Chapter 3

Second Harmonic Generation Frequency-Resolved Optical Gating in the Single-Cycle Regime

Abstract

The problem of measuring broadband femtosecond pulses by the technique of second-harmonic generation frequency-resolved optical gating (SHG FROG) is addressed. We derive the full equation for the FROG signal, which is valid even for single-optical-cycle pulses. The effect of the phase-mismatch in the second-harmonic crystal, the implications of the beam geometry and the frequency-dependent variation of the nonlinearity are discussed in detail. Our numerical simulations show that under carefully chosen experimental conditions and with a proper spectral correction of the data the traditional FROG inversion routines work well even in the single-cycle regime.
3.1 Introduction

Recent progress in complete characterization of ultrashort pulses reflects the growing demand for detailed information on pulse structure and phase distortion. This knowledge plays a decisive role in the outcome of many applications. For instance, it has been recognized that pulses with identical spectra but different spectral phases can strongly enhance efficiency of high-harmonic generation [1], affect wavepacket motion in organic molecules [2,3], enhance population inversion in liquid [4] and gas [5] phases, and even steer a chemical reaction in a predetermined direction [6]. Moreover, a totally automated search for the best pulse was recently demonstrated to optimize a pre-selected reaction channel [7]. Then, by measuring the phase and amplitude of the excitation pulses, one can perform a back-reconstruction of potential surfaces of the parent molecule.

The complete determination of the electric field of femtosecond pulses also uncovers the physics behind their generation as has been demonstrated in the case of fs Ti:sapphire lasers [8,9]. Such information is invaluable to determine the ways of and ultimate limits for further pulse shortening. Last, owing to the great complexity of broadband phase correction required to produce spectrum-limited pulses with duration shorter than 5 fs [10-13], the characterization of the white-light continuum as well as compressed pulses becomes mandatory.

A breakthrough in the full characterization of ultrashort pulses occurred six years ago with the introduction of frequency-resolved optical gating (FROG) [14,15]. FROG measures a two-dimensional spectrogram in which the signal of any autocorrelation-type experiment is resolved as a function of both time delay and frequency [16]. The full pulse intensity and phase may be subsequently retrieved from such a spectrogram (called FROG trace) via an iterative retrieval algorithm. Notably, no a priori information about the pulse shape, as it is always the case for conventional autocorrelation measurements, is necessary to reconstruct the pulse from the experimental FROG trace.

In general, FROG is quite accurate and rigorous [17]. Because a FROG trace is a plot of both frequency and delay, the likelihood of the same FROG trace corresponding to different pulses is very low. Additionally, the great number of data points in the two-dimensional FROG trace makes it under equivalent conditions much less sensitive to noise than the pulse diagnostics based on one-dimensional measurements, such as the ordinary autocorrelation. Last but not least, FROG offers data self-consistency checks that are unavailable in other pulse measuring techniques. This feedback mechanism involves computing the temporal and spectral marginals that are the integrals of the FROG trace along the delay and frequency axes. The comparison of the marginals with the independently measured fundamental spectrum and autocorrelation verifies the validity of the measured FROG trace [9,18,19]. To date, FROG methods have been applied to measure a vast variety of pulses with different duration, wavelength and complexity [20].
A number of outstanding features make FROG especially valuable for the measurement of extremely short pulses in the range of 10 fs and below.

First, since FROG utilizes the excite-probe geometry, common for most nonlinear optical experiments, it is ideally suited to characterize pulses that are used in many spectroscopic laboratories. Unlike other pulse diagnostics [21-25], FROG does not require splitting of auxiliary laser beams and pre-fabrication of reference pulses. This fact is of great practical relevance, since the set-up complexity in many spectroscopic experiments is already quite high [26-32]. Therefore, it is desirable to minimize the additional effort and set-up modifications that are necessary for proper pulse diagnostics. FROG directly offers this possibility. Pulse characterization is performed precisely at the position of the sample by simply interchanging the sample with a nonlinear medium for optical gating. The last point becomes especially essential for the pulses consisting of only several optical cycles [10-13,33] currently available for spectroscopy. The dispersive lengthening that such pulses experience even due to propagation through air precludes the use of a separate diagnostics device. Thus, FROG is the ideal way to measure and optimize pulses on target prior to carrying out a spectroscopic experiment.

Second, it is still possible to correctly measure such short pulses by FROG even in presence of systematic errors. Several types of such errors will inevitably appear in the measurement of pulses whose spectra span over a hundred nanometers or more. For example, a FROG trace affected by wavelength-dependent detector sensitivity and frequency conversion efficiency can be validated via the consistency checks [9]. In contrast, an autocorrelation trace measured under identical conditions may be corrupted irreparably.

Third, the temporal resolution of the FROG measurement is not limited by the sampling increment in the time domain, provided the whole time-frequency spectrogram of the pulse is properly contained within the measured FROG trace. The broadest feature in the frequency domain determines in this case the shortest feature in the time domain. Therefore, no fine pulse structure can be overlooked [20], even if the delay increment used to collect the FROG trace is larger that the duration of such structure. Thus, reliability of the FROG data relies more on the proper delay axis calibration rather than on the very fine sampling in time, which might be troublesome considering that the pulse itself measures only a couple of micrometers in space.

Choosing the appropriate type of autocorrelation that can be used in FROG (so-called FROG geometries [18,20]), one must carefully consider possible distortions that are due to the beam arrangement and the nonlinear medium. Consequently, not every FROG geometry can be straightforwardly applied to measure extremely short pulses, i.e. 10 fs and below. In particular, it has been shown that in some $\chi^{(3)}$-based techniques (for instance, polarization-gating, transient grating etc.) the finite response time due to the Raman contribution to nonlinearity played a significant role even in the measurement of 20-fs pulses [34]. Therefore, the FROG with the use of the second harmonic generation in transparent crystals...
[35-37] and surface third-harmonic generation [38], that have instantaneous nonlinearity, presents the best choice for the measurement of the shortest pulses available to date.

Another important experimental concern is the level of the signal to be detected in the FROG measurement. Among different FROG variations, its version based on second harmonic generation (SHG) is the most appropriate technique for low-energy pulses. Obviously, SHG FROG [35] potentially has a higher sensitivity than the FROG geometries based on third order nonlinearities that under similar circumstances are much weaker. Different spectral ranges and polarizations of the SHG FROG signal and the fundamental radiation allow the effective suppression of the background, adding to the suppression provided by the geometry. The low-order nonlinearity involved, combined with the background elimination, results in the higher dynamic range in SHG FROG than in any other FROG geometry.

In general, the FROG pulse reconstruction does not depend on pulse duration since the FROG traces simply scale in the time-frequency domain. However, with the decrease of the pulse duration that is accompanied by the growth of the bandwidth, the experimentally collected data begin to deviate significantly from the mathematically defined ideal FROG trace. Previous studies [8,9] have addressed the effect of the limited phase-matching bandwidth of the nonlinear medium [39] and time smearing due to non-collinear geometry on SHG FROG measurement which become increasingly important for 10-fs pulses. The possible breakdown of the slowly-varying envelope approximation and frequency dependence of the nonlinearity are the other points of concern for the pulses that consist of a few optical cycles. Some of these issues have been briefly considered in our recent Letter [40].

In this Chapter we provide a detailed description of SHG FROG performance for ultrabroadband pulses the bandwidths of which correspond to 3-fs spectral-transformed duration. Starting from the Maxwell equations, we derive a complete expression for the SHG FROG signal that is valid even in a single-cycle pulse regime and includes phase-matching in the crystal, beam geometry, dispersive pulse-broadening inside the crystal and dispersion of the second-order nonlinearity. Subsequently, we obtain a simplified expression that decomposes the SHG FROG signal to a product of the ideal SHG FROG and a spectral filter applied to the second harmonic radiation. Numerical simulations, further presented in this Chapter, convincingly show that the approximations made upon the derivation of the simplified expression, are well justified.

The outline of the Chapter is the following: in Section 3.2 we define the pulse intensity and phase in time and frequency domains. In Section 3.3 the spatial profile of ultrabroadband pulses is addressed. The complete expression for SHG FROG signal for single-cycled pulses is derived in Section 3.4. We discuss the ultimate time resolution of the SHG FROG in Section 3.5. The approximate expression for the SHG FROG signal, obtained in Section 3.6, is verified by numerical simulations in Section 3.7. In Section 3.8 we briefly comment on Type II phase-matching in SHG FROG measurements. Possible distortions of the
3.2 Amplitude and phase characterization of the pulse

The objective of a FROG experiment lies in finding the pulse intensity and phase in time, that is \( I(t) \), \( \varphi(t) \) or, equivalently, in frequency \( \tilde{I}(\omega) \), \( \tilde{\varphi}(\omega) \). The laser pulse is conventionally defined by its electric field:

\[
E(t) = A(t) \exp(i\varphi(t)),
\]

where \( A(t) \) is the modulus of the time-dependent amplitude, and \( \varphi(t) \) is the time-dependent phase. The temporal pulse intensity \( I(t) \) is determined as \( I(t) \propto A^2(t) \). The time-dependent phase contains information about the change of instantaneous frequency as a function of time (the so-called chirp) that is given by [41,42]:

\[
\omega(t) = \frac{\partial \varphi(t)}{\partial t}.
\]

The chirped pulse, therefore, experiences a frequency sweep in time, i.e. changes frequency within the pulse length.

The frequency-domain equivalent of pulse field description is:

\[
\tilde{E}(\omega) = \int E(t) \exp(i\omega t) dt \equiv \tilde{A}(\omega) \exp(i\tilde{\varphi}(\omega)),
\]

where \( \tilde{E}(\omega) \) is the Fourier transform of \( E(t) \), and \( \tilde{\varphi}(\omega) \) is the frequency-dependent (or spectral) phase. Analogously to the time domain, the spectral intensity, or the pulse spectrum, is defined as \( \tilde{I}(\omega) \propto \tilde{A}^2(\omega) \). The relative time separation among various frequency components of the pulse, or group delay, can be determined by [42]

\[
\tau(\omega) = \frac{\partial \tilde{\varphi}(\omega)}{\partial \omega}.
\]

Hence, the pulse with a flat spectral phase is completely “focused” in time and has the shortest duration attainable for its bandwidth.

It is important to notice that none of the presently existing pulse measuring techniques retrieves the absolute phase of the pulse, i.e. pulses with phases \( \varphi(t) \) and \( \varphi(t) + \varphi_0 \) appear to be totally identical [43]. Indeed, all nonlinear processes employed in FROG are not sensitive to the absolute phase. However, the knowledge of this phase becomes essential in the strong-
field optics of nearly single-cycled pulses [44,45]. It has been suggested [46], that the absolute phase may be assessable via photoemission in the optical tunneling regime [47].

In fact, the full pulse characterization remains incomplete without the analysis of spatio-temporal or spatio-spectral distribution of the pulse intensity. In this Chapter we assume that the light field is linearly polarized and that each spectral component of it has a Gaussian spatial profile. The Gaussian beam approximation is discussed in detail in the next Section.

### 3.3 Propagation and focusing of single-cycle pulses

The spatial representation of a pulse which spectral width is close to its carrier frequency is a non-trivial problem. Because of diffraction, lower-frequency components have stronger divergence compared with high-frequency ones. As a consequence, such pulse parameters as the spectrum and duration are no longer constants and may change appreciably as the beam propagates even in free space [48].

We represent a Gaussian beam field in the focal plane as:

\[
\tilde{E}(x, y, \omega) = \tilde{E}(\omega) \sqrt{\frac{2\ln 2}{\pi}} \frac{1}{d(\omega)} \exp \left[-2 \ln 2 \frac{x^2 + y^2}{d^2(\omega)} \right],
\]

where \( d(\omega) \) is the beam diameter (FWHM) of the spectral component with the frequency \( \omega \) and \( x \) and \( y \) are transverse coordinates. The normalization factors are chosen to provide the correct spectrum integrated over the beam as measured by a spectrometer:

\[
\tilde{I}(\omega) \propto \iint \left|\tilde{E}(x, y, \omega)\right|^2 \, dxdy
\]

We now calculate the beam diameter after propagating a distance \( z \):

\[
d(\omega, z) = d(\omega, z = 0) \sqrt{1 + \left(\frac{2cz}{d^2(\omega, z = 0)\omega}\right)^2},
\]

where \( c \) is the speed of light in vacuum. To avoid the aforementioned problems, we require diameters of different spectral components to scale proportionally as the Gaussian beam propagates in free space, i.e.

\[
d^2(\omega, z = 0)\omega = \text{const}
\]
The constant in Eq.(3.8) can be defined by introducing the FWHM beam diameter $d_0$ at the central frequency $\omega_0$. Therefore, the electric field of the Gaussian beam given by Eq.(3.5) becomes

$$\tilde{E}(x, y, \omega) = \tilde{E}(\omega) \sqrt{\frac{2 \ln 2}{\pi}} \frac{1}{d_0} \sqrt{\frac{\omega}{\omega_0}} \exp \left[-2 \ln 2 \frac{x^2 + y^2}{d_0^2} \frac{\omega}{\omega_0} \right]$$

(3.9)

At this point, the question can be raised about the low-frequency components the size of which, according to Eq.(3.9), becomes infinitely large. However, the spectral amplitude of these components decreases rapidly with frequency. For instance, the spectral amplitude of a single-cycle Gaussian pulse with a central frequency $\omega_0$ is given by

$$\tilde{A}(\omega) = \exp \left[-\frac{\pi^2}{2 \ln 2} \left(1 - \frac{\omega}{\omega_0} \right)^2 \right]$$

(3.10)

Consequently, the amplitude of the electric field at zero frequency amounts to only 0.1% of its peak value.

The spatial frequency distribution was observed experimentally with focused terahertz beams [49] and was discussed recently by S. Feng et al. [50]. Note that our definition of transversal spectral distribution in the beam implies that confocal parameters of all spectral components are identical:

$$b = \frac{d^2(\omega, z = 0)\omega}{4 \ln 2} = \frac{d_0^2 \omega_0}{4 \ln 2}$$

(3.11)

This is totally consistent with the beam size in laser resonators where longer wavelength components have a larger beam size. The spatial distribution of radiation produced due to self-phase modulation in single-mode fibers is more complicated. First, the transverse mode is described by the zero-order Bessel function [51]. Second, near the cut-off frequency the mode diameter experiences strong changes [52]. However, for short pieces of fiber conventionally used for pulse compression and reasonable values of a normalized frequency $V$ [51] it can be shown that a Gaussian distribution given by Eq.(3.9) is an acceptable approximation. The situation with hollow waveguides [53] is quite different since all spectral components have identical radii [54].

Another important issue concerns beam focusing which should not change the distribution of spectral components. Since the equations for mode-matching contain only confocal parameters [55], the validity of Eq.(3.9) at the new focal point is automatically fulfilled provided, of course, the focusing remains achromatic.
Fig.3.1: Spatial parameters of an ideal single-cycle Gaussian pulse centered at 800 nm. (a) Spatial intensity profiles of two spectral components that are separated by FWHM \( \Delta \omega \) from the central frequency \( \omega_0 \). (b) Intensity spectra as a function of transverse coordinate \( x \). (c) Dependence of pulse central frequency (solid curve) and pulse duration (dashed curve) on transverse coordinate \( x \). The beam axis corresponds to \( x=0 \).

Although Eq.(3.9) ensures that different spectral components scale identically during beam propagation and focusing, it also implies that the pulse spectrum changes along the transversal coordinates. Fortunately, this effect is negligibly small even in the single-cycle regime. Figure 3.1a shows the spatial intensity distribution of several spectral components of a Gaussian single-cycle pulse with a central wavelength 800 nm. As one moves away from the beam axis, a red shift is clearly observed (Fig.3.1b), since the higher frequency spectral components are contained in tighter spatial modes. However, the change of the carrier frequency does not exceed 10\% (Fig.3.1c, solid line), while the variation of the pulse-width is virtually undetectable (Fig.3.1c, dotted line). Therefore, this kind of spatial chirp can be disregarded even for the shortest optical pulses.

3.4 The SHG FROG signal in the single-cycle regime

In this Section, the complete equation is derived which describes the SHG FROG signal for pulses as short as one optical cycle. We consistently include such effects as phase-matching conditions in a nonlinear crystal, time-smearing effects due to non-collinear geometry,
spectral filtering of the second harmonic radiation, and dispersion of the second-order nonlinearity.

We consider the case of non-collinear geometry in which the fundamental beams intersect at a small angle (Fig.3.2). As it has been pointed out [39] the pulse broadening due to the crystal bulk dispersion is negligibly small compared to the group-velocity mismatch. This means that the appropriate crystal thickness should mostly be determined from the phase-matching conditions. For instance, in a 10-µm BBO crystal the bulk dispersion broadens a single-cycle pulse by only by ~0.1 fs while the group-velocity mismatch between the fundamental and second-harmonic pulses is as much as 0.9 fs.

![Fig.3.2: Non-collinear phase matching for three-wave interaction.](image)

$k(\omega)$ and $k(\Omega - \omega)$ are the wavevectors of the fundamental fields that form an angle $\alpha$ with z axis. $k_{SH}(\Omega)$ is the wave-vector of the second-harmonic that intersects z axis at an angle $\beta$.

We assume such focusing conditions of the fundamental beams that the confocal parameter and the longitudinal beam overlap of the fundamental beams are considerably longer than the crystal length. For instance, for an ideal Gaussian beam of ~2-mm diameter focused by a 10-cm achromatic lens the confocal parameter, that is, the longitudinal extent of the focal region, is ~1.2 mm. This is considerably longer than the practical length of the nonlinear crystal. Under such conditions wavefronts of the fundamental waves inside the crystal are practically flat. Therefore, we treat the second harmonic generation as a function of the longitudinal coordinate only and include the transversal coordinates at the last step to account for the spatial beam profile (Eq.(3.9)). Note that the constraint on the focusing is not always automatically fulfilled. For example, the use of a 1-cm lens in the situation described above reduces the length of the focal region to only 12 µm, and, in this case, it is impossible to disregard the dependence on transverse coordinates.

We assume that the second-harmonic field is not absorbed in the nonlinear crystal. This is well justified even for single-cycle pulses. Absorption bands of the crystals that are transparent in the visible, start at ~200 nm. Consequently, at these frequencies the field amplitude decreases by a factor $\exp\left(-\frac{\pi^2}{2\ln 2}\right) \approx 0.001$ (Eq.(3.10)) compared to its maximum at 400 nm. We also require the efficiency of SHG to be low enough to avoid depletion of the fundamental beams. Hence, the system of two coupled equations describing nonlinear interaction [56] is reduced to one. The equation that governs propagation of the
second harmonic wave in the $+z$ direction inside the crystal can be obtained directly from Maxwell’s equations [57]:

$$\frac{\partial^2}{\partial z^2} E_{SH}(z,t) - \varepsilon_0 \mu_0 \frac{\partial^2}{\partial t^2} \int_{-\infty}^{t} \varepsilon(t-t') E_{SH}(z,t') \, dt' = \mu_0 \frac{\partial^2}{\partial t^2} P^{<2>}(z,t), \quad (3.12)$$

where $E_{SH}(z,t)$ is the second harmonic field, $\mu_0 \varepsilon_0 = 1/c^2$, $\varepsilon$ is the relative permittivity, and $P^{<2>}(z,t)$ is the induced second-order dielectric polarization. By writing both $E_{SH}(z,t)$ and $P^{<2>}(z,t)$ as a Fourier superposition of monochromatic waves, one obtains a simple equivalent of Eq.(3.12) in the frequency domain:

$$\frac{\partial^2}{\partial z^2} \tilde{E}_{SH}(z, \Omega) + k_{SH}^2(\Omega) \tilde{E}_{SH}(z, \Omega) = -\mu_0 \Omega^2 \tilde{P}^{<2>}(z, \Omega), \quad (3.13)$$

where $\tilde{E}_{SH}(z, \Omega)$ and $\tilde{P}^{<2>}(z, \Omega)$ are Fourier transforms of $E_{SH}(z,t)$ and $P^{<2>}(z,t)$, respectively, $\Omega$ is the frequency and $k_{SH}(\Omega)$ is the wave-vector of the second harmonic field: $k_{SH}^2(\Omega) = \Omega^2 \varepsilon_0 \mu_0 \tilde{\varepsilon}(\Omega)$, with $\tilde{\varepsilon}(\Omega)$ being the Fourier-transform of the relative permittivity $\varepsilon(t)$.

In order to simplify the left part of Eq.(3.13), we write the second harmonic field as a plane wave propagating along $z$ axis:

$$\tilde{E}_{SH}(z, \Omega) = \tilde{E}_{SH}(z, \Omega) \exp(ik_{SH}(\Omega)z), \quad (3.14)$$

whence Eq.(3.13) becomes:

$$2ik_{SH}(\Omega) \frac{\partial}{\partial z} \tilde{E}_{SH}(z, \Omega) + \frac{\partial^2}{\partial z^2} \tilde{E}_{SH}(z, \Omega) = -\mu_0 \Omega^2 \tilde{P}^{<2>}(z, \Omega) \exp(-ik_{SH}(\Omega)z) \quad (3.15)$$

So far we have made no simplifications concerning the pulse duration. Now we apply the slowly-varying amplitude approximation [57], i.e.

$$\left| \frac{\partial}{\partial z} \tilde{E}_{SH}(z, \Omega) \right| \ll |2k_{SH}(\Omega)\tilde{E}_{SH}(z, \Omega)|, \quad (3.16)$$

in order to omit the term $\frac{\partial^2}{\partial z^2} \tilde{E}_{SH}(z, \Omega)$.

Note, that the use of the time-domain description of the signal wave propagation results in a second-order differential equation, similar in its structure to Eq.(3.15). Unlike Eq.(3.15), though, simplification of the time-domain expression requires a rejection of the second-order
temporal derivative of the envelope, i.e. \[ \frac{\partial^2}{\partial t^2} E(t) \ll \left| \frac{4\pi}{T_{\text{per}}} \frac{\partial}{\partial t} E(t) \right| , \]

where \( T_{\text{per}} \) is the characteristic period of light oscillation. Such a move implies the assumption of the slow envelope variation as a function of time. This condition is not fulfilled for the pulses that carry only a few cycles, since the change of the envelope within one optical period is comparable to the magnitude of the envelope itself. Brabec and Krausz [58], who explored the time-domain approach for the propagation of nearly monocycle pulses, found out that the rejection of the second-order derivative term is warranted in the case when the phase and the group velocities of light are close to each other. To this point we notice that the application of non-equality (3.16) to the frequency-domain Eq.(3.15) does not require any assumptions on the change of the temporal envelope altogether. Therefore, non-equality (3.16) is safe to apply even to monocyclic pulses, provided there is no appreciable linear absorption at lengths comparable to the wavelength. The only point of concern is related to the lowest frequencies for which \( k_{\text{SH}} \) becomes close to zero. However, as we have already mentioned in Section 3.3, the amplitude of such components does not exceed 0.1% of the maximum and therefore can be disregarded. Consequently, Eq.(3.15) can be readily solved by integration over the crystal length \( L \):

\[
\tilde{E}^-(L, \Omega) = i \frac{c \mu_0 \Omega}{2n_{\text{SH}}(\Omega)} \int_0^L \tilde{P}^{<\alpha}(z, \Omega) \exp(-k_{\text{SH}}(\Omega) z) dz
\]

where \( n_{\text{SH}}(\Omega) = \sqrt{\epsilon(\Omega)} \) is the refractive index for the second harmonic wave. Now we should calculate the second-order polarization \( \tilde{P}^{<\alpha}(z, \Omega) \). We assume that two fundamental fields cross in the \( xz \) plane at a small angle \( 2\alpha_0 \) (Fig.3.2). The inclination with the \( z \) axis of each beam inside the crystal is then \( \alpha(\omega) = \arcsin[n(\omega) \sin \alpha_0] \approx \alpha_0 n(\omega) \). We denote the relative delay between the pulses as \( \tau \). An additional delay for off-axis components of the beam due to the geometry can be expressed for a plane wave as \( \tau'(x) = x n(\omega) \sin \alpha(\omega) / c = x n(\omega) \sin \alpha_0 / c = x \alpha_0 / c \) for the beam propagating in \( +\alpha \) direction, and \( \tau'(x) = -x \alpha_0 / c \) for the beam in \( -\alpha \) direction. The electric fields in the frequency domain can be found via Fourier transforms:

\[
\begin{align*}
\tilde{E}_1(\omega) &= \tilde{E}(\omega) \exp(i\omega(x \alpha_0 / c)) \\
\tilde{E}_2(\omega) &= \tilde{E}(\omega) \exp(i\omega(-x \alpha_0 / c - \tau))
\end{align*}
\]

In order to calculate the second-order dielectric polarization induced at frequency \( \Omega \) by the two fundamental fields, we should sum over all possible permutations of fundamental frequencies:

\[ \tilde{P}^{<\alpha}(z, \Omega) \]
In Eq. (3.19) we included frequency-dependence of the nonlinear susceptibility \( \tilde{\chi}^{(2)}(\Omega, \omega, \Omega - \omega) \) and represent the fundamental field analogously to Eq. (3.14). The electric field of the second harmonic therefore becomes

\[
\tilde{E}_s^\approx (L, \Omega) = i\frac{\varepsilon \mu_0 \Omega L}{2n(\Omega)} \exp(i\Omega(\tau + x\alpha_0 / c)) \int \tilde{\chi}^{(2)}(\Omega, \omega, \Omega - \omega) \tilde{E}_z^\approx (\omega) \times \exp[i(k_\perp(\omega)z + k_z(\Omega - \omega)z + \omega(\tau + 2x\alpha_0 / c))] \, d\omega, \tag{3.20}
\]

where \( \Delta k(\omega, \Omega - \omega) \) is the phase mismatch given by the equation:

\[
\Delta k(\omega, \Omega - \omega) = k(\omega) \cos(\alpha_0 n_1(\omega)) + k(\Omega - \omega) \cos(\alpha_0 n_2(\Omega - \omega)) - k_{SH}(\Omega) \cos(\beta(\Omega, \Omega - \omega)), \tag{3.21}
\]

with \( n_1 \) and \( n_2 \) being the refractive indices of the fundamental waves, and \( \beta(\omega, \Omega - \omega) \) being the angle between \( k_{SH}(\Omega) \) and the \( z \) axis inside the crystal. The appearance of this angle can be easily understood from Fig. 3.2. The momentum conservation law determines the direction of emitted second harmonic field:

\[
k(\omega) + k(\Omega - \omega) = k_{SH}(\Omega), \tag{3.22}
\]

where \( k(\omega) \) and \( k(\Omega - \omega) \) are the wave-vectors of the incident fundamental waves. In the case \( k(\omega) \neq k(\Omega - \omega) \), \( \beta \) is non-zero and it can be found from the following equation\(^*\):

\[
\sin \beta(\omega, \Omega - \omega) = \sin \alpha_0 \frac{k(\omega)n_1(\omega) - k(\Omega - \omega)n_2(\Omega - \omega)}{k_{SH}(\Omega)} \tag{3.23}
\]

\(^*\) In fact, if the second harmonic is an extraordinary wave, the magnitude of \( k_{SH}(\Omega) \) in Eq. (23) is a function of \( \beta(\omega, \Omega - \omega) \). The problem of finding the exact values of both \( k_{SH}(\Omega) \) and \( \beta(\omega, \Omega - \omega) \) could be easily solved by employing the relations of crystaloptics and Eq. (3.23). However, Eq. (3.23) alone gives an excellent approximation for \( \beta(\omega, \Omega - \omega) \) if one chooses \( k_{SH}(\Omega) \) if one chooses \( k_{SH}(\Omega) \).
Since $\beta$ is of the same order of magnitude as the intersection angle, the correction $\cos\beta(\omega,\Omega-\omega)$ is required only in the $\Delta k$ expression (Eq.(3.21)). Elsewhere this correction can be dropped.

The values of the wave-vectors and refractive indices in Eqs.(3.21) and (3.23) depend on the actual polarization of the three interacting waves. Thus, for Type I we obtain:

$$\Delta k(\omega,\Omega-\omega) = k_o(\omega)\cos(\alpha_o n_o(\omega)) + k_o(\Omega-\omega)\cos(\alpha_o n_o(\Omega-\omega)) - k_e(\Omega)\cos\beta(\omega,\Omega-\omega)$$

(3.24)

and for Type II:

$$\Delta k(\omega,\Omega-\omega) = k_e(\omega)\cos(\alpha_e n_e(\omega)) + k_o(\Omega-\omega)\cos(\alpha_o n_o(\Omega-\omega)) - k_e(\Omega)\cos\beta(\omega,\Omega-\omega)$$

(3.25)

Here indices $O$ and $E$ correspond to the ordinary and extraordinary waves, respectively.

To calculate the total FROG signal, one should integrate the signal intensity

$$I_{SH}(L,\Omega) = \varepsilon_o n_{SH}(\Omega) \frac{1}{c} \left| \tilde{F}_{SH}(L,\Omega) \right|^2$$

(3.26)

over the transverse coordinates $x$ and $y$. Hence, for the second-harmonic signal detected in FROG we obtain:

$$S(\Omega,\tau,L) = \frac{\Omega^2 L^2 \sqrt{Q(\Omega)}}{2 c^3 \varepsilon_o n_{SH}(\Omega)} \left[ \ln \frac{2}{\pi \omega_0} \right]^{3/2} \int \exp \left[ -4 \ln 2 \left( \frac{x}{\omega_0} \right)^2 \right] \int_0^\Omega \tilde{X}^{c2\tau}(\Omega,\omega,\Omega-\omega) \sqrt{\omega \left( 1 - \frac{\omega}{\Omega} \right)} \times$$

(3.27)

$$\tilde{E}^\omega(\Omega-\omega)\tilde{E}^\omega(\omega) \exp \left( i \frac{\Delta k(\omega,\Omega-\omega)L}{2} + i\omega \left( \tau + \frac{2 \pi a_o}{c} \right) \right) \text{sinc} \left( \frac{\Delta k(\omega,\Omega-\omega)L}{2} \right) d\omega \right|^2 dx$$

In Eq.(3.27), $Q(\Omega)$ is the spectral sensitivity of the photodetector. We also took into consideration transverse profiles of the fundamental beams as given in Section 3.3.

Thus far we have limited our discussion to the case of low-efficiency second-harmonic generation, i.e. when the depletion of the fundamental waves can be disregarded. In the high conversion efficiency regime, however, additional effects play an important role. While the second-harmonic intensity depends quadratically on the crystal length $L$ in the case of undepleted pump [59], in the high-efficiency regime, conversion efficiency “saturates” for more intense spectral modes but remains proportional to $L^2$ for the weaker ones. Consequently, the FROG traces measured in a Type I SHG crystal in presence of significant pump depletion typically have both spectral and temporal marginals broader compared with
the low conversion efficiency case. Hence, despite seemingly increased bandwidth in the high-efficiency regime, the FROG trace is intrinsically incorrect. The case of the high-efficiency SHG in a Type II crystal [60,61] is more complex than in the Type I and can result in both shortening and widening of the temporal width of the FROG trace. Another important example of the second-harmonic spectral shaping in the high-conversion-efficiency regime is the nonlinear absorption of the frequency-doubled radiation inside the SHG crystal [62]. Therefore, the high-efficiency second-harmonic conversion is a potential source of systematic errors in a FROG experiment and should be avoided.

To conclude this Section, we would like to make a remark on the frequency – as opposed to time – domain approach to the wave equation Eq.(3.12) in the single optical cycle regime. Clearly, the former has a number of advantages. The spectral amplitude of a femtosecond pulse is observable directly while the temporal amplitude is not. The frequency representation allowed us to include automatically dispersive broadening of both fundamental and second-harmonic pulses as well as their group mismatch, frequency-dependence of the nonlinear susceptibility, frequency-dependent spatial profiles of the beams, and the blue shift of the second-harmonic spectrum (analog of self-steepening in fibers [51]). Furthermore, we have made a single approximation given by Eq.(3.16), which is easily avoidable in computer simulations. Eq.(3.20) can also be used to describe the process of SH generation in the low pump-depletion regime to optimize a compressor needed to compensate phase distortions in the SH pulse. Extension of the theory to the high conversion efficiency by including the second equation for the fundamental beam is also straightforward. Note that a similar frequency-domain approach to ultrashort-pulse propagation in optical fibers [63] helped solve a long-standing question of the magnitude of the shock-term [51,64].

3.5 Ultimate temporal resolution of the SHG FROG

In the general case of arbitrary pulses, the complete expression for the SHG FROG signal given by Eq.(3.27) must be computed numerically. However, for the limited class of pulses, such as linearly-chirped Gaussian pulses Eq.(3.27) can be evaluated analytically. Such analysis is valuable to estimate the temporal resolution of the SHG FROG experiment.

The geometrical smearing of the delay due to the crossing angle is an important experimental issue of the non-collinear multishot FROG measurement of ultrashort pulses. As can be seen from Eq.(3.27) the dependence on the transverse coordinate $x$ yields a range of delays across the beam simultaneously which “blurs” the fixed delay between the pulses and broadens the FROG trace along the delay axis. Analogously to Taft et al. [9], we assume Gaussian-intensity pulses and, under perfect phase-matching conditions, obtain the measured pulse duration $\tau_{\text{meas}}$ that corresponds to a longer pulse as given by

$$\tau_{\text{meas}}^2 = \tau_p^2 + \delta t^2,$$

(3.28)

where $\tau_p$ is the true pulse duration, and $\delta t$ is the effective delay smearing:
with \( d_f \) being the beam diameter in the focal plane, and \( 2\alpha_0 \) the intersection angle of the fundamental beams.

We consider the best scenario of the two Gaussian beams separated by their diameter \( d \) on the focusing optic. In this case the intersection angle \( 2\alpha_0 = d / f \), and the beam diameter in the focal plane \( d_f = f\lambda / \pi d \), where \( f \) is the focal length of the focusing optic. Therefore, the resultant time smearing amounts only to \( \delta t = \lambda / 2\pi c = 0.4 \text{ fs} \) at \( \lambda = 800 \text{ nm} \). This value presents the ultimate resolution of the pulse measurement in the non-collinear geometry. Interestingly, this figure does not depend on the chosen focusing optic or the beam diameter \( d \), since the beam waist is proportional whereas the intersection angle is inversely proportional to the focal distance \( f \). It should be noted that the temporal resolution deteriorates if the beams are other than Gaussian. For instance, if the beams of the same diameter with a rectangular spatial intensity profile replace the Gaussian beams in the situation described above, the resultant temporal resolution becomes 0.7 fs.

Additional enhancement of the temporal resolution could be achieved either by placing a narrow slit behind the nonlinear medium [65], as will be discussed in Section 3.9, or by employing a collinear geometry [66,67].

### 3.6 Approximate expression for the SHG FROG signal

In this Section, our goal is to obtain a simplified expression for SHG FROG that can be used even for single-cycle optical pulses. We start from the complete expression given by Eq.(3.27) and show that the measured signal can be described by an ideal, i.e. perfectly phase-matched SHG FROG and a spectral filter applied to the second-harmonic field. Throughout this Section we consider Type I phase-matching.

In order to simplify Eq.(3.27), we make several approximations. First, as was shown in the previous Section, under carefully chosen beam geometry the effect of geometrical smearing is negligibly small. For instance, it causes only a 10% error in the duration measurement of a 3-fs pulse, and can be safely neglected. With such approximation, the integral along \( x \) in Eq.(3.27) can be performed analytically. Second, we assume that \( \omega = \Omega / 2 \) and apply this to modify the factor that is proportional to the overlap area between different fundamental frequency modes: \( \sqrt{\omega(1-\omega/\Omega)} = \sqrt{\Omega / 2} \). Third, we expand \( k_\omega(\omega) \) and \( k_\omega(\Omega-\omega) \) into a Taylor series around \( \omega = \Omega / 2 \) and keep the terms that are linear with frequency\(^*\). Hence, for Type I phase-matching we write:

\[ \delta t = \alpha_0 d_f / c, \quad \text{(3.29)} \]

\(^*\) Alternatively, one can perform Taylor expansion around the central frequency of the fundamental pulse \( \omega = \omega_0 \) [22,39,43]. However, in this case the first derivative terms do not cancel each other and must be retained. Our simulations also prove that the expansion around \( \omega = \Omega / 2 \) provides a better approximation when broadband pulses are concerned. The practical implications of both approximations are also addressed in Section 4.3.
\[ \Delta k(\omega, \Omega - \omega) = 2k_o(\Omega/2)\cos(\alpha_o n_o(\Omega/2)) - k_o(\Omega) = \Delta k(\Omega/2, \Omega/2) \]  

(3.30)

Fourth, we estimate dispersion of the second treat the second-order susceptibility \(\tilde{\chi}^{(2)}(\Omega, \omega, \Omega - \omega)\) from the dispersion of the refractive index. For a classical anharmonic oscillator model [56], 

\[ \tilde{\chi}^{(2)}(\Omega, \omega, \Omega - \omega) \approx \tilde{\chi}^{(2)}(\omega)\tilde{\chi}^{(2)}(\Omega) - \Omega \]

where \(\tilde{\chi}^{(2)}(\Omega) = n^2(\Omega) - 1\). Equation (3.27) can now be decomposed to a product of the spectral filter \(R(\Omega)\), which originates from the finite conversion bandwidth of the second harmonic crystal and varying detector sensitivity, and an ideal FROG signal \(S_{\text{SHG FROG}}(\Omega, \tau)\):

\[ S(\Omega, \tau, L) \approx R(\Omega) S_{\text{SHG FROG}}(\Omega, \tau), \]  

(3.31)

where

\[ S_{\text{SHG FROG}}(\Omega, \tau) = \left| \int \tilde{E}^- (\Omega - \omega)\tilde{E}^- (\omega) \exp(i\omega \tau) d\omega \right|^2, \]  

(3.32)

and

\[ R(\Omega) = Q(\Omega) \frac{\Omega^3}{n^2(\Omega)} \left[ n^2(\Omega)/2 - 1 \right]^2 \text{sinc}^2 \left( \frac{\Delta k(\Omega/2, \Omega/2)L}{2} \right). \]  

(3.33)

In Eqs.(3.31-33) we retained only the terms that are \(\Omega\)-dependent.

The FROG signal given by Eq.(3.32) is the well-known classic definition of SHG FROG [14,18,35] written in the frequency domain. The same description is also employed in the existing FROG retrieval algorithms. Note that the complete Eq.(3.27) can be readily implemented in the algorithm based on the method of generalized projections [68]. However, relation (3.31) is more advantageous numerically, since the integral Eq.(3.32) takes form of autoconvolution in the time domain and can be rapidly computed via fast Fourier transforms [69]. It is also important that the use of Eq.(3.31) permits a direct check of FROG marginals to validate experimental data.

The spectral filter \(R(\Omega)\), as given by Eq.(3.33), is a product of several factors (Fig.3.3). The \(\Omega^3\)-term (dotted line) results from \(\Omega\)-dependence of the second-harmonic intensity on the spatial overlap of the different fundamental frequency modes*, and from the \(\Omega^2\) dependence that follows from Maxwell’s equations. The meaning of the latter factor is that the generation of the higher-frequency components is more efficient than of the lower-frequency ones. The combined \(\Omega^3\) dependence leads to a substantial distortion of the second-

*This dependence should be disregarded for the output of a Kerr-lens mode-locked laser [70] and for a hollow fiber [43,54]
harmonic spectrum of ultrabroadband pulses. For instance, due to this factor alone, the up-conversion efficiency of a spectral component at 600 nm is 4.5 times higher than of a 1000-nm one.

Fig.3.3: Constituent terms of spectral filter $R(\Omega)$ given by Eq.(3.33): the $\Omega^3$ dependence (dotted line), estimated squared magnitude of second-order susceptibility $\chi^{(2)}$ (dash-dotted line), the crystal phase-matching curve for a Type I 10-µm BBO crystal cut at $\theta=29^\circ$ (dashed line), and their product (solid curve). The second-harmonic spectrum of a 3-fs Gaussian pulse is shown for comparison (shaded contour).

The variation of the second-order susceptibility with frequency, expressed in Eq.(3.33) as the dependence on the refractive indices, plays a much less significant role than the $\Omega^3$ factor (dotted line). According to our estimations for BBO crystal, the squared magnitude of $\tilde{\chi}^{(2)}$ for the 600-nm component of the fundamental wave is only 1.3 times larger than for the 1000-nm component. Such a virtually flat second-order response over the immense bandwidth is a good illustration of the almost instantaneous nature of $\tilde{\chi}^{(2)}$ in transparent crystals. Nonetheless, the estimation the contribution of the $\tilde{\chi}^{(2)}$ dispersion is required for the measurement of the optical pulses with the spectra that are hundreds of nanometers wide.

The last factor contributing to $R(\Omega)$ is the phase-matching curve of the SHG crystal (Fig.3.3, dashed line). The shape and the bandwidth of this curve depend on the thickness, orientation and type of the crystal. Some practical comments on this issue will be provided in Section 4.2.

3.7 Numerical simulations

In this Section we verify the approximations that were applied to derive Eqs. (3.31–33). In order to do so, we numerically generate FROG traces of various pulses using the complete expression Eq.(3.27) and compare them with the ideal FROG traces calculated according to
Eq.(3.32). To examine contributions of different factors to pulse reconstruction, we compare FROG inversion results with the input pulses.

Fig.3.4: Simulation of SHG FROG signal for an ideal 3-fs Gaussian pulse for Type I phase-matching. (a) ideal FROG trace, as given by Eq.(3.32). (b) complete FROG trace as given by Eq.(3.27). (c) spectral filter curve $R(\Omega)$ computed according to Eq.(3.33) (shaded contour) and the ratio of FROG traces given in (b) and (a) at several delays (broken curves). (d) spectral marginal of the traces shown in (b) (solid curve) and autoconvolution of the fundamental spectrum (dashed curve). The FROG traces here and further on are shown as density plots with overlaid contour lines at the values 0.01, 0.02, 0.05, 0.1, 0.2, 0.4, and 0.8 of the peak second harmonic intensity.

Two types of pulses with the central wavelength at 800 nm are considered: 1) a bandwidth-limited 3-fs Gaussian pulse, and 2) a pulse with the same bandwidth that is linearly chirped to 26 fs. We assume that the fundamental beam diameter in the focus is $d_f = 20 \mu m$ and the beams intersect at $2\alpha_0 = 2^\circ$. Therefore, the geometrical delay smearing that was defined in Section 3.5 [Eq.(3.29)] amounts to $\delta t = 1.2$ fs. The thickness of the Type I BBO is $L = 10 \mu m$. As we pointed out in Section 3.4, such a short crystal lengthens the pulse less than 0.1 fs, and, therefore, dispersive pulse broadening inside the crystal can be
disregarded. The crystal is oriented for the peak conversion efficiency at 700 nm\(^*\). The spectral sensitivity of the light detector \(Q(\Omega)\) is set to unity.

*SHG FROG in the single-cycle regime*

**Fig. 3.5**: Simulation of SHG FROG signal for a linearly-chirped 26-fs Gaussian pulse. The conditions are the same as in Fig.4. (a) ideal FROG trace, as given by Eq.(3.32). (b) complete FROG trace as given by Eq.(3.27). (c) spectral filter curve \(R(\Omega)\) computed according to Eq.(3.33) (shaded contour) and the ratio of FROG traces given in (b) and (a) at several delays (broken curves). (d) spectral marginal of the traces shown in (b) (solid curve) and autoconvolution of the fundamental spectrum (dashed curve).

The results of FROG simulations for each type of pulses are presented in Figs.3.4 and 3.5. The ideal traces calculated according to Eq.(3.32) are shown in Figs.3.4a and 3.5a, while the traces computed using Eq.(3.27) are displayed in Fig.3.4b and 3.5b. The FROG trace of the

\[ \theta = \theta_{\text{linear}} + \alpha_0 / n, \]

where \(n\) is the refractive index of the fundamental wave at the phase-matching wavelength. For instance, the 800-nm phase-matched cut of a BBO crystal for \(2\alpha_0=2^\circ\) becomes \(\theta=29.6^\circ\) instead of \(\theta_{\text{linear}}=29^\circ\) for collinear SHG. This fact should be kept in mind since the phase-matching curve is quite sensitive to the precise orientation of the crystal.

---

\(\text{\textsuperscript{*}}\) The phase-matching angle is slightly affected by the non-collinear geometry. Due to the fact that the fundamental beams intersect at an angle \(2\alpha_0\), the equivalent phase-matching angle is different from that in the case of collinear SHG: \(\theta = \theta_{\text{linear}} + \alpha_0 / n\), where \(n\) is the refractive index of the fundamental wave at the phase-matching wavelength.
3-fs pulse is also noticeably extended along the delay axis as the consequence of the geometrical smearing. For the 26-fs pulse, as should be expected, this effect is negligible. The spectral filtering occurring in the crystal becomes apparent from the comparison of the spectral marginals that are depicted in Figs. 3.4d and 3.5d. Calculated marginals are asymmetric and substantially shifted toward shorter wavelengths.

By computing a ratio of the FROG signals given by Eq. (3.32) and Eq. (3.27) we obtain delay-dependent conversion efficiency, as shown in Figs. 3.4c and 3.5c. The spectral filter \( R(\Omega) \) calculated according to Eq. (3.33), is shown as shaded contours. Clearly, at the small delays \( \tau \) the conversion efficiency is almost exactly described by \( R(\Omega) \). With the increase of pulse separation, the approximation given by Eq. (3.33) worsens, as both the conversion peak position and the magnitude change. The rapid ratio scaling at non-zero delays for the 3-fs pulse (broken curves in Fig. 3.4c) is mostly determined by the geometrical smearing rather than by the phase matching, as in the case of the chirped pulse (Fig. 3.5c). On the other hand, the deviations from \( R(\Omega) \) at longer delays become unimportant because of the decreasing signals at large pulse separations.

To estimate the significance of the spectral correction of the distorted FROG traces and feasibility of performing it in the case of extreme bandwidths, we examined FROG inversion results of the numerically generated traces using the commercially available program from Femtosoft Technologies. Four different cases were considered for each type of pulses: a) an ideal phase-matching (zero-thickness crystal); b) a 10-µm BBO crystal with the parameters defined above; c) the trace generated in the case (b) is corrected by \( R(\Omega) \); and, last, in d) geometrical smearing is included as well. In its essence, the case (d) is similar to (c), but in (d) the FROG trace was additionally distorted by the geometrical smearing. The results of the FROG inversion of the cases (a) - (d) are presented in Fig. 3.6.

In the case (a), the \( \Omega^3 \) dependence is exclusively responsible for the spectral filtering that substantially shifts the whole FROG trace along the frequency axis. Both the bandwidth-limited and the chirped Gaussian pulses converged excellently to their input fields, but around a blue-shifted central frequency. In (b), where the phase-matching of a 10-µm BBO crystal is taken into account as well, the central wavelength is even more blue-shifted due to spectral filtering in the crystal. A small phase distortion is obtained for both types of pulses. The retrieved 3-fs pulse is also artificially lengthened to ~3.4 fs to match the bandwidth narrowed by the spectral filtering in the crystal. The results of FROG retrieval of the same trace upon the correction by \( R(\Omega) \) (case (c)) indicate an excellent recovery of both the bandwidth-limited and the chirped pulses.

Finally, in the case (d) the geometrical smearing had a negligible effect on the 26-fs pulse. However, the FROG of the shorter pulse converged to a linearly chirped 3.3-fs Gaussian pulse. This should be expected, since the FROG trace broadens in time and remains Gaussian, while the spectral bandwidth is not affected. In principle, like the spectral correction \( R(\Omega) \), the correction for the temporal smearing should also be feasible. It can be
implemented directly in the FROG inversion algorithm by temporal averaging of the guess trace, produced in every iteration, prior to computing the FROG error.

Fig 3.6: Retrieved pulse parameters in the time and frequency domains for various simulated FROG traces. (a) perfectly phase-matched crystal, no geometrical smearing. (b) Type I 10-µm BBO crystal cut at θ=33.4°, no geometrical smearing. (c) same as in (b), the FROG trace is corrected according to Eq.(3.33). (d) same as in (c) but with the geometrical smearing included. Dashed curves correspond to initial fields, while solid curves are obtained by FROG retrieval.
Several important conclusions can be drawn from these simulations. First, they confirm the correctness of approximations used to obtain Eq.(3.31-33). Therefore, the spectral correction given by $R(\Omega)$ is satisfactory even in the case of single-cycle pulses, provided the crystal length and orientation permits to maintain a certain, though not necessarily high, level of conversion over the entire bandwidth of the pulse. Second, a time-smearing effect does not greatly affect the retrieved pulses if the experimental geometry is carefully chosen. Third, the unmodified version of the FROG algorithm can be readily applied even to the shortest pulses. Forth, it is often possible to closely reproduce the pulse parameters by FROG-inversion of a spectrally filtered trace without any spectral correction [43]. However, such traces rather correspond to similar pulses shifted in frequency than to the original pulses for which they were obtained.

![Graph](image)

**Fig.3.7**: Dependence of the systematic FROG trace error on the pulse duration. FROG matrix size is 128x128. The dotted curve corresponds the trace after the spectral correction given by Eq.(33). The error due to geometrical smearing of a perfectly phase-matched trace is shown as a dashed curve, while the error of a spectrally corrected and geometrically smeared FROG is given by the solid curve. The parameters of the crystal and of the geometrical smearing are the same as above. The central wavelength of the pulse is kept at 800 nm.

In order to quantify the distortions that are introduced into the SHG FROG traces by the phase-matching and the non-collinear geometry and that cannot be removed by the $R(\Omega)$-correction, we compute the systematic error as rms average of the difference between the actual corrected FROG trace and the ideal trace. Given the form of the FROG error [19], the systematic error can be defined as follows:

$$
G = \frac{1}{N} \left[ \sum_{i,j=1}^{N} S_{FROG}(\Omega_i, \tau_j) - a \frac{S(\Omega_i, \tau_j, L)}{R(\Omega)} \right]^2,
$$

(3.34)
where $S_{FROG}^{SHG}(\Omega, \tau)$ and $R(\Omega)$ are given by Eq.(3.32) and Eq.(3.33), and $S(\Omega, \tau, L)$ is computed according to Eq.(3.27). The parameter $a$ is a scaling factor necessary to obtain the lowest value of $G$. The dependence of $G$ on the duration of a bandwidth-limited pulse for the 128×128 FROG matrix that has optimal sampling along the time and frequency axes is presented in Fig.3.7. As can be seen, the systematic error for ~5-fs pulses becomes comparable with the typical achievable experimental SHG FROG error. It also should be noted, that the contribution of geometrical smearing is about equal or higher than that due to the spectral distortions remaining after the spectral correction.

The systematic error should not be confused with the ultimate error achievable by the FROG inversion algorithm. Frequently, as, for instance, in the case of linearly-chirped Gaussian pulses measured in the presence of geometrical smearing, it means that the FROG trace continues to exactly correspond to a pulse, but to a different one. However, for an arbitrary pulse of ~3 fs in duration it is likely that the FROG retrieval error will increase due to the systematic error.

### 3.8 Type II phase matching

So far, we limited our consideration to Type I phase-matching. In this Section we briefly discuss the application of Type II phase-matching to the measurement of ultrashort laser pulses.

In Type II the two fundamental waves are non-identical, i.e. one ordinary and one extraordinary. This allows the implementation of the collinear SHG FROG geometry free of geometrical smearing [67]. The FROG traces generated in this arrangement in principle does not contain optical fringes associated with the interferometric collinear autocorrelation and, therefore, can be processed using the existing SHG FROG algorithms. However, the fact that the group velocities of the fundamental pulses in a Type II crystal become quite different, has several important implications. First, the second-harmonic signal is no longer a symmetric function of the time delay [39]. Second, because the faster traveling fundamental pulse can catch up and pass the slower one, some broadening of the second-harmonic signal along the delay axis should be expected [39].

In order to check the applicability of the collinear Type II SHG FROG for the conditions comparable to the discussed above in the case of Type I phase matching, we performed numerical simulations identical to those in the previous Section. The same pulses were used, i.e., the bandwidth-limited 3-fs pulse at 800 nm and the pulse with the same bandwidth stretched to 26 fs. The thickness of the Type II BBO is $L=10 \, \mu m$, and the crystal oriented for the peak conversion efficiency at 700 nm ($\theta=45^\circ$). The expression for the spectral filter, adapted for Type II, is given by:

$$R(\Omega) = Q(\Omega) \frac{\Omega^3}{n_E^2(\Omega)} \left[ n_E^2(\Omega) - 1 \right] \left[ n_O^2(\Omega / 2) - 1 \right] \left[ n_E^2(\Omega / 2) - 1 \right] \text{sinc}^2 \left( \frac{\Delta k(\Omega / 2, \Omega / 2) L}{2} \right),$$

(3.35)
where the phase mismatch is

\[ \Delta k\left(\Omega/2,\Omega/2\right) = k_o\left(\Omega/2\right) + k_E\left(\Omega/2\right) - k_E\left(\Omega\right) \]  

(3.36)

The results of FROG simulations are presented in Figs. 3.8 and 3.9. The FROG trace of the 3-fs pulse (Fig. 3.8b) is practically symmetrical along the delay axis. However, despite the fact that no geometrical smearing has occurred, this trace is evidently broadened along the delay axis. Consequently, the FROG inversion of this trace after the spectral correction yields a longer ~3.3-fs pulse. The elongation of the trace is due to the temporal walk-off of the fundamental waves, which in this case is about 1 fs.

**Fig. 3.8:** Simulation of SHG FROG signal for an ideal 3-fs Gaussian pulse for Type II phase-matching. (a) ideal FROG trace, as given by Eq. (3.32). (b) complete FROG trace as given by Eq. (3.27). (c) spectral filter curve \( R(\Omega) \) computed according to Eq. (3.33) (shaded contour) and the ratio of FROG traces given in (b) and (a) at several delays (broken curves). (d) spectral marginal of the traces shown in (b) (solid curve) and autoconvolution of the fundamental spectrum (dashed curve).

\[ ^* \text{Unlike in the case of Type I phase-matching, the first derivative terms do not cancel each other but they have been disregarded anyway.} \]
The magnitude of this temporal distortion is approximately equal to the geometrical smearing discussed in the previous Section. The trace of the chirped pulse, produced under the same conditions (Fig. 3.9b), is much more severely distorted than in the case of the bandwidth-limited pulse. The straightforward use of this trace is virtually impossible because of its strong asymmetry.

**Fig. 3.9**: Simulation of SHG FROG signal for a linearly-chirped 26-fs Gaussian pulse. The conditions are the same as in Fig. 3.8. (a) ideal FROG trace, as given by Eq. (3.32). (b) complete FROG trace as given by Eq. (3.27). (c) spectral filter curve $R(\Omega)$ computed according to Eq. (3.33) (shaded contour) and the ratio of FROG traces given in (b) and (a) at several delays (broken curves). (d) spectral marginal of the traces shown in (b) (solid curve) and autoconvolution of the fundamental spectrum (dashed curve). Note the skewness of the FROG trace in (b).

As in the Type I case, the conversion efficiency, obtained as a ratio of the ideal and simulated FROG traces, continues to correspond nicely the spectral filter $R(\Omega)$ (Figs. 3.8c and 3.9c, shaded contours) at near-zero delays. Conversion efficiency at other delays, however, sharply depends on the sign of the delay $\tau$. Similar to Type I phase-matching, the frequency marginals (Figs. 3.8d and 3.9d) are substantially blue-shifted. It is also apparent from Figs. 3.8c and 3.9c, that the phase-matching bandwidth in this case is somewhat broader than in the analogous Type I crystal.
We can conclude from our simulations that Type II SHG FROG offers no enhancement of the temporal resolution and is less versatile compared to the non-collinear Type I arrangement. Additionally, the collinear Type II SHG FROG requires a greater experimental involvement than in the Type I SHG FROG. However, for some applications such as confocal microscopy, where the implementation of the non-collinear geometry is hardly possible due to the high numerical aperture of the focusing optics, the use of Type-II-based FROG appears quite promising [67].

3.9 Spatial filtering of the second-harmonic beam

In this Section, we show how spatial filtering of the second-harmonic beam can corrupt an autocorrelation or FROG trace. Unfortunately, this type of distortion can pass undetected since the FROG trace may still correspond to a valid pulse, but not the one that is being measured.

As it was already mentioned in Section 3.4 [Eq.(3.23)], the direction in which a second harmonic frequency is emitted varies because of the non-collinear geometry. Even though the intersection angle of the fundamental beams is small, this effect becomes rather important for
the measurement of broadband pulses due to the substantial variation of the wave-vector magnitude across the bandwidth.

Let us consider a certain component of the second-harmonic signal that has a frequency of \(2\omega_0\) (Fig. 3.10). This component can be generated for several combinations of fundamental frequencies, for example, such as the pairs of \(\omega_0\) and \(\omega_0\), and of \(\omega_0 + \delta\omega\) and \(\omega_0 - \delta\omega\). The direction in which the \(2\omega_0\) component is emitted for each pair can vary, as determined by the non-collinear phase-matching. Therefore, as can be seen from Fig. 3.10, the direction of the second-harmonic beam changes as a function of delay between the fundamental pulses. This phenomenon is utilized in the chirp measurement by angle-resolved autocorrelation [71,72].

To illustrate the effect of spatial filtering of the second-harmonic beam, we examine the same Gaussian pulses linearly chirped to 26 fs, which were used in the numerical simulations described above. We keep the same geometrical parameters as in the previous sections of this Chapter, i.e. \(d_f = 20 \, \mu m\) and \(2\alpha_0 = 2^\circ\). The resulting dependence of the autocorrelation intensity as a function of the second-harmonic angle in the far field is depicted in Fig. 3.11a. The tilt of the trace clearly indicates the sweep of the second-harmonic beam direction. The signal beam traverses approximately half the angle between the fundamental beams, and the magnitude of this sweep scales linearly with the intersection angle. The autocorrelation trace obtained by integration over all spatial components of the second-harmonic beam is depicted in Fig. 3.11b (solid curve). The FROG trace corresponding to this autocorrelation, i.e. measured by detecting of the whole beam, is entirely correct and allows recovery of the true pulse parameters.

**Fig.3.11:** Angular dependence of the non-collinear second-harmonic signal for a linearly-chirped Gaussian pulse in the far field. (a) autocorrelation intensity as a density plot of delay between the fundamental pulses and the second-harmonic angle. (b) autocorrelation intensity trace obtained by integration over all spatial components of the second-harmonic beam (solid curve) and the traces detected through a narrow slit at the second-harmonic angle of \(0^\circ\) (dashed curve) and \(0.4^\circ\) (dotted curve). The pulse is stretched to ~5 times the bandwidth-limited pulse duration. The intersection angle of the fundamental beams is \(2^\circ\)
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The situation, however, becomes different if only a portion of the second harmonic beam is selected. In the considered example, the autocorrelation or FROG, measured through a narrow slit placed on the axis of the second harmonic beam, would “shrink” along the delay axis, as shown in Fig.13b (dashed curve). The width of this trace is ~10% narrower than the true autocorrelation width. Positioning of the slit off the beam axis (Fig.3.11b, dotted curve) leads to the shift of the whole trace along the delay axis, and, for some pulses, to asymmetry in the autocorrelation wings. In the case of Gaussian pulses examined here, the FROG traces measured with such spatial selection remain self-consistent, disregarding the delay shift. The spectral marginal of such FROG traces is exactly the same as in the case of the whole-beam detection. Consequently, the FROG retrieval of the spatially filtered traces yields pulses of correct bandwidth but less chirped than in reality.

The described effect should not be identified alone with the pulses that are much longer than the bandwidth limit, since even the bandwidth-limited pulses with asymmetric spectra carry a chirp in time. Therefore, careful collecting of all spatial components of the second harmonic field is extremely essential. We also underline importance of measuring an independent autocorrelation trace in front of spectrometer, since its comparison with the temporal marginal of the FROG trace might signal improper spatial filtration occurring in the FROG detection.

In Section 3.5 we have already mentioned the desirability to enhance the temporal resolution of a non-collinear measurement by placing a slit behind the nonlinear medium. This reduces the effective spot of the second harmonic beyond the size of the diffraction-limited focus. However, placing a slit into the collimated beam would cause the spatial selection considered above. To avoid such undesirable distortion, one should position the slit behind the crystal within the Rayleigh range, or, alternatively, into the scaled image of the crystal plane projected by an achromatic objective lens. The realization of both these options is rather difficult and becomes really necessary only if the beams are poorly focusable.

3.10 Conclusions

In this Chapter, we have developed the SHG FROG description that includes the phase-matching in the SHG crystal, non-collinear beam geometry, and dispersion of the second-order nonlinearity. The derived master equation is valid down to single-cycle pulses.

Subsequently, thorough numerical simulations have been performed to estimate the separate roles of the crystal phase-matching, geometrical smearing and spatial filtering of the SHG signal. These simulations have shown that the conventional description of FROG in the case of Type I phase-matching can be readily used even for the single-cycle regime upon spectral correction of the FROG traces, provided the beam geometry, the finite crystal thickness and phase-matching bandwidth are chosen correctly.

The SHG FROG of very short pulses with Type II phase-matching in a BBO crystal is shown to be rather impractical, since the group velocity mismatch between the two
fundamental waves of different polarization causes a delay smearing similar to the one originating from geometrical blurring in the non-collinear measurement.

We also show that, while the spectral correction of the FROG traces helps the recovery of true pulse characteristics, the systematic error of the FROG trace, nonetheless, increases with the increase of the spectral breadth of the pulse. This is due to the fact that the effective spectral filter applied by the second harmonic generation process on the FROG trace varies for different delay values. Consequently, higher FROG trace retrieval errors should be expected from the inversion algorithm for the bandwidths supporting durations shorter than 5-6 fs.
Chapter 3

References

SHG FROG in the single-cycle regime


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