Chapter 2

All-Solid-State Cavity-Dumped Sub-5-Fs Laser

Abstract

We discuss in detail a compact all-solid-state laser delivering sub-5-fs, 2-MW pulses at repetition rates up to 1 MHz. The laser system employed is based on a cavity-dumped Ti:sapphire oscillator the output of which is chirped in a single-mode fiber. The resulting white-light continuum is compressed in a high-throughput prism/chirped mirror/Gires-Tournois-interferometer pulse compressor. The preliminary pulse duration measurement is carried out by a collinear fringe-resolved autocorrelation. The temporal and spectral phase of the sub-5-fs pulses are deduced from the measured autocorrelation trace and optical spectrum.
2.1 Introduction

Ever since pulsed lasers were invented there has been a race toward shorter optical pulses [1]. Next to the fact that the breaking of any record is a challenge, a major scientific driving force came from dynamical studies showing that ultrashort pulses were essential to the exploration of elementary processes in chemistry, photobiology and physics. For instance, the primary step in bond-breaking reactions (femtochemistry) [2], the rate of electron-transfer in photosynthetic reaction centra [3,4], and the time scales of relaxation processes in condensed phase [5,6] could only be time-resolved with femtosecond excitation pulses. On the road toward femtosecond pulse generation a better grasp of the underlying physics proved to be essential. A milestone here was the development of the colliding pulse modelocked (CPM) laser [7]. When the importance of a careful balance between the group delay and dispersion on pulse formation was recognized [8,9], sub-100 fs optical pulses became feasible [10,11]. Further development ultimately led to the prism-controlled CPM laser [12,13] which delivered pulses of - 30 fs. It was this CPM laser that laid the foundation for many groundbreaking experiments in the past decade, from the observation of wavepacket motion in chemical reactions [14] to the exploration of carrier dynamics in semiconductors [15,16].

Another crucial invention for ultrashort optical pulse generation was the technique of fiber pulse compression [17]. In this method, a relatively long pulse is injected into a single mode fiber. Via the combined action of self-phase modulation [18] and dispersion it becomes spectrally broadened, carrying an almost linear chirp [19]. This spectrally and temporally broadened pulse is subsequently compressed by a pair of gratings [20-22], prisms [10,23] or their combination [24,25] to a much shorter pulse. The compressor’s action is to retard in a well-defined manner the frequencies of the pulse that have advanced. Pulse compression of the amplified output of the CPM laser culminated in the generation of optical pulses of 6 fs (assuming a hyperbolic secant pulse profile) in 1987 [25]. The electric field of such a pulse exhibits only 4.5 oscillations at its FWHM. With these ultrashort pulses photon echoes in solution could be studied for the first time [28-30], while their large spectral width turned out to be very useful for pump-probe experiments in photobiology [31,32].

A new era in ultrafast laser technology began with the development of the fs modelocked Ti:sapphire laser [33] which routinely generates pulses of about 10-15 fs [34-39]. In addition, this laser exhibits low amplitude noise and is extremely reliable. It is not surprising that in the past five–seven years Ti:sapphire based lasers have replaced the CPM lasers in many laboratories as new ultrafast light sources. Sub-10-fs pulse formation from a Ti:sapphire laser also looked promising since the fluorescence bandwidth of the lasing material [40] supports pulses as short as 4 fs. The efforts to construct an oscillator matching the whole bandwidth continue. Just in a

\[^{\dagger}\] The frequently cited number of 3 oscillations [6,25] refers to the duration of the intensity envelope, which, in contrast to the electric field, contains no oscillation at the optical frequency. This misleading notation has also been applied to very recent results [26,27].
couple of years the duration of pulses obtained directly from a laser dropped from 7-8 fs [41-48], what seemed to be a practical limit at the time [44,49,50], to about 5.5 fs in 1999 [26,27].

Despite the fact that a relatively simple oscillator producing very large-bandwidth sub-10-fs pulses seems to be an attractive option, its employment for nonlinear spectroscopy is rather impossible. Here the problem of repetition rate reduction becomes detrimental, since no shot-to-shot sample refreshment is feasible for the pulse trains generated at the typical 80-100 MHz repetition rates. Pulse picking the output of such a laser outside the cavity is very problematic owing to the large bandwidth. For example, the spectral content of such short pulses would be appreciably dispersed in space in case an acousto-optic pulse picker is employed. Alternatively, an electro-optical switch would modify the output spectrum because the polarization of different wavelength components could not be turned for the whole spectral interval simultaneously to the same degree. Additionally, a combination of a Pockels cell plus polarizers introduces a large amount of bulk dispersion, which is difficult to compensate. The repetition rate reduction by up-scaling the cavity length is also unfeasible as it crucially affects the laser stability and makes mode-locking operation more difficult. Clearly, other alternatives should be sought.

With the development of a 13-fs cavity-dumped laser, pulse compression was shown to be a viable route toward pulses of less than 6 fs [51]. Another very promising development was the use of a hollow fiber for spectral broadening of ultrashort pulses [52]. A distinct advantage of a hollow fiber is that it can stand high intensities, allowing pulses of millijoule energy to be compressed.

Another spectacular development took place in the ultrashort pulse generation from non-collinear optical parametric amplifiers pumped by the second harmonic of Ti:sapphire. Utilizing the uniquely broad phase-matching bandwidth of the Type II BBO crystal [53], tunable sub-10-fs pulses have been produced in the visible and infrared [54,55]. The shortest pulses obtained by this technique in the visible measure only 4.5 fs. (See Table 1 in Chapter 1.)

In our early attempts to compress the fiber-chirped output of a cavity-dumped Ti:sapphire laser, we succeeded in the production of about 5-fs pulses at repetition rates of up to 1 MHz [56,57]. While the shortest pulses were attained using a prism–grating compressor, slightly longer pulses were obtained from the higher throughput prism–chirped mirror compressor. It was also suggested that with custom-designed chirped mirrors, shorter pulses with higher pulse energies should be possible. Nisoli et al. recently showed that by using a hollow fiber - 20-µJ, sub-5-fs pulses can be generated at a 1-kHz repetition rate [58].

In this Chapter, we report the generation of sub-5-fs pulses from a cavity-dumped Ti:sapphire laser using a prism/chirped mirror/Gires-Tournois interferometer compressor. Group delay measurements of the generated continuum, which served as input for the design of this novel compressor, are discussed. It is shown that the pulse shape and spectral phase can be determined from the collinear autocorrelation function in combination with the optical spectrum. The similar pulse shape is calculated when the optical spectrum and phase difference between the pulse compressor and continuum is used as input.

Precise knowledge of the amplitude and phase of ultrashort pulses is extremely important in many experiments, especially when dynamics occur on the time scale of the pulse width. An
example is femtosecond photon echo in solution, where explicit use of the pulse shape in calculations of the echo relaxation has shown to be essential [59,60]. Another example is coherent control of wave packet motion and bond-breaking reactions [61-63]. More generally, a detailed description of a molecule–light field interaction requires full knowledge of the electric field. The applicability of the so-called slowly varying envelope approximation in experiments with ultrashort pulses becomes questionable [64]. In fact this approximation may break down and in that case new effects are to be expected.

Chapter 2 is organized as follows. In Section 2.2 we discuss the cavity-dumped laser. Generation of the continuum to be compressed is described in Section 2.3. In Section 2.4 the spectral phase of the white light is discussed, while in Section 2.5 the temporal shape of the continuum is dealt with and compared to calculations based on spectral phase measurements. In Section 2.6 the spectral and temporal shape of the continuum are commented on. Section 2.7 deals with the compressor. In Section 2.8 the pulse duration is measured by fringe-resolved autocorrelation. In Section 2.9 we demonstrate how the amplitude and phase of the compressed pulse can be reconstructed from measurement of the interferometric autocorrelation and the spectrum of the pulse. Section 2.10 outlines experimental uncertainties of the interferometric autocorrelation. Section 2.11 provides a summary and suggests some applications of this compact sub-5-fs 2-MW laser.

2.2 Cavity-dumped Ti:sapphire laser

Figure 2.1 displays the schematic of the self-mode-locked cavity-dumped Ti:sapphire laser used for continuum generation. It represents the next version of an earlier reported design [51]. Compared to the conventional Ti:sapphire oscillator [34,38,65], its cavity dumped counterpart incorporates an additional mirror fold around an acousto-optic modulator [66]. In this way the intracavity pulse energy is stored in a relatively high-Q cavity, which can be switched out of the resonator at any desired repetition rate. The maximal pulse energy of a cavity-dumped Ti:sapphire laser is typically a factor of 10 higher than that from its non-cavity-dumped counterpart. A careful cavity design ensures the Kerr-lens self-mode-locking action is not disturbed by the extra fold and by the added dispersion due to the Bragg cell. The best performance of the system is achieved when the fold mirrors of the cavity dumper are separated by nearly a confocal distance and the mirror fold around the Ti:sapphire crystal is set to the inner edge of the second stability zone [51]. This configuration allows the system to operate under soft-aperture Kerr-lens mode-locking conditions, thus making the oscillator less sensitive to perturbations caused by the cavity-dumping process and mechanical instabilities [51,67-69] than in the first stability zone. Besides, in the latter case the hard aperture needed to initiate the mode-locking reduces the intracavity power.

Compared to the earlier reported design [51] the current version of cavity-dumped Ti:sapphire laser has been significantly improved and presents a more versatile and compact master oscillator. First, the argon-ion pump laser has been replaced by an intracavity doubled Nd:YVO₄ laser (Spectra Physics “Millennia”). The superior beam pointing stability and noise characteristics of this diode-pumped solid-state laser allow the pump power to be reduced to
~4 W. Second, by introducing a high reflector (HR1) in the prism arm, the cavity has been folded, which led to a more compact laser. Third, the output coupler OC has been placed at the non-dispersive end of the cavity, providing an additional output at 82 MHz. Finally, we have saved space by replacing the output (cavity-dumped) pulse pre-compressor, consisting of four prisms [57], with two chirped mirrors [70].

Fig.2.1: Schematic of an all-solid-state sub-5-fs laser. Ti:Sa: 4-mm long Ti:Sapphire crystal (Union Carbide); L1: \( f = 12.5 \) cm lens; M1-M4: \( r = -10 \) cm cavity mirrors; HR1, HR2: high reflectors (CVI); OC: \( T = 2\% \) output coupler (CVI); M5: pick-off mirror (Newport BD2); IP1, IP2: intracavity 69° fused silica prisms; CM1, CM2: chirped mirrors for pre-compression (R&D Lezer Optika); WLG - white-light generator; OAP: 30° off-axis parabolic mirror (Kugler) with low dispersion overcoated silver coating; GTI1, GTI2: Gires-Tournois interferometers (R&D Lezer Optika); CM3, CM4: chirped mirrors for pulse compression (R&D Lezer Optika); P1, P2: 45° fused silica prisms; RM: low dispersion overcoated silver roof mirror (R&D Lezer Optika). The cavity-dump beam that in reality is ejected in the vertical plane is depicted here as being displaced in the horizontal plane. The compressor output beam passes just above GT2. The solid arrow through the OC shows the 82-MHz output used in cross-correlation experiments. The whole set-up occupies a work space of 1×1.5 m² on an optical table.

With a 3-mm thick Bragg cell (Harris), driven by a 5-W electronic driver (CAMAC Systems), the laser dumps 13–15-fs, 40-nJ pulses at a 1-MHz repetition rate. Pulses in excess of 45 nJ are generated when the RF signal is amplified to 16 W using a RF power amplifier (CAMAC Systems). Even higher pulse energies are available at lower repetition rates. Figure 2.2a presents the dynamics of the pulse train inside the cavity seen through the output coupler by a high-speed photodiode. In this illustration, every 82nd pulse is cavity-dumped (rapid drops), which leads to an interval of intracavity energy recovery lasting a few tens of cavity roundtrips. Notably, the oscillations on the intracavity pulse train go on even after the intracavity pulse energy has been fully recovered. This, however, does not affect the pulse-to-pulse stability of the cavity-dumped
output, since the modulation of the intracavity train automatically adjusts itself to the periodicity of RF bursts on the Bragg cell. (Compare the trace levels at the points immediately preceding the two successive RF bursts in Fig.2.2a.) Pulses with energies below 40 nJ can be stably generated with repetition rates reaching to 1.5-1.7 MHz. At even higher repetition rates the period between successive RF bursts becomes insufficient for the full recovery of the intracavity pulse energy, and, therefore, the RF power must be lowered to sustain stable cavity-dumping operation.

The use of the CAMAC RF driver provides an excellent contrast ratio between the preceding/trailing and dumped pulses. The contrast of the pulse switching between the cavity-dumped and the preceding by ~13 ns pulse has been found to be better than 1:1000 (Fig.2.2b). Unfortunately, it is impossible to measure the contrast with the trailing pulse due to electronic noise on the photodiode signal (right-hand part of Fig.2.2b). The given above figure of 1:1000 is similar to the contrast ratio achieved using an electro-optical cavity-dumper [71], be it that in the latter case the repetition rate is limited by ~10 kHz.

![Fig.2.2: Oscilloscope traces of the cavity-dumping dynamics. (a) Intracavity pulse train corresponding to cavity dumping of ~40-nJ pulses at the repetition rate of 1 MHz. (b) A cavity-dumped pulse. The vertical arrow indicates the location of the pulse preceding the cavity-dumping event. The sensitivity of the trace shown in the inset is enhanced 250 times. The numbers below the traces indicate the value of the vertical and horizontal grid, respectively.](image)

### 2.3 White-light continuum generation

The pre-compressed pulses from the cavity dumped laser, with 75 nm spectral bandwidth around 790 nm, were launched into a single-mode quartz fiber (Newport, F-SV, 2.75 μm core diameter) through a 18/0.35 microscope objective (Melles Griot). The optimal fiber length calculated according to Ref. [19] was ~1 mm; however, for practical reasons connected to mounting of the fiber, we used a piece of ~2–3 mm. Angular alignment of the fiber along the longitudinal axis proved necessary to prevent polarization rotation of the light passing through the fiber as a result of chromatic anisotropy. A 3D piezo-driven (Piezo-Jena) fiber positioning stage is used to simplify the alignment procedure. To keep the fiber tip dust-free, a constant flow of dry nitrogen was applied to the focusing area. No damage to the fiber was observed for up to 40-nJ pulses.
Other types of fibers from different manufacturers were also tested; however the ability to withstand high input intensities (~10 TW/cm\(^2\)) seems a unique property of Newport single-mode fibers. As of now, we have no explanation for this phenomenon.

The fiber output is collected by an off-axis parabolic mirror (OAP, Fig.2.1), which ensures achromatic and nearly dispersion-free beam recollimation. The focal length of our custom-manufactured parabola (Kugler) is ~7 mm and the inclination angle to the parabola axis is 30°. Note, that the production of such a mirror with a high optical surface quality is a great technological challenge, since the required post polishing of a diamond-turned parabola is difficult due to the high curvature of the aspherical profile. The OAP is made of aluminum and is coated by a silver- and a low-dispersion protective dielectric coatings (R&D Lezer-Optika, Hungary).

![Fig.2.3](image)

Fig.2.3: Fiber output (solid line) and cavity-dumped laser (filled contour) spectra. The inset shows the pulse obtained by Fourier-transforming the fiber output spectrum assuming constant spectral phase.

The white light continuum, generated by self-phase modulation exhibits approximately a fourfold spectral broadening compared to initial spectrum (Fig.2.3). The optimal pulse energy for injection into the fiber was found to be ~35 nJ, as judged by the quality of the generated continuum. In this case, the pulse energy measured after recollimation of the continuum is about 18 nJ. The long-term stability of the continuum intensity measured at several wavelengths varies from ~0.7% rms at the edges of the spectrum (below 500 nm and above 1100 nm) to less than 0.5% rms near the central frequencies.

The blue-shifted wing of the continuum reaches into the UV, and the red-shifted part stretches into the near infrared, even beyond the spectral cut-off of the silicon detector used for the spectral measurements (Fig.2.3). The shortest pulse attainable by compression of this continuum is obtained by Fourier transformation of the spectrum assuming a flat spectral phase. This yields pulse duration of ~3.7 fs (Fig.2.3, inset). Note that despite the irregular spectrum of the continuum the ideally compressed pulse looks very clean. It is also worth pointing out, that the low intensity wings of the continuum – excluded in the compression scheme to be discussed later – carry enough
intensity for a variety of spectroscopic applications. Moreover, the use of a long piece of fiber enables delivery of the pulse to a remote point in applications where the spectral bandwidth rather than the pulse width is important [72].

2.4 Measurement of spectral phase

Pulse compression aims at the removal of spectral phase distortions accumulated by self-phase modulation and propagation through dispersive media. Precise knowledge of the phase characteristics of a chirped pulse is therefore vitally important to the design of an appropriate pulse compressor. In order to fully characterize a pulse one needs to know its spectrum $I(\omega)$ and spectral phase $\Phi(\omega)$ or its time dependent intensity $I(t)$ and temporal phase $\phi(t)$. The temporal and spectral descriptions are complementary and follow from each other by Fourier transformation. Since no detector is fast enough to resolve the temporal shape of a pulse on a fs time scale, indirect methods have to be used to resolve the exact intensity profile of a fs pulse.

In recent years a number of techniques of indirect phase and pulse shape retrieval have been proposed [73-84]. For instance, in various implementations of frequency resolved optical gating (FROG, see for example [77-79] and Chapter 3 of this thesis) a spectrally dispersed signal of the autocorrelation-type is recorded. When a well-known phase retrieval algorithm is used to analyze FROG traces $I(t)$ and $\phi(t)$ can be recovered.

Another approach to phase retrieval is spectrally resolved up-conversion [25,74-76,81,82,84,85] or down-conversion [86]. In this method, the analyzed pulse (further called probe pulse) is mixed with a well-characterized reference pulse in a nonlinear crystal. The resulting signal is a (spectrally-resolved) cross-correlation. The knowledge of the parameters of the reference (gate) pulse in this case dismisses the fundamental problem of pulse retrieval from autocorrelation or FROG, where the gate pulse is unknown since it is a replica of the pulse to be characterized.

The resulting signal at the sum frequency is dispersed through a monochromator and can be expressed as

$$S(\Omega, \tau) \propto R(\Omega) \Omega^2 \left| \int \tilde{E}_r(\omega) \tilde{E}_p(\Omega - \omega) e^{i\omega \tau} \frac{e^{i\Delta k(\Omega, \omega)L}}{\Delta k(\Omega, \omega)L} \, d\omega \right|^2$$  \hspace{1cm} (2.1)

where $R(\Omega)$ stands for the spectral sensitivity of the detector, $\tilde{E}_r(\omega) = \tilde{A}_r(\omega) \exp[i\tilde{\Phi}_r(\omega)]$ and $\tilde{E}_p(\omega) = \tilde{A}_p(\omega) \exp[i\tilde{\Phi}_p(\omega)]$ are the (complex) amplitudes of the reference and probe pulses, respectively, $\tau$ is a delay between them, and $L$ is the interaction length. Here we assume that the nonlinearity is instantaneous. A non-instantaneous response will be considered in the Chapter 3. The phase mismatch for Type I (oo–e) interaction [87] is given as

$$\Delta k(\Omega, \omega) = k_O(\Omega - \omega) + k_O(\omega) - k_E(\Omega).$$  \hspace{1cm} (2.2)
with \( k \) denoting the wavevectors for ordinary \((O)\) or extraordinary \((E)\) waves. Similarly to FROG, the cross-correlation signal \( S(\Omega, \tau) \) contains explicit information about the complex electric field of the probe pulse, provided that the reference pulse has been fully characterized.

In principle, the knowledge of the reference field permits direct recovery of the probe field through a so-called Wigner deconvolution \([76,84]\). The implementation of a FROG-like iterative inversion algorithm, however, is also well justified \([85,86]\) given experimental uncertainties of the measured cross-correlation spectrogram.

A valuable asset of the spectrally-resolved up-conversion technique is that in some special cases a time-consuming algorithm of phase retrieval can be replaced by a straightforward analysis. For instance, if the spectrum of the reference pulse is sufficiently narrow and its spectral phase is constant, Eq.\((2.1)\) simplifies significantly and becomes:

\[
S(\Omega, \tau) \approx R(\Omega)\Omega^2 \text{sinc}^2 \left( \frac{(k_O(\Omega - \omega_r) + k_O(\omega_r) - k_E(\Omega))L}{2} \right) 
\times \tilde{A}_p^2(\Omega - \omega_r) \tilde{A}_r^2 \left( \tau - \frac{d\bar{\varphi}_p(\Omega)}{d\omega} \right)
\]  

(2.3)

where \( A_r(t) \) stands for the temporal amplitude of the reference pulse. Note that the magnitude of the measured signal is proportional to \( \Omega^2 \) \([88]\) (See Chapter 3). This often omitted factor gains importance with increased spectral bandwidth of the probe pulse.

Eq.\((2.3)\) shows that if the delay \( \tau \) is scanned for a given setting of a monochromator \( \Omega \), the maximum of the up-converted signal directly reflects the group delay of the probe field \([81]\):  

\[
\tau_p(\Omega = \omega_p + \omega_r) = \frac{d\bar{\varphi}_p(\omega_p)}{d\omega}.
\]  

(2.4)

Another important trait of spectrally-resolved up-conversion is that all factors limiting the acceptance bandwidth, like phase-matching or spectral response of a detector, do not influence the position of the maxima. These factors only affect the signal intensity. Furthermore, the phase-matching conditions are also relaxed for spectrally-resolved up-conversion compared to second-harmonic FROG, because the necessary acceptance bandwidth of the crystal is smaller by approximately a factor of two.

The spectral phase of the chirped white light is readily obtained from \( \tau_p(\Omega) \) by integration of Eq.\((2.4)\):

\[
\bar{\varphi}_p(\omega) = \int \tau_p(\omega) d\omega
\]  

(2.5)

The aforementioned technique is valid for reference pulses whose spectral bandwidth are
appreciably narrower than those of the probe pulse. However, by choosing a spectrally infinitely narrow pulse the duration of the up-converted signal becomes infinitely long, limiting the time resolution. In the other extreme limit, when an infinitely short reference pulse is used, the up-converted signal would be detected with an infinitely broad spectrum, limiting the resolution in the frequency domain. Therefore, there is an optimal reference pulse duration, which yields a compromise between temporal and spectral resolutions.

![Fig. 2.4: Normalized probe-reference correlation signals at different wavelengths.](image)

In our experiment we cross-correlated the chirped white light pulse with a laser pulse from the output coupler (shown as a solid arrow through the OC in Fig.2.1) in a 100-µm thick BBO crystal. The importance of having an independent reference beam at 82 MHz repetition rate is derived directly from the Ti:sapphire laser. PMT - photomultiplier tube.

In our experiment we cross-correlated the chirped white light pulse with a laser pulse from the output coupler (shown as a solid arrow through the OC in Fig.2.1) in a 100-µm thick BBO crystal. The importance of having an independent reference beam at 82 MHz from the laser now becomes evident. This pulse has a suitable duration and spectral width for the reference pulse in
the cross-correlation experiment. In order to have this pulse chirp-free, it was passed through a 4-prism compressor. The interferometric autocorrelation of this pulse was found to be in excellent agreement with the one calculated from the pulse spectrum assuming a constant spectral phase.

To measure the group delay across the continuum the up-converted signal was scanned as a function of time delay between the white light and reference pulse at different wavelengths selected by a monochromator. The layout of this experiment is shown in the inset to Fig.2.4. The spectral resolution of the monochromator was ~1 nm. Due to the limited phase-matching bandwidth of the crystal, small angular tuning was necessary to obtain reliable measurements of the infrared and the visible components of the white-light spectrum. Typical normalized up-converted profiles at different settings of the monochromator are depicted in Fig.2.4. The corresponding frequencies of the white light can be obtained knowing the central wavelength of the reference pulse (800 nm). The duration of the up-converted signals increases toward the blue-shifted wing of the continuum. This is explained by a faster change of the spectral phase of the probe pulse within the spectral width of the reference pulse, compared to the relatively slow changing phase in the infrared region, where material dispersion is considerably lower. The modulation appearing in some profiles is due to intensity variations in the spectrum around the central frequency of the continuum (Fig.2.3). The up-converted signals cover the fundamental wavelengths of the white-light from 0.55 to 1.2 \( \mu \)m. Note that the bandwidth of the white-light that can be up-converted stretches much further into the infrared region than can be reliably measured (Fig.2.3) using a silicon photodiode array. This means that the real bandwidth of the white-light continuum as well as the shortest achievable pulse duration (Fig.2.3, inset) are most probably underestimated.

\[ \text{Fig.2.5: Group delay of the white-light continuum retrieved from the probe-reference cross-correlation. Solid circles denote the first momenta of the up-converted temporal profiles and the solid line is a polynomial fit to the experimental points.} \]

To obtain the group delay across the white-light spectrum weighted averages of the time-dependent up-converted traces were measured. There are two reasons why this approach is superior to evaluation of \( \tau(\omega) \) from the peak positions as given by Eq.(2.4). First, the actual peak positions might be additionally shifted due to the unevenly distributed spectral intensity in the probe pulse. Second, by calculating weighed averages one uses the information from all
experimental points and not only from the maxima [75,76,84]. The group delay of the white-light continuum is shown in Fig.2.5 as solid points. The solid line represents a low-order polynomial fit used in the further calculations of the pulse compressor. The estimated group delay dispersion is ~380 fs$^2$ at the wavelength of 600 nm and decreases to ~220 fs$^2$ at 1 μm. We will return to the discussion of the apparent nonlinearity in the group-delay in Section 2.6.

In closing to this Section we note that the measurements described in it were repeated several times using slightly different fiber lengths. The results were found to be identical - within experimental uncertainty - to those presented in Figs.2.4 and 2.5, which indicates a remarkable long-term stability of the spectral phase.

### 2.5 Temporal analysis of the white light pulse

To verify the group delay measurements, we studied the properties of the white-light continuum in the time domain. To this end, we compare the wavelength-integrated cross-correlation trace, recorded with pulses that are considerably shorter than the duration of the white pulse, with the calculated temporal profile of the continuum. The latter is obtained by Fourier transformation of the electromagnetic field, taking into account the spectral phase calculated according to Eq.(2.5). The amplitude of electromagnetic field is derived from the measured spectrum. The continuum is mixed with the reference pulse in a 15-μm thick BBO crystal and the up-converted signal detected using a photomultiplier tube (PMT) [17,89,90].

![Fig.2.6: Comparison of the experimental (open circles) and computed (solid line) cross-correlation between the white-light continuum and the reference pulse (filled contour). The solid curve was obtained by numerical correlation of the reference pulse with the white-light pulse and corrected for the spectral sensitivity $R'(\lambda_p)$. The overall spectral response of the detector and up-conversion efficiency of a 15-μm BBO crystal is displayed in the inset.](image)

The measured signal is displayed in Fig.2.6 (open circles). Negative times represent the leading and positive times the trailing edge of the pulse. Note that the direction of time is known unambiguously since no time reversal symmetry is present in a cross-correlation experiment. The red-shifted components of the spectrum are concentrated in the leading edge of the pulse and the
blue-shifted ones are trailing behind.

To compare this experimental pulse shape with the calculated one, several factors should be taken in consideration. First, the finite duration of the reference pulse needs to be taken into account. Second, the spectral response of the detector and the relevant phase-match conditions must be regarded. The up-converted signal can be calculated by integration of Eq.(2.3) over frequency:

\[
S_{CC}(\tau) = \int S(\Omega, \tau) d\Omega. \tag{2.6}
\]

Taking into consideration the fact that the probe pulse is spectrally narrower than the white-light continuum, we may assume that each given instant corresponds to a single instantaneous frequency. In this approximation Eqs.(2.6) and (2.3) yield:

\[
S_{CC}(\tau) \propto R^\prime(\omega_p(\tau)) \left| E_p(t) \right|^2 \left| E_r(t-\tau) \right|^2 dt, \tag{2.7}
\]

where \( \omega_p(\tau) \) denotes the instantaneous frequency of the probe field and the overall spectral sensitivity is:

\[
R^\prime(\omega_p(\tau)) = R(\omega_r + \omega_p(\tau)) \left| \omega_r + \omega_p(\tau) \right|^2 \times \text{sinc}^2 \left( \frac{k_o(\omega_r(\tau)) + k_o(\omega_r) - k_E(\omega_r + \omega_p(\tau))L}{2} \right) \tag{2.8}
\]

The correction term \( R^\prime(\omega_p(\tau)) \), comprising a spectrally-varying conversion efficiency, the phase-matching factor of the crystal and the spectral response of the PMT, is depicted in the inset to Fig.2.6. The main spectral distortions occur at the high-frequency part of the white-light continuum (i.e. in the trailing edge) where phase-mismatch in the nonlinear crystal increases due to the increased dispersion. The resulting temporal shape of the continuum calculated according to Eq.(2.7) is depicted in Fig.2.6 as a solid line, and agrees reasonably well with the experimentally measured data, given all the assumptions made. This also indicates that the spectral phase was measured correctly. The asymmetry of the pulse (Fig.2.6) and its spectrum (Fig.2.3) will be addressed in more detail in the following Section.

In the previous Section we described the measurement of group delay by frequency-resolved cross-correlation. The obtained spectral phase correctly describes the wavelength-integrated trace presented in this Section. The question remains, however, whether the precision of group delay estimation is satisfactory. In our approach we captured the gross features of the spectral phase distortion of the white-light pulse. As was mentioned in Section 2.4, a more complete set of parameters can be recovered if a FROG-like inversion is applied to up-converted traces. This technique has been recently called XFROG [85]. It implies that the reference pulse is separately fully characterized by FROG prior to the inversion of the cross-correlation trace. While this
approach is feasible in our case, it is not entirely welcome since the final result of the continuum characterization also depends on the error in the separate measurement of the reference pulse. Instead, in Chapter 4 we will be able to recover refined information on the spectral phase of the uncompressed, as well as the compressed pulse through a SHG FROG measurement. Lying ahead, however, is the solution of the difficulties, which were rather unimportant in the case of a narrow – compared to the probe pulse – reference bandwidth in the spectrally-resolved cross-correlation experiment. The issues of the frequency mixing of two identical ultrabroad bandwidths will be examined in detail in Chapter 3.

2.6 Fiber output: experiment vs. numerical simulations

In a single-mode fiber, spectral broadening occurs due to the self-phase modulation (SPM), while a combination of SPM and normal (or positive) group velocity delay (GVD) acts to smoothen the chirp [19]. The dynamic evolution of a pulse propagating in a single-mode fiber is described by the nonlinear Schrödinger equation (NSE) [91]. When only SPM and group-delay dispersion are considered, the solution of the NSE yields a symmetric power spectrum, which corresponds to a symmetric rectangular-like pulse in the frequency domain and an almost linear chirp over most of the pulse duration [19]. It has been shown that linear frequency chirp, corresponding to a parabolic spectral phase, can be compensated by a quadratic compressor [19]. However, experiments [22] and numerical studies [92-96] have shown that higher-order dispersion and nonlinearities become increasingly important for propagation of femtosecond pulses, even for fibers shorter than 1 cm. In order to account for the intensity dependence of the group velocity, the conventional NSE should be extended to include a nonlinear correction term involving the time derivative of the pulse envelope, the so-called optical shock term [92]. This means that the part of the pulse that has the highest peak intensity, moves at a lower speed than the low-intensity wings. This effect, named self-steepening, causes pulse asymmetry and has been widely discussed in the literature (see, for example Chapter 4 of ref. [91] and references therein). In absence of mechanisms that stabilize this self-steepening process, the latter leads to an infinitely sharp pulse edge that creates an optical shock, similar to the development of an acoustic shock on the leading edge of a sound wave. Moreover, in this case the spectral phase of the pulse undergoes fast fluctuations that are difficult to compensate in a compressor.

Significant progress in numerical modelling of pulse propagation in fibers was made by taking into consideration both the optical shock term and higher-order dispersion [92-95]. It was shown that these two effects acting together suppress severe oscillations in the chirp. The predicted strongly asymmetric pulse shape and power spectrum agree reasonably well with the measured properties of our white-light pulse\(^\text{1}\). The nonlinearity of the chirp near the leading edge

\(^{1}\) The results of simulations most relevant to our experiments, are presented in ref. [94] in Fig.2 (pulse shape and chirp) and Fig.12 (pulse spectrum and spectrum phase).
of the pulse fully agrees with our measurements. It is worth noticing that despite the fact that the spectrum is asymmetric, the bandwidth introduced by the higher-order terms can effectively be used to obtain pulses shorter than those from the purely SPM-broadened spectra [95].

2.7 Compressor design

A light pulse broadened by SPM action in a fiber and by propagation through bulk material, can be compressed by passing it through a suitable optical element with anomalous (or negative) dispersion [97]. The group delay (or the spectral phase) is conventionally expanded into a Taylor series around a central frequency \( \omega_0 \) [25]:

\[
\tau(\omega) = \frac{d\phi(\omega)}{d\omega} \Bigg|_{\omega_0} \equiv \phi''(\omega_0)(\omega - \omega_0) + \frac{1}{2} \phi'''(\omega_0)(\omega - \omega_0)^2
\]

\[
+ \frac{1}{6} \phi''''(\omega_0)(\omega - \omega_0)^3 + \ldots
\]

(2.9)

where \( \tau(\omega) \) is the group delay, \( \phi''(\omega_0) \) is the group delay dispersion (GDD), \( \phi'''(\omega_0) \) is the third-order dispersion (TOD), \( \phi''''(\omega_0) \) is the fourth-order dispersion (FOD), etc. Note that a constant (non-frequency dependent) group delay has been disregarded in Eq.(2.9). This equation shows that in first order one should match the GDD of the compressor to the GDD of the pulse, in second order the TOD’s of pulse and compressor should be matched, and so on.

The quest for optical pulse compression emerged soon after the invention of sub-nanosecond lasers. The first report on extracavity pulse compression concerned a mode-locked He-Ne laser [98]. Over the past three decades a number of compressors have been proposed and successfully implemented: resonant Gires-Tournois interferometers (GTI’s) [99], resonant vapour delay lines [17], diffraction gratings [20,100] and prism pairs [10,23]. In particular, a combination of gratings and prisms [24,25] was triumphantly used to achieve 6-fs pulses [25]. This compressor can compensate for both GDD and TOD over a very broad spectral range [101]. Recently chirped mirrors [102] revolutionized the technology of ultrashort pulse generation.

Design of an appropriate high-throughput pulse compressor becomes increasingly difficult for larger bandwidth of the chirped pulse. In addition, the spectral region over which any of the aforementioned compressors provides adequate phase compensation, narrows rapidly with the increase of the chirp rate. The requirements for compression of the white-light continuum arise from the fact that both GDD and TOD are positive as is evident from Fig.2.5. Therefore, one should aim for a compressor that exhibits both negative GDD and negative TOD. In our previous experiments [57], the spectral range of the prism-grating compressor was broadened by careful balancing the GDD against the FOD, such that nearly transform-limited 5-fs pulses were obtained. This seems to represent the current limit of this technique for pulses chirped in a fused-silica fiber. Moreover, an oscillatory residual phase remained – as a trade-off between phase corrections of different orders – which led to sidelobes on the 5-fs pulse. Another inherent drawback of the grating-prism compressor is its low throughput, typically \( \sim 25\% \) [57]. Note that a 200-nm
bandwidth of recently designed high-efficiency (~90%) gratings [103] is not sufficient for pulse compression down to 5 fs.

Fig. 2.7: Overview of optical elements used in the compressor and autocorrelator: 45° fused silica prism compressor (a), chirped mirror (b), Gires-Tournois interferometer (c), overcoated silver mirrors (d), 1 m of air (e), beam splitter in the autocorrelator (f) and total compressor (g). Group delays of various dispersive components are indicated by solid lines (left axis) while dotted lines show transmittance or reflectance (right axis). The compressor itself comprises three parts: a prism pair, chirped mirrors and Gires-Tournois interferometers. Solid circles in (g) are experimentally measured group delay depicted with the reversed sign and used as the desired group delay of the compressor. Reflection on the beam splitters is not taken into account in the overall throughput. The interprism pathlength in air is included in the data for the prism compressor.

A major advance in pulse compression technology was made by the introduction of a compressor based on chirped mirrors and prisms [104]. In contrast to gratings, chirped mirrors can be made that have a large acceptance bandwidth and a very high reflectivity at the same time. With this prism-chirped mirror compressor, pulses of 20-fs were amplified to the millijoule level [105]; more recently a similar compressor was used to generate 5.5-fs, 6-nJ pulses at 1-MHz repetition rate [57] and sub-5-fs, 20-µJ pulses at a 1-kHz repetition rate [58].
To improve on our previous compression scheme [57], we designed a novel high throughput compressor. To obtain the required negative GDD and FOD a fused-silica prism compressor was used (Fig.2.7a), which, however, overcompensates the TOD when used alone. Recently it was shown that ultra-broadband chirped mirrors can be made that exhibit negative GDD and positive TOD (Fig.2.7b), while having a reflectivity exceeding 99% over a bandwidth of 600-1100 nm [70,106]. This means that a combination of chirped mirrors and a prism compressor provides flexible control over TOD across a large spectral range [70]. For higher-order phase corrections, broadband dielectric GTI’s [107-109] have been shown to be suitable (Fig.2.7c). GTI’s counteract the FOD of the prism pair, which becomes significant above 900 nm. These ideas lead us to a compressor design that consists of a prism pair, ultra broadband chirped mirrors, and dielectric GTI’s.

We employed dispersive ray-tracing analysis [101,110] to compute the group delay of the three-stage compressor. The use of Eq. (2.9) to calculate the spectral phase by a Taylor expansion becomes impractical since the compressor should span the region from 600 to 1100 nm. Wavelength-dependent refractive indices were calculated from dispersion equations while corresponding refraction angles in the prism compressor were obtained by using Snell’s law. Subsequently, the total accumulated phase of the prism compressor and bulk material was computed at each wavelength. By numerical differentiation of the phase, the group delay of the prism part of the compressor was obtained. This group delay was added to the group delay of the reflective optics to compute the overall group delay of the compressor. The resulting group delay $\tau_{\text{COMPR}}$ is than compared to the measured group delay of the white-light continuum $\tau_{\text{WLC}}$, but taken with opposite sign. Subtracting the calculated group delay from the desired, we find the residual group delay

$$\tau_{\text{RES}}(\omega) = \tau_{\text{COMPR}}(\omega) - \tau_{\text{WLC}}(\omega),$$

by integration of which we obtain the residual spectral phase $\varphi_{\text{RES}}(\omega)$. To further characterize the compressor performance, the input white-light spectrum (Fig.2.3) modified by the compressor throughput (Fig.2.7g, dotted curves) is calculated. Taking into account the residual phase, the temporal shape and phase of the compressed pulse is then computed via a Fourier transformation.

An overview of all optical elements used in the compressor and autocorrelator is presented in Fig.2.7. The previously employed [57] unprotected gold-coated mirrors with 90% peak reflectivity and rapidly growing absorption below 600 nm, were substituted by low dispersion and higher reflectivity overcoated silver mirrors (Fig.2.7d). The dispersion due to propagation in air [111] was also found to play an essential role for 5-fs pulses (Fig.2.7e).

The compressor performance is optimized by varying the number of reflections on the dispersive mirrors, by changing the interprism spacing and by varying the prism apex angles. Optimal performance is judged by looking for the shortest pulse through second harmonic generation in the autocorrelator. Hence, the pathlength in air from the compressor output, the 0.5-mm thick beam splitter at 45° incidence (Fig.2.7f) and reflections off the autocorrelator mirrors should be included in calculations as well. Pulse broadening due to dispersion inside the
autocorrelator nonlinear crystal was not considered because of its negligible effect. Reflectivity curves (Fig.2.7, dotted lines) and group delays (Fig.2.7, solid lines) of the chirped mirrors, GTI’s, the overcoated silver mirrors and beam splitters were provided by the manufacturer (R&D Lezer-Optika, Hungary).

The angle of incidence onto the fused-silica [112] prism, being a sensitive parameter, was chosen to correspond to the least deviation angle for the sake of experimental convenience. Simulations show that the apex angles smaller than 45° are impractical because they call for unreasonably large interprism separation. Moreover, with increased prism separation the positive dispersion of air (Fig.2.7e) between the prisms becomes more important so that the whole compressor would need to be put in a vacuum. Note, that the amount of the TOD could also be reduced by employing doubled-prism pairs as has been demonstrated previously [58,65].

Optimal compression (Fig.2.7g) was obtained for 5 reflections of the chirped mirrors, 2 reflections of GTI mirrors, and use of a 45° prism compressor with the following settings: ~5.2 mm of prism material for the 800-nm wavelength ray and ~115 cm distance between apices. The root mean square error of the residual group delay amounts to ~1.5 fs. The blue wavelength cut-off of the prism compressor coincides with the abrupt reflectivity drop of the chirped mirrors, thus no additional loss of the spectral content originates from the prism part of the compressor. The compressor throughput is fairly flat between 600 and 1100 nm and amounted, at the beginning, to ~75%, mainly due to eight reflections from the non-Brewster-angle prisms. When a low-dispersive anti-reflection coating is deposited on the surfaces of the prisms, the total compressor throughput reached ~90%.

The Fourier transform of the compressor output spectrum assuming constant phase, yields a pulse of ~4.2 fs in duration, i.e. somewhat longer than the Fourier transform of the input spectrum (Fig.2.3, inset). This lengthening of the pulse occurs due to the loss of spectral components in the near infrared and visible part of the continuum (Fig.2.7g, dotted curve). Residual phase correction should be feasible by installation of a programmable phase mask [113] into the pulse compressor. The applicability of this technique has recently been demonstrated for pulses as short as 10 fs [114]. Spectral shaping would also allow manipulating of the spectrum leading to cleaner optical pulses [115].

With the compressor being set up near the cavity-dumped laser and white-light generator (Fig.2.1) the overall size of the system is 1×1.5 m². This compactness makes our sub-5-fs laser system extremely robust and ensures that the cavity alignment is retained for a long time. When starting up the laser the only thing needed is to correct for the sub-micron drift of the fiber tip. Due to the short warming-up time of the diode-pumped “Millennia”, the stable regime of sub-5-fs operation is achieved within minutes. The compactness of the laser source presents a distinct advantage in experiments because it allows building the experimental setup close to the laser thereby limiting pulse propagation through air.

2.8 Pulse duration measurement

Accurate pulse-width measurement of pulses containing only a few oscillations is quite a
challenge. An easy and informative method to judge the compression quality is the second-order interferometric autocorrelation (IAC) [97,116,117]. An additional benefit from this technique is that it can be used as an on-line tool. Of course, the technical demands to be made for a 5-fs autocorrelator are substantial. In our experiments we employ a Mach-Zehnder interferometer [57,118-120], which has the advantage of being fully symmetric with respect to both arms. Note that the “magic” 0 : 1 : 8 ratio between the minimum, the asymptotic level and maximum of the IAC trace [116] is obtained only if the intensities of two interfering beams are strictly equal. If one intensity exceeds the other one by a factor of $\beta$, the re-normalized ratio becomes $0 : 4 - 6\sqrt{\beta} / (1 + \beta) : 8$ with the asymptotic level being between 1 and 4. Imperfectness in alignment of the interferometer leads to the same result.

Fig.2.8: Schematic of Mach-Zehnder interferometer for measurements of interferometric autocorrelation. BS1, BS2: 50% ultra-broadband beam splitters centred at 800 nm; M1-M4: flat low dispersion overcoated silver mirrors; M5: $r = -10$ cm, low dispersion overcoated silver mirror; BBO: 15-µm thick BBO crystal; M6: $r = -10$-cm protected aluminum coated mirror; PD: photodiode; PMT: photo multiplier tube; PZT: piezo transducer; HVA: high voltage amplifier; DAC: digital-analog converter; ADC: analog-digital converter. All optics were obtained from R&D Lezer Optika, Budapest.

The input beam is split and recombined in such a way that each of the beams travels once through an identical beam splitter while both reflections occur on the same coating-air interfaces (Fig.2.8). To match the beam splitters [46], the initial horizontal polarization of the compressed pulse is rotated by a periscope. A 15-µm BBO crystal is used for second-harmonic generation. Such a thin crystal is required to avoid dispersion-induced pulse broadening and to ensure a sufficiently broad phase-matching bandwidth.

The moving arm of the interferometer is driven by a piezo transducer (PZT) which is controlled by a computer via a digital-analog convertor (DAC) and a high voltage amplifier (HVA). After having moved the M3-M4 arm to a new position, the measurement of the second
harmonic intensity is performed by sampling and digitalization of the photomultiplier (PMT) signal. The experimental points obtained in this way are depicted in Fig.10 by open circles. To introduce an on-line calibration of the time axis, a He-Ne laser beam is aligned in a direction opposite to the white light. The signal of the photodiode (PD) monitoring the interference fringes at the wavelength of He-Ne laser [121] is used for precise time calibration (Fig.2.9, lower panel). This allows autocorrelation measurements to be performed with ~0.2 fs accuracy throughout the whole scanning region of ~100 fs. A typical time step is ~0.1 fs, or 23 points per oscillation period at 800-nm wavelength at the rate of ~60 ms for a 100-fs scan.

The typical IAC shown in Fig.2.9 was obtained by setting the compressor according to the calculated optimal settings, whereupon the amount of prism material was balanced so as to get the shortest autocorrelation. Compared to our earlier result [57], the wing structure of the IAC is substantially reduced, which demonstrates the superior characteristics of this compressor. We verified the importance of the GTI’s for high-order dispersion correction by changing the angle of incidence from the design angle of 45° to ~15°. In this case the group delay curve shifts toward shorter wavelength (Fig.2.7c) resulting in broadening of the central part of the IAC function and an appreciable increase of the amplitude in its wings.

![Fig.2.9: Interferometric autocorrelation (IAC) of the compressed pulse. Open circles: experimental points. solid line: calculated IAC of deduced pulse shape shown in the inset. Bottom panel depicts He-Ne laser interference fringes used for on-line time calibration.](image)

When fitting the IAC to a hyperbolic secant envelope, we get a pulse of ~3.7 fs, a Gaussian a pulse of ~4.4 fs is obtained. The former value clearly violates the earlier derived spectral-limited pulse duration of ~4.2 fs (Section 2.7). Furthermore, neither of these pulse shapes reproduces the wing structure on the experimental IAC. This clearly indicates that one should be extremely cautious about fitting the IAC of a short pulse to an a priori assumed pulse profile, especially when the pulse spectrum is not smooth. It should also be noted that the standard deviation, conventionally used in fitting routines to judge the fit quality [122], can hardly serve as a criterion in favor of any particular pulse shape. Most of the experimental points in the IAC are
located at the slopes of the fringes where the gradient is too high to recognize any anomaly. As a matter of fact, only 8-10 points at the extrema of the IAC are meaningful which clearly is not sufficient to discriminate between the different pulse profiles. We will address the problem of retrieving the pulse shape from the IAC in the next Section.

2.9 Reconstruction of 5-fs pulse from the IAC and spectrum

In the previous Section we showed that a fit of the interferometric autocorrelation (IAC) to an a priori analytical pulse intensity profile is not warranted. Clearly, it would be a major step forward if the pulse shape could be retrieved from the IAC without having to rely on any assumption concerning the temporal profile of the electric field. It was pointed out by Naganuma et al. [73] that information on the phase and amplitude of the pulse is, in principle, contained in the IAC and pulse spectrum. Several algorithms have been applied over the years to treat the problem of pulse reconstruction from its autocorrelation [123-128]

The normalized interferometric autocorrelation signal can be expressed as [73]:

\[
IAC(\tau) = 1 + 2G(\tau) + 4\text{Re}[F_1(\tau)\exp(i\omega_0\tau)] + \text{Re}[F_2(\tau)\exp(-i2\omega_0\tau)],
\]

(2.11)

where, for the sake of clarity, in the expression of the complex electric field in the time domain we separated the term oscillating at the carrier frequency \(\omega_0\). The constituent terms of the sum in Eq.(2.11) are:

\[
G(\tau) = \int I(t)I(t-\tau)dt, \quad (2.12)
\]

\[
F_1(\tau) = \int \frac{I(t) + I(t-\tau)}{2} E(t)E^*(t-\tau)dt, \quad (2.13)
\]

\[
F_2(\tau) = \int E^2(t)E^{*2}(t-\tau)dt. \quad (2.14)
\]

Here \(G(\tau)\) stands for the (background-free) intensity autocorrelation, and \(F_2(\tau)\) represents the second harmonic field autocorrelation. Note that when the temporal phase is a constant the functions \(G(\tau)\) and \(F_2(\tau)\) become identical [73]. This property can be exploited to determine whether the compressed pulse carries any residual chirp. Since the carrier frequencies of \(G(\tau)\) and \(F_2(\tau)\) are different, the simplest way to extract this information from the IAC (Fig.2.9) is to Fourier transform the IAC, with the constant background (unity level in Eq.2.11) subtracted. One then obtains a spectrum composed of \(\tilde{G}(\omega)\) at zero frequency, \(\tilde{F}_1(\omega)\) at the fundamental frequency \(\omega_0\), and \(\tilde{F}_2(\omega)\) at the second harmonic frequency \(2\omega_0\) (Fig.2.10). Note that since the latter two components are projected at both positive and negative frequencies, their magnitudes are reduced.
by a factor of two compared to the ratios given by Eq.(2.11). As can be seen from Fig.2.10, the functions \( \tilde{G}(\omega) \) and \( \tilde{F}_2(\omega) \) are quite similar, which confirms our earlier conclusion that the compressor has removed most of the chirp in the white-light continuum. Nonetheless, the small asymmetry of \( \tilde{F}_2(\omega) \) indicates that there is some residual chirp in the compressed pulse\(^\dagger\).

Peatross et al. recently demonstrated temporal decorrelation of intensity autocorrelation function \( G(\tau) \) \cite{125,126} which yields the modulus of the pulse electric field in the time domain. Combining this data with the modulus of the electric field in the frequency domain (square foot of spectral intensity), in the second stage of their two-stage algorithm they extract phase information that corresponds to these two moduli.

![Fig.2.10: Fourier-transform of the experimental interferometric autocorrelation function. The mirror image of the spectrum at the negative frequencies is not shown. The close similarity between the zero and double-frequency peaks indicates that the compressed pulse is almost chirp-free.](image)

Concisely, the problem of phase retrieval from a collinear (IAC) or non-collinear (intensity) autocorrelation and the fundamental spectrum is summarized in the following integral equations written for the Fourier transforms of \( G(\tau) \) and \( F_2(\tau) \), which are denoted here as \( \tilde{G}(\omega) \) and \( \tilde{F}_2(\omega) \):\(^\dagger\)

\[
\tilde{G}(\omega) = \left[ \sqrt{S(\omega')S(\omega'-\omega)} \exp[\bar{\phi}(\omega') - \bar{\phi}(\omega'-\omega)] \right] d\omega',
\]

(2.15)

\(^\dagger\) Note that there is a distinct difference between transform-limited (or spectral-limited) and chirp-free pulses. For instance, in the case of an asymmetric spectrum, a transform-limited pulse does carry some chirp, even if its spectral phase is constant.
\[ \tilde{F}_2(\omega) = \left| \int S(\omega') S(\omega - \omega') \exp \left[ i \phi(\omega') + \phi(\omega - \omega') \right] d\omega' \right|^2, \]  

(2.16)

where \( S(\omega) = \left| \tilde{E}(\omega) \right|^2 \) is a fundamental spectrum measured by a spectrometer or, alternatively, Fourier-transformed from an interferogram [73], and \( \phi(\omega) \) is the unknown spectral phase. As can be seen from the structure of these equations, Eq.(2.15) contains a modulus square of the autocorrelation of the frequency-domain electric field, while Eq.(2.16) contains a modulus square of its autoconvolution. Regrettfully, the knowledge of only a modulus of the autoconvolution prevents the possibility of a straightforward deconvolution of the complex electric field. In fact, \( \tilde{F}_2(\omega) \) is equivalent to a directly measured second-harmonic spectrum of the pulse. While the fundamental and the second harmonic spectra in principle uniquely define the amplitude and phase of a pulse (with time-direction ambiguity) [124], the influence of different spectral phases on the shape of \( \tilde{F}_2(\omega) \) is frequently only very minute. On the other hand, the profile of \( \tilde{F}_2(\omega) \) is critically affected by the frequency-dependent conversion of the fundamental field into the second harmonic radiation (See Section 3.6 for the detailed explanation of the spectral filtering effect). Like \( \tilde{F}_2(\omega) \), the term \( \tilde{G}(\omega) \) is also susceptible to spectral filtering. However, the recognition of different pulse shapes from \( \tilde{G}(\omega) \) is much more reliable, compared to \( \tilde{F}_2(\omega) \).

To deduce the parameters of the compressed pulse, we applied a two-stage phase retrieval algorithm [126,129] to the Fourier transform of the experimental data from Fig.2.9. The resulting temporal intensity profile is depicted in the inset to Fig.2.9. The IAC corresponding the retrieved complex electric field is shown as a solid curve alongside the experimental trace. Since our procedure relied only on the treatment of the IAC part corresponding to the intensity autocorrelation, \( G(\tau) \), it is interesting to see how the calculated terms \( F_1(\tau) \) and \( F_2(\tau) \) comply with the overall measured IAC trace. Clearly, certain discrepancies appear around \( \pm 10\text{-fs} \) delay (Fig.2.9). Their most likely explanation is in the mentioned above spectral filtering effect, which was not considered in the employed phase retrieval routine.

### 2.10 Pitfalls of IAC

IAC, or collinear autocorrelation, measurement has a number of advantages and disadvantages. For example, the background-free autocorrelation or FROG measurement of sub-10-fs pulses in the non-collinear beam arrangement becomes difficult because of the need to overlap the intersecting beams at the point where the beam waist is the smallest. This is required to minimize the geometrical smearing effect, or delay blurring. (See Section 3.5). In this respect, the collinear alignment is much easier and IAC does not suffer from delay blurring. Unlike the intensity autocorrelation that is determined solely by the temporal intensity shape, whatever the phase is, IAC is phase-sensitive [116]. IAC also possesses a certain degree of intuitiveness in the form of fringes oscillating at the carrier frequency. This can almost directly be related to the number of optical cycles contained by the electric field of the pulse.

However, the very merit of IAC – its fringes – may turn against it. Unlike the background-
free autocorrelation, in which a very high dynamic range may be achieved, the IAC asymptotically approaches its background (ideally 1/8 of its peak value) in oscillatory fashion. Importantly, these oscillations are on both sides of the background level. As can be seen from Eq.(2.11), the terms comprising an IAC, have different periods of oscillations. Therefore, some IAC fringes may cancel giving a pleasing aesthetic appearance of the trace that in reality does not correspond to a nice pulse. The “lucky” interplay of the fringes is especially easy to achieve when there are merely 3-5 significant fringes on the IAC trace.

By its nature, a collinear fringe-resolved autocorrelation cannot be single-shot. Thus, to avoid the “mop-up” of the IAC wings by the statistical averaging of the fringes, the mechanics of the autocorrelator should be engineered in the way ensuring reliable interferometric stability. This, however, cannot prevent such fringe averaging if the phase and/or the spectrum of the laser output fluctuate in time. Obviously, the statistical sum of slightly different IAC’s may produce an unpredictable result.

As has been pointed out by R. Trebino [130], the inhomogeneity in the spatial distribution amplitude-phase characteristics across the beam also may result in the fringe averaging effect. Figure 2.11 gives an example of an IAC of a 4.6-fs pulse that is spectrum-limited and has a top-hat spectrum (Fig.2.11a, inset). The solid lines in Fig.2.11a and 2.11b represent the ideal IAC and intensity autocorrelation, respectively. The laser beam is assumed to be Gaussian, and all frequency components have identical sizes. We now model spatial chirp by assuming that at the position of the SHG crystal the beam is dispersed linearly with frequency in the horizontal direction to ~1.7 of its vertical size. Therefore, the spectrum is somewhat red-shifted on one side of the beam, and blue-shifted on the other. The spectrum at each point across the beam in horizontal direction now corresponds to a slightly longer pulse than the one obtained from the total spectrum, as reflected by a slight broadening of the background-free autocorrelation (Fig.2.11b, dotted line). However, the period of oscillation of IAC traces corresponding to different points across the beam now vary by about a third of the period length. This efficiently damps oscillations in the overall resulting IAC depicted as dotted line in Fig.2.11a, beginning with the third fringe. Clearly, the “new” IAC gives the impression of a wing-free pulse.

The situation discussed above is not unrealistic. For instance, it can easily take place in presence of chromatic aberrations in the focusing into the SHG crystal. The described situation is also likely to occur following a small angular “adjustment” of a prism or grating compressor while judging the perfectness of compression by the quality of IAC. Unbalancing the arm-length in a prism or grating compressor would lead to the same effect. Even if the identical spectra are measured across the beam profile, there is no guarantee that the spectral phase is identical in all points. This compromises the use of IAC unless it is backed up by other pulse measuring methods.
In the previous Section we also hinted at the importance of the spectral filtering effect which we so far disregarded in our measurement of the compressed pulse. In combination with the factors described in this Section, this might have caused under- or over-estimation of the pulse characteristics. To solve all these problems and obtain a rigorous amplitude-phase characterization, in Chapter 4 we return to the measurement of the compressed pulse by means of frequency-resolved non-collinear autocorrelation (FROG).

Summarizing our ideas about IAC, we suggest that great caution should be exercised in dealing with it and that circumstances potentially jeopardizing its validity should carefully examined. In any case, the quality of the pulse reconstruction should be judged by the ability to reproduce the wing structure of the measured IAC trace.

### 2.11 Summary and outlook

In this Chapter we have discussed a compact and robust light source that generates sub-5-fs, 2-MW pulses at variable repetition rates of up to 1 MHz, using a novel three-stage compressor. The phase characteristics of the compressor have been analyzed using dispersive ray tracing and mapped unto the measured group delay of the continuum. The fidelity of this approach has been confirmed by the fact that the pulse shape derived from the optical spectrum and the calculated residual phase, fit the measured autocorrelation function very well. It has also been shown that the interferometric autocorrelation and optical spectrum of the compressed pulse comprise sufficient information to derive the temporal pulse intensity and its phase.

We foresee several applications of this ultrafast laser. First, it is an almost ideal tool for ultrafast spectroscopy, if not for the short pulse then for the white light continuum that can be used as a probe for spectral events from the blue-green part of the spectrum to the near infrared region (500 nm to 1.3 µm). The large bandwidth of this laser may also be of use in optical coherence tomography measurements [72]. For the near future we aim for an all chirped-mirror compressor
which would enable an even more compact design of this laser. With smaller diode pumped light sources coming on the market there is every reason to believe that soon it will be possible to build a sub-5-fs cavity-dumped laser that fits onto a breadboard of only 1 m by 0.5 m. This may be an important asset for many applications.

Another replica of the 5-fs set-up that is described in this chapter has been built in our laboratory to adapt to the needs to carry out several parallel projects in nonlinear spectroscopy in solutions. Both laser systems displayed a very high performance in terms of both long- and short-term stability over more than two years of their intensive use. Typical cycles of experiments lasted longer than 24 hours of non-stop data collection. Several interesting systems have been studied using the white-light pulses, for example, solvation dynamics of small rigid ions [131], solvent-controlled electron-transfer reaction [132], and photon-echo and pump-probe studies on equilibrated solvated electron. The latter study is included in Chapters 6 and 7 of this Thesis. Meeting the demands of particular experiments [132], tunable excitation pulses of about 20-fs duration and 10-fs probe pulses were tailored from the white light to facilitate two-color experiments. Considering the overall simplicity of our set-up, favorably distinguishing it from very complex amplified laser systems with parametric generators for wavelength tuning, this remarkable versatility in our case was achieved at a relatively low expense.
References

111. C. DeWitt Coleman, W. R. Bozman, and W. F. Meggers, *National Bureau of Standards*
Chapter 2


