Hydrated electron dynamics explored with 5-fs optical pulses
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Chapter 4

FROG Characterization of Fiber-Compressed Pulses

Abstract

The technique of SHG FROG is applied to measure the white-light continuum pulses in the spectral region of 500-1100 nm. The obtained spectral phase of these pulses served as a target function for the pulse compressor design. The pulses around 800 nm produced by compression were also characterized by SHG FROG. The resulting pulse duration measures 4.5 fs which corresponds to ~2.5 optical cycles.
Chapter 4

4.1 Introduction

This Chapter puts to work the ideas about the SHG FROG pulse characterization, which have been developed in Chapter 3. We demonstrate practical application of this pulse measuring technique to strongly chirped ultrabroadband pulses and compressed 4.5-fs pulses from the cavity-dumped-laser based white-light generator that has been presented in Chapter 2. The use of SHG FROG allows us to finally obtain rigorous electric field characterization in terms of amplitude and phase, which Chapter 2 clearly lacked.

Prefacing the account on the experimental results, several practical implications of dealing with the extraordinarily broad bandwidths will be considered here. The basic experimental requirements raised to the FROG apparatus are the adequate bandwidth of phase matching of the SHG crystal, the low overall dispersion of the optical elements. Next to it, the device should be able to yield exact replicas of the pulse that do not differ from each other in their spectral content or phase.

In particular, the choice of the frequency-doubling to be employed for the FROG measurement is a very delicate issue. This is partly due to the fact that the demand of a high signal to noise ratio and, therefore, the need to employ thicker, more efficient doublers finds itself in conflict with the necessity to keep the phase-matching bandwidth broad and, consequently, the crystal thickness low. Another riddle to solve is the right angular orientation of the crystal or the wavelength it is cut for. The difficulty here comes from the fact that the “red” crystal orientation typically provides nearly flat frequency conversion efficiency over the most of the bandwidth of an ultrabroadband pulse. Such an orientation, however, dispenses with the blue-shifted wing of the spectrum where the conversion efficiency dramatically falls. On the other hand, the use of the “blue” phase-matching significantly lowers frequency-conversion efficiency in the red wing of the spectrum. To address these issues and to find a reasonable balance that satisfies the demands of various pulses, a useful criterion on crystal selection is developed in this Chapter. Further, the merits of the two most commonly used SHG crystals, BBO and KDP, are compared among themselves. We also present a case study of two contradicting recipes concerning the most suitable, for measuring sub-5-fs pulses, cut angle of the crystal.

We next focus our attention on the working of the FROG apparatus and the peculiarities of the measurement of strongly chirped and nearly fully compressed laser pulses by this technique. The spectral phase of the white light pulse measured before and after the pulse compressor permits a good verification of the ray-tracing routine employed to design it. We subsequently present valuable observations on how extra information about the level of pulse compression can be gained from a simple examination of the SHG FROG trace that is normally considered quite unintuitive.

This Chapter is organized as follows: Section 4.2 advises on the choice of the SHG crystal. Section 4.3 provides a quantitative study of the effect of phase-matching wavelength
on the outcome of SHG FROG. Section 4.4 we describe our SHG FROG apparatus. SHG FROG characterization of the white-light continuum and 4.5-fs pulses is demonstrated in Sections 4.5 and 4.6, respectively. Finally, in Section 4.7 we summarize our findings.

4.2 The choice of the SHG crystal

In this Section, we provide several guidelines for the right SHG crystal selection in the FROG measurement. On the one hand, the crystal should be thick enough to generate an appropriate level of the second-harmonic signal for a high dynamic range measurement. One the other hand, the thickness of the crystal should be sufficiently small to provide an appropriate phase matching bandwidth and minimize pulse broadening in the crystal.

Obviously, when choosing the crystal one must consider the bandwidth of the pulse that has to be characterized. We employ a simple criterion to estimate the required crystal thickness: the conversion efficiency calculated according to Eq.(3.33) must be higher than 50% of the peak conversion efficiency everywhere over the double FWHM of the FROG spectral marginal. For the pulses that are Gaussian in frequency, the ideal spectral marginal, or the autoconvolution of the fundamental spectrum, is \( \sqrt{2} \) times broader than the pulse bandwidth. Using this criterion, we evaluated BBO and KDP crystals, which are typically employed for the ultrashort pulse measurement. Both considered crystals are cut for Type I phase-matching at the wavelength of 800 nm and 600 nm. Figure 4.1 depicts the appropriate crystal thickness of the BBO (solid curve) and KDP (dashed curve) as a function of duration of a bandwidth-limited Gaussian pulse.

\[ \text{Fig.4.1: Crystal thickness required for SHG FROG measurement as a function of the pulse duration at the central wavelength of 800 nm (a) and 600 nm (b). The crystals are cut for Type I phase-matching, which corresponds to } \theta = 29^\circ \text{ for BBO (solid line) and } \theta = 44^\circ \text{ for KDP (dashed line).} \]

As can be noticed from Fig.4.1, an approximately 10-µm BBO should be employed to measure 5-fs pulses at 800 nm. The adequate thickness of the KDP crystal is approximately
2.5 times larger due to its lower dispersion. However, while clearly providing an advantage in thickness, the KDP crystal has a disadvantage in the SHG efficiency. The signal level that can be obtained with a 2.5 times thinner BBO crystal is still approximately by a factor of 6 larger than in KDP because of the higher nonlinear coefficients and lower phase-matching angle in the BBO crystal [1]. Therefore, BBO is a more suitable choice for characterization of weak-intensity pulses. For high-intensity pulses, where the low level of the second-harmonic signal is not really the issue, KDP presents a better choice [2].

With the growth of the phase-matching bandwidth of the crystal, the $\Omega^3$ dependence (Eq.(3.33)) begins to dominate the conversion efficiency. As it was shown in Section 3.6, this dependence blue-shifts the second-harmonic spectrum. In case the phase-matching bandwidth of the SHG crystal is wider than required by the pulse bandwidth, angular tuning of the crystal can effectively counteract such blue shift [2]. To illustrate the point, we consider a 10-µm BBO crystal applied to measure 8-fs Gaussian pulses at 800 nm. Figure 4.2a shows the blue-shift of the FROG spectral marginal (filled circles) with respect to the autoconvolution (solid curve) if the crystal is perfectly phase-matched at 800 nm, i.e. $\theta=29^\circ$. However, after adjusting the phase-matching angle to $\theta=24.4^\circ$ that now corresponds to the central wavelength of 970 nm (Fig.4.2b) the phase-matching curve of the crystal (dashed curve) nearly perfectly balances the $\Omega^3$ dependence (dotted curve). The overall conversion efficiency becomes almost flat and no spectral correction of the FROG trace is required. Experimentally, Taft et al. [3] demonstrated the effectiveness of the angular adjustment that enabled them to yield correct FROG data.

The mutual compensation of the $\Omega^3$ and phase-matching terms is only possible for relatively long (~10 fs) pulses. As a thinner crystal is chosen to measure shorter pulses, the high-frequency slope of the phase-matching curve becomes relatively steeper than the low-frequency one (Fig.4.2c, d). This is to be expected, since crystal dispersion is low in the infrared and increases approaching the UV absorption band. Tuning the central wavelength of the crystal from 800 nm (Fig.4.2c) to 970 nm (Fig.4.2d) substantially narrows the SH spectrum in the blue due to the crystal phase-matching. Even worse, the FROG trace can hardly be corrected for the imposed spectral filter since the conversion efficiency becomes extremely low in the blue wing (Fig.4.2d). This should be contrasted to the 800-nm-cut case when the correction is still possible (see Fig.3.6). Therefore, in order to extend the phase matching-bandwidth in the blue, one should consider using a crystal with the phase-matching wavelength blue-shifted with respect to the central frequency of the pulse. For example, a $L=10$ µm BBO crystal oriented for peak conversion efficiency at 700 nm is more suitable for the measurement of sub-5-fs pulses centered at 800 nm than the same crystal tuned to 970 nm. Although the 700-nm-cut crystal has poorer conversion efficiency in the infrared it, nonetheless, allows the extension of the phase matching below 600 nm. Consequently, this crystal has an appreciable efficiency of frequency conversion all over the spectrum of a 5-fs pulse and, therefore, the FROG traces can be validated upon spectral correction. In contrast, information about the blue spectral wing is completely filtered out if the crystal oriented for
970 nm is used. The quantitative analysis of how the poor choice of the SHG crystal can affect the FROG recovery of a sub-5-fs pulse will be given in the next Section.

Fig. 4.2: Correction of frequency conversion efficiency by crystal orientation for 8-fs (a,b) and 3-fs (c,d) bandwidth-limited Gaussian pulses. A Type I 10-µm BBO crystal is oriented for the phase-matched wavelength of 800 nm (a,c) and 970 nm (b,d). The phase-matching curve and the $\Omega^3$ dependence are shown as the dashed and dotted lines, respectively. The solid curves depict the autoconvolution of fundamental spectra, while spectral marginals of FROG traces are given by filled circles. In (b), no spectral correction of the FROG trace is required for an 8-fs pulse because of the red-shifted phase-matched wavelength. In contrast, the use of the 970-nm phase-matched crystal irreparably corrupts the second-harmonic spectrum in the case of a shorter 3-fs pulse (d). Note the difference in horizontal scales in (a), (b) and (c), (d).

In closing to this Section, we mention an interesting property of very thin crystals, i.e. those that have a thickness in order of a few microns. In such thin crystals the differentiation between the Type I (oo-e interaction) and Type II (eo-e interaction) becomes less strict. For instance, if we speak about a Type I 800-nm-cut crystal this means that the phase mismatch, $\Delta k$, for this wavelength is zero. However, if the crystal thickness, $L$, is very small then the product $\Delta k_{EO-E} L$, albeit never reaching a zero value, becomes comparable to the magnitude of $\Delta k_{oo-e} L$ for the wavelengths detuned from the phase-matching frequency. Therefore, we can no longer neglect the contribution to the SH signal produced by Type II interaction. Additionally, even for the fundamental waves that have a perfect linear polarization the
second harmonic beam, obtained in this case, becomes somewhat depolarized. This situation reciprocates for thin Type II crystals where the mixture of the oo-e contribution adds up to the total signal. This has far-reaching consequences. For instance, this means that in collinear Type II FROG experiments some fringes that are due to the interference of the SH waves, produced by each interacting fundamental wave, will always be present, no matter how perfectly orthogonal the polarizations of fundamental beams are kept. The mentioned here property is considerably stronger for BBO than for KDP for which, for the same thickness, no such effect takes place. Finally, we point out that the necessity to account for both Type I and Type II contributions applies only to sub-10-µm BBO crystals.

4.3 Case study: Two contradicting recipes for an optimal crystal

In the previous Section we outlined the issue of a proper crystal choice and offered a solution. However, no particular quantitative assessment of the damaging role of a poorly chosen crystal was given there. In this Section, we ascertain the possible distortions of the amplitude-phase measurement, which arise from the orientation of a 10-µm BBO crystal applied to measure sub-5-fs at 790 nm. Two distinctly different recommendations had been given on that account in this Thesis and in the recent publication by Chen et al. [2], respectively.

The first suggestion claims that, in order to extend the phase-matching bandwidth to cover the wavelengths below 600 nm, a 700-nm 10-µm BBO crystal should be used and the FROG trace must be necessarily corrected to the frequency-doubling efficiency, that significantly drops in the NIR region.

The second recipe, on the contrary, recommends the use of a 970-nm-centered 10-µm BBO crystal. The red-shifted cut-wavelength balances off the $\Omega^2$ term in the conversion efficiency, which gives rise to a very broad and nearly symmetric phase-matching band around the carrier wavelength of the pulse, 790 nm. Because of the large width of the latter band, no additional spectral correction of the FROG trace is necessary, according to Ref. [2].

The apparent contradiction between the two recipes (subsequently in this Section called recipes I and II) arises from the difference in the calculation of the approximate expression for the spectral filter, $R(\Omega)$, imposed by the SHG process on the FROG trace (Eq.(3.33)). To comply with the line of reasoning in Ref. [2], in the expressions throughout this Section we drop the dispersion of the second-order nonlinearity and the extra $\Omega^2$-dependence responsible for the variation in the mode sizes of different frequency components. The resulting expression for the spectral filter is then given by:

$$R(\Omega) \propto \Omega^2 \text{sinc}^2 \left( \frac{\Delta k L}{2} \right).$$

The two recipes stated above are based on two different Taylor expansions of the effective phase mismatch, $\Delta k :$ around $\Omega/2$ (see Eq.(3.30)) in the first case, and around the carrier frequency of the pulse, $\omega_0$ [2,4,5], in the second. The resulting respective approximations are:
\[ \Delta k^{(i)}(\Omega) \equiv 2k_o \left( \frac{\Omega}{2} \right) - k_e(\Omega), \quad (4.2) \]

and

\[ \Delta k^{(ii)}(\Omega) \equiv 2k_o(\omega_0) - k_e(\omega_0) + (\Omega - 2\omega_0) \left( \frac{\partial k_o}{\partial \omega} \bigg|_{\omega_0} - \frac{\partial k_e}{\partial \omega} \bigg|_{2\omega_0} \right), \quad (4.3) \]

The spectral filters calculated using these approximate expressions for 970-nm and 700-nm cut BBO crystals are plotted in Fig.4.3. Indeed, the dashed line (970-nm-cut crystal) in Fig.4.3b in the approximation given by Eq.(4.3) seems to provide a much better choice than the 700-nm one.

![Figure 4.3](image)

**Fig.4.3:** Spectral filter for 10-µm BBO crystal for two different approximations of the phase mismatch. (a) and (b) represent the approximation given by Eq.(4.2) and Eq.(4.3), respectively. Solid and dashed curves are calculated for phase-matching wavelength of 700 nm and 970 nm, respectively. The shaded contour shows autoconvolution of super-Gaussian intensity spectrum supporting 4-fs pulses.

To test the implications of these two recipes and to verify the better approximation of the phase mismatch, we simulated SHG FROG measurements of a 4.5-fs pulse at 790 nm. In order to follow a realistic scenario, we assume that the spectrum of the laser pulses is super-Gaussian and the bandwidth supports pulses as short as 4 fs. The autoconvolution (SHG FROG spectral marginal) of the spectrum is shown in Fig.4.3a,b alongside with the spectral filters.

We next assume that the pulse is not perfectly compressed, and a small amount of a quartic spectral phase (cubic group delay) broadens the pulse to ~4.5 fs. The chosen fourth-order phase distortion roughly describes the residual phase of a hypothetical pulse compressor such as, for instance, a combination of prisms and diffraction grating or chirped mirrors. We now compute the FROG traces of this pulse according to Eq.(3.27) for a 700-nm and 970-nm-cut crystal. Note that since Eq.(3.27) is exact, no approximations about the phase mismatch are made in this calculation. Following the two recipes given above, the FROG
inversion algorithm is applied to the resulting traces: with the spectral correction given by Eq.(4.1,2) for the 700-nm crystal and directly to the other trace.

The pulse reconstructions corresponding to the two recipes are given in Fig.4.4b and Fig.4.4c. (dashed curves) along with the input pulse parameters (thin lines in Fig.4.4a-c). The respective FROG errors [6] for the matrix size of 128×128 are 0.0008 for recipe I, and 0.0042 for recipe II. The main source of error in the first case originates from the fact that, as has been shown in Section 3.6, the spectral filter is somewhat delay-dependent, which is not reflected in Eq.(4.1,2). The FROG error in the second case is much more substantial. However, the pulse duration is almost accurately recovered in both these cases. There is only a minor asymmetry on the recovered pulse in Fig.4.4c and the deviation from the input phase is not significant for the portions of the pulse carrying any appreciable intensity. Clearly, better criteria should be employed to judge if the pulse reconstruction in one ore both these cases were faulty. A possible candidate for such a criterion can be the intensity-weighed phase error [6]. Another recently demonstrated approach uses a comparison of Wigner representations of the input and reconstructed pulses [7]. A particular merit of Wigner representation is that it constitutes a very intuitive two-dimensional “fingerprint” of a laser pulse field. The properties of such Wigner traces are summarized in Appendix I. Particularly, such frequency-time-domain plots, also known as chronocyclic representation [8], asymptotically show the sequencing of frequency components in time according to the group delay.

The respective Wigner traces and the group delays of the input pulse and the two recovered pulses are depicted in Fig.4.4d–f. The Wigner trace error, defined in Appendix I, amounts to 0.035 for recipe I and to 0.238* for recipe II. According to Ref. [7], the error in excess of 0.15 represents an unacceptably poor pulse reconstruction. Indeed, while the Wigner trace of the recovered following recipe I pulse (Fig.4.4e) is nearly identical to the input one (Fig.4.4d), the result produced by recipe II exhibits a clearly different behavior. The inspection of Wigner traces in Fig.4.4d and Fig.4.4f conspicuously shows difference in instantaneous frequency spectra of the two pulses, especially at times below and above the half-width of the pulse. Obviously, the intensity of frequency components belonging to the spectral wings, which are displaced in time (Fig.4.4f), is very small to affect the overall correctness of the pulse duration measurement. On the other hand, such intensities are still usable for a variety of nonlinear optical experiments, for instance, frequency-resolved optical pump–probe. Consider the example, where the pulse discussed here serves as a probe in such a spectroscopic experiment. The false phase reconstruction, using recipe II, makes us believe that the sample interacts simultaneously with the blue and red frequencies of the probe pulse. Therefore, a possible increase of the nonlinear response of the matter at positive pump-probe delays, when the blue frequencies arrive in reality, can be mistakenly interpreted as having to do with the dynamics of the sample. Consequently, misjudged phase distortions of the pulse

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* Since time-direction ambiguity is present in SHG FROG reversal, the orientation of traces in Fig.4.4c and Fig.4.4f was chosen accordingly to the smallest Wigner trace error.
can lead to an erroneous interpretation of the data obtained in a nonlinear spectroscopic experiment.

**Fig. 4.4**: Comparison of SHG FROG recovery of a 4.5-fs pulse. (a) spectrum and phase of the input pulse. (b) and (c) time-domain intensity and phase of retrieved pulses (dotted curves) and input pulse (solid curves) for two different crystal choices (see the text for details). (d),(e), and (f) Wigner traces of the input and retrieved pulses. The thick curves show group delay. The convention on contour lines in (d)-(f) is the same as the one adopted in Chapter 3, dotted contour lines represent negative values.

In summary to this Section, we performed the quantitative assessment of the effect of using differently phase-matched crystals for the measurement of sub-5-fs pulses. Our simulations confirm that, in the calculation of the spectral SHG filter, the approximation of the phase mismatch given by Eq. (4.2) is superior to the one specified by Eq. (4.3). We finally comment that the approximation by Eq. (4.2) has a clear physical sense since, due to the increase of the crystal material dispersion approaching UV resonances, the phase mismatch must grow sharply toward the high frequencies. This produces a very distorted \( \text{sinc}^2 \) shape, –
the feature, which the approximation calculated from Eqs.(4.1,3) entirely fails to reproduce. We also point out that the amount of spectral filtering introduced by a 10-µm BBO crystal in the measurement of 5-fs pulses around 800 nm does not significantly affect the figure of the pulse duration. Therefore, in the given situation, it is unlikely that the error in labeling the pulse by its intensity FWHM would rise above 10% as a consequence of varying the phase-matching angle.

4.4 SHG FROG apparatus

In our experiments, we used pulses from a self-mode-locked cavity-dumped Ti:sapphire oscillator compressed upon chirping in a single-mode fused silica fiber. We measured the white-light continuum (WLC) pulses directly at the fiber output and, again, upon the their compression performed as described in Chapter 2.

The SHG FROG apparatus (Fig.4.5) is based on a phase and amplitude balanced multi-shot autocorrelator designed for sub-5-fs short pulses [9]. The input beam was split and recombined in such a way that each of the beams travels once through an identical 50% beam splitter while both reflections occurring on the same coating-air interfaces. To match the beam splitters, the initial horizontal polarization of the laser beam was rotated by a periscope. The moving arm of the autocorrelator was driven by a piezo transducer (Physik Instrumente) which is controlled by a computer via a digital-analog converter and a high voltage amplifier. The precise time calibration was provided by an auxiliary Michelson interferometer. The photodiode monitored the interference fringes that serve as time calibration marks.

Fundamental pulses were focussed in the nonlinear crystal with an \( r = -25 \text{-cm} \) spherical mirror at near normal incidence to minimize astigmatism. Due to the low curvature of the mirrors, delay variations within each beam are less than 0.1 fs. To achieve simultaneous up-conversion of the entire fundamental bandwidth, we employed a 10-µm-thick BBO crystal cut for a central wavelength of 700 nm (EKSMA Inc.). Dispersive lengthening of a 5-fs pulse by such crystal does not exceed 0.02 fs. The blue-shifted central wavelength permits one to extend the phase-matching bandwidth below 600 nm as shown in Fig.3.4c. The cut angle of the crystal was verified with a tunable 100-fs laser. Retro-reflection of the beams from the crystal surface provided exact reference for crystal orientation. This enables us to accurately calculate \( R(\Omega) \) required for data correction according to Eq.(3.33). A visible-IR PC1000 (Ocean Optics) spectrometer was used to detect the fundamental spectra.

Two different second harmonic detection systems were employed in the measurements of the compressed and the chirped pulses. In the case of compressed pulses, a well-characterized UV-Vis PC1000 (Ocean Optics) spectrometer was used. Therefore, the FROG traces could be readily corrected by \( R(\Omega) \), as described above.

\[ For \text{ shorter pulses, one should use lower-reflectivity beam splitters that have a broader reflectivity range and flatter spectral phase.} \]
In the case of the strongly chirped pulses a combination of a scanning monochromator and a photo-multiplier tube provided the dynamic range necessary to measure the spectral wings (see next Section). The reason for this was the following: The dynamic range of the measurement in a CCD-based spectrometer is determined not only by the spectral sensitivity, which is adequately high, but by the charge spreading all over the array due to overload of some channels. To further extend the dynamic range, a lock-in amplifier was used to detect the second-harmonic signal. Because of the unknown spectral sensitivity $Q(\Omega)$, the spectral correction of the FROG traces in this case was performed according to the method suggested in Taft et al. [3], i.e., by using the ratio of the autoconvoluted fundamental spectrum and the spectral marginal.

### 4.5 SHG FROG of white-light continuum

As has been shown in Chapter 2, the study of the group delay of the chirped WLC, is the corner stone of the pulse compression. The phase measurement of the pulses leaving the fiber permits to assess the feasibility of the pulse compression in general. Understandably, the spectral phase must be sufficiently smooth to allow compensation by the existing dispersion control means. A measurement of the spectral intensity, on the other hand, provides only a limited insight and reveals the minimum duration of the would-be compressed pulse. As an example of virtually uncompressible pulses, one might consider the case of spectral broadening due to a pure self-phase modulation. Furthermore, the task of building an appropriate pulse compressor is substantially eased if the phase distortion on the pulse is measured beforehand. This becomes increasingly important with the growth of the pulse spectral bandwidth that puts severe limitations on dispersion tunability of the pulse compressor. Therefore, it is desirable to replace a great deal of “trial and error” work by measuring the phase distortion and computing the settings of the pulse compressor.

Somewhat counter-intuitively, the FROG measurement of a strongly chirped pulse is considerably more complicated compared with the case a bandwidth-limited pulse with the

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**Fig.4.5**: Schematic of the SHG FROG apparatus. Spectrometer and its coupling optics are not shown.
identical spectrum. First, the up-conversion signals are weaker due to the lower peak power. This is evident, since the second harmonic intensity of a pulse that is stretched to the ten times its initial duration drops down 100 times.

Second, a higher dynamic range is required because of the uneven temporal spread of spectral wings. This occurs due to the high-order material dispersion. To explain this, we consider two spectral components with frequencies separated by 1000 cm\(^{-1}\). The group delay accumulated between them after passing 1 mm of quartz amounts to 4 fs if these components are situated around 1000 nm and exceeds 11 fs in the case of 600 nm. Evaluating roughly, the corresponding elements of the FROG trace scale ~7 times in intensity. In our experiments, the bandwidth of the WLC that needs to be captured in the FROG trace is broader than 10000 cm\(^{-1}\), and, therefore, the signal intensity varies very strongly across the resultant FROG traces.

The third complication is purely numerical, since FROG inversion demands greater matrix sizes to provide the adequate sampling in both time and frequency domain. For the sake of speed, the FROG inversion algorithms employ fast Fourier transform (FFT) [10]. To avoid the loss of information in the change from the time to the frequency domain and vice versa, FFT requires an equal number of points \(N\) in both these domains. Therefore, if the FROG matrix covers the total delay of \(\Delta \tau N\) in the time domain, where \(\Delta \tau\) is the time step, the spectral width represented in this trace is \(N/\Delta \tau\). Compared with bandwidth-limited pulses, the pulses stretched in time require larger \(\Delta \tau\) to contain the whole time information of the FROG trace in the matrix used in the FROG inversion algorithm. This narrows the spectral window covered by the matrix. Consequently, the number of points \(N\), that in FFT is an integer power of two, must be increased to fully represent the FROG trace in the matrix used by the algorithm. This has an appreciable effect on the calculation speed. The change of \(N\) from \(2^n\) to \(2^{n+1}\), where \(n\) is an integer number, slows the FROG retrieval by a factor of \(4(1+n^{-1})\). In other words, by changing a 128x128 matrix with a 256x256 one increases the calculation time by a factor of ~4.5.

Lastly, we point out the experimental inconvenience. In the case of strongly chirped pulses the crystal alignment and the detected FROG trace become very sensitive to the delay-dependent change in the direction of the second harmonic beam, as has already been discussed in Section 3.9.

The SHG FROG traces of the chirped WLC in our experiments were recorded in 2.5-fs delay steps and converted into 256x256 matrices for processing. To reveal the conditions best suited for the compression of the WLC we varied the parameters of the pulses entering the fiber, by changing of the settings of the prism precompressor. The intensity and the chirp of the input pulses, derived by SHG FROG, are shown in Fig.4.6a. The measured and retrieved FROG traces of the WLC are depicted in Figs.4.6c and d, and the retrieved WLC spectra and the group delay are shown in Fig.4.6b. The combined action of self-phase modulation and dispersion leads to a nearly linear group delay over most of the spectrum (Fig.4.6b, solid curves).
Fig. 4.6: Experimental results of FROG measurement of the strongly chirped white-light continuum (WLC). (a) temporal intensity (shaded contours) and chirp (solid curves) of the pulses entering a single-mode fused-silica fiber. (b) measured and (c) retrieved SHG FROG traces of the WLC. (d) retrieved spectral intensity (shaded contours) and the group delay of the WLC (solid curves). The amount of bulk material (fused silica) used to pre-chirp the input pulses is indicated in right top corners of (a). Note that the input pulse energy is kept constant, while the respective scaling of the WLC spectra in (d) is preserved.

The departure of the overall group delay from a linear asymptotic can be partly explained by the bulk dispersion of the fiber, air, and the beamsplitters in the FROG apparatus. For
instance, while the optimal fiber length was estimated to be 1 mm [11], we employed a 2-mm piece for the practical convenience and in order to clean the exiting mode structure.

The WLC spectrum changes dramatically with the change of the input pulses (Fig.4.6b, shaded contours). The widest and least modulated spectrum corresponds to the almost chirp-free input pulse (Fig.4.6b, the third from the top panel). Positive as well as negative chirping leads to a substantial narrowing of the WLC spectrum. In contrast, the overall behavior of the group delays shown as solid lines in Fig.4.6b, remains virtually unaffected. This ensures efficient pulse compression under different experimental conditions.

![Graph](image_url)

**Fig.4.7:** Group delay of the designed pulse compressor. Solid curve is calculated by dispersive ray-tracing and is depicted reversed in time. The broken curves are the measured group delay of the WLC reproduced from all panels in Fig.4.6b.

Group delay measurements of the generated continuum served as a target function for the design of the three-stage, high throughput compressor, consisting of a quartz 45°-prism pair, broadband chirped mirrors and thin-film Gires-Tournois dielectric interferometers [9]. The spectral bandwidth of the compressor is 590–1100 nm and is limited by the reflectivity of the employed chirped mirrors [12]. (See Fig.2.7b.) The phase characteristics of the compressor have been analyzed using dispersive ray tracing and mapped onto the measured group delay of the continuum. Figure 4.7 depicts the measured group delay for different pulses, entering the fiber (shown as broken curves) which are reproduced from Fig.4.6b and the calculated group delay of the pulse compressor (solid line). As one can see, our design compensates the group delay of the white light everywhere across the compressor bandwidth. The adjustment of the material of the prism-pair allows final fine optimization of the compressor dispersion, as judged form the FROG trace of the compressed pulses.
4.6 SHG FROG of compressed pulses

The FROG traces of the compressed pulses were recorded by incrementing the time delay between the arms in steps of 0.5 fs. The acquired two-dimensional arrays of points were converted into a 128×128 FROG matrix. The experimental and retrieved FROG traces of compressed pulses are depicted in Figs. 4.8a and b. The FROG error amounted to 0.004 and is mainly caused by the noise in the spectral wings which scaled up after the spectral correction of the FROG trace. The temporal marginal of the FROG trace has a nice correspondence with the independently measured intensity autocorrelation (Fig. 4.8c) obtained by detecting the whole second-harmonic beam. This suggests that no spatial filtering of the second-harmonic beam has taken place. Comparison of the FROG frequency marginal and the autoconvolution of the fundamental spectrum (Fig. 4.8d) indicates that no loss of spectral information has occurred.

Fig.4.8: The results of SHG FROG characterization of compressed pulses. (a) experimental and (b) retrieved traces. (c) temporal marginal (filled circles) and independently measured autocorrelation of 4.5-fs pulses (solid curve). (d) frequency marginal (filled circles) and autoconvolution of the fundamental spectrum (solid curve).
Figure 4.9 shows the retrieved intensity and phase in the time and frequency domains. To remove the time direction ambiguity in the measurement of the compressed pulses, we performed an additional FROG measurement introducing a known amount of dispersion (a thin fused silica plate) in front of the FROG apparatus. The obtained pulse duration is 4.5 fs while variations of the spectral phase (dashed line in Fig.4.9b) is less than ±π/4 across the whole bandwidth. These results fully confirm our previous analysis based on the interferometric autocorrelation [9].

![Figure 4.9](image_url)

**Fig.4.9:** Retrieved parameters of 4.5-fs pulses in the time (a) and frequency (b) domains. The FROG-retrieved intensity and phase are shown as shaded contours and dashed curves, respectively. Independently measured spectrum (filled circles) and computed residual phase of the pulse compressor (dash-dotted curve) are given in (b) for comparison.

To additionally verify both the self-consistency of our compressor calculations and the accuracy of the FROG retrieval, we compare the obtained spectral phase of the 4.5-fs pulse (Fig.4.9b, dashed curve) with the predicted residual phase of the pulse compressor (Fig.4.9b, dash-dotted curve). The close similarity of the two reassures us of the correctness all used procedures, including the measurement of the chirped WLC, the knowledge of the dispersion of compressor constituent parts, the numerical routines employed for the ray tracing analysis, and, finally, the characterization of the compressed pulses.

The electric field of the compressed pulses is shown in Fig.4.10. Approximately 2.5 optical cycles comprise its half-width. The heavy oscillations in the wings, however, indicate the imperfection of spectral phase correction and the toll that the modulation of the spectrum takes on the temporal structure of the pulse. The importance of the pulse energy carried in the wings of the pulse is only moderately critical for some experiments, for instance, such as $\chi^{(3)}$- or higher-nonlinearity-order spectroscopies, where the signal is proportional to a certain power of intensity. This leads to a “clean-up” of the signal. Indeed, the prominent
wing structure of the electric field (Fig.4.10) is effectively “suppressed” in the intensity profile (Fig.4.9a) because of the square dependence between the field and the intensity.

![Electric field E(t)](image)

**Fig.4.10:** Reconstructed electric field of 4.5-fs pulses. The electric field of continuous light-wave at 790 nm is drawn for reference.

The SHG FROG traces are generally considered unintuitive due to their symmetry along the delay axis [13-15]. We found out that in the case of nearly bandwidth-limited pulses, one can significantly increase the amount of information available from the simple visual inspection of the trace. In order to do so, every trace in the time domain at its corresponding second-harmonic wavelength should be normalized to unity. Effectively, this represents the FROG trace as a series of normalized autocorrelations. In the case of the pulse with an arbitrary spectrum and the flat spectral phase, such representation of the SHG FROG trace would give a streak of uniform thickness around zero delay. The result of such operation applied to the FROG trace of the 4.5-fs pulse is presented in Fig.4.11a. The variation of the thickness, that is, the width of autocorrelation at a given second-harmonic wavelength\(^*$, which can be seen in Fig.4.11a indicates the non-perfect pulse compression without the necessity to run the FROG inversion algorithm.

Figure 4.11b shows two autocorrelation traces derived from the spectrogram in Fig.4.11a at two separate wavelengths. The FWHM of the autocorrelation at 350 nm is merely 6 fs which is indicative of an ~4-fs pulse duration. However, the autocorrelation at 470 nm is three times broader. Such a difference clearly illustrates the effect of the spectral filtering in nonlinear crystal as well as second harmonic detection on the autocorrelation width. This also underscores the importance of pulse characterization by frequency-resolved\(^*$

\(^*$Here we apply the term “autocorrelation” to a slice of a frequency-resolved autocorrelation of the pulse intensity purely for the sake of convenience. In an arbitrary case, such a slice in itself is not necessarily an autocorrelation function of any real non-negative distribution.
(e.g., FROG) rather than non-frequency-resolved (e.g., intensity autocorrelation) methods if one deals with such broadband pulses.

Fig. 4.11: Normalized FROG data of the 4.5-fs pulses. (a) SHG FROG trace of compressed pulses normalized along the delay axis as described in the text. (b) autocorrelation traces derived from the FROG trace at the second-harmonic wavelength of 350 nm (solid curve) and 470 (dashed curve). Note that because of spectral selection the pulse duration estimated from the autocorrelation width can be both lower and higher than the real one and differ by as much as a factor of 3.

Finally, we note that the width of the autocorrelation traces, such as the ones shown in Fig. 4.11a, can be directly related to the instrument response of a spectroscopic experiment. For instance, the temporal resolution of a kinetic trace in a frequency-resolved pump-probe experiment [16,17] detected at 950 nm will be ~12 fs, albeit the weighted average pulse duration is 4.5 fs [18,19]. Therefore, the frequency-resolved measurement (as FROG) brings invaluable information even if the correct estimation of the pulse width could be achieved by other, simpler means, such as the autocorrelation measurement.

In closing to this Section, we summarize the accumulated here knowledge about the amplitude-phase properties of the compressed pulse by constructing a “fingerprint” form of a Wigner spectrogram (see Appendix I). As has been shown above in the example discussed in Section 4.3, direct information about the time sequence of light–matter interaction with the frequencies throughout the pulse spectrum is readily available from the Wigner plot. For instance, a simple examination of the spectrum and spectral phase (or group delay) (Fig. 4.9b) gives us the idea about the peak moment of time when the heaviest presence of a certain frequency component is observed. Such an examination, however, is unable to show a relative measure of how much, at any moment, and for how long, in total, this frequency component will be felt by matter on its passage through the latter. The Wigner plot (Fig. 4.12), on the other hand, allows such assessment by a simple visual inspection.
Several useful observations can be made on the basis of the trace depicted in Fig.4.12. First, we notice that the IR wing of the pulse precedes the arrival of the main body. This feature is inherited from the chirped pulse where the red-shifted frequencies are advanced while the blue-shifted ones are delayed. The failure to properly retard the IR wing is mostly explained by the dominating role in the infrared of the reversed (above 850 nm) third-order dispersion of the prism compressor. Second, we notice that the frequency components, corresponding to the sharp peaks on the spectrum (Fig.4.6d), occupy much broader time intervals than the rest of the “well-behaved” spectrum. These peaking frequencies dominate instantaneous intensity spectra seen at times $\pm 20$ fs around the main pulse. To some extent, the behavior of such sharp spectral irregularities may be viewed as a superposition of different pulses that are distinguished by a narrower spectral and broader temporal width. The implications of this on the interpretation of pump–probe data will be addressed in Chapter 7.

4.7 Conclusions and Outlook

SHG FROG is a powerful and accurate pulse diagnostics technique that is ideally suited for the measurement of a vast variety of pulses. In particular, the instantaneous nonlinearity, high sensitivity, and broadband response allow measuring the shortest pulses available to the date. The FROG measurement of the pulses that are shorter than 5 fs is nowadays probably the
only available means to evaluate the pulse parameters and the temporal resolution of a nonlinear spectroscopic experiment. We have applied the developed theory to the SHG FROG measurement of 2.5-optical-cycle pulses with a central wavelength around 800 nm. To the best of our knowledge, these are the shortest pulses that have been completely characterized to date. We have also successfully measured strongly non-spectral-limited weak-intensity pulses generated at the fiber output. These two key experiments that are required to design, test and optimize the pulse compressor have both been performed without a single change in the SHG FROG apparatus. Under the given conditions, no other pulse measuring technique known to the present day allows similar versatility.

FROG characterization of the chirped spectrally broadened pulses offers an important shortcut in the generation of the ever-shorter pulses via external compression. The direct phase measurement of the output of glass fibers, as demonstrated in this Chapter, hollow waveguides [20] and parametric amplification [21-23] provides a rigorous target function for the pulse compressor design. In particular, we foresee clear benefits for two direct methods of pulse compression: adaptive dispersion control and all-mirror compression.

In the first case, the whole pulse compressor or one stage of it consists of the computer-controlled intensity and phase masks [24] or an acousto-optical modulator [25]. The required phase pattern can be calculated and set to match the target function measured by FROG. Such straightforward finding of the optimal conditions allows eliminating the time-consuming iterative search based on the feedback [26] and guarantees the correctness of the phase corrections.

In the second case, in which no flexible control over the resulting dispersion of the pulse compressor is permitted, the precise knowledge of the target function is even more important. The well-developed theory of the chirped mirrors [27] makes it possible to design the adequate dielectric layer structure that in many cases almost perfectly follows the required dispersion curve, measured by FROG. In general, the phase distortion of nearly any complexity can be compensated for by a mirror that is based on the gradient change of the refractive index instead of the discrete dielectric layers, as is the case in the currently available chirped mirrors [12]. No doubt that with the growing interest in the intracavity [28,29] and extra-cavity broadband dispersion control [9,21,22,30], the possibility of manufacturing the gradient-index structures will shortly become available. Therefore, the phase measurement of chirped pulses gains paramount importance.

Appendix I: Wigner representation and Wigner trace error

A Wigner representation of a pulse [8], \( W(\tau,\omega) \), which is a two-dimensional trace in the time–frequency space, is straightforwardly calculated from the complex electric field in frequency, \( \tilde{E}(\omega) \):

\[
W(\tau,\omega) = \int_{-\infty}^{\infty} \tilde{E}(\omega') E^*(\omega - \omega') \delta(\omega - \omega') d\omega'
\]
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\[ W(t, \omega) = \int E^\ast \left( \omega - \frac{\Omega}{2} \right) E \left( \omega + \frac{\Omega}{2} \right) \exp(-i\Omega t) d\Omega \]  

(4A.1)

Alternatively, in a similar fashion, one can produce a Wigner trace starting from the complex electric field in the time domain, \( E(t) \).

\[ W(t, \omega) = \int E^\ast \left( t - \frac{\tau}{2} \right) E \left( t + \frac{\tau}{2} \right) \exp(i\omega \tau) d\tau \]  

(4A.2)

where \( \tilde{E}(\omega) \) and \( E(t) \) are a Fourier pair.

Because \( W(t, \omega) \) is a function of both time and frequency, it can be conveniently plotted as a two-dimensional spectrogram in the time-frequency domain. To reflect this fact, Wigner representation of the light pulse is also called chronocyclic [8].

Integration of \( W(t, \omega) \) along \( \omega \) or \( t \) produces pulse intensity in time or pulse spectrum, respectively.

\[ I(t) \equiv |E(t)|^2 = \int W(t, \omega) d\omega \]  

(4A.3)

\[ \tilde{I}(\omega) \equiv |\tilde{E}(\omega)|^2 = \int W(t, \omega) dt \]  

(4A.4)

\( W(t, \omega) \) is a real function that can be both positive and negative. The marginals of \( W(t, \omega) \) given by Eqs.(4A.3,4) are non-negative.

The Wigner representation is very intuitive since the shape of the contour basically reflects the group delay. In fact, for each time value it gives the instantaneous spectrum of frequencies [31]. For some classes of pulses, such as double pulses, however, this intuitiveness is lost [32]. Next to the intuitive properties, the Wigner trace contains a delicate balance between the amount of phase- and amplitude-information. While an element of a Wigner trace scales accordingly to the intensity, phase information remains responsible for the precise location in the time-frequency domain of the Wigner trace element corresponding to this intensity. Because Wigner traces give a linear distribution of a field, their comparison is significantly more sensitive than a comparison of corresponding FROG traces. For instance, like Wigner traces, \( \chi^{cbr} \)-based FROG traces also provide a quite intuitive delay-versus frequency distribution of the FROG signal. However, due to the fact that FROG is based on a nonlinear frequency-mixing, the response from weaker spectral components can be hidden under the pile-up of the signal at a given frequency.

The Wigner trace error, proposed in Ref. [7] computes the error between two Wigner matrices, \( W'(t, \omega) \) and \( W(t, \omega) \), in the following form:
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\[ \varepsilon = \sqrt{\sum_{i,j}^N \left[ W_{i,j}^0(t_i, \omega_j) - \alpha W_{i,j}(t_i, \omega_j) \right]^2} / \sqrt{\sum_{i,j}^N \left[ W_{i,j}^0(t_i, \omega_j) \right]^2}, \]  

(4A.5)

where \( \alpha \) is a scaling factor that minimizes \( \varepsilon \), and \( N \) is the size of the matrix. The error \( \varepsilon \) takes values from 0 to 1, the upper limit being the worst case scenario in which the discrepancy between the two matrices equals the value of the initial matrix itself. A valuable property of \( \varepsilon \) is that it is insensitive to the matrix size \( N \) and to the sampling along the time and frequency axes. The precise lateral overlap of the two Wigner traces in the time-frequency space is required to correctly compute \( \varepsilon \). This can be easily arranged by optimizing the respective overlap of their marginals (i.e. temporal and spectral intensities).

According to Ref. [7], the error level below \( \varepsilon = 0.15 \) corresponds to a reasonable amplitude-phase reconstruction of the target pulse represented by \( W^0(t, \omega) \), and the error below \( \varepsilon = 0.03 \) is considered excellent.
References

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