phase was retrieved from autocorrelation and spectrum. This yielded $\tau_{\text{FWHM}} = 5.8$ fs; in the right part, the SPIDER (spectral phase interferometry for direct electric-field reconstruction) was used and resulted in a 5.9 fs pulse duration. These techniques will be explained in much more detail in Section 4 of this thesis. The commonly used ideal sech$^2$ fit would have resulted in a severe underestimation of $\tau_{\text{FWHM}}$ (4.5 fs, left, and 4.7 fs right part of Fig. 2.4), due to the significant deviation from an ideal sech$^2$ spectrum in both cases. A sinc$^2$ assumption gives an improved estimate for $\tau_{\text{FWHM}}$ but phase sensitive diagnostics obviously result in a much better agreement with the measured autocorrelation. Surprisingly, for both Fig. 2.4 and 2.5, the retrieved pulses are shorter than the intracavity transform limit. The key ingredients for the performance of the laser are the different types of mirrors, as discussed in the following.

### 2.2 Stabilization of Kerr-lens mode locking with a semiconductor saturable absorber mirror (SESAM)

A broadband semiconductor saturable absorber mirror (SESAM) with a low-finesse antiresonant Fabry-Perot structure was used in many of the experiments to stabilize Kerr-lens mode locking. The SESAM consists of a silver bottom mirror, followed by an AlAs spacer layer and the absorbing layers: a 20-nm In$_{0.22}$Ga$_{0.78}$As quantum well (QW) sandwiched between GaAs layers, see Fig. 2.5a. The modulation depth due to bleaching of the absorption is $\Delta R \approx 3.5\%$ over a bandwidth of 400 nm Fig. 2.5b. The saturation fluence is $F_{\text{A,sat}} \approx 180$ µJ/cm$^2$ and the absorber recovery time is $\tau_{\text{A}} \approx 2$ ps (Fig. 2.5).

As compared to pure KLM, the cavity alignment and the starting of the pulsed operation are significantly simplified by using the SESAM. The laser runs for many hours, providing stable single-pulse operation in the two-cycle regime with little changes of power or pulse width. Moreover, KLM can be sustained over a wide range of the cavity parameters. Using SESAM-assisted KLM, cavity parameter regions characterized by superior beam quality and higher average output power can be accessed where the laser would otherwise drop out of mode-locking. This improvement of beam quality is most apparent in the extreme spectral wings, where the cavity can no longer provide sufficient spatial mode confinement, mainly due to the strongly reduced reflectance of the OC mirror. This effect is most perceivable at the visible edge of the spectrum 650 nm. Figure 2.6 shows a comparison of transverse mode profiles in the yellow as observed behind one of the double-chirped resonator mirrors. The DCM coating has several sharp transmission resonances below 620 nm. After blocking the red and infrared components in the spectrally dispersed path between the external prisms, even yellow spectral parts become visible in the output beam. Behind the focusing mirror, even greenish spectral components could be made visible with appropriate filtering using laser safety goggles.
Sub-6-fs pulse generation from a Ti:sapphire laser

Fig. 2.5: SESAM design: (a) normalized standing-wave intensity distribution for wavelengths from 650 nm to 1 µm and refractive index profile; (b) reflectivity in the unsaturated regime (low incident light intensity, solid line) and under saturation (e.g. for the short intracavity pulses, dashed line). The SESAM structure is designed to show a broadband saturable loss with a modulation depth of $\approx 3.5\%$, shaded in (b). (c) and (d) Absorption bleaching of the SESAM: (c) the measured saturation fluence is $F_{A,\text{sat}} = 180$ µJ/cm$^2$; (d) the modulation depth $\Delta R$ shows an exponential absorber recovery time of $\tau_A = 2$ ps in the pump-probe trace. Both measurements were done with 150-fs, 830-nm pulses from a Spectra-Physics Tsunami laser. The pump-probe trace was recorded at a pump fluence of 75 µJ/cm$^2$ and a probe fluence of 3 µJ/cm$^2$.

Fig. 2.6: Transverse mode profiles observed on a screen behind resonator mirror M1 (Fig. 2.3): (a) with the resonator close to the stability limit, pure KLM action; (b) with resonator at center of the stability range, SESAM assisted KLM. Photographs taken with a 100ASA slide film (Kodak Elite).
2.3 Spectral Shaping for the Reduction of Pulse Durations

Haus’ master equation predicts a hyperbolic secant temporal and spectral pulse shape for KLM lasers [28]. For sub-10-fs lasers, a refined model, based on dispersion-managed solitons, predicts Gaussian or super-Gaussian intracavity spectral shapes [29]. These models, however, do not account for higher-order dispersion. Therefore, even though conclusions based on analytical models may well serve to explain some general features of the mode-locking process, a precise prediction of the pulse spectrum or shape is impossible for realistic experimental conditions. Measured pulse shapes in this regime exhibit complex multi-peaked structures caused by small imperfections in dispersion compensation (compare Fig. 2.4). These spectra are far away from any theoretically inspired analytical function and resemble a box spectrum with an additional modulation on top of it. The sharp roll-off observed at the ends of the spectra clearly sets them apart from the analytical scenarios with their smooth wings. Another important contribution to the spectral shape comes from the transmission characteristics of the output coupler. Even though an output coupler does not offer unlimited degrees of freedom, the mirror design it is based on can be adapted over quite a range of transmission characteristics. At first sight it may actually appear a little bit surprising that the shortest pulse is not reached with a spectrally flat ultrabroadband transmission. In the following, we explore how spectral shaping can be used for the generation of the shortest pulses, and we discuss the implications for the extracavity pulse shape.

2.3.1 Wavelength-dependent output coupling (OC)

To optimize the performance of the laser, OC mirrors with different bandwidths and transmissions ranging from 2% to 8% (at 780 nm) were available. Most of these output coupler designs are based on quarter-wave stacks, where the bandwidth is governed by the refractive index ratio of the dielectric coating materials. The target transmission of these designs is inversely proportional to Gaussians with transform limits of about 7 fs and centered near 800 nm, Fig. 2.7a. The reason for this will become clear in the following. For ultrabroadband OC, a thin glass plate can be placed in one resonator arm with one side anti-reflection coated and the other side uncoated to yield Fresnel reflection. Under certain circumstances, this can lead to two outputs of the same spectrum but opposite chirp [30, 31]. In the current work, we alternatively use a broadband OC mirror with a chirped dielectric coating. This mirror provides ≈5% transmission from 650 nm to 1.1 µm (Fig. 2.6b). Note that this specially designed OC has a bandwidth of nearly double that of a standard quarter wave design without introducing severe distortions on the phase of the transmitted light.
2.3.2 Optimum Amplitude Shaping

For a simple estimation on the severeness of the gain bandwidth, let us assume that the finite gain of Ti:sapphire and the finite bandwidth of the resonator mirrors support a maximum bandwidth of approximately 190 THz, beyond which the spectrum drops to zero. This box-like spectral filter would correspond, e.g., to a spectrum centered at 800 nm and a wavelength range from 650 nm to 1.1 µm. At pulse durations well above 10 fs, such spectral clipping does not result in any significant increase of pulse width. In contrast, a broad Gaussian amplitude spectrum with a FWHM of $\nu_{\text{FWHM}} = 100$ THz resulting in a transform-limit of 6.2 fs, is already stretched to $\tau_{\text{FWHM}} = 6.8$ fs with the 190 THz bandwidth limit.

Infinitely increasing the bandwidth of the Gaussian would ultimately result in a 190-THz broad box spectrum with a 4.7-fs Fourier limit. The duration can be decreased further by lifting the spectral wings via wavelength-dependent output coupling [27], resulting in M-shaped spectra. However, this decrease of $\tau_{\text{FWHM}}$ is traded for increased pedestals or satellite pulses. Going to the extremes, strongly modulated spectra like those obtained recently from an impulsively excited Raman medium [32, 33] correspond, under transform-limited conditions, to a train of extremely short pulses with a temporal spacing determined by the spectral modulation period. This makes it clear that spectral shaping has to be traded for quality of the pulse shape.
So far, spectral shaping effects were evaluated in their consequence on $\tau_{\text{FWHM}}$, which is a commonly used but somewhat arbitrary standard. The root-mean-squared pulse duration $\tau_{\text{rms}}$, i.e. the standard deviation of the temporal intensity envelope, is a more suitable criterion for the theoretical evaluation of pulse shapes. For spectral windowing as described above, $\tau_{\text{rms}}$ and therefore also the rms time-bandwidth product $\Delta v_{\text{rms}} \tau_{\text{rms}}$ are limited by $\tau_{\text{rms}}$ of the spectral windowing function. The largest possible curvature of the intensity envelope is determined by $\Delta v_{\text{rms}}$, i.e. the standard deviation of the spectral intensity, and will be reached at linear spectral phase [34]. Such a pulse is termed “transform-limited” because a linear phase will also yield the smallest rms time-bandwidth product $\Delta v_{\text{rms}} \tau_{\text{rms}}$ if it exists at all. Note that $\tau_{\text{rms}}$ may not be defined for certain theoretical pulse shapes, e.g. for a sinc$^2$ function [34]. For some applications, the rise time of the pulse, or the contrast ratio between peak and background intensity, the autocorrelation width, or the smoothness of the pulse spectrum might be more important criteria. Spectral shaping can be employed to improve either of these properties.

Fig. 2.8: Ideal amplitude shaping by wavelength-dependent output coupling: (top) The intracavity amplitude spectrum (the square root of the spectral power density) is a Gaussian with a full-width at half-maximum bandwidth of $\nu_{\text{FWHM}} = 100$ THz, clipped to 190 THz support (dotted line). A minimum transmission of 3% has been assumed. The external spectrum is box-shaped (solid line, shaded, bottom) The semi-logarithmic plot of the temporal intensity envelope reveals sinc$^2$-like side peaks. The intracavity transform-limited pulse duration is $\tau_{\text{FWHM}} = 6.8$ fs and the extracavity transform limit is $\tau_{\text{FWHM}} = 5.1$ fs. The extracavity pulse power is 6.9% and the peak intensity 8.7% of the respective intracavity values.

A practical compromise between $\tau_{\text{FWHM}}$ and pulse quality is given by a box-shaped spectrum with a sinc$^2$ temporal intensity profile. Such a spectrum is achieved when the OC is inversely proportional to the intracavity power spectrum. This neglects the influence of the OC on the intracavity pulse formation, which is a good approximation as long as the intracavity spectral width is limited mainly by the gain bandwidth. The maximum relative increase of power in the spectral wings is given by $1/T_{\text{min}}$, where $T_{\text{min}}$ is the minimum transmission of the OC mirror. For the quarter-wave-like mirror designs, intracavity spectra similar to those obtained with previous broadband DCMs were
assumed. In the example shown in Fig. 2.8, shaping a clipped Gaussian spectrum into a box results in a reduction of $\tau_{\text{FWHM}}$ to 75% of its intracavity value. However, it has to be noted that amplitude shaping is always accompanied by phase effects, as explained by the Kramers-Kronig relations that prohibit pure amplitude shaping, and additional phase compensation is required to fully exploit this reduction of $\tau_{\text{FWHM}}$.

2.3.3 Optimum Phase Shaping

The largest peak-to-average intensity ratio will be reached for transform-limited pulses. It is possible, however, that deviations from a linear phase reduce $\tau_{\text{FWHM}}$ below the transform-limited value (Fig. 2.9). In this case, more pronounced temporal pedestals arise and the rms duration $\tau_{\text{rms}}$ is increased. At the same time, the peak intensity drops, and the instantaneous frequency becomes time-dependent, inducing a chirp on the pulse.

Fig. 2.9: Phase shaping: (top) A box-shaped spectrum is Fourier-transformed assuming a constant phase (solid line) and two different non-constant phases (dash-dotted: medium chirp, dotted: strong chirp). (bottom) The temporal intensity envelope shows increasing side-peaks at decreasing peak intensity. The pulse duration is $\tau_{\text{FWHM}} = 5.1$ fs at flat spectral phase, 4.6 fs at medium chirp, and 4.0 fs at strong chirp. Normalized to the same average power, the peak intensity for medium chirp is 58% of the transform-limited pulse and 34% for strong chirp.

KLM lasers rely on the quasi-instantaneous self-amplitude modulation (SAM) of the Kerr-lens inside the crystal. Together with unbalanced dispersion and spectral shifts due to SPM, SAM causes spectral amplitude shaping. Strong shaping due to unbalanced fourth-order dispersion has been described previously [35, 36]. In fully dispersion-compensated linear resonators, a Fourier-limited pulse is obtained in the center of the gain crystal and at the end mirrors of the resonator if the dispersion is equally distributed on both resonator arms. This results in optimum SAM and consequently in shortest intracavity pulses. Therefore the laser was designed to provide equal negative group delay dispersion (GDD) in both arms.
Because the pulse is theoretically transform limited at the end of the resonator, the extracavity pulse will consequently carry a chirp induced by the transmission through the OC mirror. For the maximum attainable extracavity peak intensity, phase shaping has to be employed to remove this chirp. Currently, DCMs and prisms are to be preferred for this purpose. Nevertheless, the potential of liquid crystal devices that allow for both amplitude and phase shaping [37, 38, 39] has already been demonstrated with pulse durations slightly above those reached directly from the oscillator and may allow compression to even shorter pulse duration.

2.4 Conclusion

In the work described here, sub-6-fs pulses directly from a SESAM-assisted KLM Ti:sapphire laser have been obtained. This result shows that spectral shaping provides an efficient way to produce shorter pulses and to extend the spectrum of the modelocked pulses beyond the gain bandwidth. With the yellow light generated in the laser, a clear indication is given that intracavity white-light continuum generation plays an important role for the shortness of the generated pulse. A careful characterization of the generated pulses with SPIDER revealed durations well below the intracavity Fourier transform limit. The small residual chirp can be attributed mainly to the dispersion of the OC mirror. Phase shaping could even be used to push $\tau_{\text{FWHM}}$ further below the extracavity transform limit. Using the entire gain bandwidth of Ti:sapphire, intracavity 5.5-fs pulses appears to be achievable. Similar spectral shaping would then result in sub-4-fs pulses. This clearly demonstrates that, even with the results described here forth, there is still some potential for shorter pulses and the ultimate limit for Ti:sapphire lasers has not yet been reached. A remaining challenge in this regime is the further reduction of dispersion oscillations and the compensation of the external phase. Methods opening up new perspectives in dispersion compensation will be treated in the following chapter.

The wide frequency combs of ultrafast pulses allow for new applications in precision frequency metrology. Based on the octave-spanning spectra of two-cycle pulses, novel schemes have been proposed which give access to a measurement of the carrier-envelope offset (CEO) phase $\varphi_{\text{CEO}}$ (compare Section 5). Controlling $\varphi_{\text{CEO}}$, a predicted phase dependence of nonlinear optical processes can be explored, which is expected to be significant only at pulse durations of two-cycles or below. The scope of new applications clearly motivates the quest for shorter and shorter pulses.
2.5 References to Chapter 2

3. Chirped Mirrors

3.1. The problem of ultrabroadband dispersion compensation

Compensation of the spectral dependence of the optical path length is an ubiquitous problem in ultrashort pulse generation [1]. A coherent broad spectrum only yields a short bandwidth-limited pulse if all its spectral components have experienced the same group delay (GD)

\[
GD(\omega) = \frac{d}{d\omega} \frac{\omega}{c} n(\omega) \ell, \tag{3.1}
\]

where \( \omega \) is the angular optical frequency, \( c \) the speed of light, \( n \) the index of refraction, and \( \ell \) the length of the medium. Throughout the visible and most of the near-infrared spectral range, optical materials generally display a positive group delay dispersion (GDD)

\[
GDD(\omega) = \frac{d^2}{d\omega^2} \frac{\omega}{c} n(\omega) \ell. \tag{3.2}
\]

Consequently, “blue” spectral components will always lag behind “red” spectral components as soon as a short pulse is transmitted through some optical material, be it a lens, a beam splitter substrate or even simply air. The earliest strategy to compensate for GDD is the geometric dispersion of a parallel pair of gratings [2]. Typically, these grating pairs are used in negative first order to induce a group delay difference that exactly compensates material GDD. Quite generally, the group fronts exiting an element with angular dispersion are tilted, which induces red spectral components to lag behind blue ones. This geometric effect can be used to counteract GDD of common optical materials and can be utilized to generate negative GDD.

**Fig. 3.1a.** Dispersion compensation by grating (thick solid line) or prism sequences (dashed line). Shown is the group delay dispersion in comparison to a target curve (thin solid line). As the target, compensation for 1 cm of sapphire (e-axis) is chosen. The prism compressor uses fused silica Brewster prisms at an apex separation of 1.6 m and yields good compensation over a 100-nm interval at 850 nm. The gratings with 1000 gr./mm at a nominal distance of 0.54 mm also compensate for 1 cm of sapphire. Because of unmatched third-order dispersion, however, a grating pair provides a much smaller dispersion compensation bandwidth. **b.** Dispersion compensation by a single bounce off a chirped mirror. Here the dispersion of a combination of material and prism dispersion is used as the target curve (thin line). Extremely small residual dispersion oscillations are visible. The calculated example provides extremely broadband dispersion compensation with the technique of back-side coated mirrors.
Most notably, a pair of Brewster-cut prisms can compensate dispersion without introducing losses and has been very successfully used inside laser cavities [3]. The major shortcoming of the geometrical approach, however, is that it introduces higher-order dispersion terms (see Fig. 1). For a prism compressor, careful choice of the prism material allows for vanishing third-order dispersion

$$\text{TOD}(\omega) = \frac{d^3}{d\omega^3} \frac{\omega}{c} n(\omega) \ell$$

(3.3)

for one particular wavelength [4]. In particular, fused silica is the prism material of choice in a small part of the Ti:sapphire wavelength range, which was used for the first demonstration of sub-10-fs pulse generation with this laser [5]. Combinations of 4 and more prisms, such as the Proctor-Wise prism sequence [6] have been investigated and allow to design the ratio of second to third order dispersion within close bounds.

Gratings sequences may be used instead. They are of extreme importance for chirped-pulse amplification (CPA [7, 8]), which allows for the amplification of pulses from the oscillator to the mJ or Joule level at a reduced repetition rate. Terawatt and recently even Petawatt peak powers have been achieved [9]. To prevent damage in the amplifier chains, the oscillator pulse is stretched into the ps-range or ns-range before amplification. This reduces its peak power by the stretching ratio and also prevents nonlinear optical effects. After amplification the pulse can then be recompressed into the fs range using a grating sequence with exactly opposite dispersion of the stretcher and the amplifier material dispersion. This restores a short pulse duration and allows for the generation of extremely high peak powers. The stretcher [10] employed in CPA is typically a grating sequence, which incorporates a telescope with -1 magnification. The telescope exactly inverts the dispersion of a compressor in all orders. The trick is now to slightly unbalance a stretcher and a matched compressor and to accommodate for material dispersion in the difference between stretcher and compressor dispersion. Second-order dispersion (Eq. (3)) is adjusted by a difference in grating distances, third order dispersion (Eq. (4)) can be zeroed out by adjusting grating angles [11]. Fourth order aberrations, finally, can be compensated for by use of gratings with different groove frequencies [12]. Typically, aberrations of the telescope have to be compensated for by suitably corrected optics [13]. Other approaches exist, which introduce a controlled amount of imaging aberrations in the stretcher’s telescope to achieve compensation up to fourth order [14]. CPA has been demonstrated down to about 15 fs pulse duration [15, 16].

Grating or prism sequences can also be employed to adjust dispersion in an adaptive way. A very common set-up is the so-called 4f zero dispersion delay line [17]. This set-up is very similar to the previously described stretcher, but operates at an effective zero grating distance by placing the second grating exactly in the focus of the second grating. Thus no net negative or positive dispersion is introduced. If one places a spatial phase modulator into the Fourier plane of the stretcher set-up, this allows for the generation of arbitrary dispersion. As a spatial phase modulator either a liquid crystal array [18] or a deformable mirror [16] can be used. The adaptive approach is particularly useful if the dispersion to be compensated for is not very well known or may change over time. Recently, an adaptive pulse compressor was used to compress pulses to sub-6-fs pulse duration [19].
3.2 An introduction to chirped mirrors

Chirped mirrors [20] offer an alternative method for dispersion compensation. Other than the geometric dispersion approaches described before, they allow for compensation of arbitrary order dispersion. These dielectric mirrors consist of alternating pairs of transparent high-index and low-index layers. If the optical thickness of all layers is chosen equal to $\lambda_B/4$, interference of all Fresnel reflections generated by the index discontinuity at the layer interfaces will constructively add up for the Bragg-wavelength $\lambda_B$. Varying the optical layer thickness along the mirror structure during deposition then results in a dependence of Bragg wavelength $\lambda_B$ on penetration depth $z$, as shown in the top part of Fig. 3.2. Chirping the mirror structure therefore allows to generate a wanted group delay $GD(\omega)$. In addition the mirrors display an extended reflectivity range because the Bragg wavelength is swept over a range rather than kept constant. It is obvious, that the Bragg wavelength does not have to be varied linearly with penetration depth, but any single-valued function can be used as the chirp law $\lambda_B(z)$. In principle, this enables the compensation of material second-order dispersion together with arbitrary higher-order dispersion contributions.

![Diagram of chirped mirrors](image-url)

**Fig. 3.2:** Types of chirped mirrors. a: Simple chirped mirror structure. The desired effect of a wavelength-dependent penetration depth is accompanied by strong interference effects, mainly from a spurious reflection at the interface to the ambient medium. b: Double-chirped mirrors (DCMs) provide impedance matching with an additional anti-reflection (AR) coating on top of the mirror stack and by a duty-cycle modulation inside the mirror stack. c: Back-side coated (BASIC) mirrors use the substrate as the ambient medium. Suitable choice of the substrate material removes the need for an AR coating and provides impedance matching by double-chirping alone.
Unfortunately, the desired dispersion characteristics of a chirped mirror can be spoiled by spurious effects, which stem from multiple reflections within the coating stack. The combination of the highly reflecting chirped mirror stack and an unwanted reflection, e.g. at the interface between coating stack and ambient medium, effectively forms a Gires-Tournois interferometer (GTI, [21], see Fig. 3.2a). Such an interferometer consists of a highly and a partially reflecting mirror. An ideal GTI reflects all of the input light independent of wavelength but exhibits a periodic phase variation vs. frequency with a periodicity $\Delta \nu = c / 2L$, where $L$ is the distance between partial and high reflector. These dispersion oscillations disturb the above-described desired dispersion effects; the corresponding peak GDD of the dispersion oscillations can be much larger than the GDD from the penetration depth dependence. The already mentioned interface between air and the mirror stack was identified as the dominant mechanism causing such unwanted GTI-effects.

![Graph](image)

**Fig. 3.3:** Top Double-chirped mirror (DCM): Normalized standing wave intensity distribution inside the DCM coating (image plot: brightness corresponds to intensity). In the high-reflectance band, longer wavelengths penetrate deeper into the coating. For the green pump light, the mirror has a high transmission as shown on the reflectivity (Refl.) plot on the right. **Bottom:** Group-delay dispersion (GDD) of the double-chirped mirror for incidence angles of 5° (dash-dotted) and 20° (dotted), compared with the design target (solid line). The 20° angle is chosen for best cancellation of the GDD oscillation.

Depositing an antireflection (AR) coating on top of the chirped mirror stack appears to be a straightforward solution to the dispersion oscillation problem. Such an AR coating effectively provides matching between air and the effective impedance of the chirped mirror coating. It turns out that rather than directly matching from air to the coating, it is
far superior to split the problem into 2 steps: First, use an antireflection coating to match from ambient medium to the low-index layer material, and then use double-chirping within the layer stack to match from the low-index material to the effective impedance of the coating [22]. Double-chirping means that rather than only changing the Bragg period, the duty-cycle between high and low index material is also changed. Conventional chirped mirrors always employ equal optical lengths of high and low-index material within one period: \( L_{hi} = L_{lo} = \lambda / 4 \). Double-chirping offers the duty cycle \( \eta \) as an additional degree of freedom by only requiring \( L_{hi} + L_{lo} = (1 - \eta) \lambda_h / 2 + \eta \lambda_l / 2 = \lambda / 2 \).

This two-step approach depicted in the middle part of Fig. 3.2 considerably improves suppression of dispersion oscillations. For a given dispersion a double-chirped mirror design can be determined analytically [23]. Double-chirped mirrors have been demonstrated in Ti:sapphire lasers and optical parametric amplifiers in the sub-6-fs range. Dispersion compensation bandwidths in excess of 180 THz have been achieved in the visible and near-infrared wavelength range.

A limit in achieving even wider bandwidths of chirped mirrors is the quality of the antireflection coating. So far mode-locking in Ti:sapphire lasers was typically observed to tolerate GDD oscillations on the order of several 10 fs\(^2\). This requires that residual reflections at the top of the mirror stacks are suppressed to better than 10\(^{-4}\). Given the refractive indices of common coating materials such a low reflectivity can only be provided for a limited spectral range. This practically limits conventional chirped mirrors to slightly more than half an octave of bandwidth.

Several proposals to overcome the effective bandwidth limitation of chirped mirrors have been published. One fairly simple trick is the combination of several mirrors in such a way that dispersion oscillations cancel out [24]. Dispersion oscillations are shifted, e.g. by a change of the angle of incidence (see Fig. 3.3). Another more difficult approach is the manufacturing of matched pairs of chirped mirrors with exactly compensating dispersion oscillations [25, 26]. None of these approaches really removes the problem caused by the spurious reflection from the interface to the ambient medium. This problem could only be totally avoided if the ambient medium is chosen identical to one of the layer materials. This is the idea for the latest generation of chirped mirrors: back-side coated (BASIC) chirped mirrors [27] and shown as the bottom part of Fig. 3.2. For this type of mirror, the chirped layer sequence is deposited on the back surface of a thin optical substrate. Typically, one chooses fused silica for the substrate material, which then provides nearly ideal matching to the SiO\(_2\) used as the low-index coating material. Within the chirped mirror coating double-chirping can be used for impedance matching as this is not limited by any bandwidth restrictions. Detrimental reflections off the front surface of the substrate, which would result in GTI-type dispersion oscillations, can be suitably avoided by use of wedged or curved substrates. This allows to render surface reflections non-interfering and to completely avoid the problems of dispersion oscillations. Practical design studies based on this concept display a minimal amount of residual deviations from the desired dispersion characteristics as shown in Fig. 3.1 b. The latest generation of chirped mirrors allows for dispersion compensation over more than one optical octave with strongly reduced residual dispersion oscillations [27]. In the following the theoretical foundations for this type of chirped mirrors will be derived.
3.3 Exact coupled-mode theory and equivalent layers for the description of multilayer interference coatings

One of the most powerful analytical design tools in the theory of thin-film optical coatings is the method of equivalent layers. According to Herpin’s theorem, every symmetrical multilayer structure is equivalent, at one arbitrary wavelength, to a single homogeneous layer [28]. This layer, called the equivalent layer, is characterized by its equivalent (refractive) index \( N_e \) (Herpin index) and its equivalent (phase) thickness \( \Gamma_e \). For the design wavelength, the replacement of multiple layers by an equivalent layer conserves the optical properties of the coating and is therefore exact. Adjustment of \( N_e \) and \( \Gamma_e \) allows for the analytical design of multilayer coatings with various constraints on their spectral response characteristics [29-34].

An alternative description of multilayer coatings uses the coupled-mode theory. Originally, this approach described weakly index-modulated systems like waveguides [35, 36], (chirped) fiber gratings [37], and distributed feedback (DFB) lasers [38]. The coupled-mode equations are a differential equation system (DES) for the right- and leftward propagating waves. The solution of this DES results in a transfer matrix that relates waves at different positions, both inside and outside the multilayer structure. In coupled-mode theory, the design parameters are the coupling coefficient \( \kappa \) and the detuning coefficient \( \delta \). For systems with weak index-modulation, the standard coupled-mode approach provides an excellent approximation to the problem. In contrast, for the design of dielectric multilayer filters and mirrors, standard coupled-mode theory is regarded as insufficient because the assumption of a small perturbation is violated in the case of large index discontinuities. However, it was recently proven that by proper redefinition of the design parameters, the coupled-mode theory can be made absolutely exact even for the description of multilayer interference coatings with arbitrarily large refractive-index differences [23].

In both exact formalisms, the symmetrical multilayer structure is described by a set of two wavelength-dependent design parameters. Using these wavelength-dependent parameters in the respective formalism allows the calculation of the spectral response characteristics of the multilayer coating. One might assume there is a closer connection between coupled-mode theory and the method of equivalent layers. Here, the relationship between both methods is investigated, and relations are derived that link the parameters of coupled-mode theory \( (\kappa, \delta) \), with those describing the equivalent layer, \( (N_e, \Gamma_e) \). One of the major results is that the equivalent thickness is a linear function of \( \gamma \), the exact effective propagation constant of the Bragg structure.

The coupled-mode equations can be converted into equivalent transmission-line equations [39]. For these transmission-line equations, a normalized characteristic impedance \( Z \) is defined, and can be expressed as a function of the coupling and detuning coefficients. Finally, the equivalent index is proportional to the reciprocal characteristic impedance, i.e., \( N_e \propto Z^{-1} \).
The relations between the design parameters allow switching between the coupled-mode formalism and the method of equivalent layers, and the results derived within one formalism can be transformed into the other. The specific design problem dictates which method would be more convenient. As an example, if phase properties of the multilayer coating are a primary concern, the coupled-mode approach is advantageous. If matching of refractive index and phase thickness is most important the method of equivalent layers is favored. In the following, the formal equivalence of both methods will be used to analyze the problem of impedance matching in chirped mirrors and derive a solution that allows a nearly complete suppression of the impedance matching problem, giving rise to unwanted dispersion oscillations.

### 3.3.1 Exclusion of the ambient medium

A binary multilayer coating consists of a sequence of alternating layers with low and high refractive indices $n_1$ and $n_2$, respectively. For the theoretical considerations and examples in Sections 3.3 and 3.4, it is assumed that the refractive indices are real quantities without any wavelength dependence. The chirped Bragg structure is composed by symmetrically defined unit cells according to Fig. 3.4a [23]. The values for the refractive indices may be chosen arbitrarily and $n_2 > n_1$ is not required. Figure 3.4b shows the resulting refractive-index profile for a sequence of three unit cells. Application of standard coupled-mode theory on such a multilayer stack neglects embedding of this structure into an ambient medium, e.g., air on the top and the substrate at the bottom side of the coating. Usually the influence of index discontinuities at both ends of the coating is taken into account after the coupled-mode differential equation system has been solved.

**Fig. 3.4a:** Refractive-index profile of the symmetrically defined unit cell. $\phi_i (i = 1, 2)$ denotes the phase shift in the different layers, yielding a total optical phase shift $\phi$. The amplitude $A(m)$ refers to rightward-propagating waves, $B(m)$ to leftward-propagating waves. **b:** Resulting index-profile for a sequence of three unit cells.
An exact description for chirped Bragg gratings is given by exact coupled-mode equations of the form [23]

\[
\frac{d}{dm} \begin{pmatrix} A(m) \\ B(m) \end{pmatrix} = i \begin{pmatrix} -\delta(m) & -\kappa(m) \\ \kappa(m) & \delta(m) \end{pmatrix} \begin{pmatrix} A(m) \\ B(m) \end{pmatrix}.
\] (3.4)

In (3.4) \( A \) and \( B \) represent slowly varying normalized amplitudes of the right- and leftward propagating waves, respectively. The quasi-continuous variable \( m \) determines the position inside the mirror and counts the symmetric unit cells. By convention, \( m \) is a negative number. \( \kappa \) and \( \delta \) denote the coupling and detuning coefficients, respectively. For nonuniform grating structures these coefficients are locally defined for each unit cell. As was recently proven, a chirped mirror is exactly described by the coupled-mode equations (3.4), even for arbitrarily large refractive-index differences of the layer materials, provided the normalized coupling coefficient and detuning coefficient are properly defined. The exact coefficients can be written as [39, 40]

\[
\kappa(m) = -\alpha(\phi(m), \Delta\phi(m)) \frac{2r}{1-r^2} \sin \left( \frac{1}{2} (\phi(m) + \Delta\phi(m)) \right),
\] (3.5)

\[
\delta(m) = -\alpha(\phi(m), \Delta\phi(m)) \frac{1}{1-r^2} \left\{ \sin(\phi(m)) + r^2 \sin(\Delta\phi(m)) \right\},
\] (3.6)

with

\[
r = \frac{n_2 - n_1}{n_2 + n_1},
\] (3.7)

\[
\phi(m) = \phi_2(m) + \phi_1(m),
\] (3.8)

\[
\Delta\phi(m) = \phi_2(m) - \phi_1(m),
\] (3.9)

\[
\phi_1(m) = \frac{2\pi}{\lambda} n_1 d_{1,m},
\] (3.10)

\[
\phi_2(m) = \frac{2\pi}{\lambda} n_2 d_{2,m}.
\] (3.11)

In (3.7)-(3.11), \( r \) denotes the Fresnel reflectivity between the adjacent media 1 and 2, \( \lambda \) is the vacuum wavelength, and \( d_{1,2,m} \) the physical thickness of layer 1 and 2 of the \( |m| \)th unit cell. \( \phi_{1/2}(m) \) describes the optical phase shift in medium 1 and 2, respectively. Thus \( \phi(m) \) gives the total optical phase shift of the \( |m| \)th unit cell, and \( \Delta\phi(m) \) is a measure of the duty cycle within this unit cell. The factor \( \alpha = \gamma / \sin(\gamma) \) is defined by the exact propagation constant
3. Chirped mirrors

\[ \gamma = \sqrt{\delta^2 - \kappa^2} = \begin{cases} 
- i \cdot \ln \left( - F_R + \sqrt{F_R^2 - 1} \right); & F_R < -1 \\
\arctan \left( \frac{\sqrt{1 - F_R^2}}{-F_R} \right); & -1 \leq F_R \leq 0 \\
- \arctan \left( \frac{\sqrt{1 - F_R^2}}{F_R} \right) + \pi; & 0 < F_R \leq +1 \\
- i \cdot \ln \left( F_R - \sqrt{F_R^2 - 1} \right) + \pi; & F_R > +1 
\end{cases} \]  
\quad (3.12)

with

\[ F_R = \frac{1}{1 - r^2} \left\{ \cos(\phi) - r^2 \cos(\Delta \phi) \right\}, \]  
\quad (3.13)

using the definitions as given in [40]. The piecewise definition of \( \gamma \) allows for the distinction of different stop-band and passband regions. Complex values of \( \gamma \) (for \( |F_R| > 1 \)) indicate the stop bands. In the first case (\( F_R < -1 \)), the stop bands are centered at total phase shifts \( \phi \) that are odd multiples of \( \pi \). This includes the case of the fundamental Bragg wavelength at \( \phi = \pi \). In the fourth case (\( F_R > 1 \)) the stop bands are centered at even multiples of \( \pi \).

In coupled-mode theory the coupling and detuning coefficients are design parameters, i.e., particular spectral response characteristics of a multilayer coating can be achieved by suitable choice of these parameters. For the design problem of a chirped mirror the concept of impedance matching plays an important role. As derived in [39], the exact coefficients (3.5) and (3.6) for the individual unit cells define a characteristic impedance \( Z \) according to

\[ Z(m) = \frac{\sqrt{\delta(m) - \kappa(m)}}{\sqrt{\delta(m) + \kappa(m)}}. \]  
\quad (3.14)

The oscillations observed in the dispersion properties of a chirped mirror are caused by an impedance mismatch in the front part of the mirror. The impedance can be matched by properly adjusting the coupling coefficient as a function of penetration depth [39]. Moreover, the GDD can be independently designed by suitable choice of the detuning coefficient with the chirp law [23]. It is important to note that \( \kappa, \delta, \) and \( Z \) are not only functions of the considered unit cell \( |m| \); they also depend on wavelength \( \lambda \) (see (3.10) and (3.11)).

Alternatively, multilayer coatings that are composed of symmetrically defined unit cells can be described by the method of equivalent layers [31, 41]. According to Herpin’s theorem, every symmetrical combination of homogeneous layers is equivalent, at one arbitrary wavelength, to a single homogeneous layer [28]. This layer, called the equivalent layer, is characterized by its equivalent (refractive) index \( N_e \) (Herpin index) and its equivalent (phase) thickness \( \Gamma_e \). At the design wavelength the substitution of multiple layers by an equivalent layer conserves the optical properties of the coating and is therefore exact. In the picture of equivalent layers, \( N_e \) and \( \Gamma_e \) act as design parameters, similar to the situation described above for the coupled-mode theory. This represents an
alternative formalism that exactly describes a multilayer coating by using a set of two design parameters.

As recently shown in [40], there is a formal equivalence between coupled-mode theory and the method of equivalent layers. Relations exist that directly link the coupled-mode parameters \((\kappa, \delta)\) with those describing the equivalent layer \((N_e, \Gamma_e)\). For the three-layer combination shown in Fig. 3.4a, the following relations are valid:

\[
\frac{N_e}{n_1} = \frac{1}{Z} = \sqrt[\delta + \kappa]{\delta - \kappa} = \sqrt[\sin(\phi) + r^2 \sin(\Delta \phi) + 2 r \sin\left(\frac{\phi + \Delta \phi}{2}\right)]{\sin(\phi) + r^2 \sin(\Delta \phi) - 2 r \sin\left(\frac{\phi + \Delta \phi}{2}\right)},
\]

\[
\cos(\Gamma_e) = \cos(\gamma + \pi) = F_R
\]

\[
= \frac{1}{1 - r^2} \left\{ \cos(\phi) - r^2 \cos(\Delta \phi) \right\},
\]

where Eqs. (3.5), (3.6), and (3.12-3.14) have been used. Thus, the normalized equivalent index is the reciprocal of the characteristic impedance \(Z^{-1}\), and the equivalent thickness is essentially given by the exact propagation constant, \(\gamma\). Inversion of (3.16) results in a multi-valued solution for \(\Gamma_e\). The solution derived in [40] is compatible with the definition given in [29] and is used here. In contrast to the constant indices \(n_1\) and \(n_2\), the equivalent index shows a strong wavelength dependence. Moreover, for wavelengths in the stop-band regime the equivalent-layer parameters become complex. It follows from (3.15) that the impedance-matching problem of a chirped mirror translates into the problem of matching the equivalent index, as will be discussed in detail in Section 3.4. Additionally, finding an appropriate chirp law by adjusting the propagation constant \(\gamma\) via the detuning coefficient \(\delta\) is equivalent to a proper choice of the equivalent thickness along the chirped mirror structure according to (3.16).

### 3.3.2 Inclusion of the ambient medium

Now the index discontinuity at the ambient/coating interface is incorporated into the coupled-mode description. The refractive-index profile of the unit cell is defined as shown in Fig. 3.5a, i.e., the step of the refractive index from medium 1, \(n_1\), to the refractive index of the ambient medium, \(n_a\), is included at both ends of the unit cell. Figure 3.5b shows the refractive-index profile of a sequence of three unit cells. Inside the mirror structure, the downward steps and subsequent upward steps cancel each other. At the ends of the structure, however, an index step from material 1 to the respective ambient medium remains. This symmetric description of the unit cells presupposes that the refractive indices of both ambient media (e.g., substrate and air) are equal, which is generally not the case. However, for highly reflecting mirror coatings, as considered in this paper, this is irrelevant as almost no light passes through the coating.
Mathematically, the terminal discontinuity of the refractive index is described by Fresnel transfer matrices $S_u$ and $S_d$ for the upward and downward steps, respectively. These transfer matrices transform the amplitudes $A$ and $B$ into new amplitudes $\tilde{A}$ and $\tilde{B}$ (see Fig. 3.5a). This gives

$$
\begin{pmatrix}
\tilde{A}(m) \\
\tilde{B}(m)
\end{pmatrix} = S_u \begin{pmatrix}
A(m) \\
B(m)
\end{pmatrix} \Leftrightarrow
\begin{pmatrix}
A(m) \\
B(m)
\end{pmatrix} = S_d \begin{pmatrix}
\tilde{A}(m) \\
\tilde{B}(m)
\end{pmatrix},
$$

(3.17)

with the Fresnel matrices [42]

$$
S_u = \frac{1}{2\sqrt{n_1 n_2}} \begin{pmatrix}
n_a + n_l & n_a - n_l \\
n_a - n_l & n_a + n_l
\end{pmatrix} = S_d^{-1},
$$

(3.18)

$$
S_d = \frac{1}{2\sqrt{n_2 n_1}} \begin{pmatrix}
n_a + n_l & - (n_a - n_l) \\
-(n_a - n_l) & n_a + n_l
\end{pmatrix} = S_u^{-1}.
$$

(3.19)

Using the transformation (3.17), one yields the following new coupled-mode equations for the transformed amplitudes

$$
\frac{d}{dm} \begin{pmatrix}
\tilde{A}(m) \\
\tilde{B}(m)
\end{pmatrix} = i \begin{pmatrix}
-\tilde{\delta}(m) & -\tilde{\kappa}(m) \\
\tilde{\kappa}(m) & -\tilde{\delta}(m)
\end{pmatrix} \begin{pmatrix}
\tilde{A}(m) \\
\tilde{B}(m)
\end{pmatrix},
$$

(3.20)

where the transformed coefficient matrix is given by
Thus, the new coefficients are obtained from the original ones by a similarity transformation. Evaluating (3.21) yields the following explicit expressions for the transformed exact coupling and detuning coefficients

\[
\begin{pmatrix}
-\tilde{\delta}(m) & -\tilde{\kappa}(m) \\
\tilde{\kappa}(m) & \tilde{\delta}(m)
\end{pmatrix}
= S_u \begin{pmatrix}
-\delta(m) & -\kappa(m) \\
\kappa(m) & \delta(m)
\end{pmatrix} \cdot S_u^{-1}.
\]  

(3.21)

One can immediately see that the transformed coupling and detuning coefficients reduce to the original coefficients (3.5) and (3.6) in case of an ambient medium with a refractive index equal to the index of medium 1, i.e., \( n_a = n_1 \). Table 3.1 summarizes examples of transformation coefficients (3.24) and (3.25) that will be discussed in the next section. The coefficient \( c_1 \) is always close to unity, whereas the value of \( c_2 \) strongly varies. \( c_2 \) is a direct measure of the index discontinuity between the ambient medium and medium 1 and essentially describes difference between transformed and original coefficients.

<table>
<thead>
<tr>
<th></th>
<th>( c_1 )</th>
<th>( c_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>A. no impedance matching</td>
<td>1.08</td>
<td>0.42</td>
</tr>
<tr>
<td>B. partial impedance matching</td>
<td>1.03</td>
<td>-0.26</td>
</tr>
<tr>
<td>C. perfect impedance matching</td>
<td>1.00</td>
<td>0.00</td>
</tr>
</tbody>
</table>

**Table 3.1:** Values of the transformation coefficients \( c_1 \) and \( c_2 \) used in the following design studies.

Using (3.14) and (3.22-3.25) the transformed impedance follows to

\[
\tilde{Z}(m) = \frac{\tilde{\delta}(m) - \tilde{\kappa}(m)}{\sqrt{\tilde{\delta}(m) + \tilde{\kappa}(m)}} = \frac{n_a}{n_i} Z(m) = \frac{n_a}{n_i} N_e.
\]  

(3.26)

Finally, it should be noted that the equivalent-layer parameters are invariant under the transformation (3.17), i.e., \( \tilde{N}_e = N_e \) and \( \tilde{\Gamma}_e = \Gamma_e \).
3.4 Impedance matching to the ambient medium

Comparing the coupled-mode equations (3.3) with (3.20) one sees that both equations are exactly of the same form. Hence, all equations and conclusions derived in [22, 23, 39] still hold, provided the transformed coefficients (3.22-3.25) are used instead of (3.5) and (3.6). The conditions for the suppression of dispersion oscillations

\[
Z(m = 0) = 1 \quad \forall \delta \Rightarrow \kappa(m = 0) = 0, \quad (3.27)
\]

\[
Z'(m = 0) = 0 \quad \forall \delta \Rightarrow \kappa'(m = 0) = 0 \quad (3.28)
\]

provide perfect impedance-matching of the chirped mirror structure to an ambient medium with the same refractive index as medium 1 (see [39]). The prime in (3.28) denotes the derivative with respect to \( m \). Condition (3.27) states that, according to (3.15), the equivalent index must equal the index of medium 1 at the mirror front. Moreover, for perfect matching, condition (3.28) requires that the equivalent index be ramped up or down as slowly and smoothly as possible.

The same conditions also hold for the transformed coupled-mode parameters, i.e.,

\[
\tilde{Z}(m = 0) = 1 \quad \forall \tilde{\delta} \Rightarrow \tilde{\kappa}(m = 0) = 0, \quad (3.29)
\]

\[
\tilde{Z}'(m = 0) = 0 \quad \forall \tilde{\delta} \Rightarrow \tilde{\kappa}'(m = 0) = 0 \quad (3.30)
\]

According to (3.26), these impedance-matching conditions are equivalent to matching of the equivalent index of the unit cell to the refractive index of the ambient medium. Generally, for an arbitrary ambient medium, finding a broadband solution satisfying these conditions is very difficult because of the strong wavelength dependence of the equivalent index.

In the following subsections, the matching condition (3.29) is investigated for three ambient media with different refractive indices \( n_a \). Without loss of generality, the investigation is restricted to Eq. (3.29). If this condition is satisfied, condition (3.30) can be always fulfilled by a sufficiently slow change of the design parameters. Large differences in the quality of the design are found, depending on the degree of impedance matching achieved and indicated by the magnitude of dispersion oscillations. In the following examples, the refractive indices \( n_1 = 1.5 \) and \( n_2 = 2.5 \) are chosen for the layer materials, leading to \( r = 0.25 \). This choice was motivated by the fact that these values are close to the indices of SiO\(_2\) and TiO\(_2\), commonly used as dielectric coating materials.

3.4.1 No impedance matching

In the first example, it is assumed that the ambient medium is air, i.e., \( n_a = 1.0 \). In terms of dispersion oscillations this is discussed as the worst possible case. No attempts to improve impedance matching at the interface to air have been done and a plain simple-chirped mirror structure is considered in the following. Simple-chirped mirror refers to a multilayer structure with a gradually increased Bragg wavelength but a constant 50% duty cycle, i.e., \( \phi_1(m) = \phi_2(m) \forall m \).
Figure 3.7 shows the response characteristics of a simple-chirped mirror. In this example, the Bragg wavelength is chirped over the initial 20 unit cells and then kept constant for another 5 unit cells to increase the reflectance for long wavelengths. The reflectance is very high over a broad wavelength range and covers most of the Ti:sapphire gain spectrum. However, the group delay (GD) shows extremely large oscillations, similar to a Gires-Tournois interferometer, with a peak-to-peak amplitude on the order of 100 fs, which makes such a mirror useless for ultrashort pulse generation.

![Figure 3.7: Calculated amplitude (left axis) and phase (right axis) properties of a standard front-coated simple-chirped mirror as a function of wavelength. Shown are the reflectance and the group delay upon reflection, respectively. In this example the Bragg wavenumber is linearly decreased over the first 20 unit cells from the maximum value \( k_B^{\text{max}} = \frac{2\pi}{(650 \text{ nm})} \) to the minimum value \( k_B^{\text{min}} = \frac{2\pi}{(950 \text{ nm})} \) according to \( k_B(m) = k_B^{\text{max}} - (|m|-1) \cdot \frac{(k_B^{\text{max}} - k_B^{\text{min}})}{19} \). For the last 5 unit cells the Bragg wavenumber is kept constant at its minimum value \( k_B^{\text{min}} \). Air has been assumed as the ambient medium. The simple-chirped mirror provides a broad high-reflectance range, but exhibits strong dispersion oscillations over most of this range.

Obviously, the simple-chirped mirror structure does not provide any impedance matching. To further investigate this, the transformed coupled-mode design parameters \((\tilde{\kappa}, \tilde{\delta})\) and the equivalent-layer parameters \((N_e, \Gamma_e)\) are explored as two-dimensional functions of the unit cell and wavelength, depicted as contour-plots. In these plots the x-axis gives the unit cell and the y-axis gives the incident wavelength. It is assumed that the light incidents from the right. In Fig. 3.8a the contour lines of the transformed coupling coefficient (3.22) are almost horizontal lines with values far from zero at the coating surface. Condition (3.29) is not fulfilled for any given wavelength in the high-reflectance region. In fact, the transformed coupling coefficient in Fig. 3.8 is even farther from being matched to zero than the original coupling coefficient. The original coupling coefficient is always negative and approximately constant due to the simple-chirped mirror structure \((\kappa = -2r = -0.5, [40])\). According to (3.22), matching can only be achieved by positive values of \(c_2\delta\). For wavelengths above 650 nm, however, this is not fulfilled at the coating surface because of the negative detuning coefficient \(\delta\). Consequently, absolutely no impedance matching is achieved in this simple-chirped structure.
Fig. 3.8: Contour plots of the transformed coupling coefficient $\tilde{k}$ (a), the detuning coefficient $\tilde{\delta}$ (b), the equivalent index $N_e$ (c), and the equivalent thickness $\Gamma_e$ (d, in units of $\pi$) of the simple-chirped mirror of Fig. 3.7 as functions of unit cell $|m|$ and wavelength.

Figure 3.8b shows the contour lines of the transformed detuning coefficient. These contour lines are nearly linear functions over the range of chirped Bragg wavenumbers. According to (3.23), the transformed detuning coefficient essentially follows the original detuning coefficient with an offset of $c_2 / k \approx 0.2$. The qualitative behavior of the detuning coefficient is not changed by the transformation. The zero contour line of the
original detuning coefficient indicates the Bragg condition, i.e. the dependence of Bragg wavelength on penetration depth or chirp law. Compared to the original detuning coefficient, the zero contour line of the transformed detuning coefficient is shifted by \( \approx 50 \text{ nm} \) to shorter wavelengths (dashed line). This plot clearly illustrates the approximately linear increase of the GD with wavelength (negative GDD).

This scenario can be also interpreted in the picture of equivalent layers. According to Section 3.3, impedance matching requires that the equivalent index at the mirror front equals the refractive index of the ambient medium \((n_a = 1.0)\). However, Fig. 3.8c shows that the equivalent index is extremely far from being matched to unity at the mirror front, illustrating the large impedance mismatch. The broad area surrounded by contour lines with the values zero and infinity marks the stop-band region, where the equivalent index becomes complex. For this region the unit cells represent a potential barrier with an exponential behavior of the electromagnetic field. In the passband regions outside the evanescent region, the equivalent index is real and the unit cells are transparent for the light. With respect to the quantum mechanical scattering problem as introduced in [39], the upper line \((N_e = \infty)\) corresponds to right turning points and the lower line \(N_e = 0\) to left turning points of the corresponding classical motion.

Figure 3.8d shows that, for a fixed wavelength, the equivalent thickness of the unit cells monotonically increases with penetration depth illustrating the chirp of the Bragg wavelength along the mirror structure. As mentioned above, custom-tailored dispersion properties can be designed by a proper control of the equivalent thickness. For passband regions the equivalent thickness of unit cell \(|m|\) approximately corresponds to the total optical phase shift \((3.8)\) evaluated at wavelength \(\lambda\) [40]. Similarly to Fig. 3.8c, the evanescent region is surrounded on both sides by the dashed unity contour line (given in units of \(\pi\)).

3.4.2 Partial impedance matching

In the second example the case of a simple-chirped mirror is considered again, but now the refractive index of the ambient medium with best possible impedance matching and without additional matching layers (e.g., an AR coating) is searched for. It might be suspected that best matching is achieved if the index of the ambient medium is the geometric average of the indices of medium 1 and 2. Structures matched to the geometric average of the refractive indices have been used before as a starting design for chirped mirrors (see, e.g., [43]). It is shown by an example, however, that this method does not yet lead to the optimum solution for the impedance-matching problem considered and only provides partial impedance matching.

For a simple-chirped mirror with \(\Delta \phi(m) = 0 \ \forall m\), using (3.15) and (3.26) the impedance-matching condition (3.29) is rewritten as

\[
\tilde{Z}(m) = \frac{n_a}{n_i} \cdot \frac{\sin(\phi) - 2r \sin(\phi/2)}{\sqrt{\sin(\phi) + 2r \sin(\phi/2)}} = 1.
\] (3.31)

From this the ambient refractive index can be resolved as
\[ n_a = n_1 \cdot \sqrt{\frac{\cos(\phi / 2) + r}{\cos(\phi / 2) - r}}. \] (3.32)

The right-hand side of (3.32) is the equivalent index as a function of the phase variable \( \phi \). Impedance matching requires that (3.32) be fulfilled over a broad wavelength range, i.e., the equivalent index should equal the index of the ambient medium \( n_a \) independent of the total phase shift \( \phi \). The refractive index of the ambient medium is a real quantity. This requires \( \phi < 2 \cdot \arccos(|r|) \) as a necessary condition for phase shifts \( \phi \leq \pi \). The equivalent index is approximately constant only for sufficiently small phase shifts \( \phi \) close to zero, see Fig. 3.9. In particular, this figure clearly illustrates the strong wavelength dependence of the equivalent index, as mentioned in Section 3.3. In the limit \( \phi \to 0 \) (\( \lambda \to \infty \)) this yields the long-known result [33]:

\[ n_a = n_1 \cdot \sqrt{\frac{1 + r}{1 - r}} = n_1 n_2. \] (3.33)

This is exactly the geometric average refractive index of both layer materials. The limit \( \phi \to 0 \) appears to be in contradiction to the assumption of quarter-wave layers, where \( \phi = \pi \) for wavelengths near the Bragg wavelength. In this relevant case, the impedance-matching condition practically requires to start chirping at Bragg wavelengths much smaller than the minimum design wavelength of the coating. In other words, the impedance-matching condition \( \phi \to 0 \) is only fulfilled for wavelengths much larger than the Bragg wavelength of the unit cell considered.

![Graph](image)

**Fig. 3.9:** Equivalent index as a function of phase shift as given by Eq. (3.32). As in the other examples, \( n_1 = 1.5, n_2 = 2.5 \) has been assumed. For small values of \( \phi \), the equivalent index approaches the geometric average of \( n_1 \) and \( n_2 \), i.e. \( n_a = 1.94 \). The equivalent index diverges for \( \phi \to 2 \arccos(|r|) \) and is complex-valued for the fundamental stop-band region at Bragg resonance \( \phi_B = \pi \).

Let us consider the example of a simple-chirped mirror structure in Figs. 3.10 and 3.11. Other than in the previous example (Figs. 3.7 and 3.8) one now assumes an ambient medium with an index equal to the geometrical average of the two layer materials. For a
coating providing dispersion compensation from 600 nm to 1000 nm, chirping the Bragg wavelength starts at 300 nm. The high-reflectance range of the coating is extremely wide as shown in Fig. 3.10, but most of this range lies well out of the band with phase properties that might be useful for dispersion compensation. The GD oscillations with a peak-to-peak amplitude of about 10 fs are considerably reduced compared to Fig. 3.7. The dispersion oscillations decrease with an increasing wavelength but never reduce to a negligible value. The curve is far from being smooth and such oscillations are not tolerable inside an ultrashort-pulse laser. Reducing the number of layers and increasing the initial Bragg wavelength of the design further deteriorates the phase properties of the coating. On the other hand, dispersion properties at 800 nm may be improved by starting to chirp at even lower Bragg wavelengths < 300 nm. In any case, only a small part of the spectrum is covered providing high reflectance and decent dispersion properties simultaneously.

The contour plots shown in Fig. 3.11 further illustrate the partially achieved impedance matching. Still, the contour lines at the mirror front are essentially horizontal lines similar to the contour lines shown in Fig. 3.8. In contrast, matching condition (3.29) is partially fulfilled now, as indicated by the contour plots for the transformed coupling coefficient and for the equivalent index (see, Fig. 3.11a and c). These contour plots confirm that matching is improved at longer wavelengths. With increasing wavelength the coupling coefficient approximates zero, Fig. 3.11a, and the equivalent index in Fig. 3.11c approaches \( n_a \) given by (3.33).

In conclusion, assuming quarter-wave layers and an ambient medium with geometric average of the indices of medium 1 and 2, provides a partial reduction of dispersion oscillations. The quality of impedance matching, however, is directly related to the excess bandwidth with excellent reflectance but poor dispersion properties. Therefore,
this design approach makes very uneconomic use of the available number of layers, which renders this approach impracticable for many applications.

Fig. 3.11: Contour plots of the transformed coupling coefficient $\tilde{\kappa}$ (a), the detuning coefficient $\tilde{\delta}$ (b), the equivalent index $N_e$ (c), and the equivalent thickness $\Gamma_e$ (d, in units of $\pi$) of the back-side coated simple-chirped mirror of Fig. 3.10 as functions of unit cell $|m|$ and wavelength.
3.4.3 Perfect impedance matching

In the final example, it is assumed that the ambient medium has the same refractive index as medium 1, i.e., \( n_a = n_i = 1.5 \). As mentioned in Section 3.3, this reduces the transformed coupled-mode parameters to the original ones and matching condition (3.29) reduces to the old matching condition (3.27). According to [39], the DCM design technique now allows for perfect impedance-matching. This means to use very thin layers of material 2 for the initial unit cells in the mirror structure, i.e., \( n_m d_{2,m} \ll \lambda_B(m) / 4 \) and then slowly ramping up the duty cycle of the coating until 50% is reached, i.e., \( n_m d_{2,m} = \lambda_B(m) / 4 \). With the DCM technique, the coupling coefficient vanishes according to (3.5) and (3.7-3.14), and the impedance equals unity according to (3.14). Other than in a standard DCM, however, no broadband AR coating is required to match the index to the ambient medium.

Figure 3.12 shows reflectance and GD of a DCM consisting of 25 unit cells. The Bragg wavenumber is linearly chirped over the first 20 unit cells and then kept constant for the remaining 5 unit cells in the same way as in Section 3.4.1. Now, however, a material with an index identical to the low-index layer material is assumed as the ambient medium. Also, the thickness of the material 2 over the first 12 unit cells is independently chirped (double-chirping). The very smooth dispersion over the entire high-reflectance range in Fig. 3.12 clearly illustrates the resulting effect of impedance matching. This comes at the expense of a slight reduction of the high-reflectance bandwidth on the short-wavelength side, as a comparison of Fig. 3.7 and Fig. 3.12 reveals [39].

**Fig. 3.12:** Calculated reflectance (left axis) and group delay (right axis) of a BASIC double-chirped mirror as a function of wavelength. In this example the Bragg wavenumber is linearly decreased over the first 20 unit cells from the maximum value \( k_B^{\text{max}} = 2\pi / (650 \text{ nm}) \) to the minimum value \( k_B^{\text{min}} = 2\pi / (950 \text{ nm}) \) according to \( k_B(m) = k_B^{\text{max}} - (|m|-1) \cdot (k_B^{\text{max}} - k_B^{\text{min}}) / 19 \). For the last 5 unit cells the Bragg wavenumber is kept constant at its minimum value \( k_B^{\text{min}} \). The thickness of the material 2 is varied with penetration depth according to \( d_{2,m} = \pi / (2k_B(12)n_i) \cdot (|m| / 12)^{1/2} \) over the first 12 unit cells. A material with an index equal to the low-index material of the coating has been assumed as the ambient medium \( (n_a = n_i = 1.5) \). The BASIC double-chirped mirror provides a broad high-reflectance range and smooth dispersion characteristics, simultaneously.
3. Chirped mirrors

![Diagram](image.png)

**Fig. 3.13:** Contour plots of the coupling coefficient $\tilde{k}$ (a), the detuning coefficient $\tilde{\delta}$ (b), the equivalent index $N_e$ (c), and the equivalent thickness $\Gamma_e$ (d, in units of $\pi$) of the BASIC double-chirped mirror in Fig. 9 as functions of unit cell $|m|$ and wavelength. Note that in a and b transformed and original values for the coupling coefficient and the detuning coefficient are identical.

In Fig. 3.13a, the (transformed) coupling coefficient of the DCM is plotted. At the beginning of the mirror structure, the contour lines are almost vertically oriented. Each individual unit cell provides an almost constant coupling coefficient for any given
wavelength. The coupling coefficient vanishes at the beginning, indicating excellent broadband impedance matching. The absolute value of the coupling coefficient increases along the grating structure until it reaches its maximum value. The behavior of the (transformed) detuning coefficient shown in Fig. 3.13b generally resembles the first example shown in Fig. 3.8b. In contrast, however, the detuning values of Fig. 3.13b are shifted towards larger values. The zero contour line pinpoints Bragg resonance, yielding the chirp law [23].

The high degree of impedance matching achieved is also clearly illustrated by the contour plot of the equivalent index. For the passband region at the mirror front, the contour lines are essentially vertical lines with a value close to 1.5 over a broad wavelength range. This demonstrates again the almost perfect matching to the refractive index of the ambient medium. The equivalent thickness plotted in Fig. 3.13d shows the same qualitative behavior as the equivalent thickness in the first example (see, Fig. 3.8d). However, one deviation between the equivalent-layer parameters of Figs. 3.8 and 3.13 is very obvious. For short wavelengths at the mirror front the dashed contour lines, which surround the stop-band region, are narrower, indicating the reduction of the high-reflection bandwidth for short wavelengths (compare Figs. 3.7 and 3.12).

3.5 BASIC mirror design

3.5.1 Substrate properties

The theoretical findings of the previous sections now offer a convenient method to avoid dispersion oscillations of chirped mirrors by proper design. Choosing an index of refraction of the ambient medium identical to the index of one of the coating materials, perfect impedance matching at this interface is achieved by double-chirping of the chirped mirror structure. Practically, this means inversion of the layer sequence and coating the layers on the back side of a substrate. Consequently, the light beam passes through the substrate before reflection by the chirped mirror coating. Therefore, this method is termed back-side coated (BASIC) chirped mirrors.

It might appear difficult to choose a substrate material with exactly matching index because the refractive index of sputtered or evaporated materials may significantly deviate from the refractive index of bulk materials. A refractive index $n_{\text{SiO}_2} \approx 1.49$, e.g., for sputtered silica compares to a bulk value $n_{\text{FS}} \approx 1.45$ at 800 nm. Such an index discontinuity at the interface between coating and substrate, however, merely causes a reflection of $2 \times 10^{-4}$ at normal incidence. A further reduction is achieved by suitable choice of an optical glass with a refractive index closer to sputtered SiO$_2$ (e.g. BK7 $n_{\text{BK7}} \approx 1.51$ @ 800 nm). In any case, the problem of residual mismatch between substrate and coating index is far less stringent than the problem of matching to air. A residual mismatch of the order discussed above is easily resolved by numerical optimization of the layer sequence.
One might argue that the BASIC approach only transfers the impedance-matching problem to the opposite interface of the substrate. In fact, if plane-parallel substrates were used, interference with the reflection from the interface to air gives rise to pronounced satellite pulses. With the high reflector from the back and the partial Fresnel reflection from the top, such a mirror would form a GTI, causing a strong spectral variation of the mirror phase. However, these detrimental interference effects are suitably suppressed by a geometrical mismatch of the two surfaces and will be discussed in the Section 3.6.

### 3.5.2 Practical design examples

For BASIC chirped mirrors, one has to design two independent coatings, an AR coating for the top surface and the chirped mirror coating for the back surface. An 18-layer AR coating and a 60-layer BASIC coating is used to cover the gain bandwidth of Ti:sapphire (650 – 1100 nm) with both high net reflectance and smooth dispersion properties. The AR coating is designed using a commercial software [44]. Figure 3.14a depicts the residual reflectivity of about $10^{-3}$ for the given wavelength range. Note that this coating also provides high transmission for the pump wavelengths 488 and 514 nm (argon-ion laser). Figure 3.14b shows the resulting GDD of the BASIC AR coating upon transmission with nearly negligible dispersion over the entire gain bandwidth of Ti:sapphire.

The 60-layer chirped mirror coating on the back surface is designed according to [23] and is subsequently computer-optimized with a local gradient algorithm [44]. The design goals are a high reflectance and a smooth GDD over the 450-nm wavelength range of the AR coating. Additionally, the mirror has to be highly transparent for the pump laser. The mirror is designed for p-polarized light with an incidence angle of 5° in air. This corresponds to an incidence angle of $\approx 3.44°$ in the fused silica substrate. Figure 3.15 depicts the spectral response characteristics of the resulting BASIC DCM structure, calculated for fused silica as the incident medium. The chirped mirror structure provides a
99.8% reflectance from 610-1000 nm (190 THz bandwidth). Accounting for the additional 0.2% losses due to the double pass through the AR coating, this results in a net reflectivity of 99.6% over the same 400-nm bandwidth. The wide reflectance range favorably matches the unprecedented bandwidth of extremely smooth dispersion of 220 THz.

Fig. 3.15a: Calculated reflectance of a BASIC double-chirped mirror structure. The structure is designed for use in an ultrashort-pulse Ti:sapphire laser. In a computer optimization process, a highly transmissive window for the pump laser is introduced. Measured refractive index data for the coating materials are used. In these calculations, the influence of AR coating and substrate on spectral properties is not considered. The reference plane is chosen in the substrate close to the coating, isolating the effect of the DCM coating. The reflectance is plotted on two different scales to show the high reflectance in the 620 to 1050 nm range and the transmissive region at 500 nm. 

For an even more extreme example for the possible performance of the BASIC design concept, the bandwidth of a design was extended to one optical octave, reaching from 600 nm to 1240 nm (Fig. 3.16). The design exhibits simultaneous small dispersion oscillations and a high reflectivity over this ultrabroad bandwidth. It needs to be admitted, however, that such a design requires deposition accuracies of 1Å or below and is currently out of reach even using the most sophisticated deposition machines. Current state-of-the-art deposition errors would increase the dispersion oscillations from <10 fs$^2$ to the order of 100 fs$^2$, which would render such a mirror relatively useless for intracavity application in a Ti:sapphire laser. It needs to be pointed out that broadband mirrors based on a different technology but with similar large errors have already been used in sub-10-fs lasers. Nevertheless, stepping back to coverage of slightly more than the Ti:sapphire gain bandwidth, the design shown in Fig. 3.15 was used.
3. Chirped mirrors

**Fig. 3.16:** Design study of an octave-spanning BASIC DCM design. Shown are the group delay dispersion (left axis) and the reflectivity vs. wavelength. Note that this design covers the range from 600 nm to 1240 nm with more than 99.7% reflectivity and simultaneously provides dispersion compensation with as little as 3.8 fs\(^2\) rms dispersion oscillations. This is equivalent to 0.3 fs rms oscillation of the group delay. The useful bandwidth of this coating is 260 THz.

3.5.3 *Comparison to standard DCMs*

Comparison of the structure of a BASIC mirror and a standard DCM readily reveals three identical functional sections but in a different order: a substrate, a chirped mirror structure, and an AR coating (Fig. 3.2). In case of the BASIC mirror, however, the two coating sections are non-interfering, separated by the substrate in between. The non-interference of the two coating sections decouples their design, and imperfections of the AR sections do not spoil the dispersion properties of the mirror. Figure 3.14a illustrates this by comparing the reflectance of the AR coating used in the BASIC design approach with similar 14-layer coatings used in the design of previous standard DCMs [39]. Due to the high sensitivity of the DCM dispersion towards residual reflection from the AR section, the residual reflectivity has to be kept at 10\(^{-4}\) or below, which limits the bandwidth of such a coating to about 250 nm at 800 nm center wavelength. Given the layer materials and the 450-nm bandwidth of this design, the residual reflectivity generally saturates at about 10\(^{-3}\) with only marginal improvement from an increased number of layers [45]. Consequently, any attempt to further increase the bandwidth of standard DCM coatings causes a dramatic increase of dispersion oscillations.

In contrast, the BASIC approach tolerates a residual reflectance of 10\(^{-3}\) for use inside a Ti:sapphire laser, and this can be immediately traded for 200 nm of extra bandwidth. The BASIC design technique is mainly limited by the achievable net reflectivity rather than by the magnitude of dispersion oscillations as in conventional DCM designs. The insensitivity towards dispersion oscillation is illustrated in Fig. 3.17, comparing the dispersion properties of a BASIC DCM and a conventional DCM. Both coatings shown use 60 layers, of which 14 are used in the AR section of the standard DCM. This example clearly shows that the BASIC design approach reduces dispersion oscillations to a peak-
to-peak value of 5 fs$^2$ (2 fs$^2$ rms), whereas the standard DCM of significantly less bandwidth exhibits peak-to-peak oscillations of 50 fs$^2$ (15 fs$^2$ rms).

Some additional advantages of the BASIC approach should be pointed out. With the weak remaining impedance-matching problem, computer optimization of the coating structure is less demanding than in the conventional DCM design approach. Using the same number of layers in a BASIC mirror, a wider wavelength range can be covered because no layers have to be sacrificed for impedance matching to air. Finally, the AR coating of a BASIC mirror may be grown independently, using materials with higher index contrast to further increase the net reflectance.

The design in Fig. 3.15 may serve as an illustration how to achieve optimum performance given the bandwidth and other constraints of a mode-locked Ti:sapphire laser. Giving up the highly transmissive region around 500 nm used for pumping the laser allows for an additional reduction of dispersion oscillations in the design. A further extension of the dispersion-compensation bandwidth, e.g., for the compression of white-light supercontinua, is possible by allowing for a reduced net reflectance or using more layers. For applications outside a laser cavity, slightly increased losses typically pose no problem. In principle, support of single-cycle spectra with BASIC chirped mirrors is feasible in external compression schemes.

![Dispersion oscillations of BASIC double-chirped mirrors (solid line) and standard DCMs (dashed line). Data from Fig. 3.15 and [24, 46] has been used for this comparison. Shown are the differences between desired and designed values of group delay (a) and GDD (b).](image)
3.6 Mechanical and optical design of BASIC mirrors

So far design issues of the BASIC coating have been discussed without paying too much attention on how to construct such a mirror. From the discussion carried out so far, it is clear that very thin substrates are needed, which steer the unwanted reflection from the top surface into a direction that does not interfere with the main reflection off the back face of the mirror. The substrate material has to be chosen equal to one of the materials used in depositing the layer sequence. With the commonly used materials SiO$_2$ a TiO$_2$, the use of fused silica as the substrate material is straightforward. However, even when substrate and layer material are chosen chemically identical, this does not mean that the respective indices are also identical. Sputtered materials exhibit a measurable increase of the index of refraction compared to chemically identical bulk substrate materials. This may actually be compensated for by use of a higher-index glass for the substrate, e.g. BK7. In any case are the observed index differences so small ($\Delta n<0.1$) that the resulting dispersion oscillations can be decreased to a negligible level in the subsequent computer optimization. This is to be compared with the situation of matching to air with its one order of magnitude larger ($\Delta n=0.5$) index mismatch.

The geometrical mismatch to prevent reflections off the front surface to interfere with those from the back surface essentially only requires that both surfaces be not parallel [47]. One convenient way to achieve this is a wedge-shaped substrate [47, 48], the other employing thin-lens substrates [27, 47], where both surfaces do not share the same center of curvature (see Fig. 3.18). Residual interference is calculated from the overlap of the reflections in the far field. For typical applications in a laser cavity with beam divergences of a few mrad, wedge angles on the order of one degree readily reduce interference to a negligible amount for a plane mirror. To replace the concave focusing mirrors in a typical laser cavity, thin plano-convex lens substrates may be used to diminish interference effects.

![Fig. 3.18: Practical construction of a BASIC mirror [47]](image-url)
The geometrical mismatch of the substrate surfaces then allows for suppression of detrimental interference effects by any given degree, but the power losses caused by the Fresnel reflection at the top surface appear prohibitive for use inside a laser cavity. Of course, an AR coating on the front surface of the BASIC mirror readily reduces this problem. Additionally, an AR coating further suppresses dispersion oscillations caused by a small residual overlap of the beams in the far field and allows for a smaller geometrical mismatch of the two surfaces. Hence, even though it is not strictly required, the AR coating also improves the dispersion properties of the BASIC mirror.

The total dispersion of a BASIC mirror is given by the dispersion of the chirped mirror structure plus twice the dispersion of substrate and AR coating upon transmission. Typical chirped mirror coatings for use in Ti:sapphire lasers allow for the compensation of material dispersion equivalent to a path length of about 2 mm fused silica per bounce \[49\]. Therefore, it is desirable to use substrates of a few 100 µm thickness to permit the generation of a net negative GDD with a BASIC mirror. On the other hand, however, multilayer coatings of 60 or more layers are known to generate tensile or compressive stress on to the substrate, causing a deformation of extremely thin substrates. In the experiments a substrate thickness of about 0.5 mm was found a viable compromise between surface deformation and tolerable material path.

Plano-convex 0.45-mm center thickness lens substrates were used to replace the 10-cm radius concave focusing mirrors previously used in the laser. The convex side of the lens substrates has a radius of curvature of 150 mm (focal length \(f = R / 2n \approx 5 \text{ cm}\)) and is coated with the BASIC chirped mirror structure. The AR coating is applied to the plane surface. During polishing, the thin lens substrates are optically contacted to plane carrier substrates. With the support of the carrier substrates, surface deformations of the substrates can be kept below \(\lambda / 5\) (peak to valley values over 1 cm diameter) in the manufacturing process. For the coating process, the substrates are removed from the carrier. With the chirped mirror coating applied, the thin lens substrates are finally cemented into mating concave 10-mm thick carrier substrates, similar to the construction of an achromatic lens. Cementing to a carrier substrate prevents mechanical stress to the thin mirrors and also protects the chirped-mirror coatings from environmental influences. The internal stress of the coating is decreased by the reduced substrate curvature. Since the BASIC DCM is buried behind the lens, the front surface can be cleaned with no danger of mechanical damage to the chirped mirror coating. The final structure consisting of the thin BASIC DCM lens and the thick glass support forms a mechanically rugged slab with two flat surfaces. The back side of the thick supporting glass substrate can be additionally AR-coated to improve transmission of the pump light.

Using optical flats, it is expected that even thinner substrates on the order of 100 µm can be used. The BASIC coating can first be deposited on a thicker substrate. This substrate can be thinned after cementing or optically contacting the BASIC substrate on to a carrier. On the top surface, finally the AR coating can be deposited. The experiments with cementing coated substrates on carriers indicated that forces in the polishing procedure can be kept small enough to prevent the coating from being sheared off the
substrate. This type of mirror would be extremely interesting for extra cavity applications, e.g., compression of white-light continua.

Apart from use inside a laser cavity, which requires some tricks in the mechanical construction of the mirrors, there are applications where back side coating comes quite naturally. Some examples are collected in Fig. 3.19. Prism sequences in combination with the BASIC design approach can deliver adjustable dispersion. One may also use this design approach for ultracompact femtosecond lasers, as the dispersion compensation can be implemented by BASIC coatings right on the laser crystals. This shows the great versatility of the new chirped mirror design approach, which allows for many new applications that were impossible with previous generation of dispersion compensating devices.

**Fig. 3.19:** Advanced variants of a BASIC mirror [47].
3.7 Experimental results

3.7.1 Back-side coated chirped mirrors

The design presented in the previous section was grown using high-precision ion-beam sputtering [50]. Active control of layer deposition in the few-Ångstrom range [51] is indispensable because of the high sensitivity of the GDD on deposition errors [22, 23].

The dispersion of the manufactured mirrors is characterized with white-light interferometry. Figure 3.20a shows the measured GDD of a BASIC chirped mirror in comparison with the target GDD of the design. Note that these curves now include twice the GDD of a 0.45-mm substrate and twice the GDD of the AR coating upon transmission. An excellent agreement between designed and manufactured dispersion is observed, with small residual dispersion oscillations up to a wavelength of about 800 nm. For longer wavelengths, which penetrate deeper into the mirror structure, deposition errors become noticeable and the GDD oscillation amplitude exceeds the designed value. But still, up to a wavelength of about 950 nm the resulting dispersion oscillations are of the same magnitude as in previously grown DCMs, which are shown for comparison in Fig. 3.20b. Compared to standard DCMs, the BASIC mirrors again exhibit much smaller dispersion oscillations at longer wavelengths up to 1050 nm. In summary, the manufactured BASIC mirrors exhibit a similar magnitude of dispersion oscillations but cover a 100 nm wider bandwidth.

Simulations (Fig. 3.20c) indicate an rms layer growth accuracy of $\approx 2$ Å, for both the standard DCMs and the BASIC mirrors manufactured. Although this is an excellent value, it is still necessary to combine mirrors to compensate for manufacturing defects. A further reduction of manufacturing errors to about $\approx 1$ Å per layer appears necessary to use BASIC mirrors of similar bandwidth directly without compensation for manufacturing errors and to fully exploit their potential in an oscillator.
Fig. 3.20a: Desired round-trip GDD (single pass through fused silica prism sequence with 50 cm apex separation and 2.3 mm of Ti:sapphire) used as a target function for the mirror design (dotted line). The measured GDD of the BASIC chirped mirror is shown as dots. Note the net positive dispersion of the BASIC mirror at 900 nm. In comparison with Figs. 3.15 and 3.16, dispersion of the AR coating (Fig. 3.14 b) and the substrate with a measured thickness of 0.45 mm are now included. In b the differences between designed and manufactured GDD are shown (BASIC chirped mirror, solid line; standard DCM [24], dashed line). The standard DCM exhibits similar dispersion oscillations, which are, however, shifted such that they partially cancel out dispersion oscillations of the BASIC mirror. A combination of one standard DCM and two BASIC mirrors yields smooth dispersion compensation from 650 nm to 950 nm, shown as the solid line in a. The magnitude of the dispersion oscillations of the BASIC mirror coating agrees with numerical simulations, indicating a 2-Å (rms) layer deposition accuracy (c).
3.7.2 Ultrashort-pulse Ti:sapphire laser

In initial attempts to use three BASIC mirrors in a Ti:sapphire laser, the mode-locked spectrum could not be pushed beyond the positive net dispersion region at 900 nm. Previously, similar problems could be solved by combining standard DCMs mirrors from one coating run under different angles of incidence [24] (compare Section 2 and Fig. 3.3). Increasing the angle generally allows shifting the dispersion oscillations of a particular mirror towards shorter wavelengths. As flat BASIC mirror were not available for that purpose, a combination of two BASIC chirped mirrors and one standard DCM was used instead to reduce the dispersion oscillations. The standard DCM is identical to the mirrors used in [24]. All mirrors are now used at near-normal incidence. The net dispersion of the mirror combination is shown in Fig. 3.21a. Note how dispersion oscillations of the individual mirrors cancel out for the wavelength range from 780 nm to 970 nm (Fig. 3.21b). Together, two BASICS and one standard DCM yield a relatively smooth net dispersion, ranging from 650 nm to 970 nm with maximum dispersion oscillations of 80 fs² (peak-to-peak value, for a single pass through the cavity). The described mirror combination is used in a Kerr-lens mode-locked Ti:sapphire laser otherwise similar to [24] and as described in the previous Chapter.

![Graph](image-url)

**Fig. 3.21:** SPIDER measurement of pulses from a Kerr-lens mode-locked Ti:sapphire laser employing two BASIC chirped mirrors (see Figs. 3.15, 3.16, 3.20). a) Measured spectral power density (solid line) and phase (dashed line). Assuming a flat phase, the transform-limited duration of the pulse is 5.6 fs. The corresponding intracavity spectrum exhibits a transform limit of 6.7 fs. b) Reconstructed temporal profile of the pulse with a pulse duration (full width at half maximum) of 5.8 fs.
Compared to the set-up described earlier, two of the focusing mirrors inside the cavity are replaced by BASIC chirped mirrors and the flat folding mirror is taken out. To balance the average negative GDD in both resonator arms, the standard DCM with stronger negative GDD is placed in the resonator arm with the semiconductor saturable-absorber mirror (SESAM). The fused silica prism pair (50 cm apex separation) introduces negative GDD in the other resonator arm that contains the OC mirror. Compared to [24] a slightly shifted output coupling mirror design is used to optimize spectral shaping (comp. Section 2.3). Standard DCMs are also used for the external compression scheme. The pulses are fully characterized by spectral phase interferometry for direct electric-field reconstruction (SPIDER [52], comp. Section 4.3). Spectral intensity and phase and the resulting pulse profile in the time domain are shown in Fig. 3.21. A pulse duration of 5.8 fs is measured. These measurements indicate that the pulse duration is very close to the bandwidth limit of 5.6 fs and significantly lower than the intracavity transform limit of 6.7 fs. Taking into account that there is still one conventional DCM in the cavity, these results agree favorably with the 5.9 fs achieved with only conventional DCMs. The results achieved with the BASIC mirrors are within 3% of the bandwidth limit, resulting in a nearly transform-limited pulse. In a slightly different configuration, very clean pulses of about 7 fs duration were obtained. The latter pulses have strongly reduced satellites with less than 5 % of the main pulse and exhibit a nearly bell-shaped spectrum. This configuration made use of a broadband OC mirror of 5.5% transmission, surrendering spectral shaping upon output coupling [53] (comp. Section 2.3.1). In summary, the results obtained with the BASIC mirrors clearly demonstrate the potential of the new mirror design strategy. The experiments show that given the current state of the art of layer deposition accuracy and substrate manufacturing the BASIC design approach can be successfully used in a sub-6-fs laser. With a similar dispersion oscillation amplitude, the bandwidth of the BASIC mirrors is clearly enhanced compared to earlier designs.

### 3.8 Conclusions

The matching problem of chirped mirror structures to the ambient medium was reviewed. The analytical findings and numerical investigations show that the double-chirped mirror technique provides an optimum solution to the impedance-matching problem if an ambient medium with index identical to one of the layer materials is chosen. The resulting design approach of back-side coated (BASIC) chirped mirrors allows for ultrabroadband dispersion compensation with virtually no dispersion oscillations. A comparison with standard DCMs clearly demonstrates the superiority of the BASIC design approach. Over a bandwidth of 220 THz (610 - 1100 nm), GDD oscillations of 2 fs² (rms) can be achieved, which presents an order-of-magnitude improvement compared to earlier designs of less bandwidth. For an even wider bandwidth of 260 THz, which is sufficient for coverage of an optical octave, dispersion oscillations of less than 4 fs² magnitude were achieved in the mirror design. BASIC chirped mirrors were demonstrated inside a Kerr-lens mode-locked Ti:sapphire laser with near-bandwidth-limited 5.8-fs pulses. Despite the clearly extended bandwidth of the manufactured mirrors, deposition errors of BASIC chirped mirrors are still a limiting factor for the further reduction of dispersion oscillations. With expected improvements of fabrication accuracy, the BASIC technique will allow for chirped-mirror structures with negligibly small dispersion oscillations.
oscillations. Besides use inside an oscillator, BASIC mirrors appear very attractive for external white-light continuum compression schemes [54, 55] and optical parametric amplifiers [56, 57, 58], which can greatly benefit from the extended bandwidth. BASIC mirrors promise an improvement of dispersion control for octave-exceeding pulse spectra, which is a very important prerequisite to push pulse durations further into the single-cycle regime.

3.9 References to Section 3

3. Chirped mirrors

4. Methods for a full characterization of few-cycle laser pulses

With the generation of shorter and shorter laser pulses, characterization of these pulses becomes an increasingly complex problem. The traditional approach to characterizing short pulses is autocorrelation. This method has the disadvantage that it actually provides only very limited information on the pulse shape and it also only allows a rough estimate on the actual pulse duration. In principle, it is impossible to reconstruct the pulse shape from an autocorrelation measurement without additional information. For reasons that have become clear in the previous chapter, sub-10-fs laser sources generally display a strong spectral modulation, which in turn translates into a complex temporal pulse shape. Any attempt to guess the right pulse shape for deconvolution in the sub-10-fs regime, as has been the standard method with longer pulses for years, is therefore highly questionable. To illustrate this, several measurements of autocorrelations of completely different sources of sub-10-fs pulses have been compiled in the left part of Fig. 4.1. Despite the suspected difference in pulse shape and duration, the autocorrelation traces look very similar and only reveal the differences after extremely careful inspection.

![Fig. 4.1 left: Interferometric autocorrelation traces measured at different sources of sub-10-fs lasers. Bottom trace: compression of cavity-dumped pulses in a silica fiber [4], second trace from bottom: optical parametric amplification [5], third trace from bottom: pulses from a Ti:sapphire oscillator ([6]) and top trace: compression of μJ pulses in a hollow fiber filled with Krypton [7]. Dots refer to measured data; lines indicate the fit that was used to estimate pulse duration from the autocorrelation. Right: Frequency-doubled pulses from a Ti:sapphire laser (bottom trace, [8]) and fully characterized pulses from a non-collinear optical parametric amplifier (middle trace, [3]) in comparison with pulses directly generated in a Ti:sapphire laser (top trace, [9]). The frequency-doubled pulses have been characterized with a decorrelation method (comp. Section 4.1), the other results have been measured with the SPIDER technique (Section 4.3).](image-url)
This fundamental problem is long known, and many approaches to overcome this limitation have been described in literature. These methods are typically referred to as full characterization methods, as they provide amplitude and phase of the pulse. The most successful of these methods to date was frequency-resolved optical gating (FROG). The main idea behind this method is to utilize additional information from spectrally resolving the autocorrelation and then iteratively fit a pulse shape to the measured data. FROG is a very successful and widespread method, however, it has some intrinsic weaknesses, which particularly come out in the sub-10-fs regime. In the following, an approach is described, how some of these disturbing problems can be removed [10]. A quick look at Fig. 4.1 shows up the advantage of full characterization methods. Clearly, pulse shapes of different sources can be easily distinguished and compared, whereas the comparison of the autocorrelation traces only gives very diffuse information on the pulse shapes behind the measurements.

It needs to be pointed out, that this work has not the goal of promoting one or the other pulse characterization technique but to collect the most accurate and concise information on the pulses. All described techniques have specific advantages and disadvantages, and it strongly depends on pulse durations and pulse energies, which one of them is to be preferred. There are instances, where even sophisticated variants of the correlation techniques have specific advantages. This is when information on the pulses is to be collected very rapidly and pulse energies are very low. In the following, examples will be presented, where a full characterization provides insight into limiting physical mechanisms that cannot be provided by a single number as the pulse duration alone. While autocorrelation only indicates that the pulse is too long, FROG and SPIDER also give a clue on why the pulse is too long.

In the course of this work, standards in pulses characterization have heightened considerably. Before, autocorrelation was still customary and highly accepted. Nearly all recent record results, however, used one of the methods that will be described in much more detail below. The SPIDER method adapted for use at less than 10-fs pulse duration during this work [9, 11] has been copied by many other groups and been found an extremely helpful tool for pulse characterization. This method is about to define a new standard on characterization of ultrashort pulses.

4.1. Iterative reconstruction from temporal correlation and spectrum

Despite the existence of more advanced methods as FROG and SPIDER, correlation methods deserve a closer look. In this work, autocorrelation data serves as an additional crosscheck for comparing the results of either of the advanced methods with the traditional autocorrelation measurement. Moreover, attempts were made to collect as much information as possible from the autocorrelation and to reconstruct the pulse shape with additional information from spectral measurements. In the following, describe these methods will be described. However, one has to be extremely careful in interpreting the results of such decorrelation procedures. There exist experimental situations, where FROG or SPIDER measurements are difficult to perform and a correlation measurement is much simpler but less reliable. One such situation is low light levels. Reconstruction
from cross-correlation and spectrum was used in the experiments on second-harmonic generation of sub-10-fs pulses described in Chapter 6.

Autocorrelation [12, 13, 14] characterizes a laser via autocorrelation by a nonlinear interaction of overlapping replicas of a pulse. Thus, one replica of the input pulse is multiplied with a second replica and the resulting signal is recorded as a function of the delay between the two replica pulses. Technically, the multiplication of the two optical signals is done using a nonlinear optical effect such as second-harmonic generation or two-photon absorption. As a result, the autocorrelation trace

\[ AC(\delta t) = \int_{-\infty}^{\infty} I(t)I(t - \delta t) dt \]  

is measured, where \( I(t) = E(t)E^*(t) \) is the optical intensity. This type of autocorrelation is called background-free, as it will measure zero signal for large delays \( \delta t \to \pm \infty \). A background-free autocorrelator uses a noncollinear beam-geometry in such a way that second-harmonic generation requires one photon from each of the two beams while SHG from each individual beam is not detected. This background-free set-up allows for large dynamic ranges, but is typically not the preferred set-up in the sub-10-fs regime. In a noncollinear set-up, an additional problem occurs due to a reduction of temporal resolution caused by the finite crossing angle of the two beams [15]. This is a problem also for FROG measurements and will be treated in Chapter 4.2. Therefore a collinear set-up is preferred. This then yields the interferometric autocorrelation trace [16]:

\[ IAC(\delta t) = \int_{-\infty}^{\infty} [E(t) + E(t - \delta t)]^2 dt \]  

Unfortunately, no way exists to retrieve the original pulse profile from any measured autocorrelation traces without additional knowledge. Inspired by a theoretical description of the mode-locking process one can sometimes assume a certain pulse shape, and then retrieval of the pulse duration is simple. This is not an option in the sub-10-fs regime with its complex pulse shapes. In this regime simple analytical functions must not be assumed anymore for decorrelation of the measured autocorrelation function. Additionally, the sub-10-fs regime is very demanding, and pulse shaping by spectral filtering or dispersion in the beam splitters and nonlinear crystal has to be kept at a minimum. Wherever possible, this regime calls for the use of metal-coated reflective optics.

Several methods have been discussed to solve the problem of decorrelation [4, 17]. These methods only use the DC part of the IAC trace (4.2) and discard the modulation index of the interference. These methods require additional experimental information, which in the simplest form can be provided by a simultaneous measurement of the power spectrum of the laser. Decorrelation methods employ a computer optimization strategy to find a simultaneous fit to measured spectrum and autocorrelation. This removes the arbitrariness of assuming a particular pulse shape for pulse retrieval, but requires data with excellent signal-to-noise ratio for reliable operation. This is a particular problem in the sub-10-fs range, where high-dynamic-range autocorrelation methods cannot be employed without sacrificing temporal resolution.
An example for these methods is shown in Fig. 4.2, where the method described in [17] was applied to a sub-6-fs Ti:sapphire laser [1, 18]. In this case, the crude assumption of an ideal sech$^2$ to the IAC data yielded a pulse duration of 4.8 fs. While this is incompatible with the spectrum measured, it should be clearly pointed out that the IAC method cannot be held responsible for this underestimation. Rather it is the unmotivated use of a particular pulse shape. Assumption of a sinc$^2$ pulse shape, which would correspond to a box-shaped spectrum, gives a more conservative estimate of a 5.7-fs pulse duration. Using the decorrelation procedure obliterates the a priori assumption of a particular pulse shape [4]. For the data presented here, this algorithm extracts a 5.7-fs pulse duration (see Fig. 4.2), which was later confirmed with SPIDER measurements (see Chapter 4.3). As the algorithm uses only the unmodulated DC part of the IAC data, a comparison of reconstructed and measured IAC provides an independent verification of the method.

Similar algorithms can be used for decorrelation of cross-correlation measurements. In this case, however, significantly more information is available, when one of the pulses in the cross-correlation can be characterized with either SPIDER or FROG [8]. Then only the spectral phase of the second pulse in the cross-correlation has to be determined. One of the algorithms used for decorrelating cross-correlation measurements of frequency-doubled and fundamental Ti:sapphire laser pulses, as employed in the experiments on quasi-phase matched pulse compression described in Chapter 6, is depicted in Fig. 4.3. This algorithm employs the spectral phase and amplitude of the reference pulse, the spectrum of the pulse to be characterized, and the cross-correlation measurement itself as input data. The reference pulse was characterized using the SPIDER technique (comp. Chapter 4.3). The phase of the unknown pulse is parameterized in the spectral domain.
with 128 individual data points. The spectral phase is initialized with random noise. The corresponding cross-correlation is then computed and compared with the measured data. The optimization procedure employs the high-dimensional downhill simplex algorithm [19] and the mean square deviation as the error metric. Once the optimization stagnates, the error metric is switched from mean square deviation to the mean absolute deviation. In the presence of experimental noise, this strategy was found to yield a slightly improved agreement in the wings of the crosscorrelation compared to the mean square optimization alone, without sacrificing agreement around zero delay.

For a double-check on the convergence of the algorithm, additionally the same procedure was applied to a reduced input data set, consisting only of the spectral amplitudes of both, the reference pulse and the pulse to be characterized, and the crosscorrelation trace. In this method the multidimensional optimization is applied to the FH and the SH spectral phase simultaneously. With the reduced amount of input data, the algorithm is expected to be much more susceptible for stagnation or to yield incorrect results. In fact, this behavior was observed in several tests using synthetic input data. Best convergence was found in the presence of moderate noise on the input data and with a less than a factor of two difference in pulse duration between the FH and the SH. For the experimental data presented in Section 6.4, both algorithms delivered consistent results and did not depend on the starting parameters.

**Fig. 4.3:** Algorithm for the retrieval of the SH pulse from the measured crosscorrelation.
4.2. Collinear type-II frequency-resolved optical gating (FROG)

Frequency-resolved optical gating (FROG) is a characterization method based on the measurement of a spectrally resolved autocorrelation signal followed by an iterative phase-retrieval algorithm to extract the intensity and phase of the laser pulse. FROG has been used for pulse durations from few femtoseconds to several picoseconds and for pulse energies ranging from the nJ- to the mJ-regime. For a more general overview of the FROG technique, the reader is referred to Refs. [20, 21]. The nJ pulse energies of the lasers introduced in Section 2 dictate the use second-harmonic generation FROG (SHG-FROG, [21]). Consequently, the discussion will focus on this FROG variant.

In the sub-10-fs range, two serious problems with the FROG technique arise. The first is bandwidth limitation of the optics and the detection system involved, in particular the bandwidth limitation of the SHG process. Using extremely thin nonlinear optical crystals reduces this problem. In contrast to autocorrelation, FROG allows to correct for bandwidth limitations to a certain extent. With this correction, measurements of sub-4-fs pulses have been successfully demonstrated in the visible spectral range [15, 22]. Still, particular care has to be given to the accurate determination of the spectral calibration of the setup.

A second more fundamental limitation is the reduction of temporal resolution caused by the finite beam-crossing angle in the nonlinear crystal. Unlike sub-10-fs autocorrelators, a noncollinear geometry is conventionally used for FROG measurements. In the noncollinear geometry, temporal resolution has to be traded for suppression of interference fringes. Assuming Gaussian spatial and temporal profiles, ideal phase matching, and small crossing angles \( \theta_0 \) the temporal resolution \( \delta t \) is given by

\[
\delta t = \theta_0 \frac{w_0}{c}
\]

(4.3)

where \( w_0 \) is the beam radius in the focal plane and \( c \) the speed of light [15, 23]. Now, \( \theta_0 \) has to be chosen according to the required suppression of interferences in the recorded FROG trace. The apparent duration \( \tau_M \) retrieved from a distorted FROG trace is related to the effective pulse duration \( \tau_p \) by

\[
\tau_M^2 = \tau_p^2 + \delta t^2.
\]

(4.4)

To overcome limitations due to geometrical blurring and to simplify the set-up of a sub-10-fs FROG apparatus, a collinear geometry can be employed if one uses type-II phase matching [10]. This method has been first demonstrated for the measurement of 20-fs pulses in the focus of a high numerical aperture objective [24]. The latter work was targeted mainly at exploring geometric distortions of the pulse shape under strong focusing conditions and did not optimize their set-up for ultrabroadband operation.
In addition to the usual bandwidth requirements, particular care has to be taken to avoid polarization-dependent pulse shaping in a type-II setup. Otherwise an asymmetry of the measured FROG-traces will occur. To avoid polarization dependence of the beam splitting, we use near-normal 4° incidence rather than the 45° conventionally used (see Fig. 4.4). This scheme allows for a polarization-independent bandwidth of more than 400 nm. Asymmetry of the FROG trace can also be caused by group-velocity-mismatch in the nonlinear crystal. This can be easily avoided with a sufficiently thin nonlinear crystal, whose use is already dictated by the phase matching bandwidth requirements.

![Collinear type-II SHG-FROG setup](image)

**Fig 4.4:** Collinear type-II SHG-FROG setup: BS, beam splitters; PR, periscope for polarization rotation; HA, periscope for height adjustment; FM, focusing mirror (25.6 cm radius of curvature); SHG, nonlinear crystal (10 µm thick type-II ADP); OMA, optical multichannel analyzer. Dots and arrows on the beam path display the polarization state of the beam. The black box objects represent silver coated mirrors.

With the crystal thickness approaching the coherence length, non-phasematched SHG processes will produce non-negligible contributions to the observed signal. For a 10 µm BBO crystal (point group 3m) a significant contribution of second harmonic radiation is generated by the square nonlinearity tensor element $d_{31}$[25]. This light is polarized along the extraordinary axis as the type-II signal is. This gives rise to interference. To avoid these delay-dependent distortions of the FROG trace, a material with higher crystal symmetry is to be preferred. For maximum bandwidth, a 10 µm thin ADP crystal (point group: 42m) is used. Now only one undesired mixing process is contributing: The $d_{25}$ tensor element generates SHG radiation polarized along the ordinary axis. Because this light is polarized orthogonal to the FROG signal, it is easily rejected by a polarizer or subtracted as a constant background.

The ultimate limitation of temporal resolution in a type-II SHG-FROG arises from group-velocity-mismatch (GVM) in the nonlinear crystal (compare Section 6.1). In principle, GVM between the two orthogonally polarized fundamental pulses causes an effect similar to the geometrical blurring effect in type-I SHG-FROG. In a 10 µm ADP crystal, GVM limits the temporal resolution to 1.2 fs. This is either smaller or on the order of values published for type-I SHG-FROG [15, 23, 26].
4. Pulse Characterization Methods

Figure 4.4 depicts the collinear type-II SHG-FROG setup used. Note the use of all-reflective optics wherever possible and the dispersion balanced configuration of the beam-splitting scheme. Two identical dielectric beam splitters with 300 µm thickness are used. The beam is polarized better than 1:100 after the out-of-plane polarization rotation. Unwanted type-I radiation is suppressed by a broadband Glan-Thompson-polarizer. Additional filtering is required to suppress fundamental light. For this purpose, a 1 mm Schott BG3 color glass and four bounces on two broadband dielectric UV/VIS mirrors (New Focus) are employed. The spectra are recorded on a 0.3m optical multichannel analyzer equipped with a 300 gr/mm grating and a UV-enhanced 1024x128 pixel CCD camera. Wavelength dependent sensitivity of the detection system is measured using a calibrated white light source. The recorded FROG traces are corrected for spectral dependence of conversion and detection efficiency.

The ultrafast type-II FROG characterization method is demonstrated with pulses from the Kerr-lens modelocked Ti:sapphire laser described in the previous chapter [6]. These pulses have a Fourier limit of 5.3 fs. Measured and reconstructed FROG traces with a grid size of 128 x 128 points are shown in Fig. 4.5. The measured FROG trace do not exhibit any indications of excess GVM or polarization dependent pulse shaping, as favorably indicated by the symmetry of the measurement. Pulse reconstruction is performed using a commercial software (Femtosoft Technologies). The FROG error of this reconstruction amounts to 0.0078. The slight disagreement in the spectral wings of the traces near zero delay is caused by the delay-dependent second harmonic stray light. The FROG error is strongly reduced with slightly narrower input spectra. Some imperfections in the measured FROG trace can be attributed to the residual interference fringes in the unfiltered input data. Noise increases in the spectral wings due the reduced color glass filter transmission at these wavelengths.

Fig. 4.5: Measured and reconstructed FROG trace(logarithmic contour spacing, lowest contour at 0.7% of peak). For display purposes, residual fringes in the measured FROG trace have been removed by Fourier filtering. They are not strong enough to affect the reconstruction of the pulse.
In Figure 4.6, the reconstructed pulse profile and spectrum are shown. The width of the temporal intensity profile is 6.6 fs indicating uncompensated chirp. The divergence of the spectral phase below 700 nm and above 900 nm is caused by the phase properties of the output coupler. In the time domain, together with the modulated shape of the spectrum, this uncompensated high-order dispersion results in a pulse pedestal with less than 15% of the pulse peak power. The small sinusoidal oscillations observed in the spectral phase are explained by group delay oscillations of the double-chirped mirrors [27] used for external dispersion compensation.

![Figure 4.6: Reconstructed temporal intensity profile with a full width at half maximum of 6.6 fs (left), spectrum (right, solid), and spectral phase (right, dashed). Additionally, the independently measured spectrum is plotted on the right (dash-dotted). Intensity and phase are scaled on the left and right vertical axis, respectively.](image)

To supplement the internal consistency checks of the FROG method (Fig. 4.7, [20]), the results are compared with independent pulse characterization tools. Figure 4.8 shows a comparison of the reconstructed and the measured fundamental pulse spectra. The frequency-dependent mode-size introduces a significant transversal variation of the spectral structure. This is the reason for some of the discrepancies between the two spectra and some of the deviations in the spectral consistency check. Figure 4.8 depicts the excellent agreement of the interferometric autocorrelation (IAC) calculated from the FROG data with the measured IAC. The assumption of analytical pulse shapes usually leads to unsatisfying fits of the IAC and often to large errors in the estimated pulse duration. For example, a sech² fit to the IAC would yield a pulse duration of 4.5 fs, a duration below the transform-limit of the spectrum. Clearly, if the spectrum already indicates strong deviations from simple analytical shapes, deconvolution of the autocorrelation under an a priori assumption of such a particular pulse shape is not justified. This strongly confirms the need for accurate and easy-to-use characterization tools for sub-10-fs pulses.
4. Pulse Characterization Methods

Fig. 4.7: Internal consistency checks of FROG (marginals). Delay (left) and frequency marginal (right) of the FROG data in Fig. 4.5. The dashed lines are the marginals of the measured trace. The solid lines represent the DC part of the interferometric autocorrelation and the autoconvolution of the fundamental frequency-spectrum, respectively. Both have been obtained from independent measurements. The sharp modulation peaks in the delay marginal are due to residual fringes.

Fig. 4.8: Comparison of the independently measured interferometric autocorrelation (dots) with the IAC calculated from the reconstructed spectrum and phase (solid line).

At first sight, the type-II SHG-FROG geometry seems to introduce additional complications over type-I set-ups. However, suppression of polarization-dependent pulse shaping and group-velocity mismatch can be easily accomplished by proper optical design. In turn, geometrical blurring effects are completely avoided in a collinear technique. The temporal resolution can be calculated exclusively from nonlinear crystal properties. Moreover, the apparatus is very simple to align. Sub-fs resolution of a type-II SHG-FROG can be achieved using low-dispersion nonlinear optical materials with small GVM, such as KDP or ADP. Type-II SHG-FROG combines the simplicity of an interferometric autocorrelation with the phase-retrieving properties of standard FROG. This combination makes it a powerful and easy-to-use tool for the demanding characterization of few-cycle pulses.
4.3. Spectral phase interferometry for direct electric-field reconstruction (SPIDER)

Spectral phase interferometry for direct electric-field reconstruction (SPIDER) is a novel method that was first introduced by Walmsley and coworkers [28]. Other than correlation based techniques and FROG, SPIDER is based on spectral interferometry [29]. As will become clear below, nonlinear optical processes are only needed to generate a spectral shear on the data, otherwise the technique is linear and inherently avoids complications arising from optical correlation and numerical decorrelation procedures. The main step in this work is an extension of SPIDER to pulses shorter than 10 fs [9, 11].

The SPIDER apparatus is schematically explained in Fig. 4.9. First two replicas of the input pulse are generated with a fixed time delay \( \tau \). These two replicas are upconverted using sum-frequency generation (SFG) with a strongly chirped pulse that can be derived from the same input pulse. Given the strong stretching of the chirped pulse, the upconverter can be considered as a quasi cw wave in the following. Due to their temporal separation, the two pulses are upconverted with two quasi-cw slices of different wavelengths, leading to two identical versions of the input pulse that are frequency-shifted with respect to each other by a spectral shear \( \delta \omega \). This is shown in the top right corner of Fig. 4.9. Without the spectral shear, the temporal spacing of the two replicas would be independent of wavelength. Introduction of the spectral shear renders the temporal spacing a function of wavelength. Therefore, also the fringe spacing of the respective spectral interferogram becomes spectrally dependent. The resulting interferogram is recorded using a spectrometer (Fig. 4.9 and 4.10). SPIDER is a self-referencing interferometric technique, i.e., there is no need for a well-characterized reference. Note that no moving parts are required in a SPIDER apparatus and only a single interferogram \( S(\omega_c) \) has to be measured:

\[
S(\omega_c) = |E(\omega_c)|^2 + |E(\omega_c + \delta \omega)|^2 + 2|E(\omega_c)E(\omega_c + \delta \omega)|\cos\{\phi_{\omega_c}(\omega_c + \delta \omega) - \phi_{\omega_c}(\omega_c) + \omega_c \tau\}  
\]

Here \( E(\omega) \) is the frequency domain representation of the electric field and \( \phi_{\omega_c}(\omega) \) the spectral phase of the pulse. \( \omega_c \) is the passband frequency of the spectrometer. A fast non-iterative algorithm using two one-dimensional Fourier transforms extracts the phase of the oscillatory cosine term (Eq. (4.5)) of the interferogram. The linear phase term \( \omega_c \tau \) is separately measured by conventional spectral interferometry [29] and subtracted from the cosine-phase-term. This calibration measurement has to be done only once. While it can be done either with the fundamental pulse replicas or with their second harmonic, the latter is preferable because it uses the same wavelength range for calibration and measurement, and therefore avoids wavelength calibration errors. The spectral phase \( \phi(\omega) \) is obtained by adding up the appropriate phase differences \( \phi(\omega + \delta \omega) - \phi(\omega) \). Finally, the time-dependent intensity and phase are calculated from the reconstructed spectral phase and an independently measured fundamental pulse spectrum. The accuracy of
reconstruction of the spectral phase is quite insensitive to the phase-matching bandwidth of the upconversion crystal and the spectral responsitivity of the detector, since it depends only on the spacing of the spectral fringes. Therefore, even for ultrabroadband pulses no complicated spectral corrections are needed, in contrast to other techniques [15]. Furthermore, the non-iterative character of the evaluation procedure together with the absence of moving parts in the experimental set-up allows for pulse measurements at video refresh rates [30, 31].

Fig. 4.9: Spectral phase interferometry for direct electric-field reconstruction (SPIDER). Two replicas of the input pulse are mixed with a chirped pulse. The chirped pulse can be derived from the same laser. Sum frequency-generation yields two spectrally sheared replicas of the input pulse. As shown in the frequency-time representation of the pulse’s phase front, the temporal separation of the two sheared pulses is wavelength-dependent. This gives rise to a wavelength-dependent fringe spacing of the spectral interferogram, which is then used to reconstruct the spectral phase of the pulse.

Fig. 4.10: Concrete SPIDER set-up used in the experiments: GDD = SF10 glass block, PR = periscope for polarization rotation, BS = beam splitters, TS1 = translation stage to adjust delay \( \tau \), TS2 = translation stage for adjustment of temporal overlap of short pulse pair with stretched pulse, HA = periscope for height adjustment, FM = focusing mirror (30 cm radius of curvature), SFG = upconversion crystal (30 \( \mu \)m thick type-II BBO), OMA = optical multichannel analyzer. Black filled objects represent silver coated mirrors, dots and arrows on the beam path display the polarization state of the beam.
The SPIDER apparatus developed in this work is shown in Fig. 4.10 [9, 11]. Two pulse replicas with a delay of $\tau = 300$ fs are generated in a Michelson-type interferometer. The input pulse for the interferometer is the reflection from the surface of a 6.5 cm long SF10 glass block. The signal transmitted through this block is used as the strongly chirped pulse for upconversion. The group delay dispersion of the block ($10^4$ fs$^2$) together with the delay of 300 fs results in a spectral shear of 4.8 THz. This chirp is adequate to ensure that each pulse replica is upconverted with a quasi-cw field, and the spectral shear is small enough to satisfy the sampling theorem [28]. Therefore this configuration is suitable for measuring pulses up to 40 times the transform limit of our spectrum. The use of the low-cost uncoated glass block as the first beam splitter assures a large bandwidth and an appropriate power splitting ratio, allowing for an optimal upconversion signal. The Michelson interferometer uses two 300 µm thick dielectric beam splitters in a symmetric dispersion-balanced configuration. The symmetry allows for maximum fringe contrast. The two delayed pulses are mixed non-collinearly with the stretched pulse in a 30 µm thick type-II BBO crystal. The phase-matching bandwidth in the ordinary axis of a type-II crystal is usually much larger than in a type-I crystal of the same material and thickness, whereas the bandwidth in the extraordinary axis is smaller. SPIDER takes advantage of a type-II configuration since only a small part of the bandwidth of the stretched pulse, given by the spectral shear $\delta \omega$, is actually required. Consequently, the narrow-band e-axis of the crystal is oriented parallel to the polarization of the stretched pulse while the short replicas are rotated into the broadband ordinary axis using a periscope. In this configuration the upconversion efficiency in our crystal varies by less than 20% over the range from 660 nm to 1 µm (Note that using the other orientation would reduce this range to 750 nm to 880 nm). A non-collinear mixing geometry avoids the need for another beamsplitter. Geometric smearing effects caused by this non-collinear geometry and group velocity mismatch do not play a role, provided that the strongly chirped pulse can be considered as quasi-cw for the duration of the pulse to be measured. This condition is easily fulfilled.

The upconverted pulses are detected in a 0.3 m imaging spectrograph equipped with a 1200 gr/mm grating and a 1024 by 128 pixel UV enhanced CCD array allowing for rapid acquisition. Only the horizontal axis of the CCD output is needed to measure the SPIDER interferogram. To calibrate the apparatus, we measure the linear phase term arising from the delay between the replicas using a 10 µm type I KDP instead of the BBO. As an alternative one could simply rotate the BBO crystal by 45 degrees. The reduced bandwidth of this configuration causes significant shaping of the resulting interferogram but is still sufficient for accurate measurement of the linear phase. Because SPIDER reconstructs the phase information from the position of the fringes, the delay between the two short pulse replicas is chosen to obtain a large number of fringes while still being able to properly resolve the individual fringes with the spectrometer.
The described SPIDER set-up was first used to measure the amplitude and phase of sub-6-fs duration pulses from a state-of-the-art SESAM-assisted Kerr-lens modelocked Ti:sapphire laser [6]. Figure 4.11 shows the SPIDER interferogram of a pulse with a transform limit of 5.3 fs. For comparison, the individual spectra of the upconverted pulses are also displayed. Note that they are identical but shifted by the spectral shear $\delta \omega$. The spectral phase reconstructed from the SPIDER trace is plotted in Fig. 4.12, together with the independently measured pulse spectrum and the corresponding temporal intensity profile with a full width at half maximum (FWHM) of 5.9 fs. The oscillations in the central part of the phase are caused by the extracavity double-chirped mirrors [27], which are used to compensate for the dispersion of output coupler together with the higher order dispersion of the extracavity prism pair (compare Chapter 2). Note the correspondence between the phase oscillations and the shape of the spectrum, which is expected since similar double-chirped mirrors are used in the laser cavity itself. The global ‘S’-like shape of the phase stems from residual uncompensated phase of the output coupler. This clearly demonstrates that SPIDER can be used to directly reconstruct the complicated phase distortions experienced by ultrabroadband pulses.

**Fig. 4.11:** SPIDER trace of a sub-6-fs pulse (dotted). Additionally, the spectra of the individual upconverted pulses are shown (solid and dash-dotted line).

**Fig. 4.12:** Reconstructed temporal intensity profile (left) and spectral phase (right, dashed). The independently measured power spectrum of the pulse (right, solid) has a transform limit of 5.3 fs. The solid lines are referring to the left and the dashed line to the right vertical axis.
As an independent check for the accuracy of the method, we compare the interferometric autocorrelation (IAC) calculated from the SPIDER data with a separately measured IAC (Fig. 4.13). The agreement is excellent even for the low intensity structure in the wings of the IAC. The conventional but unjustified method of fitting a sech^2 pulse to the autocorrelation deceivingly yields a pulse duration of 4.5 fs. This systematic underestimation of the pulse duration affirms the need for complete characterization methods. The SPIDER measurement also confirms the analysis introduced in Section 4.1 [6], which resulted in a duration of 5.8 fs for pulses generated by this laser.

![Figure 4.13: Comparison of measured interferometric autocorrelation (IAC, dots) with the SPIDER-reconstructed IAC (solid line).](image)

It needs to be pointed out that the experiments demonstrated here did not exploit the full bandwidth of the SPIDER technique. In fact it is expected that SPIDER is applicable even to pulses with single-cycle duration. In that regard, an important advantage of SPIDER is the insensitivity towards spectral filtering, which sets it apart from autocorrelation-based techniques. SPIDER only extracts the fringe spacing from the spectral interferogram, but does not require the amplitude information of this trace. Thus, the nonlinear optical process does not need to be constant for the full pulse bandwidth but only needs to give enough signal to measure the spectrally dependent fringe period. For example a 30 µm thick BBO crystal can be used to measure pulse in the single cycle regime (see Fig 4.14). Therefore, it can be used to fully characterize ultrashort pulses with only a few femtoseconds duration. In the meantime, the SPIDER technique was already demonstrated with slightly shorter pulses of 5 fs duration, which is still far from exploiting the full potential of the method [32]. Moreover, SPIDER employs a non-collinear geometry without suffering from a reduction of the temporal resolution. This geometry also offers much better ways of suppressing unwanted fundamental light from detection, which can be a problem in the collinear autocorrelation of extremely broadband sources.

SPIDER is now an established method for the characterization and has been used for characterization of some of the shortest pulses available to date. The results of the SPIDER measurement show excellent agreement with those made by other methods. In contrast to those methods, however, SPIDER offers the unique advantage of providing complete spectral phase information with update times well below 1 s. This suggests the
possibility for its use as a complete and accurate online characterization tool for pulses down to the single-cycle range.

![Efficiency graph](image)

**Fig. 4.14:** Calculated conversion efficiency of type-II sum-frequency generation of a 30-µm thick BBO. The cut-angle was varied from 41.5° to 50.5°. The smallest angle is ideal for the Ti:sapphire wavelength range. The bandwidth of the SPIDER set-up can be extended to the entire visible and near-infrared range (50.5°). Note that the efficiency variation only reduces the dynamic range of the method, but does not influence the phase measurement. Intermediate cases are also shown (44.5° and 47.5°).

### 4.4. Cross-correlation SPIDER

For characterization of Ti:sapphire laser pulses, the self-referencing SPIDER technique is virtually ideal, as described in the previous chapter. It is a big advantage to use one and the same source of laser pulses to generate both, the upconverter pulse and the pulses to be characterized. This is not a necessary condition; it is only required that the sample pulses and the upconverter pulse are coherent in respect to each other. Therefore, fundamental and second harmonic of a pulse could be utilized, or pulses linked by a parametric generation process. Other than cross-correlation variants of FROG, the cross-correlation SPIDER does not require a well-characterized reference pulse. The method is useful, when the second harmonic of the laser pulses leads into the deep UV region and complicates detection. In collaboration with G. Cerullo and S. De Silvestri, Politecnico Milano, a variant of the above method [2, 3] was investigated that simplifies characterization of visible pulses from an optical parametric amplifier (OPA) in the 500 – 700 nm regime. Details on the OPA have been described in [3, 33-36]. Straightforward SFG as described in the previous chapter would require a minimum detection wavelength of 250 to 290 nm, depending on in which part of the OPA spectrum the upconverter pulse is extracted. However, if the upconverter pulse is derived from the Ti:sapphire laser at 780 nm, detection has to cover only the range down to 320 nm, which greatly simplifies detection. The downside of this variant is that the trick of using the SHG of the replica pulses for calibration of the set-up cannot be used anymore, as it does not fully overlap
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with the detection range. This trick made it very simple to generate a fringe pattern that was known to be equidistant in frequency. For a reliable calibration, 12 spectral lines of a mercury lamp were used. Otherwise, the set-up was very similar to the one introduced in the previous chapter. Figure 4.15 shows a recorded cross-correlation SPIDER trace and the reconstructed temporal pulse shape with a FWHM of 5.8 fs.

4.5. Spatially resolved SPIDER

In this Section the influence of spatially dependent pulse parameters on pulse characterization is discussed. A simple extension of the previously introduced sub-10-fs SPIDER setup allows recording spatial variations [37]. The SPIDER technique is particularly attractive for this purpose because the spectral phase measurement consists of only a single acquisition of a spectrum. The noniterative reconstruction algorithm enables rapid processing of the large amount of data resulting from the spatial resolution. The intrinsically fast acquisition and reconstruction have previously been demonstrated in real-time variants of SPIDER with refresh rates up to 20 Hz [38, 39]. In addition, without any prior knowledge of the FDMS, autocorrelation measurements require spatial filtering before the nonlinear process, which strongly reduces signal strength. In contrast,
SPIDER obtains a spatially resolved pulse characterization even after the nonlinear process without any prior knowledge of the FDMS. Again the previously described sub-10-fs laser is used for a first demonstration of spatially resolved SPIDER measurements.

Spatial distortions of femtosecond pulsed beams occur, for example, in pulse stretchers, amplifier chains, and frequency-domain pulse shapers. Generally these effects become more severe with larger pulse bandwidth. Apart from these imperfections, free-space diffraction of broadband beams is an inevitable source of spatial structure causing an FDMS. For Gaussian beams, the radius \( w \) as a function of the propagation distance \( z \) is given as

\[
w(z) = \sqrt{\frac{\lambda z_0}{\pi}} \sqrt{1 + \left(\frac{z}{z_0}\right)^2}
\]

(4.6)

with the wavelength \( \lambda \) and the confocal parameter \( z_0 \). For beams originating from a laser cavity the confocal parameter \( z_0 \) is a constant. Therefore, the mode area is proportional to the wavelength at a fixed distance \( z \) from the focus. For the nearly one-octave-bandwidth spectra of sources in the 5-fs-regime [40], this effect can lead to spectral variations of the mode area of more than 50%.

For Kerr-lens mode-locked (KLM) lasers, complex FDMS effects have been reported [41]. Inside the gain medium of such a laser, the time-dependent Kerr-lens translates the temporal intensity profile of the pulse into an accompanying spatial structure and vice versa. Thus, in such a laser the FDMS is pulse-shape dependent. Figure 4.16 shows measurements of the FDMS for sub-10-fs pulses from a KLM Ti:sapphire laser [6]. The mode areas have been measured using a beam profiler and 10-nm-wide interference filters. The variation of spectral power density and mode area with wavelength reflects the dispersion oscillations [40] of the double-chirped mirrors used inside the cavity (compare Chapter 3). Small adjustments of intra-cavity dispersion alter the temporal pulse shape in the Kerr medium, resulting in significant variations of the FDMS.
In autocorrelation or cross-correlation techniques using second-harmonic generation (SHG), such as SHG-FROG, each frequency-component of a pulse is mixed with each frequency-component of a gating pulse in a convolution-like manner (Fig. 4.17a). Ideally all these mixing processes occur with equal efficiency. However, even provided perfect phase-matching conditions, the FDMS produces a frequency-dependent conversion efficiency. In the simple case of pure SHG of quasi-monochromatic slices of the spectrum, for example, a 41% increase in mode area (Fig. 4.16) already reduces the second-harmonic signal by a factor of two. Such efficiency variations can result in erroneous pulse characterization. Previously, the effect of the FDMS of an ideal Gaussian beam (Eq. 4.6) has been taken into account in SHG-FROG measurements [15]. In principle, any given FDMS can be accounted for in the reconstruction algorithm. However, this becomes increasingly difficult as the complexity of the mode structure increases. This is especially true if the mode structure changes significantly with minor adjustments of the laser, as is the case for the structures shown in Fig. 4.16.

**Fig. 4.16:** Wavelength-dependent mode area of a sub-10-fs Kerr-lens modelocked Ti:sapphire laser. The spectral variation of the power density (solid) and the mode area (dashed line and filled circles) reflect the dispersion oscillations of the double-chirped mirrors used inside the cavity. The dash-dotted line shows the qualitative behavior expected from Eq. 4.6 (up to a scaling factor). Shown are two measurements for slightly different spectral shapes. Despite the small change in the spectral shape, both measurements exhibit a significantly changed frequency-dependent mode-size.

**Fig. 4.17:** Schematic picture of the mixing processes in broadband second-harmonic generation (SHG) (a) and in sum-frequency generation (SFG) of a broadband pulse with a quasi-cw spectral slice (b). In SHG many different input wavelengths with differing spatial patterns are contributing to the signal at a given wavelength, whereas in SFG each individual spectral component of the signal is generated by a single mixing process.
The SPIDER technique uses sum-frequency generation (SFG) of two broadband input pulses delayed with respect to each other and two quasi-cw slices of a strongly linearly chirped pulse. In the spectral domain, each of these SFG processes corresponds to a convolution of a broadband spectrum with delta-function-like spectrum (Fig. 4.17b). As a result, the input beam gets shifted in frequency space by the constant frequency and multiplied by the spatial mode pattern of that cw slice. The different frequencies of the two cw slices generate a spectral shear between the two broadband input spectra. A FDMS of the strongly chirped pulse does not cause a frequency-dependent efficiency as the same cw slice is mixed with each frequency-component of the broadband input pulse. The SPIDER signal, which consists of the spectral interference of the two spectrally sheared broadband pulses, is only affected in its spatial intensity pattern, fringe contrast, and an undetermined phase constant but not in spectral fringe spacing. Because SPIDER uses only the latter for spectral phase reconstruction, spatially resolved SPIDER works correctly even in the presence of significant FDMS effects. It should be noted, that this implementation of SPIDER, like all self-referenced pulse measurement techniques demonstrated to date, cannot extract the linear phase term. Based on the initial description of the spatially resolved SPIDER [37], recently a further extension has been published that allows to also extract a phase front tilt, which is important for characterizing amplifier systems [42]. Demonstration of this method with unamplified pulses from an oscillator is still outstanding.

The spatially resolved SPIDER set-up images the SPIDER signal beam and the fundamental beam from the SFG crystal on the entrance slit of a 0.3-m imaging spectrograph equipped with a 600-groove/mm grating and a two-dimensional 1024 by 128 pixel CCD camera. Otherwise the set-up is widely identical to Fig. 4.10, except that now 2-dimensional information is to be extracted from the camera. SPIDER signal and fundamental beam were measured independently. The entrance slit provides the spatial resolution along one axis while the vertical CCD dimension resolves the beam along the other axis. With the slit and the CCD camera, an absolute spatial resolution of roughly 80 µm is achieved in each direction. The number of spatial sample points across the beam diameter can be adjusted by the choice of the magnification of the imaging optics. With this set-up the spatially resolved spectral phase along one lateral beam axis is measured in a single acquisition. To access off-axis points, the beam can be either translated sideways or rotated around its axis. The beam rotation can be achieved with a Dove prism or by out-of-plane reflection in an equivalent arrangement of mirrors.

Figure 4.18 shows the fundamental spectrum of the sub-10-fs Ti:sapphire laser measured at three spatial positions relatively close to the beam center. With increasing distance from the center the spectra shift to longer wavelengths, which agrees favorably with the trend expected from Eq. 4.6. For different operating conditions of the laser, however, the KLM-effect may even reverse the tendency expected from free-space diffraction. Under typical operating conditions of our laser, the time-dependent Kerr-lens appears to be the dominant contribution to the FDMS (see Fig. 4.16). In a similar measurement shown in Fig. 4.19, a highly irregular structure in the short wavelength part of the spectrum shows up. This complicated pattern can be explained with diffraction
effects due to slight clipping at one of the prisms, a situation that might go unnoticed in the absence of spatial resolution.

![Normalized spectrum](image)

**Fig. 4.18:** Three spectra measured at different lateral positions inside the beam (solid: at 97%, dashed: at 50%, and dash-dotted: at 23% of the peak intensity). For comparison the spatially integrated spectrum is also shown (shaded).

![Contour plot](image)

**Fig. 4.19:** Contour plot of the spatially resolved pulse spectrum. The contours are evenly spaced on a logarithmic scale and start at 0.7% of the maximum value. Note the complicated structure on the short wavelength side.

The combination of the spatially resolved spectrum and the spectral phase measurement gives access to the full lateral dependence of the pulse shape (Fig. 4.20). The situation shown in Figs. 4.18 - 4.20 corresponds to a spatially averaged transform limited pulse of 9.2 fs duration, slightly chirped to an averaged duration of 11.6 fs. A clear trend is observed even for the relatively insensitive full width at half maximum (FWHM) duration. On-center, the pulse has a duration of 12.3 fs in contrast to an 11-fs duration in the wings. It should be noted that these variations result in systematically erroneous spatially integrated measurements with an error depending on the specific technique and the power law of the nonlinear process. The data in Fig. 4.20 can be further processed to determine the variation of beam diameter within the pulse. This allows to measure beam diameter variations in the time domain, caused by a temporally varying Kerr lens of the laser. This is shown in Fig. 4.21. For the range of ±10 fs displayed in Fig. 4.21, the beam diameter varies ±10% of the diameter at zero delay ($w_0=445 \mu m$).
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Fig. 4.20: Left: Laterally varying temporal pulse shapes normalized to the spectrally integrated beam intensity. Right: To facilitate a direct comparison, two pulses (dotted and dash-dotted line) are also shown normalized to the same peak intensity. The differences are most pronounced in the temporal wings of the pulses. Nevertheless, the full width at half maximum of the two pulses shown already differs by more than 10%.

The spatially resolved SPIDER demonstrates very impressively the flexibility of this new method towards advanced characterization problems. These cannot be easily addressed with any other technique. Autocorrelation may well be rapid enough for such a problem, but fails in fully resolving pulse amplitude and phase. FROG, however, succeeds in the latter, but requires time-consuming reconstruction algorithms, which simply overload with the wealth of data. With the recently demonstrated high immunity of the SPIDER technique towards experimental noise, an accurate pulse characterization is possible even in the low-intensity spatial or spectral wings of a beam [43]. The accuracy of the method can be further improved by suitable adaption of the integration times to the local intensity. Spatially resolved amplitude and phase characterization should enable a quantitative analysis of spatial distortions and propagation effects of femtosecond pulses. Additionally, this tool should simplify the interpretation of experimental results obtained with spatially structured beams.

Fig. 4.21: Visualization of the dependence of the beam diameter vs. delay measured with 12-fs pulses from a Ti:sapphire laser. At pulse center, a \( w_0 \) of 445 µm was measured. Two FWHMs ahead, \( w_0 = 470 \) µm, and two FWHMs behind, \( w_0 = 390 \) µm. These beam diameter variations are on the order of 30% and clearly illustrate the combined action of chirp and Kerr-lens inside the laser, translating into beam diameter variations.

\[
w_0(0 \, \text{fs}) = 445 \, \text{µm}
\]

\[
w_0(-12 \, \text{fs}) = 390 \, \text{µm}
\]

\[
w_0(12 \, \text{fs}) = 470 \, \text{µm}
\]
4.6. Comparison of the characterization techniques

In the previous Sections, several methods for characterization of sub-10-fs pulses have been discussed. Three different methods have been applied to pulses from a Ti:sapphire laser and were found to provide consistent results. These measurements are compiled in Fig. 4.22. Every characterization method required some adjustments for operation with such short pulses, but was found to operate reliably and provide accurate data. All internal consistency checks were passed, and comparisons with independently recorded autocorrelations indicated proper operation of the method. In particular, SPIDER turned out to be a flexible tool that can easily be extended towards spatial resolution or rapid acquisition. At this point, one may conclude that both, SPIDER and FROG are well suited for the task of sub-10-fs pulse characterization, whereas decorrelation from autocorrelation and spectrum is to be used with some more caution. Sorting out decorrelation methods because of their susceptibility to experimental noise, this calls for a more direct comparison of FROG and SPIDER [11].

For a fair experimental comparison of collinear type-II SHG-FROG and SPIDER, an experimental situation was chosen that allowed for optimum long-term stability of the Ti:sapphire laser and kept fluctuations at an absolute minimum. This requires operating the laser slightly farther away from the stability limit and at longer pulse duration. For the experiments described in this section, the laser exhibited a transform limit of 7.4 fs. Under these conditions, 10 independent measurements were recorded, both with FROG and SPIDER. The FROG traces were recorded at a 1.46-fs step size and a spectral resolution of 1.1 nm. The data is arranged on a (128 x 128)-grid. All traces were corrected according to Section 4.2. For pulse-retrieval, a commercial software (Femtosoft Technologies) was used, and 600 iterations of the algorithm were used to ensure convergence. The SPIDER data was acquired with a spectral resolution of 0.27 nm. The delay of the short pulse replicas was 302 fs, resulting in a spectral shear of 0.029 fs⁻¹. The group delay dispersion in the dispersive SPIDER arm was 10400 fs². The phase was retrieved from the interferogram applying the procedure described in Ref. [44].

Even though SPIDER can acquire data more rapidly, the same time span was deliberately used for each set of measurements to keep laser drift at a comparable magnitude. Moreover, the two setups were designed to have nearly identical dispersion. However, due to the very short pulse duration, even small deviations caused by slightly unequal air paths became noticeable. Slight differences between the results obtained with the two techniques are attributed mostly to unequal dispersion in the beam paths.
Fig. 4.22: Different methods of pulse characterization demonstrated with sub-10-fs pulses from a Ti:sapphire laser. **a.** Spectral phase interferometry for direct electric field reconstruction (SPIDER). The data shown is a spectral interferogram between two spectrally sheared replicas of the input pulse. The spectral phase is encoded as the fringe distance vs. optical frequency. **b.** Pulses from a Ti:sapphire laser measured by SPIDER.  
**c.** Frequency-resolved optical gating measures the spectrally resolved autocorrelation shown and also iteratively reconstructs the pulse shape. **d.** Pulses from a Ti:sapphire laser measured by FROG. Note that the discrepancies between the results of the individual methods have to be mainly attributed to different experimental conditions. Both methods, however, retrieve pulse durations of about 6 fs or slightly below. **e.** Interferometric autocorrelation. **f.** Power spectrum of the fundamental. **g.** The data of IAC and spectrum can be also used to reconstruct the pulse profile, however, with notably reduced accuracy, comp. Section 4.1.
Fig. 4.23: Interferometric autocorrelations calculated from the retrieved pulses (solid lines) and the independently measured data (circles). Only one half of the autocorrelation is shown to better resolve the details of the traces. The result obtained from the FROG measurement is shown on the left and the data reconstructed with the SPIDER method is plotted on the right.

Before going into a detailed analysis of statistical errors of one or the other method, a check on systematical errors can be achieved by comparing results deduced from both methods with an independent third measurement. An important crosscheck is a comparison of the retrieved pulses with simultaneously recorded interferometric autocorrelations (Fig. 4.23). The agreement between the measured data and the autocorrelations calculated from the retrieved pulses is excellent, as was observed in earlier crosschecks performed on shorter pulses (see Fig. 4.9. and 4.14.). From these checks, systematic errors are expected to be relatively small for both techniques. For the pulses displayed in Fig. 4.23, FROG and SPIDER both measure a pulse duration of about 9 fs, while fitting a sech$^2$ pulse shape to the experimental autocorrelation yields a pulse duration of 7.5 fs. As discussed before, the assumption of a particular pulse shape can lead to large errors in the estimation of the pulse duration. Often, this tendency is indicated by the strong deviations of the spectral shape from a sech$^2$ shape. The same arguments hold for the a priori assumption of other analytical pulse shapes.

4.6.1 Statistics of the temporal pulse parameters

For a statistical analysis, a set of 10 measurements is used. The complete set of FROG measurements displays FROG errors ranging from 0.0032 to 0.0054. The FROG error allows sorting out some outliers, which significantly improves statistical performance of the method. Figure 4.24 contains temporal intensity profiles reconstructed with both FROG and SPIDER. The strongest fluctuations are present in the FROG evaluation. One trace strongly deviates from all other measurements in the wings, and this trace also corresponds to the highest FROG error. Generally, the FROG error appears to be well suited to compare the quality of FROG reconstructions taken at identical conditions. However, the FROG error might be a misleading criterion for a more general comparison.
of measurements because it depends on the grid size of the FROG trace as well as on the percentage of the grid being covered by the actual FROG data.

Fig. 4.24: Temporal intensity profiles reconstructed with FROG and SPIDER.

Restricting the evaluation to the first five SPIDER measurements results in a slight reduction of statistical fluctuations. This suggests a small but systematical temporal drift in the SPIDER data, which may be attributed either to a drift of the laser system or of the SPIDER apparatus itself. The greater fluctuations of the FROG setup prohibit the observation of such a small drift.

One can now compare the results of the FROG and SPIDER measurements in terms of the full width at half maximum (FWHM) duration of the temporal intensity profile. This disregards a lot of the information in Fig. 4.24, and the pulse duration turns out to be relatively insensitive to phase fluctuations. Averaging over the complete set of 10 measurements, we obtain a duration of $8.92 \pm 0.23$ fs with FROG and a pulse duration of $8.91 \pm 0.11$ fs with SPIDER. The pulse duration derived from both methods is in excellent agreement.

4.6.2 Statistics in the spectral domain

Reconstructed spectra from the FROG trace confirm the trend of the analysis in the temporal domain. Generally, those FROG traces with a large FROG error reproduce an independently recorded spectrum far worse than those with small FROG errors. This gives an additional criterion for selecting data, but it also indicates a relatively low robustness of the FROG method towards experimental noise. Figure 4.25 contains a plot of the average spectral phase determined by each technique for each set of measurements. The error bars represent standard deviations. To estimate the correlation between spectral
power density and phase error, the independently measured pulse spectrum is included in each graph.

**Fig. 4.25:** Spectral phases measured with FROG and SPIDER in ten successive acquisitions (solid line). Statistical fluctuations are indicated by error bars. For comparison, the spectral power density is also depicted as a dashed line. The data corresponds to the temporal pulse profiles shown in Fig. 4.24.

Inspection of this figure makes it immediately clear, that FROG shows significantly stronger statistical fluctuations in the reconstructed spectral phase than SPIDER. The relatively poor performance is not only caused by the single outlier of the FROG traces in Fig. 4.24, but also persists when the data set is reduced to those with smallest FROG errors. For the FROG technique we obtain an averaged statistical phase error of 0.122 rad, for SPIDER 0.044 rad. It should also be noted that the statistical uncertainty of the phase in the SPIDER measurements is correlated with signal strength. This trend is not so clear in the FROG measurements, and strong fluctuations are already present when the spectral signal strength is reduced to about 10% of the maximum signal. In these measurements, SPIDER outperforms FROG by a factor of 3. The higher accuracy of SPIDER for spectral phase measurements can be understood from the fact that it is an interferometric technique that directly measures this quantity. One obvious explanation for the poorly converging data sets, giving rise to the outliers, is the much longer acquisition time required for multi-shot FROG. For a single FROG trace, 128 spectra are recorded and temporal drift of the laser might result in a slight inconsistency of the data acquired. In contrast, the data required for the SPIDER technique can be recorded in a single measurement on a CCD array.

### 4.5. Summary and conclusion

The discussion in the preceding Sections makes it clear, that pulse characterization with sub-10-fs pulses is by far a too complex a problem that a single number could simply characterize pulses from a laser. In the last few years, sophisticated tools have been developed that allow for much more insight than the traditional approach of using an autocorrelation and fitting some analytical but incorrect function to it. The spectral phase
determined by FROG or SPIDER contains valuable additional information and gives direct insight into imperfections, e.g., of the dispersion compensation scheme. This was useful to recognize the output coupler phase in the Ti:sapphire laser and dispersion oscillations of the mirrors to cause the small but non-negligible deviation from the Fourier limit in such lasers. Moreover, in the experiments performed in the Milano group, a filter incorporated into the mirror design was clearly recognized to be responsible for phase distortions at larger wavelengths. With this information, an improved mirror design without the incorporation of a seed-rejection filter promises a significant increase in bandwidth. Finally, in some experiments that were done in collaboration by E. Riedle in Munich [45], information of a SPIDER measurement was used to optimize an adaptive dispersion compensation scheme, which used a flexible membrane mirror. In that experiment, the SPIDER set-up greatly simplified adjustment of dispersion, change of operating wavelength and compensation for higher-order dispersion in particular. All these examples show that a more thorough characterization immediately improves experimental capabilities and allows for experiments that were not possible before.

It is an ongoing discussion, which measurement method is to be preferred in which situation. In particular, there is a great divide between supporters of the FROG technique and supporters of the SPIDER technique. It clearly needs to be pointed out that no general decision on that debate can be derived from the experiments described in this work. It is not the goal of this work to promote one or the other characterization technique, but to arrive at an accurate characterization of the Ti:sapphire pulses described in Chapter 2. For this purpose, it was extremely useful to have several techniques at hand and to compare their respective results in a testbed situation. If any conclusion on a characterization technique can be drawn at all, then it is only valid for characterization of the very shortest pulses. For any of the measurement tasks described throughout this thesis, SPIDER was found to be an accurate and simultaneously very rapid technique. Additionally, SPIDER uses nonlinear conversion bandwidth more efficiently than FROG. However, FROG was also used by other authors very successfully to characterize pulses shorter than 4 fs [46]. In the meantime, the SPIDER technique was also demonstrated with pulses as short as 5 fs [32].

Whatever full characterization technique is employed, the wealth of additional information clearly pays off. At this point the traditional approach of fitting analytical functions to autocorrelation traces is clearly not acceptable anymore and can only serve to derive a rough estimate on pulse duration. During the course of this work, the complexity of performing full characterizations has significantly decreased while the options have increased, establishing new standards for measuring the shortest controlled man-made events.
4.6. References in Chapter 4

5. Carrier envelope offset measurement and control

Current state-of-the-art laser sources deliver optical pulses with a duration of about 5 fs, which corresponds to two optical cycles in the near-infrared [1]. At this duration, the peak electric field strength of such a pulse shows a pronounced dependence on the relative phase between carrier and envelope of the pulse. In the following, this relative phase will be referred to as the carrier-envelope offset (CEO) phase. For longer pulse durations, this previously inaccessible parameter does not play a role. For few-cycle pulses, however, a dependence of the conversion efficiency on the CEO phase is obvious for processes like above-threshold ionization (ATI, [2]). In this process, the ionization probability shows a step-like behavior with electric field strength. Similarly, the strongly fluctuating photon numbers of high harmonics have been explained by a lack of control of the CEO phase [3]. Controlling the CEO phase is also an important prerequisite for several proposed attosecond generation schemes. A recent study on the duration of an isolated high harmonic pulse revealed a pulse duration of about 1.5 fs [4]. The absence of the anticipated attosecond time signature in these experiments was partly attributed to temporal averaging over different values of the CEO phase. These examples clearly identify $\phi_{\text{CEO}}$ as an important parameter in a new regime of nonlinear optics. Because of the extremely short pulses required to see these effects, this regime has been termed extreme nonlinear optics [5]. At pulse durations of two cycles and below, the conversion efficiency starts to exhibit a noticeable dependence on the CEO phase. The higher the order of the nonlinear optical process, the more pronounced this effect is expected to be.

5.1. The carrier-envelope offset frequency and phase

The physical origin of the CEO phase is the difference between phase velocity $u_p$ and group velocity $u_g$ inside an oscillator cavity. If this cavity does not contain any dispersive elements, both velocities are equal and the position of a maximum of the electric field stays fixed relative to the maximum of the envelope. Intracavity dispersion, however, induces a per-roundtrip phase offset

$$\Delta \phi_{\text{GPO}} = \frac{2\pi}{\lambda} \int_0^L (n_g(z) - n(z)) \, dz = \frac{\omega^2}{c} \int_0^L \frac{dn(z)}{d\omega} \, dz$$

(5.1)

between the envelope and carrier of an optical pulse. Here the coordinate $z$ is chosen along the propagation axis inside the cavity and $L$ is the effective length of the cavity, i.e. twice the geometrical length for a linear cavity. $n = c / u_p$ is the refractive index, $n_g = c / u_g$ the group index, and $\omega$ the angular frequency. For a typical laser cavity [6], the combined GPO effect of the Ti:sapphire crystal, the air path, and the prism compressor can be estimated as about 250 optical cycles. If the carrier is advanced by an integer number of cycles relative to the envelope, the electric field structure exactly reproduces itself from pulse to pulse (compare Fig. 5.1). In the following, only the subcycle part of Eq. (5.1) will be considered. This quantity is defined as the pulse-to-pulse CEO phase change.
5. Carrier-envelope offset measurement and control

\[ \Delta \varphi_{\text{CEO}} = \Delta \varphi_{\text{GPO}} \mod 2\pi. \]  \hfill (5.2)

For the case of a laser oscillator, the CEO frequency is given by

\[ f_{\text{CEO}} = \frac{1}{2\pi} \frac{d\varphi_{\text{CEO}}}{dr} = \frac{\Delta \varphi_{\text{CEO}}}{2\pi T_R} = \frac{\Delta \varphi_{\text{CEO}}}{2\pi} f_{\text{rep}}. \]  \hfill (5.3)

The first experimental approach to the measurement of the carrier-envelope offset relied on an interferometric cross-correlation of two subsequent pulses [7]. However, even the slightest dispersive asymmetry between the two correlator arms introduces an offset to the measurement of \( f_{\text{CEO}} \), which makes this method inappropriate for any long-term stabilization of \( \varphi_{\text{CEO}} \).

**Fig. 5.1:** Electric field structure of pulses from a laser oscillator. The temporal delay between the envelopes of two successive pulses is given by \( 1/f_{\text{rep}} \) whereas the underlying carrier experiences an additional temporal shift caused by the difference of group and phase velocity in the laser cavity.

**Fig. 5.2:** Mode comb of a mode-locked laser oscillator in the frequency domain. The equidistantly spaced modes are represented by thick lines. Extending the comb towards zero frequency (thin lines) reveals an offset at zero frequency. This offset frequency can be measured by taking a frequency from the low-frequency wing of the spectrum, frequency-doubling it, and comparing it to a neighboring mode in the high-frequency wing of the spectrum.
5.2 Extreme nonlinear optics

5.2.1 Estimation of the sensitivity of nonlinear effects on the CEO phase in the perturbative regime

For pulses of a few cycles duration, it is expected that the conversion efficiency of nonlinear optical processes starts to depend on $\varphi_{\text{CEO}}$. In the following, some calculations will be presented that allow for a rough estimation on when the regime of extreme nonlinear will be reached. These calculations are based on a reconstructed electric field profile of a Ti:sapphire laser, as it was measured by the SPIDER technique of the previous chapter. This electric field profile is shown in Fig. 5.3 for two extreme values of the carrier-envelope phase $\varphi_{\text{CEO}}=0$ and $\varphi_{\text{CEO}}=\pi/2$. Even though the intensity envelope of this pulse has been measured with a pulse duration of 5.9 fs, which corresponds to only 2.3 optical cycles, the difference in peak electric field strength amounts to only 2%. In construction of the electric field structure, care has been taken to ensure a vanishing DC component and energy conservation, i.e. $\int E(t) \, dt = 0$ and $\int |E(t)|^2 \, dt = \text{const.} |\varphi_{\text{CEO}}|$.

In the following, the pulse duration will only be referred to in optical cycles FWHM of the intensity envelope of the pulse, which accounts for the natural scaling properties of extreme optical phenomena. For the simulations shown here, hyperbolic secant and Gaussian pulse shapes with durations of 1 to 4 cycles have been assumed. For an estimate of the conversion efficiency of an $n$-photon process the following expression is evaluated

$$\eta^{(n)}(\varphi_{\text{CEO}}) = \int |E(t, \varphi_{\text{CEO}})|^{2n} \, dt$$

(5.4)

for different values of the CEO-phase. Examples for the integrand are depicted in Fig. 5.4. Here the pulse shape in Fig. 5.3 was assumed, showing that the satellite structure vanishes with higher order of the process. This justifies the use of simple analytic pulse...
shapes in the following. Carrying out the integration, one is particularly interested in the difference of conversion efficiency at the two extreme values of the carrier envelope phase. The quantity $\Delta$ allows for an estimate of the resulting modulation

$$\Delta^{(n)} = \frac{\eta^{(n)}(0)}{\eta^{(n)}(\pi/2)} - 1$$

(5.5)

The result of this analysis is shown in Fig. 5.5. For pulses of only cycle duration, a strong modulation of the conversion efficiency of any given process is expected. According to the simulations, even SHG has a pronounced dependence on the order of $\Delta^{(2)} = 0.01$. Closer to currently available pulse durations of about 2 optical cycles, the modulation is already strongly reduced. Currently the most promising experimental arrangement to see such an effect with an oscillator would be a 3-photon or a 4-photon process with pulses of two to three cycles duration. A resulting modulation of $10^{-6}$ should be detectable, even given the low conversion efficiency of such processes and the low numbers of generated photons/electrons imposing a shot-noise limit on the detection. In the Figures, a lower limit of $\Delta = 10^{-6}$ has been included as a guidance. To reach this detection limit, at least $2/\Delta^2$ quanta have to be converted and detected in an experiment. Some first estimations have been derived from multiphoton-ionization at a CsTe surface of a solar blind photo multiplier. This process is nominally a 4-photon process and currents at the photocathode in the pA-regime corresponding to roughly $10^7-10^9$ electrons per second have been
observed in some initial experiments. To collect the necessary $2 \times 10^{12}$ electrons, an experimental observation time corresponding to about half an hour to 2 days would be required for evidence on the phase dependence. The maximum flux with the correspondingly smallest observation times would already be within an order of magnitude of the specified absolute maximum current of such a tube in one-photon operation. So the given detection limit of $10^{-6}$ should not be misunderstood as a hard barrier; still it will be already very difficult to see effects that far down.

**Fig. 5.5:** Numerical estimations of the sensitivity of the conversion efficiency of nonlinear optical processes on the CEO phase for few-cycle pulses. Left: assuming a Gaussian pulse shape, right: assuming a hyperbolic secant pulse shape. Shown is the range from 0.75 to 4 optical cycles duration and process orders ranging from 2 (SHG) to 6. The marked region in this plot above two cycles duration and $10^{-6}$ modulation indicates currently experimentally accessible regions.

### 5.2.2 Enhanced sensitivity in the tunneling regime

So far, the sub-cycle sensitivity of nonlinear optical processes has been analyzed in a way that was automatically assuming that only one order contributes to the signal and processes can be analyzed in a perturbative way. A conclusion from this analysis is that the effects are weak with pulses of two optical cycles duration or above. Even with the shortest pulses that have been ever generated from Ti:sapphire oscillators, detection of a dependence of the conversion efficiency of a nonlinear optical effect may be at the very limit of detectability, even under optimistic assumptions.

It has been argued before that the sensitivity can be greatly enhanced if the perturbative regime is left and several order processes contribute to the nonlinear optical signal [8]. In the following, it is tried to find an order-of-magnitude for the onset of the tunneling regime. These estimations are based on three different models: the Keldysh parameter [9], the Ammosov-Delone-Krainov (ADK) model to estimate ionization rates in multiphoton processes [10], and the barrier-suppression model (BSI, [11]). These models are well-established in the field of strong-field physics and have mostly been applied to interaction of light and atomic media, in particular noble gas targets.
Nevertheless, some of the original work [9] was devoted to impact ionization in semiconductor materials. Later this work was transferred to atomic media, which formally equates the work function of bulk media and the ionization energy of atomic media. Furthermore, the models have to be scaled down to ionization energies of a few eV, as ionization of the inert gases is not an option with peak powers of oscillators. As a model case, multi-photon electron generation from commonly used uv photocathode materials will be considered. The work function of materials such as gold [8] or CsTe is about 4.5 eV. Scaling of the ADK model to small ionization energies has been considered to the very related case of ionization of cesium (3.9 eV, [13]); the Keldysh parameter was originally proposed for use at even smaller work functions. A work function of 4.5 eV requires at least 3 photons from a Ti:sapphire laser (1.2-1.8 eV) for excitation of an electron. This type of process can be excited with typically available peak powers from oscillators with decent conversion efficiencies.

For bound-free transitions in a semiconductor, the original analysis of Keldysh [9] yielded a scale parameter

$$\frac{1}{\gamma} = \frac{e E_a a_B}{\hbar \omega_0}. \quad (5.6)$$

Here, $e$ is the electron charge; $E_a$ is the peak-to-peak electric field (twice the amplitude), and $a_B$ Bohr’s radius. $\hbar \omega_0$ is the photon energy. Typically, the theoretical description of the hydrogen atom is utilized with the connection $a_B = \hbar / \sqrt{2mW_b}$ between Bohr’s radius and binding energy $W_b$. Here $m$ is the electron mass [11]. A Keldysh parameter $\gamma >> 1$ is typically associated with an adequate perturbative description of the system, while $\gamma << 1$ calls for a non-perturbative analysis. The parameter range $1-10$ is typically referred to as tunneling regime. In this regime, already a strong enhancement of the sensitivity on the carrier-envelope offset phase is expected. Assuming $W_b=4.5$ eV and a Ti:sapphire laser with a 775-nm center wavelength, the tunneling regime would be reached at an intensity level of about $10^{12}$ to several $10^{13}$ W/cm$^2$ (see Fig. 5.6). This is very high for oscillators but has been demonstrated before [12].

The second model based on the original publication of Ammosov, Delone, and Krainov (ADK, [10]) has found widespread use to calculate ionization probabilities in multiphoton processes. The ADK-model is typically used to estimate intensities of focused amplified beams according to the ionized species observed. The basis for the ADK theory is a semi-classical calculation of the tunneling rate. For the ionization rate $W$ the following expression has been derived [10]

$$W = \left[ \frac{3e}{\sqrt{\pi}} \frac{Z^2}{n^*} \left( \frac{4eZ^3}{E_a n^* 3} \right)^{2n^* - 3/2} \exp \left( \frac{-2Z^3}{3n^* 3 E_a} \right) \right]. \quad (5.7)$$

Here $Z$ is the effective nuclear charge, i.e. 1 for single ionization as will be considered in the following. $n^*$ is the effective quantum number $n^* = Z / \sqrt{2W_b}$. Note that this
calculation presupposed units according to $h = m_e = e^2 = 1$, i.e. the charge has to be expressed in units of elementary charges, the electric field strength in units $5.142 \times 10^{11}$ V/m, energy in units $4.359 \times 10^{-18}$ J, and the time in units $2.4189 \times 10^{-17}$ s. For an ionization energy of 4.5 eV yielding an effective quantum number $n^* = 1.73$, one calculates a complete ionization within one optical cycle for an intensity of $10^{13}$ W/cm² (comp. Fig. 5.6). Such a strong effect is not required, but below $10^{11}$ W/cm² no excitation is to be expected. For the generation of reasonably large multi-photon currents from a photocathode, intensities on the order of $10^{11}$ W/cm² are required according to the ADK model. In this regime, an enhanced sensitivity to the carrier-envelope offset phase is anticipated. Given the widely different approach used in [9] and [10], the results of both theories agree very well and pinpoint intensities of a few $10^{12}$ W/cm² as a necessary condition for a tunnel-enhanced phase sensitivity.

Fig. 5.6: Ionization rate as a function of optical intensity as predicted by the ADK model. Shown are several traces for work function / bound energies ranging from 1 to 10 eV. The ionization rate is given in units inverse seconds, i.e. a rate of $0.5 \times 10^{15}$ s⁻¹ corresponds to complete ionization within a single optical cycle. Depending on the geometry of the target, ionization rates of a few s⁻¹ to several $10^9$ s⁻¹ are required to generate multi-photon photocurrents of 1 nA. Additionally, the borders of the tunneling regime ($\gamma \approx 1-10$) as predicted by the Keldysh model are shown. Given a work function of 4.5 eV, intensities on the order of $10^{12}$ W/cm² or slightly below are required to reach the tunneling regime.

Let us finally address the barrier-suppression-ionization (BSI) model (see Fig. 5.7). This model simply calculates at which electrical field strength $E_a$, the superposition of an atomic Coulomb potential and the exterior electric field
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\[ E_a V(x) = V_{\text{Coulomb}}(x) + V_{\text{ext}}(x) = -\frac{e^2}{2\pi \varepsilon_0 |x|} - eE_a x \]  

(5.8)

converts an inner atomic bound state into a continuum state. Here \( \varepsilon_0 \) is the vacuum dielectric constant. From straightforward analysis, one can derive that

\[ E_a = \frac{\pi \varepsilon_0 W_b^2}{2e^3}. \]  

(5.9)

Assuming \( W_b = 4.5 \text{ eV} \), the BSI model yields an estimate for barrier suppression at about \( 5 \times 10^{11} \text{ W/cm}^2 \) (see Fig. 5.7). This is in good agreement with the other model’s prediction. It should be pointed out that the order-of-magnitude estimate of about \( 10^{12} \text{ W/cm}^2 \) extracted from all three models is well compatible with ion yield measurements in Cesium vapor [13].

\[ \text{Fig. 5.7: } \text{Barrier-suppression ionization. Shown is the suppression of the Coulomb barrier in electron volts as induced by a light field of a given intensity. The situation is depicted in the right inset. The external field deforms the nuclear potential, which converts bound states above a threshold value into continuum states. To ionize a state that is bound with 4.5 eV, e.g., an electric field corresponding to an intensity of about } 5 \times 10^{11} \text{ W/cm}^2 \text{ is required. This corresponds to 3-photon excitation of electrons off a gold photocathode [8]. Other cases as photo ionization of Helium and Xenon, as discussed extensively in literature [11], are also shown for reference. These regions at } > 4 \text{ photons energy can clearly be only accessed with amplified sources.} \]
In conclusion, one can state that an enhanced sensitivity to the carrier-envelope phase is theoretically possible, but at the edge of focused intensity levels of oscillators. It has to be made sure, that no thermal interaction takes place. One can therefore discard process with any type of spurious responses beside the desired multi-photon characteristics. This makes a gold photo cathode with its small but non-negligible absorption at the fundamental Ti:sapphire wavelength an unsuited material to detect the phase-sensitivity [8]. Multi-photon ionization in bulk materials, such as in diamond photo-diodes, is probably also not very well suited because of spurious two- and three-photon contributions. These considerations make it very clear that it is very challenging to find any detectable phase-sensitivity with two cycle pulses. Nevertheless these effects may become dramatic as soon as single-cycle pulses become available.

5.3 CEO frequency measurement schemes

5.3.1 Basic scheme based on heterodyning comb harmonics

An approach to measurement of $f_{\text{CEO}}$ that does not suffer from the offset problems of the early cross-correlation measurements was first introduced in 1999 [14]. This method can be most suitably understood in the spectral domain (see Fig. 5.2). Fourier transforming the repetitive pulse train of Fig. 5.1 yields a comb of equidistant spectral lines $v_m = f_{\text{CEO}} + mf_{\text{rep}}$. Notably, this comb does not include zero frequency unless the pulse-to-pulse phase slip $\Delta \phi_{\text{CEO}}$ is exactly zero. The frequency comb must not be confused with the modes of the linear cavity, which are only equidistant in the absence of intracavity dispersion. For a modelocked laser, the spacing of the comb lines is determined by the group velocity, i.e., the cavity round-trip time of the envelope. Experimentally, the equidistance of these comb lines has been checked to a relative uncertainty better than $10^{-15}$ [15]. Any irregularity of the comb frequencies would automatically induce different repetition rates of the spectral components of the comb and eventually cause temporal spreading of the pulses. The fact that the comb only has two degrees of freedom, $f_{\text{CEO}}$ and $f_{\text{rep}}$, has found widespread applications in metrology. Knowledge of these two frequencies provides one with a set of reference frequencies throughout the spectral coverage of the mode-locked laser. Beating an unknown optical frequency with the comb then allows one to derive the frequency of an optical transition from the measurement of three radio frequencies [16, 17, 18].

The picture in Fig. 5.2 also provides the key to measurement of $f_{\text{CEO}}$ by heterodyning harmonics from different parts of the mode-locked spectrum [14]. Taking the $N$-th harmonic of a comb line $Nv_{m_1} = Nf_{\text{CEO}} + Nm_1f_{\text{rep}}$ and beating it with the $M$-th harmonic of another comb line $Mv_{m_2} = Mf_{\text{CEO}} + Mm_2f_{\text{rep}}$ yields

$$Mv_{m_2} - Nv_{m_1} = (M - N)f_{\text{CEO}}, \quad (5.10)$$

which requires $Nm_1 = Mm_2$. This condition requires a certain minimum spectral width of the comb $\Delta f / f = 2(N - M) / (N + M)$; e.g., beating of the fundamental and second-harmonic requires an optical octave of bandwidth with $\Delta f / f = 0.67$. This situation is
illustrated in Fig. 5.2. Equation (5.10) is the key to any measurement of the carrier envelope-offset. This beat note delivers the carrier envelope phase slippage rate, either directly or as one of its harmonics.

5.3.2 Advanced schemes for measurement of the CEO frequency – transfer oscillators and interval bisection

The scheme introduced in Eq. (5.10) works very well for octave-filling spectra. In principle, this scheme could be applied to arbitrarily narrow spectra, if the conversion efficiency for harmonic generation of laser oscillators would not be extremely small for orders 4 and higher. However, there exist ways to circumvent this bottleneck. These methods rely on transfer oscillators and interval bisection. The required span of the comb can be reduced if two nonlinear-optical processes (SFG and N-th harmonic generation) and one additional transfer oscillator at $\nu_{\text{trans}}$ are combined, see Fig. 5.8 for the example of SHG. The fundamental of the frequency $\nu_{\text{trans}}$ is used for a collective upconversion of modes from the low-frequency to the high-frequency end of the comb. If $\nu_{\text{trans}} = n f_{\text{rep}}$ then $\nu(m+n+i) = \nu(m+i) + \nu_{\text{trans}}$ for all $i$ leading to modes within the phase-matching range of the SFG. With active control of $\nu_{\text{trans}}$, the frequency shift can be forced to an integer number of modes $n$, canceling out the beat note between the modes of the comb-like SFG signal and the comb modes in the vicinity of $\nu(m+n)$. Simultaneously, the $N$th harmonic at $N \nu_{\text{trans}}$ is compared with the frequency of the nearest comb mode $\nu(Nn) = Nn f_{\text{rep}} + f_{\text{CEO}}$,
\[
\nu(Nn) - N \nu_{\text{trans}} = (Nn f_{\text{rep}} + f_{\text{CEO}}) - Nn f_{\text{rep}} - f_{\text{CEO}}. \tag{5.11}
\]
As in the direct SHG process, all modes within the phase-matching range of this process contribute to the measured signal. In principle, $\nu_{\text{trans}}$ can be chosen arbitrarily but must not exceed the frequency span of the comb. For the case of SHG of the transfer oscillator, the required relative comb width for this method ranges from 0.5 with $2 \nu_{\text{trans}}$ chosen at comb center to 0.4 with $2 \nu_{\text{trans}}$ chosen at the low-frequency wing of the comb.

![Fig. 5.8: Scheme for measuring the CEO frequency based on optical transfer oscillators](image)

Given sufficient power, the high mixing efficiency of a combination of powerful laser diode pumped solid-state lasers and quasi-phase-matched nonlinear crystal allows for third harmonic generation (THG). Because THG is usually accomplished by SHG and sum frequency mixing of the second harmonic with the fundamental, three nonlinear
optical processes are required in total. For THG, a relative comb width of at least 0.29 is required.

Using two additional lasers as transfer oscillators with frequencies $\nu_a$ and $\nu_b$, the second and third harmonics of these two lasers can be phase-locked at $3 \nu_a = 2 \nu_b$ with a servo loop. In a subsequent step, $\nu_a$ is phase-locked to the nearest mode $\nu(2m)$, and the beat note between $\nu_b$ and its nearest comb mode at $\nu(3m)$ provides the desired information about $f_{\text{CEO}}$.

$$\nu_b - \nu(3m) = \frac{3}{2} (2mf_{\text{rep}} + f_{\text{CEO}}) - (3mf_{\text{rep}} + f_{\text{CEO}}) = \frac{f_{\text{CEO}}}{2}. \quad (5.12)$$

This scheme has the same efficiency constraints as the frequency doubling and tripling schemes and again, it might be necessary to employ a third oscillator for the THG process.

Frequency interval bisection [19] can be employed to divide the required comb width. The simplest scheme of this type uses a one octave interval divider stage that generates the frequency $\nu_a$ at the midpoint of $\nu_b$ and $2\nu_b$. This can be done by phase-locking the second harmonic of $\nu_a$ to the sum frequency of $\nu_b$ and $2\nu_b$ to yield

$$\frac{\nu_a}{2\nu_b} = \frac{3\nu_b / 2}{2\nu_b} = \frac{3}{4} \quad (5.13)$$

In a way similar to the SHG and THG of auxiliary oscillators, $\nu_a$ is phase-locked to the nearest line, $\nu_a = 3mf_{\text{rep}} + f_{\text{CEO}}$, and the beat note between $2\nu_b$ and its nearest comb line at $\nu(4m)$ provides the desired information, namely,

$$2\nu_b - \nu(4m) = \frac{4}{3} (3mf_{\text{rep}} + f_{\text{CEO}}) - (4mf_{\text{rep}} + f_{\text{CEO}}) = \frac{f_{\text{CEO}}}{3}. \quad (5.14)$$

Yet again, an additional laser at $2\nu_b$ might be needed to obtain sufficiently strong SFG signals.

<table>
<thead>
<tr>
<th>Scheme</th>
<th>Eq.</th>
<th>additional oscillators</th>
<th>nonlinear conv. steps</th>
<th>$\nu_{\text{low}}:\nu_{\text{high}}$</th>
<th>$\Delta \nu/\nu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. direct SHG/DFG</td>
<td>(5.10)</td>
<td>0/1</td>
<td>1</td>
<td>1:2</td>
<td>0.67</td>
</tr>
<tr>
<td>2. doubled transfer osc.</td>
<td>(5.11)</td>
<td>1</td>
<td>2</td>
<td>2:3</td>
<td>0.4..0.5</td>
</tr>
<tr>
<td>3. tripled transfer osc.</td>
<td>(5.11)</td>
<td>1</td>
<td>3</td>
<td>3:4</td>
<td>0.29..0.33</td>
</tr>
<tr>
<td>4. aux. oscillators</td>
<td>(5.12)</td>
<td>2/3</td>
<td>3</td>
<td>2:3</td>
<td>0.4</td>
</tr>
<tr>
<td>5. interval bisection</td>
<td>(5.14)</td>
<td>2/3</td>
<td>3</td>
<td>3:4</td>
<td>0.29</td>
</tr>
</tbody>
</table>

Table 5.1: Comparison of the different schemes in terms of additional oscillators, nonlinear conversion steps, and required comb bandwidth.

Table 5.1 summarizes the number of necessary auxiliary oscillators, the number of nonlinear steps and the required comb width of the proposed schemes. In principle, all
five schemes are sufficient for determining $f_{\text{CEO}}$. Excluding the direct SHG or DFG schemes with their very demanding $\Delta v/v=0.67$, all schemes can be potentially implemented with state-of-the-art KLM Ti:sapphire lasers.

For a successful implementation of any of the schemes proposed here, it is mandatory that a cycle-free phase-lock between different parts of the spectrum be maintained. This condition is equivalent to rms phase jitters of $\sigma_p=0.3$ rad [20], corresponding to CEO timing jitters in the range from 0.1 fs to 0.4 fs, depending on the scheme employed. In the following, we will explore the feasibility of the phase-locks, using experimental data from our laser.

5.3.3 Experimental bandwidth aspects

From the discussion in the preceding section a trade-off between available bandwidth of the laser source and complexity of the scheme has become obvious. Figure 5.9 shows a careful look at the spectrum of the Ti:sapphire laser introduced in Chapter 2. In this laser, self-phase modulation inside the Ti:sapphire crystal already causes spectral broadening beyond the gain bandwidth. Because of the strongly reduced reflectivity of the mirrors in the extreme parts of the spectrum, this light is immediately coupled out and not spatially filtered by multiple passes through the cavity. Consequently, the beam quality may deteriorate in the extreme spectral wings of the comb. Therefore one had to ensure that the beam is diffraction limited by using the spatial filtering of a 5-\textmu m diameter pinhole, which is about the mode size in typical single-mode fibers at 800 nm. The spectrum was recorded with a 30-cm double monochromator in additive configuration and determined the wavelength dependent sensitivity of the monochromator-detector combination with a calibrated white-light source. This set-up allowed for an usually high dynamic range of the measurement depicted on a logarithmic scale in Fig. 5.9. For most of the wavelength range, the spectrum is fitted by a Gaussian with a 55 THz FWHM, corresponding to a pulse duration of 8 fs, while the transform limit of the diffraction-limited part of the spectrum yields 6.6 fs.

**Fig. 5.9**: Measured spectral flux density of the KLM Ti:sapphire laser under diffraction limited conditions. The spectrum is calibrated in units photons per second and mode. TEM$_{00}$ power is 100 mW. Solid line: measured spectrum. The Fourier transform of this spectrum has a width of 6.6 fs. The minima in the extreme spectral wings correspond to resonances of the mirrors used inside the laser. Dashed line: Gaussian fit with 55 THz FWHM, corresponding to an 8-fs duration of a bandwidth limited pulse.
Additionally, the data in this Figure has been converted into units photons per individual mode of the mode-locked laser. Assuming a detection bandwidth of 100 kHz, \(10^5\) photons/s are needed for a shot-noise limited S/N-ratio of 1. To avoid cycle slips, the number of detected photons must be at least 100 times this value [20]. Imperfect mode matching, limited quantum efficiency, and other losses may account for an additional factor of 100, which leads to a conservative requirement of \(10^9\)-photons/s per mode. The measured spectrum in Fig. 5.9 indicates that the spectral range from 650 to 1000 nm, corresponding to a width of more than 160 THz, satisfies this criterion. This gives a \(\Delta v/v = 0.42\). This makes it immediately clear that the simplest experimental schemes described in Section 5.3.1 are hard-pressed for direct application at the laser (compare Table 5.1). Second harmonic generation within the comb, e.g., requires \(\Delta v/v = 0.67\). Even if one succeeds in generating the needed additional bandwidth by even stronger intracavity continuum generation, the S/N ratio of the beat note may be barely above the minimum tolerable levels. Recently, such experiments have been reported [21]. The authors could only detect the CEO beat note with \(\approx 20\) dB above noise, which turned out to be clearly insufficient for a meaningful stabilization of the CEO phase. They only achieved stabilization of the CEO frequency to about a MHz residual rms jitter. This clearly illustrates the importance of signal-to-noise considerations for selection of the most appropriate scheme. Basically, there are two options, which will be discussed in the following paragraphs: (i) one can use one of the more advanced schemes described in Section 5.3.2, increasing complexity with less available bandwidth. (ii) Another option is the use of extra-cavity spectral broadening, e.g. in microstructure fibers. As will be shown in the following, intra- and extracavity broadening are not equivalent in terms of introducing phase jitter. But let us first analyze applicability of the advanced measurement schemes.

For direct application with the Ti:sapphire laser, schemes 2, the frequency-doubled transfer oscillator, and 5, interval bisection, appear to be the most promising candidates for the CEO phase control of our modelocked laser (compare Table 5.1). Scheme 2, the frequency-doubled transfer oscillator requires a comb span of about 150 THz. Laser diodes in this range are not readily available and provide small power and large emission linewidths. The power in the spectral wings is insufficient to bridge larger frequency ranges in the 200-THz range, where convenient powerful lasers are readily available (1.5 \(\mu\)m Er fiber lasers). Of the two schemes, one can more easily meet the requirements of scheme 5, the interval bisection. Choosing the strong 1.338 \(\mu\)m Nd:YAG line as the fundamental of the octave-interval, one gets the second harmonic at 669 nm and the midpoint frequency at 892 nm. This 669-nm to 892-nm interval would then have to be bridged by the comb and Fig. 5.9 shows that a 6-fs Ti:sapphire laser provides sufficient power in this wavelength range. A further advantage of this scheme is that several mW of the power required at 669 nm can be directly generated by SHG from the fundamental, thus eliminating the need for an additional laser. The field at 895 nm can be generated by narrow linewidth DBR laser diodes commercially available for spectroscopy of the Cesium D\(_1\) transition. Furthermore, all nonlinear processes can be carried out with highly efficient mixers, including periodically poled KTP for SHG of 1338 nm and non-critically phase-matched K\(\text{NbO}_3\) for SHG of 892 nm and SFG of 1338 nm and 669 nm.
The alternative way of overcoming bandwidth restrictions is the use of additional spectral broadening by self-phase modulation in optical fibers [16]. With the advent of microstructure fibers, white-light continuum generation has become very simple even with the limited pulse energies of optical oscillators [22]. Spectral widths exceeding two optical octaves have been demonstrated in such fibers (compare Fig. 5.10). The dominant effect behind the continuum generation process inside the fiber is self-phase modulation. Other effects, such as Raman contributions to the fiber nonlinearity, may also be present and wash out the phase coherence between remote spectral components of the continuum. As in the anti-Stokes Raman process a phonon and a photon mix to generate a new photon of shorter wavelength, the phonon contribution may add a random contribution to the phase. Several groups reported difficulties using white-light continua for CEO frequency measurements when the input pulse duration was on the order of 100 fs. A possible way to explain these findings is that the Raman contribution is effectively reduced when the laser pulse bandwidth is below the Raman bandwidth (approx. 10 THz). In a similar way, the threshold for stimulated Brillouin scattering in fibers is reduced by the ratio of the laser bandwidth over bandwidth of the scattering process [23]. If this argument also holds for Raman scattering with its approximately 10 THz bandwidth, pulse durations of <20 fs are to be preferred over 100 fs pulse durations and explain a potential loss of phase coherence in the continuum. With an input bandwidth of 100 THz or more, no detrimental dephasing from Raman contributions is expected.

![Modal flux density of a white-light continuum. Data was taken from [22], but converted to units photons per second and mode. A 100-MHz repetition rate and 30 mW launched into the fiber were assumed. Even though peak values are below those in Fig. 5.9, the continuum stretches over nearly two optical octaves with enough flux to realize any of the schemes discussed in Section 5.3.1 and 5.3.2.](image)

Similar effects may arise from other high-order processes, e.g. multi-photon-ionization (MPI) processes [24], which have been observed at higher pulse energies in bulk continuum generation. With the relatively low pulse energies in the fibers, these processes are not expected to play a role here. These processes, however, may well play a role in CEO phase measurements at amplified systems, which utilize continuum generation in hollow-fibers.

Another important issue is amplitude-to-phase coupling in the continuum generation
process. The physical mechanism behind this coupling will become much clearer in the experimental analysis in the next section, suffice it to say that a fluctuating peak power in the spectral broadening mechanism of self-phase modulation directly translates into a fluctuating group-phase offset. Now, depending on the experimental scheme, this can either happen inside [21] or outside [16] the cavity. Inside the cavity power fluctuations will change \( f_{\text{CEO}} \), i.e. the induced phase change cumulates. Outside the cavity, the pulse GPO is only influenced in a single passage through the nonlinear medium. Extracavity broadening therefore does not affect the average values of the frequency comb parameters \( f_{\text{rep}} \) and \( f_{\text{CEO}} \). On short time scales, deviations from the average values may well be apparent and cause fluctuations of the CEO phase. An analysis of this problem will be presented in the following Section.

In conclusion, the two major experimental constraints of avoiding phase-destroying mechanisms and generating sufficient bandwidth are most easily met by employing white-light continuum generation in microstructured fibers. The advanced schemes of Section 5.3.2 could be used instead of the additional spectral broadening, but definitely increase experimental complexity with their need for additional single-mode lasers. Generally, spectral broadening outside the laser is to be preferred over [24] excessive intracavity broadening because of amplitude-to phase conversion mechanisms. As all sub-100-fs lasers rely on the Kerr effect, the coupling mechanism cannot be totally avoided, but need to be kept at the minimum level. This will be addressed in more experimental detail in Section 5.4.

5.3.4 CEO phase and frequency stabilization schemes

So far only measurement of the CEO frequency or phase has been addressed. For many applications, it is desirable not only to determine the CEO phase but also to control it and set it to a desired value. On top of a measurement scheme, this requires a servo loop that adjusts the CEO frequency inside the laser and a mechanism to control the CEO frequency inside the laser cavity. For the latter purpose, tilting the end mirror after an intracavity prism sequence has been suggested [25]. The different Fourier components of the laser pulse are geometrically separated at this location. Tilting the end mirror therefore induces linear dispersion, which is equivalent to a group-phase offset. Of course, this tilt has to be negligibly small compared to the numerical aperture of the beam at this location. The travel range is nevertheless more than sufficient to change the CEO-frequency by one free spectral range of the laser cavity. For this a tilt on the order of 10 \( \mu \)rad is required [26]. The advantage of this scheme is that the CEO frequency can be influenced without affecting other laser parameters. If the pivot point of the laser tilt assembly is carefully chosen at the center of gravity of the spectrum, the repetition rate will stay unaffected by the tilt. However, tilting the mirror requires mechanical action and is ultimately limited to a few kHz. In some early stabilization experiments a bandwidth of less than 300 Hz was reached, which is insufficient to counteract the CEO fluctuations in such a laser. Therefore, electro-optic [27] or acousto-optic schemes, as they will be explained in the following sections, have to be preferred.

In literature, several different approaches to stabilization of the carrier-envelope have
been described [21, 27, 28]. It is very important to distinguish between simple frequency locks and phase locks. A frequency lock can be relatively easily implemented by converting the CEO-frequency into a proportional voltage and then forcing this voltage to a desired value. Such a lock can still be realized even if the oscillator to be stabilized has excessive frequency fluctuations. The problem with such a lock is that a small offset in the generated voltage will accumulate into large deviations of the phase, as will become clearer in the discussion in the following section. Clearly, this is not acceptable for metrology purposes, where the number of cycles has to be evaluated over long periods of time. A phase lock, as depicted in Fig. 5.11, generates a voltage proportional to the phase difference between the CEO-frequency and a reference signal generator. While this allows for much tighter tracking of the CEO-frequency, such a scheme cannot compensate for sudden phase distortions larger than $\pi$. Therefore particular care is required for maximum bandwidth available for the servo loop and a minimization of noise generating mechanisms in the laser itself.

Fig. 5.11: Schematic illustration of the phase lock utilized in this research. The APD detects the beat note generated with one of the schemes described above. Spurious components such as intermode beats are filtered out, and the signal is then mixed with a reference oscillator. The amplitude of the generated mixing signal is carefully adjusted to preserve the phase margin of the servo loop. The signal is then fed into an acousto-optic modulator that utilizes amplitude-to-phase coupling in the laser as a control mechanism for the CEO frequency. In principle, other mechanisms can be used. Care has to be taken for sufficient bandwidth of the scheme. The depicted scheme was found to work well up to frequencies of 30 kHz.

5.4 Experimental characterization of carrier-envelope offset noise in unstabilized and stabilized oscillators

From the considerations in Section 5.3.3, it is clear that with additional broadening in microstructure fibers, the spectral span is more than sufficient for implementing the scheme from Section 5.3.1 or 5.3.2. With the octave-spanning spectra, the simplest variant employing fundamental and second harmonic can be utilized ($N=1$ and $M=2$). To further investigate the behavior of the carrier-envelope offset phase in Ti:sapphire lasers the heterodyne detection scheme of Sect. 5.3.1 is set up, see Figs. 5.2 and 5.12. In most of
For frequency-doubling of the long-wavelength components at 1060 nm a 1-cm long LBO crystal is used. Noncritical phase matching of SHG to yield 530 nm is achieved at a crystal temperature of 155 degrees Celsius. Given the 1-cm length of the crystal, one calculates a phase-matching bandwidth of about 4 nm, accommodating about 40,000 laser modes. Fundamental and SHG components at 530 nm are heterodyned in a Michelson interferometer. Careful optimization of the arm lengths in this interferometer is required for detection of the beat signal. Note that all modes within the phase-matching bandwidth contribute to the beat signal. Experimental parameters of the laser were set to ensure maximum intensity and stability of the continuum components at 1060 nm and at 530 nm.
Figure 5.13 shows a typical rf spectrum of the signal detected by the avalanche photodiode in Fig. 5.12. The CEO beat notes are clearly visible at 35 and 65 MHz at 45 dBc in a 300 kHz bandwidth. It needs to be pointed out that the quality of this signal is the most important prerequisite for any attempt to lock the carrier-envelope offset. In the unstabilized laser with prism dispersion compensation the frequency of this beat note may change very rapidly by up to several MHz in one second. This tendency is already greatly reduced by enclosing the laser in a box and carefully avoiding atmospheric turbulence. Excursions of the CEO frequency are further decreased by switching over to a prismless laser set-up. With these improvements, the CEO frequency stays within a 500-kHz interval for minutes of observation and without any active stabilization.

![RF laser spectrum containing CEO beats at 35 and 65 MHz as measured by the APD in Fig. 5.12. Resolution bandwidth was set to 300 kHz. The spectrum also shows some spurious components caused by intermodulation in the avalanche photodiode. These spurious components did not affect the measurements.](image)

For a more thorough characterization of the fluctuations of $f_{CEO}$ in the free-running oscillator, the frequency $f_{CEO}$ is electronically converted into a proportional voltage. The control voltage of the phase-locked loop of the frequency-to-voltage converter is then analyzed by a dynamic signal analyzer (HP3562A). This allows one to determine the single-sideband frequency noise density $\sigma_{f_{CEO}}(f)$ in units Hz / $\sqrt{\text{Hz}}$ as a function of offset frequency $f$. Figure 5.14 depicts measurements of the CEO noise of the unstabilized laser with intracavity prisms and the prismless laser with and without stabilization. All measurements are composed of several sweeps with different spectral resolutions and are combined in a logarithmic plot, covering the range from 1 Hz to 100 kHz. The noise spectra typically show some discrete components at line frequency harmonics and a broad background reaching up to several kHz offset frequency. The laser with intracavity prisms shows by far the worst noise behavior with a pronounced maximum centered at about 500 Hz. The prismless laser shows a more than 10 times improved passive stability. A further reduction of the noise can be achieved with an active stabilization. This servo loops utilizes the dependence of the CEO frequency on intracavity peak power and adjusts this quantity by regulating the pump power with an
acousto-optic modulator. The physical mechanisms behind this servo loop will become clear below.

Fig. 5.14: Frequency noise spectrum of the CEO frequency. Shown are three traces: Unstabilized laser with intracavity prisms (top trace), laser using only chirped mirrors for dispersion compensation with (bottom trace) and without (middle trace) active stabilization. For active stabilization, the scheme shown in Fig. 5.11/5.12 was used. Note that the bottom trace is already very close to the detection limit for most of the range shown.

From the measured frequency noise density we calculate rms values using

\[ \delta f_{\text{CEO}} = \sqrt{2 \int_{f_{\text{low}}}^{f_{\text{high}}} \sigma_{f_{\text{CEO}}}^2(f) \, df} \quad (5.15) \]

Ideally, the integration should be extended up to the repetition rate of the laser. As can be seen in Fig. 5.14, the noise rolls off very rapidly at high frequencies, and the 100-kHz bandwidth of our analyzer does not preclude significant noise contributions to Eq. (5.15). Integration over the entire range displayed in Fig. 5.14 yields \( \delta f_{\text{CEO}} \approx 100 \) kHz for the unstabilized laser with prisms. This is reduced to 10 kHz in the prismless set-up and to 0.7 kHz with activation of a phase-lock to a reference oscillator (compare Fig. 5.11/5.12). The increase of noise at frequencies above 30 kHz agrees well with the measured bandwidth of the servo loop. Using Eq. (5.3) one can now estimate typical pulse-to-pulse jitters of \( \Delta \phi_{\text{CEO}} \) and finds that they are negligibly small compared to \( \pi \). For most applications in extreme nonlinear optics, however, it is mandatory to estimate fluctuations of \( \phi_{\text{CEO}} \) on much longer time scales than the roundtrip time. One example could be the temporal delay between firing the flash lamps of a pump laser in an amplifier and selecting a pulse with a particular CEO phase for amplification in a pulse picker. In this case, one needs to be able to predict the phase evolution of the CEO for,
e.g., several 10 µs, i.e. an $f_{\text{low}}$ of a few 10 kHz. Other experiments may demand for longer intervals of a stable phase of seconds or even minutes and accordingly lower values of $f_{\text{low}}$. An estimation of the rms phase jitter is possible by calculating from the measured noise data

$$
\delta \phi_{\text{CEO}} = 2\pi \int_{f_{\text{low}}}^{f_{\text{high}}} \sigma^2(f) \, df = 2\pi \sqrt{2 \int_{f_{\text{low}}}^{f_{\text{high}}} \left( \frac{\sigma_{\phi_{\text{CEO}}}(f)}{f} \right)^2 \, df}. \quad (5.16)
$$

As the lower bound of the integration is dictated by the time that an application requires a stable CEO phase, Eq. (5.16) is plotted as a function of $f_{\text{low}}$ with fixed upper bound ($f_{\text{high}}=100$ kHz). The integrated phase jitter $\delta \phi_{\text{CEO}}(f_{\text{low}})$ is displayed in Fig. 5.15 for the unstabilized lasers of Fig. 5.14.

![Fig. 5.15: Integrated CEO phase noise spectra as a function of lower integration bound $f_{\text{low}}$. The top two traces for the unstabilized lasers (intracavity prisms, dashed and prismless, dotted) are deduced from the data in Fig. 5 employing Eq. (5.16). The bottom trace displaying the much lower phase noise of the stabilized laser (solid) is measured with an rf lockin using the same reference oscillator as for stabilization. The upper cutoff $f_{\text{high}}$ is 100 kHz in the top two measurements and 10 kHz in the bottom measurement.](image)

This plot clearly reveals the dominant contributions to the rms phase jitter as steps. For the unstabilized lasers, the most severe noise contributions are centered at 1 kHz. Integration over the entire frequency range, e.g., yields an rms phase jitter $\delta \phi_{\text{CEO}}$ of 10,000 rad in the unstabilized laser with intracavity prisms. For characterizing the quality of the phase-lock a different method is used. When deviations of the phase from the desired value are small, the phase jitter can be directly and very sensitively evaluated with an rf lockin amplifier. For measurement of the bottom trace in Fig. 5.15 a Stanford Research SR844 lockin amplifier provides a phase-proportional voltage at an update rate of 20 kHz and allows to directly evaluate $\sigma_{\phi_{\text{CEO}}}(f)$ in Eq. (5.16). In particular, this
method also extends the noise measurements to well below 1 Hz. The phase noise
measurements provide an independent proof of the successful phase lock and indicate
\( \delta \varphi_{\text{CEO}} = 0.02 \) rad for the frequency range from 0.01 Hz to 10 kHz. At the high frequency
side, these measurements can be augmented by the less reliable data derived from
frequency noise density measurements, which provide an upper estimate of
\( \delta \varphi_{\text{CEO}} \leq 0.2 \) rad.

It needs to be pointed out that the noise characterization presented here is fully
independent of the stabilization circuitry, i.e., the servo loop error signal is not used to
extract that information in a somewhat simpler but less reliable way. Similar
measurements of the CEO phase noise were reported in [27]. These authors also
established a phase lock, but did not further characterize residual phase jitter for low
frequencies. They estimate \( \delta \varphi_{\text{CEO}} \leq 0.3 \) rad for frequencies above 100 Hz from a
measurement of pulse energy fluctuations. This method, however, presupposes a strict
correlation of power and CEO frequency in the laser. Recently, stabilization of the CEO
phase of two independent lasers was also reported [28]. Those authors reported loss of
interference fringe contrast between the two lasers, i.e. a \( \delta \varphi_{\text{CEO}} \) on the order of \( \pi \), at
observation times below 20 ms corresponding to \( f_{\text{low}} = 50 \) Hz. Apart from methodical
differences in the measurements, these examples show that so far negligible \( \delta \varphi_{\text{CEO}} \ll \pi \)
was not reported for extended observation times of seconds or more.

The difficulties experienced in reaching a long-term stabilization of the CEO phase
and the strong difference between the noise in lasers with intracavity prisms and
prismless lasers clearly demonstrate the necessity of reaching a deeper understanding of
the underlying physical processes causing fluctuations of the carrier-envelope offset. This
will be addressed in the following Section.

5.5. The physical origin of carrier-envelope fluctuations

Several coupling mechanisms between carrier-envelope offset phase and intracavity
power have been proposed. One of the main applications of such a mechanism is the
ability to control \( f_{\text{CEO}} \) externally by adjusting the pump power. Xu and coworkers
observed a power-dependent shift of the mode-locked spectrum [7]. The influence of this
spectral shift on the carrier frequency of the pulse was used to explain the power
dependent change of the carrier envelope frequency also observed in these experiments.
Later, this amplitude-to-phase conversion mechanism was explained by self-phase
modulation [31]. An additional coupling mechanism due to beam pointing variations in
the prism compressor was also suspected by Morgner [21]. However, with the excessive
noise of the laser used in this investigation, the authors were unable to provide any
experimental evidence for this coupling mechanism.

To explore amplitude-to-phase coupling mechanisms in the laser, an acousto-optic
modulator (AOM) is introduced between the pump laser and the Ti:sapphire oscillator.
The experiments described in this section have been carried out with the laser with intracavity prisms. The AOM deflects a small portion of the pump power into the first diffraction order while the zeroth order of the AOM is used to pump the Ti:sapphire laser. The drive power to the modulator is periodically modulated with a reference signal, which is also used for phase-synchronous detection of the resulting modulation of \( f_{CEO} \) and beam pointing, see Fig. 5.12. Thermal and environmental contributions to these quantities have been minimized by use of modulation frequencies of several kHz. The CEO frequency is measured as described in the previous section, and the beam position is monitored with a position-sensitive detector (Sitek 1L2.5SP) using one of the residual reflections off the intracavity Brewster prism surfaces.

Using a lock-in technique, the change of the CEO frequency induced by a modulation of the intracavity peak power was carefully measured. In these experiments a change in pulse duration was also monitored but was found to be small. Using several different modulation frequencies, one finds that the CEO frequency changes by \( 5 \times 10^{-4} \) Hz per W/cm\(^2\) change of intracavity intensity. Similarly, modulation of the intracavity power also induces a beam displacement on the order of \( 10^{-15} \) m per W/cm\(^2\) change of intensity inside the laser crystal. The beam movement is measured at the location shown in Fig. 5.12. Note that a 1\% modulation of the intracavity intensity translates into a beam movement of 4.5 \( \mu \)m, corresponding to a pointing variation of several microradians.

It is obvious from Eq. (5.1) that only fluctuations of the linear dispersion \( \frac{dn}{d\omega} \) have a strong effect on the CEO frequency of a laser cavity. Cavity length fluctuations, however, may contribute only by affecting \( f_{rep} \) in Eq. (5.3). Typically, these fluctuations are found to be less than 100 Hz per second in the unstabilized laser. This cannot account for the MHz sweeps of the CEO frequency observed in some of the early measurements without proper environmental shielding. In comparison to the problem of stabilizing the frequency of a continuous single-mode laser, the contribution of acoustic and thermal length variations via displacement of the cavity mirrors is clearly negligible. This leaves dispersion variations of the intracavity elements as the major mechanism causing CEO frequency fluctuations. These variations can be thermally or acoustically induced or they can be caused by nonlinear refraction [31]. For our laser cavity, a change \( \Delta f_{CEO} \approx 1.4 \) MHz for a 1 K temperature change of the laser crystal is estimated [32]. Similarly, one calculates \( \Delta f_{CEO} \approx 20 \) kHz for an air pressure variation of 1 Pa [33]. These mechanisms can be easily shielded or will only cause a slow drift of the CEO frequency. Therefore nonlinear-optical refractive mechanisms are exclusively considered in the following.

The most straightforward idea is to explore a direct change of the refractive index via the Kerr effect. According to Eq. (5.1), however, only the linear dispersion of self-refraction plays a role. This effect is also known as self-steepening [23]. With the formalism developed in [34] one can estimate the self-steepening coefficient of sapphire as \( \frac{\omega \cdot \Delta n \cdot \epsilon}{d\omega} = 8 \times 10^{-17} \) cm\(^2\) / W. This formalism also estimates self-refraction as
The change of the beam orientation inside the intracavity prism compressor is accompanied by a change of the group-phase offset. For the fused-silica Brewster prism sequence with 30 cm apex separation used in the laser, the beam pointing sensitivity of the CEO frequency is estimated as \( \frac{\partial f_{\text{CEO}}}{\partial \vartheta} = 2.5 \times 10^{12} \text{ Hz} / \text{rad} \). While this value has been derived from a full analysis of the spectral phase of the prism compressor [36], the main effect can be easily understood from a change of the effective material insertion in the prism compressor. A change of beam direction, for example, reduces material insertion at the first prism and increases insertion at the second prism. These effects only cancel out if the pivot point of the beam movement is right at the midpoint between the two prisms (compare Figs. 5.16 and 5.17). Finally, combining both calculations of nonlinear beam steering and the pointing sensitivity yields an estimate of
\[ \frac{df_{\text{CEO}}}{dI} = 7 \times 10^{-4} \text{ Hz cm}^2 / \text{W}. \] This number agrees well with the measured value of \[ 5 \times 10^{-4} \text{ Hz cm}^2 / \text{W}. \]

Similar mechanisms based on thermally induced index-changes and their dispersion may also be present but have not been studied here. It is expected that they play a strong role for frequencies below 1 kHz. All amplitude-to-phase conversion mechanisms work both ways: First they allow the control of the CEO frequency of a cavity by simply modulating intracavity peak power. Second, stabilization of the CEO frequency will also strongly suppress pulse energy fluctuations via the described coupling mechanisms. This inverse coupling explains the reduction of pulse energy noise found in a CEO-stabilized laser. It should be pointed out that our explanation of the coupling mechanism via self-steepening is consistent with the spectral shift observed by Xu et al. [7]. Self-steepening is known to cause a spectral asymmetry of the nonlinear refractive broadening [37], even though it may be difficult to give a quantitative estimate for the carrier-envelope offset change expected for a certain spectral shift.

5.6. Implications for maintaining the CEO-stabilization in chirped-pulse amplifiers

As outlined in the previous section, an understanding of the mechanisms behind the CEO-fluctuations allows for the design of oscillators with the least possible coupling of amplitude to phase. For many applications of CEO-stabilized sources, it is necessary to amplify pulses to the µJ or mJ level and maintain the stabilized phase. In the following, we will analyze how the mechanisms that have been found important in the oscillator influence the amplification of pulses.

The most severe effect in the oscillator was found to be nonlinear beam steering in combination with the angular sensitivity of geometrical dispersion schemes. Therefore an estimation of similar effects in amplifier chains lies at hand. External femtosecond amplification schemes nearly exclusively rely on chirped pulse amplification (CPA, [38]). This requires the pulse to be stretched before and recompressed after amplification. For this purpose, grating sequences based on the Treacy compressor have found widespread use [39]. In the standard Treacy grating compressor the group-phase offset induced by the grating phase [40] at the second grating is a particular concern. Beam pointing variations leading to a displacement by a single groove spacing at a grating translate into a 2\pi change of group-phase offset. This effect is equivalent to the additional material insertion in prism sequences induced by beam pointing variations. However, gratings sequences are much more sensitive to a change of beam direction. For example, a typical grating distance of a few 10 cm and a grating period of 1000-2000 grooves/mm restricts tolerable beam movements to much less than a microradian. To analyze the net effect in complex stretcher and compressor sequences, the more illustrative picture of prisms is used in Fig. 5.17. Equivalently, the change of material path in this Figure can always be interpreted as a change of group-phase offset (GPO) in a grating sequence. In Figs. 5.17a and 5.17c, a compressor and a stretcher sequence are schematically drawn.
with the principal center-wavelength ray as a straight line. Angular dispersion in the first prism leads to an increased path length for wavelengths shorter or longer than the principal ray wavelength. This parabolic phase front gives the dominant contribution to the negative second order dispersion of such an arrangement. Angular movement of the principal ray (dotted line in Fig. 5.17b) now changes the group-phase offset at each of the 4 prisms. The material/grating GPO is reduced at prisms 1 and 4 and increased at prisms 2 and 3. From inspection of Fig. 5.17b, it becomes clear that these effects cancel out if \( d_{12} \) is chosen to be identical to \( d_{34} \), with an arbitrary choice of \( d_{23} \). A translational movement (dashed line) does not play a role. The typical compressor arrangement uses only 2 prisms in double pass, which automatically ensures \( d_{12} = d_{34} \). In this case, compressor grating sequences are not expected to be susceptible to beam movement induced effects on the CEO phase. Note that this treatment does not automatically hold for intracavity prism compressors but also depends on the exact cavity configuration of the laser. Also, the sensitivity towards beam pointing changes dramatically as soon as imaging elements are introduced into the prism/grating sequences.

**Fig. 5.17:** Illustration of the beam pointing effects in sequences of angular dispersive elements. The principal center-wavelength ray is drawn as a straight line. The changes of material insertion in this figure can be interpreted as a change of group-phase offset in a grating sequence. 

- **a:** compressor sequence with three rays of different wavelength, as designed. This set-up generates negative second-order dispersion as the outer rays experience an increased optical path length compared to the principal ray.
- **b:** Translated (dashed line) and rotated (dotted line) principal ray in a stretcher. Net material insertion remains constant when \( d_{12} = d_{34} \), i.e. the GPO is not affected.
- **c:** stretcher sequence with three rays of different wavelength, as designed. The telescopes invert the phase front curvature compared to **a**, yielding positive second-order dispersion.
- **d:** Translational (dashed) and rotational (dotted) movement of the principal ray. Beam rotation influences material insertion, resulting in a net effect on the GPO of the stretcher sequence.

A typical stretcher arrangement is inspected in Fig. 5.17c and 5.17d. In a stretcher, the diverging bundle of rays is converted into a converging one by means of a 1:1 telescope. This inverts the phase front curvature of the beam and allows one to generate positive second-order dispersion exactly canceling out the dispersion of an equivalent compressor arrangement. In such an arrangement, the direction of the beam is changed at each of the telescopes when the beam is moved out of centration (see Fig. 5.17d). This applies to both a translational (dashed) or rotational (dotted) movement of the beam. If the pivot
point of the beam movement is exactly known, $d_2$ could be chosen such that the beam always crosses the principal design ray at the location of the rear mirror (dash-dotted line in Fig. 5.17). Knowledge of the geometry of beam pointing variations may then at least allow a reduction of the effect. As several different mechanisms may contribute, it is expected that beam pointing variations still have to be kept in the few-µrad range to avoid degradation of a stabilized CEO phase in a grating stretcher.

The second concern derived from the analysis of the oscillator is amplitude-to-phase conversion via self-steepening. As in the oscillator, this effect seems to be much less of a problem because typically amplifiers are designed such that the $B$-integral, i.e. the cumulated nonlinear phase, is smaller than $2\pi$. Using the numbers derived in the previous section, the total self-steepening contribution to the CEO phase is then expected to be smaller than 0.5$\pi$. This demands a 6% shot-to-shot energy stability of the amplifier to keep effects on the CEO phase below 0.1 rad.

Amplitude-to-phase coupling is expected to be a much more severe problem when additional external compression [41] is employed. Because of the complex nonlinear mechanism in femtosecond continuum generation, it is very difficult to derive a reliable estimate of the severity of amplitude-to-phase coupling. These effects are at least one order of magnitude stronger than the intrinsic nonlinearities of the amplifier itself. This would exclude amplifier systems with strong (>10%) shot-to-shot energy fluctuations but CEO stabilization might still be manageable with amplifiers with state-of-the-art stability.

With the much lower repetition rates of amplifier systems, environmental influences deserve a closer inspection. With the numbers derived in the previous section and assuming a typical path length of 10 cm of Ti:sapphire in the amplifier, a temperature change of 1K would affect a 1.9 rad change of the CEO phase. Similarly, an air pressure variation of 1 Pa would induce about a 1-mrad CEO phase change in a 10m path length.

In summary, stabilization of the CEO phase in a chirped-pulse amplifier is a very challenging problem, in particular because of the severity of beam pointing effects. These effects do not play a role if only plane gratings are considered, as in the compressor. However, as soon as gratings and imaging elements are combined, a severe coupling mechanism between beam pointing and CEO phase is introduced. This explains the observed effects in the oscillator itself, and it disfavors the use of stretcher grating arrangements. Moreover, we also expect 4f shapers for adaptive pulse compression [42] to experience similar problems. The most viable option for avoiding detrimental effects in the stretcher seems to be the use of a bulk material for this purpose, as described in [43]. This solution may not allow for the shortest possible pulse duration but it is insensitive to beam pointing induced fluctuations of the CEO phase. As the pulses have to be compressed in any case to reach the regime of extreme nonlinear optics, the limited pulse duration does not appear to be a major problem. Using a heavy-flint glass stretcher, thermal influences on the CEO phase will require additional care. Thermal changes in the amplifier and nonlinear optical effects in a subsequent hollow-fiber compressor may well add up. At this point, the only practical solution for phase-sensitive experiments with
amplified pulses seems to be monitoring of the CEO phase [44, 45] together with binning of the experimental results.

5.7 A scheme for self-stabilizing CEO phase

Because of the difficulties in stabilizing the phase in an amplifier, an alternative path to CEO phase control may be interesting in particular for low repetition rates systems. This method automatically cancels out fluctuations of the carrier-envelope offset phase on the input pulses. It is based on difference-frequency generation of different spectral parts of broadband continua and generates an output pulse train with fixed carrier-envelope phase. The carrier-envelope phase can be directly electronically controlled without any servo loops. The scheme can be applied both to oscillators and to amplifiers with a few Hertz repetition rate. Use of the self-cancellation scheme may highly simplify experiments on the phase dependence of high-harmonic generation and attosecond pulse generation.

Difference frequency generation between two individual modes $m_1 > m_2$ of one and the same mode comb generates spectral components at

$$\nu_{m_1} - \nu_{m_2} = (f_{\text{CEO}} + m_1 f_{\text{rep}}) - (f_{\text{CEO}} + m_2 f_{\text{rep}}) = (m_1 - m_2) f_{\text{rep}} .$$

(5.17)

Note that the difference frequency comb does not display a carrier-envelope offset at zero frequency, regardless of an offset frequency of $\nu_{m_1}$ and $\nu_{m_2}$. With Eq. (5.17) it is immediately clear that the CEO phase of such a signal remains constant, whatever fluctuations may be present in the CEO phase of the generating signals. This situation is represented in Fig. 5.18. The mode frequencies of the DFG signal are all exact harmonics of the laser repetition rate $f_{\text{rep}}$. Clearly, this simplifies measurements of optical frequencies, as only one parameter of the mode-locked laser needs to be monitored. At the same time, a shift to a desired wavelength range in the infrared may also be achieved. The method is also interesting for applications in extreme nonlinear optics as the CEO phase slippage rate of the generated DFG pulse is zero, i.e. the CEO phase will stay constant.
5. Carrier-envelope offset measurement and control

Fig. 5.18: Scheme for generating a pulse with self-stabilizing CEO phase. Given the fact that all modes within the comb signal are equally spaced by the repetition rate, the generated difference frequency signal consists of exact harmonics of the repetition rate without offset frequency. This may serve to generate a signal with fixed CEO phase, regardless of the fluctuations of the CEO phase of the input pulse.

For some applications, it may be desirable to tune the phase of the CEO phase. This can be accomplished in a versatile way by frequency-shifting one of the two pulses with an acousto-optic modulator. As these devices can only be operated at carrier frequencies of several 10 MHz, operation close to $f_{\text{rep}}$ can be employed for a slow modulation of the CEO phase. Another method may be a dispersive delay. At 800 nm, a path length of 10 cm in air causes a 1-rad change of the CEO phase.

5.8 Conclusion

In this chapter, several methods how to measure and stabilize the carrier-envelope phase/frequency of an oscillator have been introduced. Despite the fact that the fundamental problem is very well known, it took until 1999 before a feasible concept how to access this parameter in a laser was introduced. In the meantime, this measurement concept of heterodyning harmonics from different parts of the mode-locked spectrum has been used in manifold variations, in particular for precision frequency metrology. Stabilization of the CEO frequency is a difficult task and requires understanding of the underlying physical mechanisms and mode-locked laser designs that suppress fluctuations of the CEO frequency to a minimum amount. In this work, several mechanisms were identified that cause a conversion of amplitude fluctuations into a change of the cavity group-phase offset. The most serious concern, both in oscillators and in amplified systems, is a translation of beam pointing variation into a phase change, which is a particular problem of geometrical dispersion compensation schemes. In principle, this effect can be avoided by suitable choice of the dispersion compensating elements. These results strongly suggest the use of prismless dispersion compensation in oscillators and bulk stretchers for chirped pulse amplification if the carrier-envelope...
phase is a concern. Nevertheless, nonlinear effects, such as self-steepening, and environmental effects like air-pressure variations and temperature changes cannot be totally avoided.

From the discussion carried out so far, the impression may arise that amplitude-to-phase coupling effects have to be avoided at any expense. However, the same effect that converts amplitude fluctuations into phase noise can also be used to control carrier-envelope fluctuations. Most suitably, this is done by a fast electro-optic or acousto-optic modulator between pump laser and oscillator. Here we utilized this scheme to establish a phase lock of the CEO frequency to a reference oscillator. A careful analysis revealed a residual jitter of the CEO phase as low as 20 mrad in a 0.01 Hz to 10 kHz bandwidth. This unprecedented low value should allow for long integration times, which greatly enhance the sensitivity of experiments in extreme nonlinear optics. Even though there is no doubt that these effects start to play a role in the two-cycle regime, so far no direct experimental evidence for phase sensitivity has been published. Control of the CEO phase opens a door into a new regime of nonlinear optics that cannot be described anymore employing the traditional envelope/carrier concept and the slowly-varying envelope approximation.

Another interesting aspect of CEO control is synchronization of two lasers. Recalling that the mode comb of any mode-locked laser is fully represented by only two parameters, $f_{\text{CEO}}$ and $f_{\text{rep}}$, two absolutely incoherent laser sources are rendered coherent, when their mode combs are made identical. This requires a servo lock and small phase jitters for both parameters. Clearly, the methods described throughout this chapter offer a new tool for synthesis of optical waveforms throughout the optical frequency range, with a degree of control similar to standard rf and microwave technology. While all previous synchronization efforts have only yielded control within a few 10 or 100 optical cycles, the methods described here enhance control down to fractions of a single optical cycle.

Access to the CEO frequency has also clearly boosted frequency metrology. Measurements of optical frequencies have already been demonstrated with a precision of $10^{-14}$. It is expected that this will be further improved by several orders of magnitude. Other efforts are underway, miniaturizing frequency counters with unprecedented accuracy. While such apparatus filled huge halls a few years ago, they now require merely a few square meter table space and may soon be rack-mounted portable devices. The ongoing quest to higher and higher precision may allow to tackle fundamental physical questions, such as the hypothesis of a small drift in elementary constants (e.g. the Rydberg constant). With the current precision, a test of this hypothesis would still require several years. Soon, with potential precisions of frequency measurements of $10^{-16}$ and below, this may actually require only a few days. The mode-locked laser, which underwent constant improvements in terms of pulse duration, was never a match to the superstable single-mode lasers used in metrology. With its new function as an optical gearbox it nevertheless made a strong impact in ultraprecise measurements.
5.9 References to Section 5

5. Carrier-envelope offset measurement and control

6. Frequency conversion of ultrashort pulses

So far, the method of direct generation of ultrashort pulses in a Ti:sapphire laser was discussed. With Kerr-lens mode-locking Ti:sapphire lasers can nowadays reliably produce sub-10-fs pulses and found widespread applications. Even though this laser material could only be reliably manufactured with high doping concentrations for some 10 years, it has displaced nearly all other broadband laser materials for generation of the shortest pulses. In the time before Kerr-lens mode-locking, researcher sought laser materials for different ranges such as dyes for the visible and color centers for the infrared and individual methods were developed to generate short pulses with a particular material. This strategy has been nearly completely replaced by starting with pulses from a Ti:sapphire laser and a subsequent wavelength conversion step. This allows to access spectral regions outside the 650 nm to 1100 nm gain bandwidth of Ti:sapphire.

The most straightforward example for a nonlinear optical wavelength conversion process is second-harmonic generation (SHG). This process generates a pulse proportional to the square of the input pulse and doubles its optical frequency. Second-harmonic generation of ultrafast pulses can also be understood in the spectral domain as a spectral autoconvolution of the fundamental spectrum. As an autoconvolution is typically spectrally broader than the fundamental input spectrum, SHG allows for an increase of bandwidth and consequently for a shorter SHG pulse. This, however, requires perfect phase-matching over the entire bandwidth of the pulse. One particular problem in ultrafast SHG is the group-velocity mismatch (GVM) between fundamental and second-harmonic pulse. Because of their unequal group velocities, second harmonic and fundamental pulse will separate while propagating through the crystal. This way, an SHG pulse with longer duration than the fundamental pulse is generated. In birefringent phase matching, this restricts the useful length of the nonlinear crystal to the so-called walk-off length. Typically, GVM limits the useful length of nonlinear optical crystals to about 10 µm for the conversion of a 5-fs pulse in the visible or near-infrared spectral range. While such crystals are extensively used for characterization methods, they suffer from extremely low conversion efficiencies. In this chapter, an alternative approach based on quasi-phase-matching will be introduced.

6.1. Birefringent phase-matching

In a material with $\chi^{(2)}$ nonlinearity, the field of the second-harmonic grows according to

$$\frac{dE_{\text{SHG}}}{dz} \propto dE_{\text{fund}}^2 \exp(-i\Delta k z). \quad (6.1)$$

Without any measures to compensate for the phase velocity mismatch between fundamental driver wave at $\omega$ and the second harmonic at $2\omega$, the two fields will rapidly start to dephase, as depicted in Fig.6.1. The critical length of the dephasing process is called the coherence length

$$L_c = \frac{\pi}{\Delta k}. \quad (6.2)$$
where \( \Delta k = 4\pi \omega \left( n(\omega) - n(2\omega) \right) / c \) represents the wave vector mismatch between fundamental and second harmonic. Without matching of the phase velocities, the SHG field will increase during propagation only for one coherence length. From there on, the SHG field will be back-converted into the fundamental (see Fig. 6.1), and will sinusoidally oscillate between zero and a maximum value. For commonly used nonlinear optical materials, the coherence length for doubling of visible and near-infrared wavelengths is evaluated to a few microns. Therefore, output intensities are extremely small without any phasematching. It can be immediately seen from Eq. 6.1, that in the case of a vanishing \( \Delta k \), in contrast, the intensity of the second harmonic will exponentially grow.

The classical approach to provide phase matching is birefringent phase matching, as shown in Fig. 6.1. This places, e.g., the fundamental wavelength in the ordinary axis, and the second-harmonic in the extraordinary axis. An example for phase-matching of 800 nm, fundamental to 400 nm second harmonic is shown in Fig. 6.1. While the cut-angle of the crystal is optimized for this particular pair of wavelengths, inspection reveals that, e.g., frequency components at 300 nm and 600 nm are not phase-matched. For ultrafast frequency conversion, one needs to find a criterion to evaluate dephasing in a broadband spectrum. To leading order this effect has been identified as group walk-off. It could be totally avoided, if not only the indices of refraction, but also their slopes were matched. Otherwise, fundamental and second-harmonic pulse have separated by one full width at half maximum \( \tau_{\text{pulse}} \) after propagation of the so-called walk-off length

\[
\ell_{\text{walk-off}} = \frac{\tau_{\text{pulse}}}{\nu_{\text{group}}^{-1}(\omega) - \nu_{\text{group}}^{-1}(2\omega)},
\]
with the group velocity $v_{\text{group}} = c / \left(n + \omega \frac{dn}{d\omega}\right)$. For ultrafast frequency conversion, therefore, the crystal length is typically chosen as the walk-off length or shorter.

![Fig. 6.2: Walk-off length for sum-frequency generation of different pairs of wavelength, as required for ultrabroadband second-harmonic generation of a Ti:sapphire laser. A pulse duration of 5 fs has been assumed. ADP was chosen as one of the least dispersive nonlinear materials. Again the cut-angle was chosen for phase-matching at 800 nm (comp. Fig. 6.1, [1]).](image)

Application of the walk-off criterion to frequency doubling of 5-fs Ti:sapphire pulses is depicted in Fig. 6.2. As the process is very broadband, also non-degenerate cases, i.e. general sum-frequency generation processes, have to be considered. Figure 6.2 displays walk-off lengths ranging from 14 µm to 30 µm. As dispersion generally increases with photon energy, the shortest walk-off lengths are found for SHG of the extreme short wavelengths of the spectrum to be considered. This figure makes it clear, that ultrabroadband frequency conversion demands extremely thin crystals, and therefore results in relatively low conversion efficiencies. In particular, birefringent phase matching cannot be scaled to arbitrarily wide bandwidths without greatly sacrificing efficiency.

Using birefringent phase-matching, blue pulses with durations of 10 fs (Ref. [2]) and 8 fs (Ref. [3]) were generated recently by doubling of ~10-fs pulses from a Ti:sapphire oscillator. In the latter case, focusing the FH tighter than confocally into a 100-µm-thick BBO crystal reduced the effective nonlinear interaction length and alleviated SH pulse broadening owing to GVM. 80 mW of SH light was generated, with 950 mW FH power and an efficiency of 0.7%/nJ. Very recently, a novel technique has been proposed that should allow compressing the fundamental pulse into a few-fs second-harmonic pulse using birefringent type-II phase-matching in a thick nonlinear crystal [4]. This method requires strong depletion of the first harmonic (FH). However, for this method there exists no experimental data in the sub-10-fs regime so far.
Several proposals exist for broadband second-harmonic conversion schemes [5, 6]. These schemes use sophisticated prism schemes to adjust the phase-matching angle with wavelength. These concepts are well suited for tunable continuous wave lasers and eliminate the need for adjusting the crystal. However, they do not compensate for the severe GVM issues in ultrafast optics.

6.2. Quasi–phase–matched second–harmonic generation and compression

Quasi-phase matching (QPM, e.g. [7-10]) is an alternative approach to frequency conversion. As will be shown, this approach allows to overcome crystal length limitations and group-velocity mismatch problems. In principle, this concept is fully scalable with pulse width and inherently only limited by the transparency range of nonlinear optical materials. The general idea for quasi-phase matched second-harmonic generation of continuous wave lasers is shown in Fig. 6.3. Without phase-matching, the second-harmonic light is backconverted to the fundamental in the interval from $\ell_c$ to $2\ell_c$. This effect can be reverted by flipping the sign of the nonlinear coefficient in this range. This possibility was already recognized in the early days of nonlinear optics [7], but discarded as an utterly unrealistic way of achieving phase matching. With modern technology employing very clean nonlinear materials, the sign of the nonlinear coefficient can be manipulated by applying strong electric fields to ferroelectric materials. Examples for nonlinear optical materials that can be microstructured with ferroelectric poling are LiNbO$_3$ and LiTaO$_3$. These materials have been used for microstructuring of the domain structure allowing for a resolution of a few microns.

Fig. 6.3: Growth of the second harmonic intensity along the axis of propagation for ideal phase-matching, quasi-phase matching, and without phase matching (top). The sign of the corresponding nonlinear coefficient $d_{\text{eff}}$ is shown in the bottom.
The resulting growth of a continuous wave second harmonic wave in a quasi-phase matching crystal with a homogeneous grating structure is depicted in Fig. 6.3. In the first half period, ranging from $\ell_c$ to $2\ell_c$, the SH field grows as in the unmatched case. At $2\ell_c$, the sign of the crystal field is inverted, and the SH field continues to grow. At $3\ell_c$, the sign is flipped back to the original, and so forth, flipping the sign of the nonlinear coefficient every $\ell_c$. As a result, the intensity of the second harmonic shows roughly an exponential behavior with propagation distance, with a slight additional modulation on top (see Fig. 6.3). One may now argue that the growth of the SH is reduced compared to the ideal phase matching situation. However, quasi-phase matching still performs favorably compared to birefringent phase-matching because it allows using nonlinearities that are not phase-matchable in birefringent materials. Therefore, quasi-phase matching often greatly enhances the conversion efficiency in frequency-doubling.

The discussion so far concentrated discussed on conversion of continuous-wave light by quasi-phase matching. In this situation a uniform grating is used with grating period $\Lambda$ of the chirped QPM-grating structure adapted for conversion of one particular fundamental optical frequency $w_{LD} = 2p/k_D$, $\Delta k = 2k(\omega) - k(2\omega)$. (6.4)

Here $\Delta k$ is the wavenumber mismatch between FH and SH as imposed by the material dispersion of the nonlinear optical material. In analogy to dispersion compensation by chirped mirrors, the idea for dispersion compensation with quasi-phase matching is clear at hand: If one succeeds in localizing the conversion process along the axis of propagation, the long-wavelength parts of the input spectrum will be converted into their respective SH at a different position inside the crystal than the short-wavelength part of the spectrum [11, 12]. This requires a chirped grating structure, rather than a periodic. Further, if one succeeds in engineering the total group delay experienced in all generation and propagation processes for any given Fourier component, one can compensate for dispersive effects in the material itself and one also compensate for phase distortions on the input pulse. This method is called quasi-phase matching pulse compression [11, 12] and will be explained in the following.

### 6.3 QPM-SHG pulse compression

#### 6.3.1 Simple time-domain picture

QPM-SHG pulse compression can be most intuitively explained in the time-domain under assumption of spatial localization of the frequency conversion process [9-13]. This assumption means that any given wavelength of the input pulse is frequency-doubled at one particular position inside the crystal. It has been shown that this treatment holds under a more rigorous analysis of the nonlinear propagation and conversion and that it allows to correctly compute the spectral phase of the output pulse [14]. Let us now start
with the additional simplifying assumption that material dispersion beyond GVM is negligible. We will later relieve this assumption and extend the discussion to arbitrary dispersion order.

Figure 6.4 schematically illustrates the basic principle of QPM-SHG pulse compression in the time domain. In this example, it is assumed that a positively prechirped fundamental pulse is to be converted into a transform limited SH pulse. The FH pulse enters the QPM crystal from the left and propagates at the fundamental group velocity. First the long wavelengths in the front section of the pulse become frequency-doubled as soon as they reach the position where the local QPM grating period phase-matches this process. The frequency-doubled components propagate at the SH group velocity, which in this case is assumed to be smaller than the fundamental one. Because of this difference in propagation speed, the short wavelengths in the trailing edge of the input pulse catch up with the previously frequency-doubled portions. If the chirp of the QPM grating matches the chirp of the input pulse, the SH of this short wavelength components can be made to exactly temporally overlap the previously generated SH pulse. As a result, one observes a compressed SH pulse at the output of the QPM crystal.

Converting a positively chirped fundamental into an unchirped SH pulse is only one of many different chirp configurations that can be achieved with this approach. Figure 6.5 represents several situations in the Fourier domain. It needs to be pointed out that a linear chirp on both, the frequency of the fundamental pulse and the grating period are equivalent to parabolic phases in the Fourier domain. A positive chirp (i.e. a concave phase) on the input pulse has to be cancelled out by a negative chirp (i.e. a convex phase) on the \( d_{\text{eff}} \) of the QPM grating. Both phases add, resulting in an SH pulse with flat phase
Nonlinear frequency conversion of few-cycle pulses

(top row in Fig. 6.5, this situation is identical to the one represented in Fig. 6.4). Inverting both chirps, as shown in the second row of Fig. 6.5 yields the same result. Finally, engineering of the grating chirp also allows to generate pulses with a desired phase, which can be used to precompensate for additional material in the SH beam path (bottom row in Fig. 6.5).

The possibility of designing such chirp configurations is especially interesting because conventional dispersion compensation schemes generally fail for wide bandwidths and short wavelengths. It is important to note here that the magnitude of the SH chirp does not affect the conversion efficiency, which for confocal focusing depends solely on the FH chirp and material properties [15]. Finally, it is not only possible to design the phase response of a QPM-SHG pulse compression device but one can control the spectral amplitudes of the SH pulse by adjusting the duty-cycle of the QPM domains. The discussion so far presented a simplified analysis of the QPM pulse compression, which was only carried up to the second-order phase of the pulses and also neglected intrinsic dispersion of the QPM device. In the following Section, the discussion will be generalized to arbitrary dispersion, including effects in the device itself.

Fig. 6.5: Schematic illustration of QPM pulse compression in the Fourier domain. The left column shows the phase of the input pulse, the middle column the phase of the grating structure. Note that a linear chirp on the grating frequency is equivalent to a parabolic phase. The phase of the output SH pulse is shown in the right column. Shown are three cases, row by row. (i) A positively chirped input pulse together with a negatively chirped grating gives an unchirped SH pulse. (ii) Reverting both, the grating and the FH pulse chirp gives again a flat phase of the SH pulse. (iii) Using the grating structure of (ii) with an input pulse of less chirp yields an output pulse with positive chirp. This way output pulses can be generated, which are precompensated for propagation through additional material.

6.3.2 Analytical QPM grating design theory

A rigorous theory of QPM-SHG pulse compression in the regime of negligible higher-order dispersion at the FH was given in Ref. [15]. This theory was recently extended to account for arbitrary dispersion [14] and thus enables the application of this technique to
much shorter pulses. In the following this design procedure will be presented in an abbreviated form. This design procedure relies on the undepleted pump approximation, the slowly varying amplitude approximation, and the localization of the conversion process. Furthermore, the equations are given for a noncritical beam geometry, i.e. for collinear wave-vectors and energy-flow. This is the geometry that is usually chosen with QPM. We define the function $z_0(\omega)$, describing the position of conversion of a particular Fourier component of the SH pulse. Equation (6.4) allows to calculate the grating period $\Lambda(z_0)$ from $z_0(\omega)$, i.e. knowledge of $z_0(\omega)$ and the dispersion of the nonlinear optical material fully defines the grating design function $\Lambda(z_0)$. The goal is now to find a function $z_0(\omega)$ such that the total group delay experienced during propagation and conversion yields the desired spectral phase of the second harmonic (up to an unimportant constant phase).

Calculation of the total group delay has to include the phase of the input pulse, propagation of the nonlinear polarization driven by the fundamental pulse up to the conversion point $z_0$ and the linear propagation of the SH light itself. The spectral phase of the SH components generated at $z_0$ can be calculated from the phase of the nonlinear polarization $P_{NL}$, which can be written as a spectral autoconvolution

$$P_{NL}(z_0, \omega) = \int_{-\infty}^{\infty} E^{(\text{fund})}(z = 0, \omega') E^{(\text{fund})}(z = 0, \omega - \omega') \times$$

$$\times \exp[-i(k(\omega') + k(\omega - \omega'))z_0]d\omega'.$$

(6.5)

Here the dispersion of the nonlinear optical material is included as $k(\omega) = \frac{dn}{d\omega}$ to arbitrary order. An arbitrary prechirp on the FH pulse is considered in the phase of the complex input field $E^{(\text{input})}(z = 0, \omega)$. From the conversion point $z_0$ on, dispersion acting on the generated SH has to be added, giving rise to the total group delay, i.e. the first derivative of the spectral phase with respect to angular frequency, at the output facet of the device

$$\tau_G(z_0, \omega) = \frac{\partial}{\partial\omega} \text{Arg}[P_{NL}(z_0, \omega)] + \frac{\partial k(\omega)}{\partial\omega}(L - z_0).$$

(6.6)

Selection of a particular dependence $z_0(\omega)$ now allows to calculate the group delay of the generated SH pulse from Eq. (6.6). This relationship can be inverted to determine $z_0(\omega)$ for a desired spectral phase of the SH output by demanding $\tau_G(z_0(\omega), \omega) = \frac{\partial}{\partial\omega} \Phi^{(\text{SH})}(\omega)$. To obtain a transform limited SH output pulse, for example, the relation $\tau_G(z_0(\omega), \omega) = \text{const.}$ must be fulfilled. As already indicated, the inverse function $\omega(z)$ can finally be used to compute the poling period $\Lambda(z)$ from Eq. (6.4).
This design procedure allows compensating for an arbitrary chirp on the input pulse and for arbitrary material dispersion of the nonlinear material. It requires knowledge of the spectral phase of the input pulse and, more importantly, precise data for the dispersion of the materials employed. In practice, knowledge of the input spectral phase is not crucial because it is a good approximation to assume that the QPM grating adds a given phase independent of the input pulse [14]. In closing, it should be noted that the design procedure can easily be generalized to sum-frequency generation of pulses with adjacent spectra and reverted for the opposite case of difference-frequency generation.

6.4 Experiments

QPM-SHG pulse compression was first theoretically and experimentally demonstrated in the regime of negligible dispersion beyond GVM [11, 13]. Such nonlinear devices soon found application in chirped-pulse-amplification based fiber amplifiers [16]. These experiments were all performed at pulse durations longer than about 100 fs. In this regime higher-order dispersion could be neglected. Using the generalized theory described in the previous section and derived in Ref. [14], we were able to reduce pulse durations in QPM-SHG pulse compression by more than one order of magnitude.

For the first experimental demonstration of this new theory, a first-order QPM grating was designed that converts a ~20 times positively chirped FH pulse into a compressed SH pulse. This grating was fabricated in a 300-µm-thick congruent LiTaO₃ substrate by electric-field poling. LiTaO₃ was chosen instead of the more common LiNbO₃, because it has a deeper UV absorption edge, which makes two-photon absorption of the SH of less concern. Additionally, small QPM periods can be fabricated in this material. The Sellmeier-equation of LiTaO₃ is accurately known for the UV through infrared spectral range [17]. The group-velocity dispersion (GVD) coefficients of LiTaO₃ are 305 fs²/mm at the FH and 1140 fs²/mm at the SH and hence GVD effects are substantial already for modest path lengths inside the nonlinear medium. The temporal walk-off length for the 8.6-fs pulses at 810 nm employed in our measurements amounts to only ~6 µm. A chirped grating length of 310 µm was used to achieve pulse compression. The local grating $k$-vector was changing nonlinearly with propagation distance, covering QPM periods from 6.5 µm to 1.8 µm. The nonlinearity of this chirp is a direct consequence of the higher-order dispersion terms accounted for in the design procedure. The grating design is depicted in Fig. 6.6. For this grating the expected conversion efficiency was ~2 %/nJ assuming confocal focusing and ideal poling quality. Other gratings, designed for a smaller FH prechirp, offer as much as 20 %/nJ theoretical efficiency, but were not explored in these experiments. This was done in order to avoid potential complications with two-photon-absorption or self-phase-modulation in this first experimental demonstration of our technique.
The experimental setup for the SH pulse generation and characterization is shown in Fig. 6.7. The fundamental pulse source was a Kerr-lens modelocked Ti:sapphire oscillator with a repetition rate of 88 MHz and average power of 280 mW as described in Section 2. The output from the oscillator passed though a stretcher composed of two identical broadband antireflection-coated fused silica prisms (wedge angle of 10°) separated by 1 mm. Variable insertion of the prisms provided an adjustable path length and hence positive chirp on the FH pulses without beam displacement. The beam was loosely focused into the chirped-period poled lithium tantalate (CPPLT) by a 1.42-mm-thick, 21-mm-focal length BK7 lens, to a spot size of 20 µm. Due to the compounded losses of the mirrors used for beam routing and the Fresnel reflection of the uncoated LiTaO₃ sample, only about 90 mW of the laser output arrived inside the CPPLT. The FH pulse acquired a total estimated chirp of ~180 fs² before entering the sample. The generated SH beam was collimated with a concave spherical aluminum-coated mirror of 50 mm radius of curvature. The SH output power was 0.41 mW inside and 0.35 mW after the CPPLT corresponding to an internal conversion efficiency of 0.45 %/nJ. We attribute the difference in efficiency compared with the theoretical value of 2%/nJ to poling imperfections and the loose focusing employed.

For the characterization of the generated SH pulses, the CPPLT was placed in one arm of the crosscorrelation setup. For the FH reference pulse, 8% of the FH power was
split off before the stretcher with a 1-mm-thick fused silica beam splitter oriented at 45° and passed through a delay line mounted on a piezoshaker. The SH beam and the FH reference beam were focused together by a concave 30-cm radius of curvature aluminum-coated mirror into a <10-µm-thick KDP crystal. The generated sum-frequency signal at around 270 nm passed through a Glan-Laser polarizer and an iris to reject the SH and the FH light and was then detected by a solar-blind photomultiplier. The position of the piezoshaker was calibrated with helium-neon laser interference fringes in a separate Michelson interferometer.

Before measurement of the crosscorrelation, the chirp of the FH reference pulse was minimized at the location of the crosscorrelation crystal, which was achieved by replacing the crosscorrelation crystal with a 15 µm thick ADP crystal and adjusting dispersion between the laser and the crosscorrelation setup to maximize the SH signal of the reference. Afterwards the dispersion of the prism stretcher before the CPPLT was adjusted for minimum crosscorrelation width. The crosscorrelation signal was recorded in a single sweep of the piezoshaker with an effective 11-bit digitization (Fig. 6.8). A high dynamic range is required in order to assure proper convergence of the decorrelation algorithms described above. The full width at half maximum (FWHM) of the trace shown in Fig. 6.8 is 12.7 fs.

Fig. 6.8: Measured crosscorrelations (solid curves) on a linear (top) and a logarithmic scale (bottom). In addition to the measured data, the crosscorrelations reconstructed by in the decorrelation procedure are shown (dashed-dotted curve, algorithm employing the amplitude and phase information on the fundamental reference pulse; dotted curve, algorithm employing only information on the spectral amplitude of the reference pulse).

The spectrum of the SH pulses was recorded with a spectrograph equipped with a 600 groove/mm grating and a UV-enhanced CCD camera (Fig. 6.9). The transform limit of this 220-THz-bandwidth spectrum is 4.8 fs. The measured SH spectral shape deviates from the theoretically expected SH spectrum, that is determined by the self-convolution of the FH spectrum, which we tentatively attribute to poling imperfections in the QPM structure. It is important to note that such poling imperfections generally do not affect the phase of the generated SH pulse, but only its amplitude [14], with the latter manifesting itself in the observed lower than ideal conversion efficiency. The origin of the rapidly
varying spectral features is not fully understood but is not necessarily due the QPM nature of the device since a similar behavior has been observed using birefringent phase-matching [3]. We observed no indications of two-photon absorption or self-phase-modulation in the CPPLT.

![Figure 6.9](image)

**Fig. 6.9:** Measured SH power spectrum (solid curve) compared with the theoretically expected spectrum (dashed curve).

The measured data was fed into the decorrelation algorithms described in Section 4.1. The SPIDER measurement of the reference pulse yielded a FWHM duration of 8.6 fs and a transform limit of 8 fs. This measurement is shown in Fig. 6.10 together with the fundamental spectrum. For comparison the fundamental pulse shape as determined by the reduced input-data algorithm of Section 4.1 is also included in this figure. The good agreement between the measured and reconstructed pulse shape is indicative of proper convergence of the algorithm. The reconstructed spectral phase of the SH pulse is depicted in Fig. 6.11. Excellent agreement was observed between the results from the two different algorithms. The rapid small-scale variation on the phase is a direct consequence of the experimental noise on the input data. A Fourier transform of the reconstructed spectral phase together with the measured SH spectrum yields the corresponding temporal pulse shape (Fig. 6.12). The FWHM durations of the SH pulse shapes retrieved by the two algorithms were 5.2 fs and 5.4 fs. The shorter pulse duration was measured with the full SPIDER information on the fundamental input pulse. Without the phase information of the fundamental pulse, the slightly longer pulse duration is determined. To our knowledge, these are the shortest pulses ever generated by SHG.
Fig. 6.10: Fundamental power spectrum (top) and temporal pulse shape (bottom). The temporal pulse shape has been measured using spectral phase interferometry for direct electric-field reconstruction (solid curve). For comparison the fundamental pulse reconstructed by decorrelation algorithm with the reduced input data is also shown (dotted curve).

Fig. 6.11: Spectral phase of the SH pulse as retrieved by the two decorrelation algorithms described in the text. The two phase-functions agree excellently over the full wavelength range covered by the SH spectrum (solid curve). (dashed-dotted curve, algorithm with full input data; dotted curve, algorithm with reduced input data)
6. Frequency conversion of ultrashort pulses

![Image of temporal pulse shapes](image)

Fig. 6.12: Temporal pulse shapes of the SH pulse reconstructed by the two decorrelation algorithms. The pulse duration corresponds to 5.2 fs for algorithm with full input data (dashed-dotted curve) and 5.4 fs for algorithm with reduced input data (dotted curve).

6.5 Conclusion

QPM-SHG pulse compression is an entirely scalable and engineerable approach to the frequency conversion of broadband pulses. No element providing negative dispersion other than the QPM crystal is needed to obtain short SH pulses. Thus, the experimental complexity is even lower than for conventional SHG using birefringent phase-matching. Stretching of the FH input pulse is achieved simply by passing the beam through an appropriate length of dispersive material. Using QPM-SHG pulse compression pulses of less than 6 fs duration could be generated in the blue spectral region. The demonstrated approach allows scaling to even shorter blue pulses by using shorter FH pulses, because bandwidth and phase response are engineerable parameters. Larger conversion bandwidths can easily be obtained by scaling the QPM grating length. This is in strong contrast to birefringent phase-matching, where the crystal length generally limits the available bandwidth.

The conversion efficiency demonstrated so far is on the order of the values demonstrated for ultrashort-pulse birefringent SHG. But the FH pulses used in our experiment were relatively strongly chirped before entering the CPPLT. Efficiency of the SHG process scales with the FH peak power and can be improved by reducing the FH chirp in the grating design. Efficiencies of ~20%/nJ are predicted for such designs. A particularly attractive design allows for the generation of negatively chirped SH pulses. These pulses can then be compressed externally to the nonlinear medium by passing through an appropriate length of positive dispersion material. An increased SH output chirp reduces the risk for two-photon-absorption and, thus, should enable the scaling to higher output powers. QPM-SHG pulse compression opens up a new perspective for frequency conversion of ultrashort laser pulses. Beyond the demonstration of sub-10-fs pulses in this paper, this method can be adapted through a wide range of pulse energies, wavelengths, bandwidths, and chirp configurations.
6.5 References to Chapter 6

7. Summary

Ti:sapphire lasers offer a gain bandwidth ranging from 650 nm to well above 1000 nm. In this work, nearly the entire bandwidth of this material could be exploited for the generation of sub-6-fs pulses. Two important mechanisms for maximum performance of the Kerr-lens mode-locked laser have been identified: spectral shaping upon output coupling and intracavity white-light generation. These mechanisms offer a means to extend mode-locked operation even beyond the limitation of gain bandwidth.

A new generation of chirped mirrors was introduced for dispersion compensation. The approach of back-side coating (BASIC) the chirped mirror structure inherently avoids the impedance matching problem of previous chirped mirror generations. It holds the potential to achieve dispersion compensation for spectra covering more than one optical octave, which makes this technique particularly interesting for compression of white-light continua. The BASIC design approach was carefully analyzed theoretically and then applied to the example of the mode-locked Ti:sapphire laser. This yielded pulses of 5.8 fs duration, which at the time are still the shortest well-characterized laser pulses from an oscillator.

Measurement of such short pulses is as challenging a task as the generation process itself. Several methods were tested to measure the Ti:sapphire laser pulses. The earliest approach to this problem was autocorrelation together with a deconvolution procedure. Even though this method was found to work, other methods such as frequency-resolved optical gating (FROG) and spectral phase interferometry for direct electric-field reconstruction (SPIDER) were found to be more reliable. The FROG technique was adapted to the few-cycle regime in a collinear variant based on type-II phase matching. This allows one to avoid the geometrical smearing artifact of noncollinear approaches. SPIDER was also adapted to the sub-10-fs regime. A spatially resolved variant of SPIDER was employed, which allows one to resolve the effects of the spectrally dependent mode size. Another variant was developed that is based on cross-correlation that is of particular interest for characterization of visible or uv femtosecond pulses. Finally, a careful comparison of the different techniques showed that SPIDER is a particularly well-suited characterization method for the sub-10-fs range, combining accuracy, acquisition speed, and experimental simplicity.

With pulses as short as two optical cycles, the relative phase between carrier and envelope starts to become an important parameter in optics. The method described in this work was the first feasible proposal to measure this entity and has now found widespread acceptance. Based on this measurement scheme, which relies on harmonic generation out of different part of the mode-locked laser comb and subsequent heterodyning,
stabilization of the carrier-envelope offset phase was realized. Mechanisms introducing a phase-jitter of this quantity in the laser were carefully analyzed and could be partly eliminated in a redesigned laser. This laser could be phase-locked to a reference oscillator with a residual rms phase jitter of only about 20 mrad. This was actually the first demonstration of a low-jitter phase lock over extended observation times of minutes. Based on the experience with the oscillator several important conclusions were derived concerning the construction of amplifiers with low carrier-envelope offset phase jitter and a self-stabilizing scheme to generate phase jitter-free pulses out of noisy pulse trains without any need for active stabilization.

In the final part of this thesis, a novel method is described to generate short pulses by frequency doubling them in an aperiodic quasi-phase matching grating structure. After reviewing the theory, an experimental demonstration yielding pulses as short as 5.3 fs was presented. The pulses generated in this new approach to femtosecond pulse compression and dispersion compensation clearly set a new record for the shortest pulse ever generated by second-harmonic generation and at oscillator repetition rates in the blue spectral range.
8. Outlook

Research in ultrafast optics has always been driven to reach shorter and shorter pulse duration. While this trend will certainly continue, a point is reached where technological improvement of individual lasers or mode-locking techniques will only cause marginal improvements. The example of the Ti:sapphire laser discussed in this work already exploited nearly the entire bandwidth of the gain material, yielding about two optical cycles pulse duration. More bandwidth can only be accessed by intracavity or extracavity continuum generation. This may slowly push pulse durations below two optical cycles. Much shorter pulses potentially in the attosecond regime can only be generated going to shorter wavelengths, which is one of the current trends in ultrafast research.

A trend that received somewhat less attention than the quest for the shortest pulse is the increased degree of optical waveform control achieved in the last few years of ultrafast research. Both the work on full characterization methods and the work on carrier-envelope phenomena greatly improved the access to the optical phase of a femtosecond pulse. Employing these two novel measurement tools in femtosecond optics, one can truly achieve optical waveform synthesis. First and foremost, this has a strong impact in frequency metrology and allows the measurement of optical transitions to unprecedented precision. The femtosecond laser may well serve as an “optical gearbox” in future generations of atomic clocks that are 1000 times more accurate than the current microwave standard. This may also give access to a more direct detection of the suspected drift of fundamental constants within reasonably short observation times. This “optical gear box” allows for phase coherent links between rf signals and optical waves, introducing the concept of phase-locked loop frequency multiplication into optics. On the more practical side, the same means of control also allow one to combine incoherent laser sources to coherent superpositions. On the conjugate side, timing control of laser pulses on the sub-cycle level appears possible. This would mean a reduction of several orders of magnitude compared to conventional schemes.

The first generation of femtosecond lasers was truly only a laboratory tool that often required several skilled operators who had to devote most of their time for maintenance of the dye laser system. Application of these sources therefore was mainly restricted to pump-probe spectroscopy. With the vast expansion of femtosecond laser sources over the entire visible spectrum and deep into the uv and x-ray range, this field has generated strong new impacts. Particularly fascinating are femtosecond x-ray sources, allowing the structural probing of the solid state with unprecedented temporal resolution. High-harmonic generation, with somewhat smaller photon energies, offers coherent photons at previously inaccessible wavelengths and holds the potential to probe innershell dynamics with attosecond time resolution. Solid-state based femtosecond systems have developed
to a maturity that opens up a new perspective of applications. Use of femtosecond lasers is vividly discussed even in fields that demand uncompromising reliability such as applications in medicine, both diagnostic and therapeutic. Laser material processing with femtosecond pulses will stay a small and specialized technique that will only be used for extreme precision ablation or processing, for microstructuring metals or ceramics or to produce waveguide structures in transparent dielectrics. It is this wealth of previously unthinkable new applications that makes femtosecond photonics a hot topic for years to come.