Ultrabroadband spectral phase measurements by two-dimensional spectral shearing interferometry (2DSI)

Extrem breitbandige Messung der spektralen Phase durch two-dimensional spectral shearing interferometry

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CFEL SCIENCE
DESY
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Abstract

This work presents an implementation and optimization of a pulse characterization technique for ultrashort laser pulses. Namely the technique of two-dimensional spectral shearing interferometry (2DSI) is set up with the development a full automatic MATLAB code to acquire, store and reconstruct the complex pulse in frequency and time domain. The performance of the retrieval code itself is tested with experimental consistency checks and simulations. Prominent characterization techniques such as SPIDER and FROG are presented and compared in a discussion.

A Ti:sapphire laser oscillator with a Fourier-transform-limited FWHM duration of 4 fs serves as a source for the first application of our device. Compression of the pulse with double chirped mirrors (DCMs) and BaF$_2$ wedges yields a measured pulse duration of 4.2 fs at FWHM.

Within the frame of this thesis, a (noncollinear) optical parametric amplifier (OPA) is built. The spectrum spans from 1.2 $\mu$m to 2 $\mu$m and supports a Fourier-transform-limited FWHM duration of 8.2 fs. The uncompressed pulse is fully characterized and found to have a smooth phase that should be compressible rather evenly.

We found that the apparatus presented here gives reliable results from low intensity pulses in the visible spectrum up to high intensity ones in the infrared regime and can be considered as an easy to use, high precision and flexible pulse characterization technique.
Zusammenfassung

Kurze Laserpulse erfordern ebenso breitbandige Spektren, deren Erzeugung zu einem komplexen Verlauf der Phase und der spektralen Intensität führt. Die vorliegende Arbeit widmet sich der experimentellen Charakterisierung ultakurzer Laserpulse, insbesondere der Messung der spektralen Phase. Im Fokus steht die Implementierung und Optimierung einer auf Interferenz und Summenfrequenzmischung basierenden Technik (two-dimensional spectral shearing interferometry, 2DSI), die anschließend mit dem nahestehenden SPIDER Verfahren und der FROG Methode verglichen wird. Eine in MATLAB geschriebene graphische Oberfläche ermöglicht die intuitive Durchführung, Speicherung und Auswertung der Messung.

Die erste Anwendung der Messmethode findet an einem Titan:Saphire Laseroszillator mit einer Pulsdauer von 4 fs im Fourierlimit statt. Durch dispersionskompensierende Spiegel (double chirped mirror, DCM) und BaF$_2$ Keilfenster komprimiert, ergibt die vollständige Charakterisierung eine Pulsdauer von 4.2 fs.

Desweiteren wird der Aufbau eines (nichtkollinearen) optisch parametrischen Verstärkers präsentiert, dessen Bandbreite von 1.2 μm bis 2 μm reicht. Auch dieser Puls im infraroten Spektralbereich weist eine kurze Pulsdauer von 8.2 fs im Fourierlimit auf. Die vollständige Charakterisierung des noch unkomprimierten Pulses zeigt einen kontinuierlichen Phasenverlauf und lässt auf mühelose Kompression schließen.

Die vorgestellten Messungen von breitbandigen Pulsen geringer Intensität im sichtbaren Spektrum bis zu Pulsen hoher Intensität im infraroten Spektrum sprechen für die Universalität und Präzision des Messaufbaus.
Acknowledgement

The time in Hamburg and the work at the Center for Free-Electron Laser Science (CFEL) and Deutsches Elektronen-Synchrotron (DESY) would have been less enjoyable without good friends and colleagues whom I would like to thank.

I thank Professor Franz X. Kärtner for granting me access to his research group and introducing me to the world of ultrafast optics. His long term support and encouragement motivated me to focus on this field of research.

Happy to know that Dr. Oliver D. Mücke is my second supervisor, I would like to thank him for all explanations and his luck while I was searching for the right delay.
I say hi to Dr. Giovanni Cirmi, who I could interrupt any time to raise a question.
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Temporally aligned with the start of my thesis, Louisa became the most beloved part in my life. Thank you for all the adventures you have planned for and your understanding when I lost my head in lab from time to time. I am looking forward!
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<td>2DSI</td>
<td>Two-dimensional spectral shearing interferometry</td>
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<tr>
<td>AC</td>
<td>alternating current, here an oscillating signal</td>
</tr>
<tr>
<td>AR</td>
<td>anti-reflection</td>
</tr>
<tr>
<td>BK7</td>
<td>borosilicate glass</td>
</tr>
<tr>
<td>BBO</td>
<td>beta barium borate</td>
</tr>
<tr>
<td>BS</td>
<td>beam splitter</td>
</tr>
<tr>
<td>CEP</td>
<td>carrier envelope phase</td>
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<tr>
<td>CPA</td>
<td>chirped pulse amplification</td>
</tr>
<tr>
<td>DC</td>
<td>direct current, here a constant signal</td>
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<tr>
<td>DCM</td>
<td>double chirped mirror</td>
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<tr>
<td>DFG</td>
<td>difference frequency generation</td>
</tr>
<tr>
<td>DOPA</td>
<td>degenerate optical parametric amplifier</td>
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<tr>
<td>fps</td>
<td>frames per second</td>
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<tr>
<td>FT</td>
<td>Fourier transform</td>
</tr>
<tr>
<td>FFT</td>
<td>fast Fourier transform</td>
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<tr>
<td>FWHM</td>
<td>full width at half maximum</td>
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<td>FROG</td>
<td>frequency-resolved optical gating</td>
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<td>GDD</td>
<td>group delay dispersion</td>
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<tr>
<td>GTI</td>
<td>Gires-Tournois interferometer</td>
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<tr>
<td>GUI</td>
<td>graphical user interface</td>
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<tr>
<td>GVM</td>
<td>group velocity mismatch</td>
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<tr>
<td>HCF</td>
<td>hollow core fibre</td>
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<tr>
<td>HHG</td>
<td>high harmonic generation</td>
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<tr>
<td>IAC</td>
<td>interferometric autocorrelation</td>
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<tr>
<td>IF</td>
<td>interference filter</td>
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<tr>
<td>IR</td>
<td>infrared</td>
</tr>
<tr>
<td>KLM</td>
<td>Kerr-lens mode locking</td>
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<tr>
<td>MI</td>
<td>Michelson interferometer</td>
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<tr>
<td>NIR</td>
<td>near infrared</td>
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<tr>
<td>NOPA</td>
<td>noncollinear optical parametric amplifier</td>
</tr>
<tr>
<td>OPA</td>
<td>optical parametric amplifier</td>
</tr>
<tr>
<td>PI</td>
<td>Physik Instrumente (brand)</td>
</tr>
<tr>
<td>PMOC</td>
<td>phase matched output coupler</td>
</tr>
<tr>
<td>PUT</td>
<td>pulse under test</td>
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<tr>
<td>SFG</td>
<td>sum frequency generation</td>
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<td>SVEA</td>
<td>slowly varying amplitude approximation</td>
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<td>THG</td>
<td>third harmonic generation</td>
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<tr>
<td>TOD</td>
<td>third order dispersion</td>
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<tr>
<td>SEA</td>
<td>spatially encoded arrangement (SPIDER)</td>
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<tr>
<td>SHG</td>
<td>second harmonic generation</td>
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<tr>
<td>SPIDER</td>
<td>spectral phase interferometry for direct electric-field reconstruction</td>
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<tr>
<td>SPM</td>
<td>self-phase modulation</td>
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<tr>
<td>TL</td>
<td>transform limit</td>
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<tr>
<td>WKS</td>
<td>Whittaker, Kotelnikow und Shannon (sampling theorem)</td>
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<tr>
<td>WLC</td>
<td>white light continuum</td>
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<tr>
<td>ZAP</td>
<td>zero additional phase (a variant of SPIDER)</td>
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1 Motivation

Ultrafast events evolving faster than our eye can capture them are fascinating. The limited frames per second that a human eye and brain can process are around 25 frames per second (fps) and many details stay hidden to our perception, for instance sports, bursting objects, single frames on the television. In the 18th century, Edward Muybridge developed a system of synchronized photo cameras that allowed a time-lapse recording. With that setup, he introduced two characteristic times: The (1) flash light duration, that has to be short enough to ensure a high quality picture of an almost single point in time and (2) the refresh rate or synchronization time between following shots that has to be much shorter than a cycle of a motion or the dynamics of interest to not loose any information that stays hidden between the frames. The apparatus of Edward Muybridge was applied on various fast macroscopic dynamics and were the first proof that a galloping horse lifts all feet at once for a short sequence [2].
On a microscopic scale, ultrafast motions of electrons and ultrashort events as in chemical reactions make it necessary to adapt the tools of observation and increase the temporal resolution by several orders of magnitude. The attempt to capture, understand and control nature’s fastest dynamics in cells, proteins and molecules is happening on timescales in the range of femtoseconds \((10^{-15} \text{ s}, \text{ fs})\) that are a million of a million times shorter than the exposure time E. Muybridge needed to capture the motion of the galloping horse \((1/2000 \text{ sec})\). Today, lasers are the tools that replace films, cameras and flash lights in order to illuminate and capture the molecules of a sample at the frontline of science. Not only probing but also driving ultrafast experiments towards attosecond timescales is in the scope of ultrafast laser science. The development of coherent sources using hollow core fibre techniques or broadband optical parametric amplification [3] [4] applied to high harmonic generation processes in noble gases evolved to sub 5 fs driver pulses that generate XUV pulses in the attosecond regime. The creation of the shortest man built events in form of those light pulses implies that there are no shorter controlled processes and therefore a lack of appropriate detection systems. Ideas that evolved over the past decades are mostly self-referencing techniques where the pulse is used to characterize itself. It is the topic of this thesis to built and measure few cycle optical pulses experimentally by a Two-dimensional spectral shearing interferometer (2DSI) [5].
2 Optical Fields

The classical description of light that propagates through media, getting reflected and finally interacts with the Rhodopsin in our eye is based on electro-magnetic (EM) fields. The frequency and corresponding wavelengths of those optical fields rank between the radio frequencies (\(\lambda > 10\) mm) and the X-ray radiation (\(\lambda < 10\) nm). The treatment of ultrashort optical pulses makes it necessary to involve a broad range of frequencies and therefore higher orders of wavelength dependent quantities have to be taken into account. The following sections form the basis of understanding and describing pulse propagation in media.

2.1 Linear pulse propagation

The properties of EM fields are governed by Maxwell’s equations following the equations 2.1.1. The set of Maxwell equations connect the physical laws named according to their inventors.

\[
\begin{align*}
\mathbf{\nabla} \cdot \mathbf{D} &= \rho \quad \text{Gauss’ Law} \\
\mathbf{\nabla} \cdot \mathbf{B} &= 0 \quad \text{Gauss’ Law for magnetism} \\
\mathbf{\nabla} \times \mathbf{E} &= -\frac{\partial \mathbf{B}}{\partial t} \quad \text{Faraday’s Law of induction} \\
\mathbf{\nabla} \times \mathbf{H} &= \mathbf{J} + \frac{\partial \mathbf{D}}{\partial t} \quad \text{Maxwell-Ampere Law}
\end{align*}
\]

Gauss’ law relates the displacement field \(\mathbf{D}\) to a source of charges, the density of charges \(\rho\). Because of magnetic monopoles do not exist, the divergence of the magnetic field \(\mathbf{B}\) equals zero in Gauss’ law for magnetism. Faraday’s law relates a time-varying magnetic field to a spatially-varying electric field \(\mathbf{E}\). The electric current through a loop is related to the magnetic field strength \(\mathbf{H}\) by Ampere’s law.

In SI units, the magnetic field strength \(\mathbf{H}\) relates to the magnetic field as

\[
\mathbf{H} = \frac{1}{\mu_0} (\mathbf{B} - \mathbf{M})
\]

(2.1.2)
where $\mu_0$ is the permeability and the magnetization $\vec{M}$ accounts for the amount of magnetic dipoles per unit volume. The displacement field $\vec{D}$ includes the properties of media and depends on the vacuum permittivity $\varepsilon_0$ and a polarization density $\vec{P}$ that expresses the density of electric dipoles in a medium.

$$\vec{D} = \varepsilon_0\vec{E} + \vec{P}$$  \hspace{1cm} (2.1.3)

$\vec{P}$ has units of coulombs per square meter and is often simply named polarization. The polarization plays a major role in nonlinear optics and accounts for the interaction of the light wave with media - the crystals, atoms and molecules. In a classical picture, the time varying electrical field acts on the trajectories of the valence electrons in the media and induces microscopic dipoles $\vec{p}(t,r)$ with a density $N$ giving again the polarization $\vec{P}(t,r)$ of the medium

$$\vec{P} = N \cdot \vec{p}.$$  \hspace{1cm} (2.1.4)

In general, the polarization displays the behaviour of bound charges where $\rho$ accounts for the density of free charges. In a linear regime the polarisation depends linearly on the electric field that drives the dipoles and is coupled via the linear susceptibility $\chi$ as a proportional constant.

$$\vec{P} = \varepsilon_0\chi\vec{E}$$  \hspace{1cm} (2.1.5)

A solution to Maxwell’s set of differential equations that describe the field in optics is derived with some constrains on the supporting media. A perfect dielectric media has no free charges ($\rho = 0$) and currents ($\vec{J} = 0$). Moreover the dielectric material is taken to be non-magnet ($\vec{M} = 0$). By taking the curl of Faraday’s law in equation 2.1.1 and implying the constitutive relations 2.1.2 and 2.1.3 one gets with Ampere’s law to

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E} = \vec{\nabla} \times \left( -\frac{\partial \vec{B}}{\partial t} \right) = - \left( \frac{\partial}{\partial t} \right) (\vec{\nabla} \times \vec{B}) = -\mu_0\frac{\partial^2 \vec{D}}{\partial t^2}. $$  \hspace{1cm} (2.1.6)

The vector identity states

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E} = \vec{\nabla}(\vec{\nabla} \cdot \vec{E}) - \vec{\nabla}^2 \vec{E} = -\vec{\nabla}^2 \vec{E}$$  \hspace{1cm} (2.1.7)

where Gauss’s law of the Maxwell equation 2.1.1 states $\nabla \cdot \vec{E} = 0$ when assuming no free charges.

Placing the equation 2.1.7 in equation 2.1.6, the wave equation reads as

$$\vec{\nabla}^2 \vec{E} - \frac{1}{\epsilon^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}$$  \hspace{1cm} (2.1.8)

where we used the speed of light in vacuum $c = 1/\sqrt{\varepsilon_0\mu_0}$ and the displacement field as given in equation 2.1.3.
A solution to the wave equation is given below with a propagation in z direction and the polarization is chosen to be linear along the x direction. Furthermore, the plane wave approximation describes the field to be constant on the plane perpendicular to the direction of propagation.

\[ \vec{E}(z,t) = \mathcal{R}\{A_0 e^{i(kz-\omega t)}\hat{x}\} \]  

(2.1.9)

The field is characterized by an amplitude \( A_0 \), the angular frequency \( \omega \) and the propagation constant \( k = \frac{\omega}{c} \) and we already used \( n(\omega) = \sqrt{1+\chi(\omega)} \), the refractive index. The fast oscillating exponential function is known as the carrier field. When dealing with pulsed laser light, the constant amplitude \( A_0 \) becomes a spatially and temporally varying envelope \( A(z,t) \) with the carrier field beneath.

\[ \vec{E}(z,t) = \mathcal{R}\{A(z,t)e^{i(kz-\omega t)}\hat{x}\} \]  

(2.1.10)

### 2.2 Susceptibility

The refractive index in the plane wave solution links the propagation of the wave to the supporting media. The broad wavelength range and high peak intensities of ultrashort pulses makes it necessary to have a closer look on the interaction of the fast oscillating EM field with the supporting dielectric that are coupled by the susceptibility \( \chi \).

We follow the principle of the classical oscillator model where the optical field is driving dipoles in the media. The microscopic motion of the dipoles shows a linear dependence on the driving field up to a certain amplitude. The related macroscopic quantity is the so called linear polarization.

\[ \vec{P}(\omega) = \varepsilon_0 \chi^{(1)}(\omega)\vec{E}(\omega) \]  

(2.2.1)

The proportional constant \( \chi^{(1)} \) is the linear susceptibility. Already the linear susceptibility tensor of rank two includes nine material dependent values that account for all possible orientations of the media with respect to the polarization of the light if the medium does not show any symmetry. For an isotropic medium, the remaining linear susceptibility is a scalar. Regarding equation 2.1.3, the effect of the polarization on the propagating light wave is related as follows

\[ \vec{D} = \varepsilon_0 \vec{E} \vec{P} = \varepsilon_0 (1 + \chi)\vec{E} = \varepsilon_0 \varepsilon_r(\omega)\vec{E} \]  

(2.2.2)

where \( \varepsilon_r(\omega) \) is the relative permittivity or complex dielectric constant. The propagation constant for a dielectric writes as

\[ k = \omega \sqrt{\varepsilon\mu_0} = \omega \sqrt{\varepsilon_r\varepsilon_0\mu_0} = \frac{n\omega}{c} \]  

(2.2.3)
and includes the relation between the refractive index and the susceptibility as

\[ n = \sqrt{\epsilon_r} = \sqrt{1 + \chi}. \] (2.2.4)

The susceptibility is zero in vacuum and is advantageous compared to the refractive index when dealing with light matter interaction and describing the polarization where it isolates the material dependent propagation. A description of the propagation with linear polarization and corresponding linear susceptibility holds as long as the optical fields are weak in a sense that the dipoles describe an harmonic motion. The stronger the fields are, the more distortion is added to the sinusoidal trajectory of the valence electrons and we enter the regime of nonlinear propagation and nonlinear susceptibilities.

### 2.3 Nonlinear propagation

The nonlinear propagation description is applied when the restoring force of the dipoles differ from a linear behaviour. The anharmonic potential and the nonlinear dependence of the macroscopic polarization with the electric field are described by a Taylor expansion of the polarization

\[ \vec{P} = \epsilon_0 \left( \chi^{(1)} \vec{E} + \chi^{(2)} \vec{E}^2 + \chi^{(3)} \vec{E}^3 + \ldots \right) \] (2.3.1)

with \( \chi^{(n)} \) being the nth-order susceptibility. It is common to split the linear and nonlinear part of the polarization

\[ \vec{P} = \vec{P}_{LIN} + \vec{P}_{NL} \] (2.3.2)

and extend the wave equation by the nonlinear part.

\[ \nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \left( \frac{\partial^2 \vec{P}_{LIN}}{\partial t^2} + \frac{\partial^2 \vec{P}_{NL}}{\partial t^2} \right) \] (2.3.3)

With the linear polarization from equation 2.2.1 and the relative permittivity, one can isolate the nonlinear polarization to the right hand side

\[ \nabla^2 \vec{E} - \frac{\epsilon_r}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2}, \] (2.3.4)

that acts as a source term in the inhomogeneous wave equation. The nonlinear susceptibilities contain a huge amount of entries \( (n+2)^2 \), as they are of rank \( n+1 \) for the nth-order. Symmetry concepts and fundamental constraints reduce the complexity of the higher order susceptibilities.

So far, we did not stress on the frequency dependence of the polarization and electric fields in equation 2.3.1. The involved electric fields can be of different origin, with different
frequency and different polarization. Taking equation 2.3.1 with a focus on the second order susceptibility and including a frequency dependence

$$\tilde{P}^{(2)}(\omega) = \epsilon_0 \tilde{\chi}^{(2)}(\omega; \omega_1, \omega_2) \tilde{E}(\omega_1) \tilde{E}(\omega_2)$$  \hspace{1cm} (2.3.5)

or in vectorial components

$$P_i^{(2)}(\omega) = \epsilon_0 \sum_{jk} \chi^{(2)}_{ijk}(\omega; \omega_1, \omega_2) E_j(\omega_1) E_k(\omega_2)$$  \hspace{1cm} (2.3.6)

yields the fundamental polarization for second order nonlinear processes. The angular frequencies $\omega_1$ and $\omega_2$ are free of choice and the configuration of the experiment and the crystal define the nonlinear process. Some possible interactions are already given in table 2.1 and will be emphasized in the chapter on nonlinear processes.

<table>
<thead>
<tr>
<th>susceptibility</th>
<th>resulting frequency</th>
<th>nonlinear process</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\chi^{(2)}(\omega_3; \omega_1, \omega_1)$</td>
<td>$\omega_3 = 2\omega_1$</td>
<td>frequency doubling</td>
</tr>
<tr>
<td>$\chi^{(2)}(\omega_3; \omega_1, \omega_2)$</td>
<td>$\omega_3 = \omega_1 + \omega_2$</td>
<td>sum frequency generation (SFG)</td>
</tr>
<tr>
<td>$\chi^{(2)}(\omega_3; \omega_1, (-)\omega_2)$</td>
<td>$\omega_3 = \omega_1 - \omega_2$</td>
<td>difference frequency generation (DFG)</td>
</tr>
<tr>
<td>$\chi^{(2)}(\omega_3; \omega_1, (-)\omega_1)$</td>
<td>$\omega_3 = 0$</td>
<td>optical rectification</td>
</tr>
</tbody>
</table>

Table 2.1: Second order optical processes and corresponding susceptibilities.

To simplify the description of the second order susceptibility, symmetries in the mathematical description and in the media are exploited. First of all it does not matter if we put the first field as second or the second field as first and therefore

$$\chi^{(2)}_{ijk}(\omega_1 + \omega_2; \omega_1, \omega_2) = \chi^{(2)}_{ikj}(\omega_1 + \omega_2; \omega_2, \omega_1)$$  \hspace{1cm} (2.3.7)

the intrinsic permutation is valid. The intrinsic permutation leads to a degeneration of the polarization as several permutations can result in the same frequency as for example in a three wave mixing process:

$$P_i(\omega) = \epsilon_0 \sum_{jk} \chi^{(2)}_{ijk}(\omega_3; \omega_1, \omega_2) E_j(\omega_1) E_k(\omega_2) + \chi^{(2)}_{ikj}(\omega_3; \omega_2, \omega_1) E_j(\omega_2) E_k(\omega_1)$$

$$= 2 \cdot \epsilon_0 \sum_{jk} \chi^{(2)}_{ijk}(\omega_3; \omega_1, \omega_2) E_j(\omega_1) E_k(\omega_2)$$  \hspace{1cm} (2.3.8)

The frequency dependence and losses in nonlinear crystals can be neglected in order to limit the amount of nonlinear susceptibilities. This holds as resonances of the $\chi^{(2)}$ are usually far from the optical spectrum for the media of interest in this thesis. The Kleinman symmetry follows this simplification and the frequencies or vice versa the indices of the susceptibilities can be permuted.

7
\[\chi_{ijk}^{(2)}(\omega_3; \omega_1, \omega_2) = \chi_{ijk}^{(2)}(\omega_2; \omega_1, \omega_3) = \chi_{ijk}^{(2)}(\omega_1; \omega_3, \omega_2)\]  \hspace{1cm} (2.3.9)

The Kleinman symmetry is further used for a contracted notation of the three dimensional tensor in the case of second order resulting in an effective nonlinear coefficient \(d_{\text{eff}}\). At this point we want to refer to the book of Boyd [6] where an ongoing classification of the susceptibility according to the crystal classes can be found. For centro-symmetric materials, we can cut the second and every even order of the polarization completely as those crystals show the same susceptibility under spatial inversion. While \(\chi^{(2)}\) stays the same, the electric field \((\vec{E} \to -\vec{E})\) and the polarization \((\vec{P} \to -\vec{P})\) follow the spatial inversion \((\vec{r} \to -\vec{r})\) so that
\[
\chi^{(2)}\vec{E}^2 \to -\chi^{(2)}\vec{E}^2
\]  \hspace{1cm} (2.3.10)

and
\[
\chi^{(2)}\vec{E}^2 \to \chi^{(2)}\vec{E}^2
\]  \hspace{1cm} (2.3.11)

have to be consistent with each other when \(\chi^{(2)} = 0\).

### 2.4 Time Frequency Representation

The description of optical pulses requires an effort in both, the time and frequency representation, as the affects on pulse propagation originate in both domains. The handling and representation of an optical pulse in frequency and time domain is treated in this chapter before we continue with propagation effects such as dispersion, broadening, and compression of the pulse.

#### 2.4.1 Fourier Transform

The Fourier transform (FT) is basically the tool to transform and display the optical field in both domains. With the FT one can retrieve details of a signal in time domain out of a signal in frequency domain. In terms of ultrashort pulses, we are interested in the temporal duration that is way too short for direct acquisition with detectors because of their limited temporal resolution. Experimentally access is only given in the frequency domain where the connection of frequency with time is most important in the generation, optimization and measuring of ultrashort pulses and ultrafast laser physics. We use the FT as follows

\[\vec{E}(t) = \frac{1}{2\pi} \int \vec{E}(\omega)e^{i\omega t}d\omega\]  \hspace{1cm} (2.4.1)

\[\vec{E}(\omega) = \int \vec{E}(t)e^{-i\omega t}dt.\]
For some signals an analytical representation of the FT exist, but usually numerical calculations are employed by means of the fast FT (FFT) for arbitrarily or experimentally obtained signals as we will do later.

Since a measurement reveals only the real part of a signal that can be the real valued signal in time domain $E(t)$ or the spectral resolved intensity $I(\omega)$ the FT is symmetric.

$$E(\omega) = E^*(\omega)$$

(2.4.2)

and the positive part of the spectrum is defined as

$$E^+(\omega) = E(\omega) \text{ for } \omega \geq 0 \text{ and } 0 \text{ for } \omega < 0$$

(2.4.3)

where the negative part contains the same information

$$E^-(\omega) = E(\omega) \text{ for } \omega < 0 \text{ and } 0 \text{ for } \omega \geq 0.$$  

(2.4.4)

The FT of a purely real signal does only return the envelope of a field $A$ without the phase information. It is the goal of this thesis to retrieve the phase information of an optical pulse for a complete pulse reconstruction.

Gaussian and secant hyperbolic pulse shapes are displayed in table 2.2 for several durations. The most common definition of the duration is the full width at half maximum (FWHM) of the intensity, where the intensity is given by

$$I = \frac{n\varepsilon_0 c}{2} |E_0|^2$$

(2.4.5)

in dependence of the electric field and the material dependent refractive index. For a nicely shaped Gaussian or secant pulse, the concept of FWHM works well and scales with the actual pulse duration. A FWHM duration might be misleading when pulses are more structured and show side peaks left or right from the central pulse position. In that case, the energy kept in the wings of the pulse is not quantified and the overall quality of the pulse does not enter the FWHM pulse duration. A concept for the pulse duration that takes into account the full shape of the pulse is based on the root mean square (RMS) error [7].
The FWHM, both in time and frequency domain is ultimately linked by the FT and results in a fixed time-bandwidth product $\Delta \nu \Delta \tau$ for various pulse shapes. A rough estimation from the plots in table 2.2 indicates that a pulse with a smaller bandwidth results in a longer pulse in the time domain and vice versa. The limited value of the time bandwidth product has an analogy in Heisenberg’s uncertainty principle saying that arrival time and energy of a particle can not be measured with arbitrarily precision. In case of EM waves or the photon particle, the energy is given by Planck’s equation $E = h\nu$.

Apart from the pulse shape, the carrier frequency and additional phase terms are described in the exponential function of equation 2.1.10. Figure 2.1 shows a Gaussian pulse with a duration of FWHM=5 fs and a carrier frequency according to $\lambda_0 = 800$ nm.

$$\hat{E}(z,t) = \mathcal{R}\{A(z,t)e^{i(kz-\omega t + \phi_{CE})}\}$$ (2.4.6)

The carrier envelope phase (CEP) $\phi_{CE}$ does not change the envelope but the position of the carrier field below the envelope and - as can be estimated from the plots - the maximum value of the electric field changes. This in particular is important when it comes to high harmonic generation and synthesis of optical pulses when processes are not only sensitive to the envelope of a pulse but to the electric field directly[3].

Table 2.2: Characteristics of Gaussian and hyperbolic secant pulses. The envelope $A(t)$ is given with dependence of the FWHM duration $\tau$. The pulse envelope in frequency domain $A(\omega)$ is plotted to the left. The time-bandwidth product $\Delta \nu \Delta \tau$ relates the bandwidth to the minimal pulse duration. The ratio $\tau_{IAC}/\Delta \tau$ is used to calculate the pulse duration from an intensity autocorrelation measurement if the pulse shape of an unknown signal can be estimated.
Figure 2.1: Gaussian pulse of 5 fs duration with varying carrier envelope phase (CEP) (a) \( \phi_{CE} = 0 \) (b) \( \phi_{CE} = \pi/2 \) (c) \( \phi_{CE} = \pi \) (d) \( \phi_{CE} = 3\pi/2 \).

### 2.4.2 Wigner Representation

The Wigner distribution originates in quantum mechanics where it was supposed by Eugene Wigner to replace Schrödinger’s probability functions. It became popular in the field of optics with the capability to display a three dimensional time-frequency plot. The combined representation of time and frequency domain makes it possible to fully observe the effects of pulse propagation. We follow the derivations of J. Paye in[8].

\[
W(E; t, \omega) = \int E(t + t') E^*(t - t') e^{i\omega t'} dt'
= \text{FT}\left\{ E\left(t + \frac{t'}{2}\right) E^*\left(t - \frac{t'}{2}\right) \right\}
\]

(2.4.7)

The representation can be calculated from the time domain or from the spectral domain from the complex spectrum \( E(\omega) \)

\[
W(E; t, \omega) = \frac{1}{2\pi} \int \tilde{E}\left(\omega + \frac{\omega'}{2}\right) E^*\left(\omega - \frac{\omega'}{2}\right) e^{-i\omega' t'} d\omega'
= \text{FT}^{-1}\left\{ E\left(\omega + \frac{\omega'}{2}\right) \tilde{E}^*\left(\omega - \frac{\omega'}{2}\right) \right\}
\]

(2.4.8)

The classic pulses of Gaussian and hyperbolic secant shape are displayed in figure 2.2 in means of the Wigner representation. Variations of those pulses are discussed within the scope of dispersion where additional Wigner diagrams are shown.
Figure 2.2: Wigner representation of transform limited Gaussian and hyperbolic secant pulses. (a) Gaussian pulse of 5 fs FWHM duration (b) Hyperbolic secant pulse of 5 fs FWHM duration (c) Gaussian pulse of 10 fs FWHM duration (d) Hyperbolic secant pulse of 10 fs FWHM duration (e) Gaussian pulse of 100 fs FWHM duration (f) Hyperbolic secant pulse of 100 fs FWHM duration. The marginals giving the pure temporal and spectral shape are projected on the wall of the plot box.

The spectral intensity of the pulse $I(\omega)$ and also the temporal intensity $I(t)$ are obtained by integrating over frequencies respectively time.
\[
\int W(E; t, \omega) dt = |E(\omega)|^2 = I(\omega) \quad (2.4.9)
\]
\[
\int W(E; t, \omega) d\omega = |E(t)|^2 = I(t)
\]

They are plotted in figure 2.2 as projections on the wall of the plot box.

2.5 Dispersion

In optics, dispersion is the dependence of the properties of an optical field on their wavelength. Solely in vacuum where the speed of light is constant, the propagation is free of dispersion. The wavelength dependent properties of an optical field in a medium result in a modulation of the phase. The mathematical construct of a pulse tells us that the phase should be flat in order to obtain the shortest pulse duration. The fundamental material dispersion originates in the wavelength dependent refractive index of a media. Shaping the material in from of a prism adds Snellius’ law of refraction at the interfaces and introduces angular dispersion, where the wavelengths follow different paths in space.

It is one of the main concerns in ultrafast optics to compensate for positive dispersion that piles up in materials like laser crystals, lenses or filters where the redder frequencies travel faster than the blue ones. The rearrangement of wavelengths so that they are ‘in phase’ is called compression and decreases the pulse duration. First of their kind compressors like prism or grating configurations make the redder frequencies travel a longer path and induce negative dispersion. Huge improvements on dispersion management were achieved with the concept of Gires–Tournois interferometers and dielectric double chirped mirrors (DCM). The compression techniques are covered in an own section.

The effect of dispersion on the wave equation is studied in the frequency domain. Fourier transforming the equation 2.1.8 and limiting the direction of propagation in \( \hat{z} \) gives

\[
\frac{\partial^2 E(z, \omega)}{\partial z^2} + \frac{\omega^2}{c^2} E(z, \omega) = -\omega^2 \mu_0 P_{LIN}(z, \omega) \quad (2.5.1)
\]

where we used the ease of the Parseval’s theorem by writing a derivation in time to a factor of \( i\omega \) in the Fourier domain

\[
\text{FT} \left\{ \frac{\partial^n f(t)}{\partial t^n} \right\} = (i\omega)^n \text{FT} \{ f(t) \}. \quad (2.5.2)
\]

For the evolution of the plane wave

\[
E(z, \omega) = A(z, \omega)e^{-ikz} \quad (2.5.3)
\]
in the wave equation 2.5.1 we calculate the spatial derivatives according to

\[
\frac{\partial E(z, \omega)}{\partial z} = \frac{\partial A(z, \omega)}{\partial z} e^{-ikz} - A(z, \omega) ke^{-ikz}
\]
\[
\frac{\partial^2 E(z, \omega)}{\partial z^2} = \frac{\partial^2 A(z, \omega)}{\partial z^2} e^{-ikz} - 2i k \frac{\partial A(z, \omega)}{\partial z} e^{-ikz} + k^2 A(z, \omega) e^{-ikz}
\]  

(2.5.4)

At this point it is convenient to apply the slowly varying amplitude approximation (SVEA) that basically neglects the variation of the pulse amplitude on the length scale of the wavelength.

\[
\left| \frac{\partial^2 A(z, \omega)}{\partial z^2} \right| \ll \left| 2 k \frac{\partial A(z, \omega)}{\partial z} \right|
\]

(2.5.5)

Note that SVEA is questionable when it comes to ultrashort pulses with a duration below 10 fs and breaks down when the envelope approaches the electric field in the single cycle regime [9]. Nevertheless, the approximation is well established to study the pulse evolution analytically.

An expression for the polarization divided in envelope and complex part similar to the electric field is given to

\[
\vec{\Pi}_{LIN}(z, \omega) = \Pi_{LIN}(z, \omega) e^{-ikz}
\]

(2.5.6)

where the envelope \( \Pi \) of the polarization is given with equation 2.2.1 to

\[
\Pi_{LIN}(z, \omega) = \varepsilon_0 \chi^{(1)}(\omega) A(z, \omega).
\]

(2.5.7)

Inserting equations 2.5.7 and 2.5.4 with the assumption of SVEA into the wave equation 2.5.1 yields

\[
\frac{\partial A(z)}{\partial z} = -ik A(z, \omega).
\]

(2.5.8)

It is obvious that \( k(\omega) \) defines the propagation and the shape of the envelope of our pulse.

The expansion of the wave number with respect to the center frequency \( \omega_0 \) is shown below

\[
k(\omega) \approx k(\omega_0) + \frac{dk}{d\omega} \bigg|_{\omega_0} (\omega - \omega_0) + \frac{1}{2} \frac{d^2k}{d\omega^2} \bigg|_{\omega_0} (\omega - \omega_0)^2 + \frac{1}{3} \frac{d^3k}{d\omega^3} \bigg|_{\omega_0} (\omega - \omega_0)^3
\]

\[
\approx k(\omega_0) + \frac{1}{v_g} (\omega - \omega_0) + \frac{1}{2} \text{GVD}(\omega - \omega_0)^2 + \frac{1}{3} \frac{d^3k}{d\omega^3} \bigg|_{\omega_0} (\omega - \omega_0)^3
\]

(2.5.9)

The group velocity \( v_g \) gives the velocity of the wave packet at a certain frequency, which is also the speed of information or energy and for sure \( v_g < c \) is valid. The group velocity dispersion (GVD) describes the spreading of the group velocity as it depends on the frequency itself. Usually the different velocities of the frequencies lead to a longer pulse with a separation of the colors. In the field of ultra short pulses the third order dispersion (TOD) must also be taken into account when the dispersion cannot assumed to be constant over frequency.
The propagation through media with refractive index \( n(\omega) \) directly defines the wave number \( k(\omega) = \frac{\omega}{c} n(\omega) \). Especially in experimental situations it is even more common to talk about the spectral phase in order to imply the thickness/length \( L \) of a media

\[
\phi(\omega) = k(\omega) \cdot L. \tag{2.5.10}
\]

The dispersion relation of the phase reads as

\[
\phi(\omega) \approx \phi(\omega_0) + \frac{d\phi(\omega)}{d\omega} |_{\omega_0} (\omega - \omega_0) + \frac{1}{2!} \frac{d^2\phi(\omega)}{d\omega^2} |_{\omega_0} (\omega - \omega_0)^2 + \frac{1}{6!} \frac{d^3\phi(\omega)}{d\omega^3} |_{\omega_0} (\omega - \omega_0)^3 + \ldots \\
\approx \phi(\omega_0) + T_g(\omega - \omega_0) + \frac{1}{2!} \text{GDD}(\omega - \omega_0)^2 + \frac{1}{6!} \text{TOD}(\omega - \omega_0)^3 + \ldots \tag{2.5.11}
\]

and the characteristic quantities of group delay \( T_g \) and group delay dispersion GDD are introduced where it is apparent that GDD = GVD \cdot L. It is most common in the field of fiber optics to call the GDD as D_2 for second order dispersion. The units are \( \frac{fs^2}{rad} \) for GDD and \( \frac{fs^3}{rad^2} \) for TOD but often written as \( fs^2 \) and \( fs^3 \). The electric field written with a frequency dependent phase is

\[
E(L, \omega) = |E(\omega)|e^{-i\phi(\omega)}. \tag{2.5.12}
\]

The frequency dependent phase reshapes the pulse in different orders and forms a chirped pulse. The term chirped might be borrowed from chirping birds with a sound of a musical chromatic scale. When redder frequencies are faster than blue ones, the chirp \( v_g(\text{red}) > v_g(\text{blue}) \) is termed positive which is the case for most media. A negative chirp is defined vice versa \( v_g(\text{red}) < v_g(\text{blue}) \).

We pick up the concept of the Wigner representation with the help of FT’s to study chirped pulses for various dispersions in table 2.3. A constant group delay \( T_g \) does not affect the pulse shape, it solely adds a delay equivalent to a longer path. The GDD stretches the pulse symmetrically in time due to the wavelength dependent phase that organizes the frequencies with different arrival times. The TOD is marked by a highly modulated signal in the time domain. The phase \( \phi(\omega) \) is not monotone and the corresponding frequency components in the left and right part of the spectrum get partly cancelled as can be seen on the intensity \( I(t) \).

The dispersion parameters for common materials used here and for the following simulations are derived from the refractive index \( n(\lambda) \). The refractive index can be called from the Sellmeier equation 2.5.13 where a huge collection is given in reference [10]. Since the Sellmeier equation is an empiric formula, the wavelength has to be put in in units of \( \mu m \).

\[
n = \sqrt{1 + \frac{B_1\lambda^2}{\lambda^2 - C_1} + \frac{B_2\lambda^2}{\lambda^2 - C_2} + \frac{B_3\lambda^2}{\lambda^2 - C_3}}. \tag{2.5.13}
\]
<table>
<thead>
<tr>
<th>Dispersion</th>
<th>Time</th>
<th>Frequency</th>
<th>Wigner</th>
</tr>
</thead>
<tbody>
<tr>
<td>3mm BK7</td>
<td><img src="image1" alt="Graph" /></td>
<td><img src="image2" alt="Graph" /></td>
<td><img src="image3" alt="Graph" /></td>
</tr>
<tr>
<td>50fs$^2$ GDD</td>
<td><img src="image4" alt="Graph" /></td>
<td><img src="image5" alt="Graph" /></td>
<td><img src="image6" alt="Graph" /></td>
</tr>
<tr>
<td>100fs$^3$ TOD</td>
<td><img src="image7" alt="Graph" /></td>
<td><img src="image8" alt="Graph" /></td>
<td><img src="image9" alt="Graph" /></td>
</tr>
</tbody>
</table>

Table 2.3: Dispersion of a 5fs FWHM Gaussian pulse centered at 800nm calculated after passing 3mm of BK7 glass and exemplary cases of 50fs GDD and 100fs TOD.

With $k(\omega) = \frac{\omega n(\omega)}{c}$ and the expansion in equation 2.5.9, the second order dispersion parameter GDD is calculated to

$$\text{GDD} = \frac{L}{c} \frac{\omega}{\omega^2} \frac{d}{d\omega} n(\omega) + \frac{\omega}{c} \frac{d^2}{d\omega^2} n(\omega).$$  \hspace{1cm} (2.5.14)$$

To match the wavelength dependence of the Sellmeier equation we use

$$\frac{d}{d\omega} = -\frac{\lambda^2}{2\pi c} \frac{d}{d\lambda},$$

$$\frac{d^2}{d\omega^2} = -\frac{\lambda^2}{(2\pi c)^2} \left( \lambda^2 \frac{d^2}{d\lambda^2} + 2\lambda \frac{d}{d\lambda} \right),$$  \hspace{1cm} (2.5.15)$$

to write

$$\text{GDD} = \frac{\lambda^2 L}{2\pi c^2} \frac{d^2 n}{d\lambda^2}. $$  \hspace{1cm} (2.5.16)$$

Following the same principle, the TOD reads

$$\text{TOD} = - \frac{\lambda^2 L}{2\pi c} \frac{d}{c} \left( \frac{d^2 n}{d\lambda^2} + \frac{d^3 n}{\lambda d\lambda} \right). $$  \hspace{1cm} (2.5.17)$$
2.5.1 Instantaneous frequency

We saw that laser light of frequency $\omega$ or wavelength $\lambda$ can result in complex pulses where a description with a single frequency is not appropriate any more. For a broadband pulse with possibly an octave spanning spectrum the center frequency $\omega_c$ gives the center location of the pulse in the spectral range. The temporal arrangement of frequencies in the pulse is defined depending on the phase and the concept of instantaneous frequency is defined as

$$\omega_{\text{inst}}(t) = \left. \frac{\partial \phi(t')}{\partial t'} \right|_{t'=t}$$

(2.5.18)

The concept of an instantaneous frequency is rather well established in optics and communication theory but has to be regarded with care. A plot of (instantaneous) frequency versus time claims a precise single frequency for an absolute precise point in time. This is not compatible with Heisenberg’s fundamental uncertainty principle. Using the instantaneous frequency, the representation should be only used over the full pulse where picking a single point in time or frequency is meaningless to our perception. Same applies for the Wigner distribution that connects time and frequency.

2.5.2 Spatial Properties

So far, our descriptions are focused on the temporal or spectral properties but spatially limited to a single coordinate $z$. Especially broadband pulses need a careful handling because imaging errors are much worse compared to narrow bandwidth or monochromatic beams.

Aberrations describe the deviation from ideal Gaussian optics. Spherical aberration accounts also for monochromatic beams, resulting in different focal points for rays close to the central axis compared to those far from it. Figure 2.3 sketches the longer focal distance for on axis rays where the off axis rays lead to blurred rings. The spherical aberration can be reduced by using plano-convex lenses. Those are used throughout the experiments in this work. The chromatic aberration resulting from dispersion adds to the spherical aberration for broadband pulses.
Figure 2.3: Imaging error properties of optical lenses. (a) Chromatic aberration where blue light focusses on shorter scale than redder frequencies (b) Spherical aberration in a bi-convex lens. This can be avoided using a plano convex lens and facing the plane side towards the focal point. (c) A rotated lens shifts the focal points and the picture is dossed like a coma. Off axis placement of the lens shows displaced focal points. (d) Astigmatism where the focus of the sagittal and meridian plane is different. [11]

The coma originates in an asymmetric misalignment of the beam with respect to the lens. A lens, positioned out of the normal, differs from the centrosymmetric focussing. Same for an off-axis object as can be seen in figure 2.3.

Off-axis objects and off-axis beams undergo astigmatism as well. The visualization is given in figure 2.3 with two planes, where the meridional plane focuses before the sagittal does, resulting in a blurred image.

2.5.3 Angular Dispersion

Angular dispersion terms a frequency dependent direction of propagation, usually induced by prisms or gratings and is typically used for a controlled manipulation or compression of the pulse. Angular dispersion (1) translates in a temporal dispersion as the colors in figure 2.4 show delayed arrival times and (2) a pulse front tilt is generated. From the scattering of a broadband light wave at an interface, the angular dispersion introduces a path difference for the colors along the wave vector $\tilde{k}(\omega)$ that translates in a phase
difference $\phi(\omega) = \vec{k}(\omega) \cdot L$ where $L$ is the distance in the medium. A detailed geometrical study is given in [12].

Figure 2.4: The origin of a pulse front tilt. Top: Using the angular dispersion of a prism to generate a pulse front tilt from an undistorted input pulse. Bottom: Generating a pulse front tilt using solely chromatic dispersion of a media with a spatially chirped pulse at the input. [13]

Geometries using angular dispersion have evolved into powerful tools for stretching, compressing or shaping a pulse because of the invention of configurations where the angular dispersion overcomes the material dispersion and the pulse leaves with a net negative chirp.

2.6 Dispersion Control

The control of the dispersion of an optical pulse gives the possibility to stretch, compress and shape the pulse to a desired form. In high power laser physics, dispersion control is used to (1) stretch the pulse in order to lower the peak intensity and therefore preventing damage in a laser amplification crystal and (2) recompress the pulse after amplification. The method was introduced in 1985 by Strickland and Mourou as chirped pulse amplification (CPA) [14]. In medium power laser systems, the dispersion control is used to account for the positive dispersion of the prior optics. Further efforts of dispersion control is the individual pulse shaping. Active components are used to tailor the waveforms according to the experiment where for example in the process of HHG a flat phase does not necessarily yield the most efficient one [15]. In the frame of ultrafast lasers it is mainly about to reduce the pulse durations by compressing the pulse towards a flat spectral phase. In common optical materials like lenses, laser crystals, windows and even air, the pulse piles up a chirp that stretches it in time. The aim of a dispersion control is to counteract
on that chirp by introducing anomalous dispersion in the optical path to counteract the rearrangement of frequencies. The common compression techniques are presented while only chirped mirrors are used for a final pulse compression in the experimental part of this thesis.

2.6.1 Prism Compressor

An arrangement of prisms is capable to introduce a net negative dispersion [16] and shows low losses on the one hand but is limited in the amount of dispersion due to limited angular dispersion. Nevertheless they are cost effective and used in the moderate pulse energy regime, for instance on laser oscillators. The laser pulse undergoes normal dispersion while propagating through the prism material which is mostly glass while the free-space travelling between the prisms is the regime of anomalous dispersion. In adjusting the distance between the prisms, one can easily compensate for the material dispersion and generate a net negative chirp. A typical configuration is shown in figure 2.5 where it is most common to use the retro-reflector at the end in order to pass four times through a prism and to recombine the angular dispersed colors spatially.

![Prism Compressor Diagram](image)

**Figure 2.5:** Prism compressor with a retro-reflective mirror to the right side and make the light passing through the prisms twice in order to recombine the pulse spatially.

The calculation of the phase raised by a sequence of prisms is followed in [17]. Starting from scattering the wave at the interface of the first prism, the calculation incorporated the passage of the prism under the angle of minimum deviation and the prisms apex angle $\alpha$ might be chosen to match the Brewster’s angle at the input and output facet

$$\tan \phi_1 = \tan \phi_2 = n \quad (2.6.1)$$

where $\phi_1$ and $\phi_2$ are the in- and output angles and $n$ is the refractive index of the prism’s material. A calculation of the path length for separated colors gives the second order dispersion effects where the material dispersion of the prism is taken into account in the first term

$$GDD = \frac{d^2 \phi_{4pris}}{d\omega^2} \approx \frac{\lambda^3 l_{mat}}{2\pi c^2} \frac{d^2 n}{d\lambda^2} - \frac{4L\lambda^3}{\pi c^2} \left( \frac{dn}{d\lambda} \right)^2 \quad (2.6.2)$$
and \( l_{mat} \) is the mean path length of the pulse in the material and \( L \) is the propagation length of the center wavelength in the prism geometry.

### 2.6.2 Grating Compressor

Grating compressors are widely used in the amplification of laser light [18]. A typical setup of a first pair of gratings stretches the pulse before it is launched in an amplifier and afterwards a second pair of gratings recompresses the pulse. Grating compressors benefit from the fact that no material has to be propagated and they can resist higher pulse energies compared to prisms. Then again they lack in efficiency and part of the pulse energy gets lost.

The angular dispersion relies on the well known grating equation

\[
\sin \theta_i + \sin \theta_D = \frac{m \lambda}{d}
\]

(2.6.3)

where \( \theta_i \) is the angle between the incident beam and the normal of the grating surface, \( \theta_D \) is the angle between the angular dispersed colors with wavelength \( \lambda \) and depends on the period of the grating \( d \). It can be indicated from figure 2.6 that the redder frequencies travel a longer distance and therefore, the setup creates a negative chirp.

![Figure 2.6: A grating compressor with a retro reflective mirror to the right side. The incident angle \( \theta_i \) and the dispersed angle \( \theta_D \) with respect to the normal on the grating surface that measures the distance \( l_\perp \) between the gratings.](image)

The second order dispersion of a pair of gratings is

\[
GDD = \frac{d^2 \phi}{d \omega^2} = -\frac{\lambda^3}{\pi c^3 d^2} \left( 1 - \left( \frac{\lambda}{d} - \sin \theta_i \right)^2 \right)^{-\frac{3}{2}} \cdot l_\perp
\]

(2.6.4)

with \( l_\perp \) the distance between the gratings measured along the normal. The third order dispersion of a grating pair reads as

\[
TOD = \frac{d^3 \phi}{d \omega^3} = -\frac{d^2 \phi}{d \omega^2} \frac{3 \lambda}{2 \pi c} \left( 1 + \frac{\lambda \tan \theta_D}{d \cos \theta_D} \right).
\]

(2.6.5)
The last factor in the TOD is always positive and therefore TOD in a grating setup always counteracts with respect to the second order dispersion. As we usually want to compensate for normal GDD, we unavoidably add normal TOD to the pulse and can state that a grating compressor can not account for TOD on its own. A combination of prism and grating compressor overcomes the shortcomings of both and is capable to compress a pulse from a mode locked laser down to 6 fs [19].

2.6.3 Chirped Mirrors

Chirped mirrors form a class of complex constructed dielectric mirrors consisting of multiple layers. The dispersion of such mirrors is based on interference effects between the layers. Apart from an easy alignment in contrast to prism and grating geometries, chirped mirrors are able to act on higher order dispersion simultaneously, that makes them favourable in the field of delicate ultrashort pulses. A starting point for dispersing mirrors is the Gires-Tournois interferometer (GTI) [20] shown in figure 2.7. The GDD depends on the reflectivity of the partially reflective mirror at the input facet and the distance between the mirrors resulting in a round-trip time depending on the refractive index of the intermediate material. GTI mirrors limit out at ultrashort pulses with large bandwidth as the distance and therefore material between the mirrors has to increased to account for large dispersion. Unavoidable higher order dispersion effects become significant.

High reflecting dielectric mirrors using stacks of alternating media on a scale of a quarter wavelength serve as high performance mirror and those Bragg stacks are the basis for double chirped mirrors (DCM). The alternating media give an alternating refractive index, say $n_H$ for high and $n_L$ for low. With increasing number of layers, the reflectivity tends to unity rather fast. The design is given by the Bragg wavelength $\lambda_B$

$$\lambda_B = 2(n_Hd_H + n_Ld_L)$$

(2.6.6)

where the thickness or optical path $P$ of the layers is chosen to be $P_H = P_L = \lambda_B/4$ in order to constructively interfere the reflections of all index discontinuities.

The fixed quarter-wave layer design is varied and a chirped distance between the index discontinuities result in chirped mirrors as shown in figure 2.7. Larger layer thickness towards the mirrors backside result in a longer path for redder frequencies whereas bluer frequencies are reflected by smaller layer structures near the front facet, leading to a negative chirp in the pulse.
Figure 2.7: From Top to Bottom: Gires-Tournois interferometer with a high reflectivity on the side of the substrate (gray) and a modal reflectivity on the input side. Bragg reflector where interference enhance the reflectivity. Chirped mirror with increasing layer thickness to propagate the redder frequencies deeper in the mirror. Double chirped mirror where the entrance layers enable an impedance matching to the free space propagating wave. An anti-reflection (AR) coating enhance the penetration in the layered structure. Furthermore backside coated DCM where the substrate is placed between AR coating and layered structure optimize the impedance matching and decouple the layer structure from the AR coating.

A problem regarding the group delay in plot 2.8 are imprinted oscillations on a designed dispersion curve. Those oscillations originate from reflections and interference at front and back side of the mirror and can be suppressed by adiabatically switching on the reflectivity using a slowly increasing thickness of the high index layer. This can be termed as a second chirp of the structure giving the name double chirped mirror (DCM) [21]. Additionally an anti-reflection (AR) coating is layered on the surface to enhance the propagation in the DCM.
2.7 Nonlinear Processes

Nonlinearities ultimately come along with ultrashort laser pulses because of their high peak intensity. In high power optical parametric amplification the nonlinearity is mandatory for today’s tunable ultrashort laser pulses from the UV to IR spectrum. Associated setups, using nonlinear crystals that show a high nonlinear coefficient and a high damage threshold are developed to mix, modify and amplify ultrashort pulses. Based on the nonlinear propagation in section 2.3, the fields originate from the nonlinear polarization. The efficiency of the process is determined by the phase matching between the fields involved.

2.7.1 Second Order Process

A typical geometry is using a second order nonlinear crystal with two input beams, say $E_1(\omega_1)$ and $E_2(\omega_2)$ where the frequencies $\omega_1$ and $\omega_2$ can be different or equal. The

Figure 2.8: Comparison of the reflectivity and the group delay of chirped mirrors with 25 layer pairs with refractive indices $n_l = 1.5$, $n_h = 2.5$. The upper portion shows the enlarged top percent of the reflectivity. The dotted curves show the result for a simply chirped mirror. The dashed and solid curves show the result for double-chirped mirrors, where in addition to the chirp in the Bragg wave number $k_B$ the thickness of the high-index layers is also chirped over the first 12 layer pairs from zero to its maximum value for a linear chirp (dashed curves) and for a quadratic chirp (solid curves). [22]
frequency at the output of the crystal depends on the input fields and the chosen phase matching constraint. The second order nonlinear polarization generated by those fields reads according to 2.3.6 as

$$P_{NL}^{(2)} = \chi^{(2)} \left\{ E_1^2 e^{2i(\omega_1 t - k_1 z)} + E_2^2 e^{2i(\omega_2 t - k_2 z)} + 2E_1E_2 e^{i((\omega_1 + \omega_2)t - (k_1 + k_2)z)} + 2E_1E_2^* e^{i((\omega_1 - \omega_2)t - (k_1 - k_2)z)} + E_1E_2^* + E_1^*E_2 + c.c. \right\}$$

oscillates @ $\omega_1$ oscillates @ $\omega_2$
oscillates @ $\omega_1 + \omega_2$
oscillates @ $\omega_1 - \omega_2$ DC part.

A nonlinear crystal supporting those processes is therefore a powerful tool for tuning the frequency of an optical pulse from $\omega_1 - \omega_2$ up to $\omega_1 + \omega_2$. The following schematics show the possible processes. Nonlinear processes have to maintain the energy conservation. Taking the particle picture of a photon, two photons of lower energy, say $E_1$ and $E_2$ result in a photon of higher energy $E_3 = E_1 + E_2$, considering the general case of a lossless media.

$$\begin{align*}
\frac{h \cdot v_3}{E_3} &= \frac{h \cdot v_1}{E_1} + \frac{h \cdot v_2}{E_2} \\
\omega_3 &= \omega_1 + \omega_2
\end{align*}$$

Figure 2.9: Second order nonlinear processes. (a) The sum frequency generation where $\omega_1$ and $\omega_2$ are the frequencies of the input pulse. They can also be of the same frequency in one pulse that corresponds to frequency doubling. (b) Optical parametric amplifier with a strong pump (blue arrow) and a signal (red arrow) that shall be amplified. The residual energy leaves with the idler (green). The arrows indicate a noncollinear geometry for visibility but a collinear propagation is possible, too and the usual case.

A calculation of the generated field uses the equations of propagation from chapter 2.3 and is shown for the case of second harmonic generation (or frequency doubling) in the undepleted pump regime. The undepleted pump regime assumes a strong input pulse as pump with a constant envelope $\frac{dA_p}{dz} = 0$. Strictly speaking, the pump has to lose energy when the second harmonic field builds up in order to fulfill the energy conservation.
The nonlinear wave equation in frequency domain is extended from equation 2.5.1 by the nonlinear polarization

\[ \frac{\partial^2 E(z, \omega)}{\partial z^2} + \frac{\omega^2}{c^2} E(z, \omega) = -\omega^2 \mu_0 P_{LIN}(z, \omega) + \omega^2 \mu_0 P_{NL}(z, \omega) \]  

(2.7.3)

With the plane wave approximation 2.1.10 substituted, the wave equation transforms to

\[ \frac{\partial^2 E(z, \omega)}{\partial z^2} + \frac{\omega^2}{c^2} \epsilon(\omega) E(z, \omega) + \omega^2 \mu_0 P_{NL}(z, \omega) = -\frac{\omega^2}{\epsilon_0 c^2} e^{ikz}. \]  

(2.7.4)

The SVEA is applied to obtain the simple relation between the nonlinear polarization and the envelope of the pulse

\[ \frac{\partial A(z)}{\partial z} = \frac{i\omega}{2\epsilon_0 c} P_{NL}(z, \omega) e^{-ik_2wz}. \]  

(2.7.5)

At this point we can insert the nonlinear polarization of the second harmonic process

\[ P_{NL}(z, 2\omega) = \epsilon_0 \chi^{(2)}(2\omega; \omega, \omega) A^2(\omega, z) e^{-i2k(\omega)z} \]  

(2.7.6)

and get the second harmonic field at twice the frequency of the fundamental

\[ \frac{\partial A_{2\omega}(z)}{\partial z} = \frac{i(2\omega)}{2\eta_2(\omega)} d_{\text{eff}} A_{\omega}^2 e^{i(k_2\omega - 2k_\omega)z}. \]  

(2.7.7)

The effective nonlinear coefficient \( d_{\text{eff}} \) is replacing the second order susceptibility in order to exploit the Kleinman symmetry. \( d_{\text{eff}} \) is tabulated for each crystal class and fixed geometries of the beams [6] and gives a single quantity compared to 27 in \( \chi^{(2)} \). The integration from \( z = 0 \) to \( z = L \) delivers in the undepleted pump regime (\( \frac{dA_\omega(z)}{dz} = 0 \)) and assuming \( A_{2\omega}(z = 0) = 0 \)

\[ A_{2\omega}(L) = \frac{i2\omega d_{\text{eff}} A_{\omega}^2}{2n_2(\omega)} \frac{\Delta k}{2} \sin \left( \frac{\Delta k L}{2} \right) e^{ik_2\omega L}. \]  

(2.7.8)

The intensity of the generated field at \( 2\omega \) that builds up over a distance \( z \) is calculated using the intensity relation 2.4.5.

\[ I_{2\omega}(L) = \frac{2\omega^2 d_{\text{eff}}}{\epsilon_0 n_2^2 n_{2\omega} c^2} \sin \left( \frac{\Delta k L}{2} \right) I_\omega^2 L^2 \]  

(2.7.9)
To identify the sinc and respectively sin function, the relation

\[
\left| \frac{e^{i\Delta k L} - 1}{\Delta k} \right|^2 = \left( \frac{e^{-i\Delta k L} - 1}{\Delta k} \right) \left( \frac{e^{i\Delta k L} - 1}{\Delta k} \right) = \frac{2(1 - \cos(\Delta k L))}{(\Delta k)^2} = \frac{\sin^2(\Delta k L/2)}{(\Delta k/2)^2} = L^2 \text{sinc}^2(\Delta k L/2)
\]

(2.7.10)
is used to derive from equation 2.7.7 to 2.7.9.

The energy transfer from the fundamental wave at \( \omega \) to the second harmonic at \( 2\omega \) holds as long as the fields are in phase. The phase relation \( \Delta k = 2k(\omega) - k(2\omega) \) is known as the phase mismatch and limits a consequent energy flow from fundamental to second harmonic wave. Chromatic dispersion of the nonlinear material limits an ideal phase matching of \( \Delta k = 0 \) for any frequency and a continuous build up of the second harmonic wave is usually limited to a certain distance. Figure 2.10 depicts the efficiency for various values of \( \Delta k \).

![Figure 2.10: Efficiency of the SHG process. The calculation is based on equation 2.7.9 and a fundamental intensity of \( I_\omega = 10^{15}\text{W/m}^2 \). A perfect phase matching shows an exponential growth of the second harmonic intensity. A phase mismatch is calculated with respect to the propagation length \( L \). With increasing phase mismatch the length over which a second harmonic field builds up decreases and the back conversion start after decreasing distance.](image)

The efficiency of the SHG scales with the intensity of the input beam \( I_\omega \), the nonlinear coefficient \( d_{\text{eff}} \), and increases exponentially when a perfect phase matching is guaranteed. The intensity chosen for the plot in figure 2.10 is \( I_\omega = 10^{15}\text{W/m}^2 \).

### 2.7.2 Phase Matching

In addition to the energy conservation, it is the phase matching that has to be fulfilled for an efficient nonlinear process. The phase matching relation

\[
\vec{k}_3 = \vec{k}_1 + \vec{k}_2
\]

(2.7.11)
has to be fulfilled together with equation 2.7.2. The nonlinear crystal used during this thesis is a $\beta - \text{BaB}_2\text{O}_4$ (BBO) crystal but the following argumentation is generally valid. BBO shows a high nonlinearity, a birefringence used for phase matching and a reasonably high damage threshold. It can be used from low to high power applications and the transparency range from 190nm to 3300nm covers the common optical range from the UV to near IR but lacks in mid to far IR applications.

Phase matching in this case is done by employing the birefringence of the medium and using the different phase velocities along the extraordinary and ordinary direction. It is the aim to tune the crystal’s axis and the cutting angle $\theta$ with respect to the wavelength that are involved in a certain nonlinear process. In birefringent media, the phase between fundamental and generated wave can be matched in two ways for the collinear interaction:

\[
\begin{align*}
\vec{k}_\theta(\omega) + \vec{k}_\theta(\omega) &= \vec{k}_o(2\omega) \quad \rightarrow \quad n_\theta(\omega) &= n_o(2\omega) \\
\vec{k}_o(\omega) + \vec{k}_o(\omega) &= \vec{k}_\theta(2\omega) \quad \rightarrow \quad n_o(\omega) &= n_\theta(2\omega) \\
\vec{k}_\theta(\omega) + \vec{k}_o(\omega) &= \vec{k}_o(2\omega) \quad \rightarrow \quad \frac{n_\theta(\omega) + n_o(\omega)}{2} &= n_o(2\omega) \\
\vec{k}_o(\omega) + \vec{k}_o(\omega) &= \vec{k}_\theta(2\omega) \quad \rightarrow \quad \frac{n_\theta(\omega) + n_o(\omega)}{2} &= n_\theta(2\omega)
\end{align*}
\]

Type I phase matching describes a configuration where the input fields have the same polarization (along the ordinary axis of the crystal for negative uniaxial crystals and vice versa for positive uniaxial crystals). In a collinear propagation of input and output beams, the wave vector directly translates into the refractive index that shall be matched for the fundamental and second harmonic frequency. The index ellipsoid (figure 2.11a) represents the Fresnel equation and gives the values for the extraordinary refractive index we can tune to. Note that $n_o(90^\circ)$ is again the ordinary refractive index $n_o$. From figure 2.11 the difference to type II phase matching becomes clear, where distinct input fields are employed.
Two input pulses of perpendicular polarization are focused in the nonlinear crystal resulting in an output at extraordinary polarization for negative uniaxial crystals and at ordinary polarization for positive uniaxial crystals for type II phase matching. The analytical representation of the index ellipsoid for a uniaxial crystal is given by

\[
\left( \frac{1}{n_\theta} \right)^2 = \left( \frac{\cos \theta}{n_o} \right)^2 + \left( \frac{\sin \theta}{n_e} \right)^2.
\]  

In order to estimate the phase matching angle \( \theta \) for a parametric amplification process, the wavelength is plotted over the angle \( \theta \) for a given pump wavelength, see figure 2.12.
Apart from an angle tuning, the temperature dependence of the birefringent refractive index gives the possibility of a temperature-wise phase matching. The index of refraction along ordinary and extraordinary direction changes differently with temperature and therefore a match of the phase velocities at different frequencies and polarizations might be possible. This kind of phase matching is rather limited to certain range of temperatures, same as crystals and wavelength range. On the other hand it has the advantage of a non-critical phase matching (see figure 2.13) with the advantage of no walk-off effect present and generally resulting in a higher conversion efficiency. Another technique yielding high conversion efficiencies by using longer nonlinear crystals and counteracting on the phase mismatch with a periodically poling of the crystals axis is called quasi phase matching. There, the phase mismatch increases until back conversion of the energy to the fundamental as shown in figure 2.10. At this point the inversely poled material decreases the mismatch in $\Delta k$ so that the back conversion is prevented before it grows again until the next poling period is reached.

The experiments in this thesis employ angle tuning of the crystal for the benefits of a thin crystal and for the flexibility in terms of tuning for wavelength and bandwidth. We want to extend that method by a non-collinear configuration that benefits from a larger bandwidth during the second harmonic process. The wave vector picture is given in figure 2.13.
The phase mismatch $\Delta k$ of a SFG process for the collinear arrangement can be calculated straightforward from the refractive indices. In the case of a non-collinear configuration we use the projection of the wave vectors depending on the intersection angle $\alpha + \beta$ of the input beams in equation 2.7.15. In order to match the experiment, where we want to phase match a broadband and a narrowband pulse in a type II configuration for sum frequency generation, the fields are named by

- $\omega_{PUT}$ for Pulse under Test, usually broadband
- $\omega_{AC}$ for ancillae pulses, usually narrowband
- $\omega_{SIG}$ for Signal, the resulting SFG signal being measured with a spectrometer

The angle dependent phase mismatch gives a second degree of freedom to tune and minimize $\Delta k$ for the frequency of interest and as broad as possible.

$$\Delta k_{col} = \frac{\omega_{SIG}}{c} n_\theta(\omega_{SIG}) - \frac{\omega_{AC}}{c} n_\theta(\omega_{AC}) - \frac{\omega_{PUT}}{c} n_\theta(\omega_{PUT})$$

$$\Delta k_{noncol} = \frac{\omega_{SIG}}{c} n_\theta(\omega_{SIG}) \cdot \cos(\alpha) - \frac{\omega_{AC}}{c} n_\theta(\omega_{AC}) - \frac{\omega_{PUT}}{c} n_\theta(\omega_{PUT}) \cdot \cos(\alpha + \beta)$$  \hspace{1cm} (2.7.14) \hspace{1cm} (2.7.15)

Calculations for SFG efficiencies depending on $\Delta k$ are given in figure 4.6.

### 2.7.3 Third Order Process

In terms of ultrashort pulses, the process of self-phase modulation (SPM) in solids and gases might be the most prominent one. Also some pulse characterization techniques like polarization gate (PG) or third harmonic generation (THG) FROG employ the $\chi^{(3)}$ nonlinearity of a medium where the majority employ the second order nonlinearity. Starting from the polarization in equation 2.3.1 and masking the second order effect

$$\vec{P} = \varepsilon_0 \left( \chi^{(1)} + \chi^{(3)} |\vec{E}|^2 \right) \vec{E}.$$  \hspace{1cm} (2.7.16)
we derive an intensity dependent refractive index. The refractive index, as built up from the susceptibility (equation 2.2.4),

\[
    n = \sqrt{1 + \chi^{(1)} + \chi^{(3)}|\bar{E}|^2} = \sqrt{n_0 + \chi^{(3)}|\bar{E}|^2}
\]

\[
    \approx n_0 \left( 1 + \frac{\chi^{(3)}|\bar{E}|^2}{2n_0^2} \right) = n_0 + n_2|\bar{E}|^2 = n_0 + \Delta n
\]  

(2.7.17)

is separated in the linear intensity independent refractive index \( n_0 \) and the nonlinear intensity dependent refractive index \( n_2 \) proposing a modulation \( \Delta n \) with \( I(t) \).

Considering the phase of the pulse in equation 2.1.10, we can separate the linear from the nonlinear contribution

\[
    \phi(t) = kz - \omega t = \frac{\omega n z}{c} - \omega t
\]

\[
    = \frac{\omega n z}{c} + \frac{\omega n_2 I(t) z}{c} - \omega t
\]

(2.7.18)

where the nonlinear refraction adds a phase shift \( \phi_{NL} = \frac{\omega n_2 I(t) z}{c} \). The frequency components inside the pulse are given by the instantaneous frequency as

\[
    \omega_{inst} = -\frac{\partial \phi}{\partial t} = \omega - \frac{\omega n_2}{c} \frac{\partial I(t)}{\partial t} z
\]

(2.7.19)

and it becomes clear that new frequencies will be generated by \( \phi_{NL} \). When the derivative \( \frac{\partial I}{\partial t} > 0 \) is positive for the leading edge of the pulse, the spectrum is red shifted and vice versa for the trailing part of the pulse where \( \frac{\partial I}{\partial t} < 0 \). Since the phase and frequency modulation is driven by the pulse itself, the effect is termed self-phase modulation (SPM).

Figure 2.14 plots the instantaneous frequency over its temporal position in the pulse (2.14b) and displays the heavily modulated spectra in frequency domain (2.14a).

![Figure 2.14: The simulation of a Gaussian pulse at a center wavelength of \( \lambda_c = 800 \text{nm} \) with a FWHM duration of 25fs undergoing pure SPM. (a) The clean input pulse and the self-phase modulated one in the frequency domain. (b) The intensity and the instantaneous frequency resulting from the intensity dependent refractive index.](image)

The huge modulation in the frequency domain is explained by the interference of frequency components that are equal but generated at different times dependent on the
pulse envelope. Following the way we described second order nonlinear processes, the nonlinear polarization of third order is capable of mixing four distinct fields. An overview of the processes is given in table 2.4.

<table>
<thead>
<tr>
<th>susceptibility</th>
<th>nonlinear process</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\chi^{(3)}(\omega_1;\omega_1,0,0)$</td>
<td>DC Kerr effect</td>
</tr>
<tr>
<td>$\chi^{(3)}(\omega_1;\omega_1,\omega_1,-\omega_1)$</td>
<td>SPM</td>
</tr>
<tr>
<td>$\chi^{(3)}(3\omega_1;\omega_1,\omega_1,\omega_1)$</td>
<td>frequency tripling</td>
</tr>
</tbody>
</table>

Table 2.4: Third order optical processes and corresponding susceptibilities.

An application of SPM is found in the hollow core fibre (HCF). There, a laser pulse of sufficient intensity is focussed in hollow fibre filled with a noble gas. The pressure of the gas, the initial chirp of the laser pulse and the length of fibre give two major parameters in order to enhance the SPM and frequency broadening caused by third order nonlinearity of the gas.

Phase matching in third order processes is not an issue because of the nature of the process itself. The phase matching relation for SPM for instance results in $\Delta k = k_1 - k_1 - k_1 + k_1 = 0$.

The generation of a white light continuum (WLC) is of interest for broadband parametric amplifier (OPA) and is driven by an interplay of higher order nonlinearities. The WL continuum defines a broad spectrum, that is generated from a laser pulse focused in a transparent crystal like sapphire or YAG. The laser pulse has to be sufficient short to reach high peak intensities but the spectrum compared to the WLC is rather narrowband. Because of white light is used to seed broadband OPAs in the experimental part of this thesis, we want to describe the principle in detail. In our case, the process of filamentation generates the WLC in a solid like YAG or sapphire crystals.

It is triggered when the intensity of a laser reaches a critical value where self-focusing starts. Assuming a Gaussian intensity profile, the Kerr effect results in a higher refractive index in the center part of the laser beam compared to the wings according to equation 2.7.20. The increased refractive index slows down the phase velocity $v_{ph} = c_0/n$ in the center part of the pulse and leads to converging phase fronts.

$$n = n_0 + \frac{n_2 I}{\Delta n_{kerr}}$$  \hspace{1cm} (2.7.20)

Since a pulse shows a varying intensity in the longitudinal direction, too, the slice-by-slice focusing model predicts differing focal points for each longitudinal coordinate of the pulse.
Self-focussing is the spatial and transversal aspect of the Kerr effect, where SPM is temporal aspect of the Kerr effect and occurring along the direction of propagation. SPM ensures the spectral broadening of the high intensity and self-focused pulse as derived in equation 2.7.19 and shown in figure 2.14a.

Without additional defocussing effects, the self-focusing would lead to a collapse of the pulse. But, the high intensities in the focus enables photo ionization of the media and forms a weak plasma in the area of the filament. The plasma is changing the refractive index contrary to the Kerr effect.

\[
\Delta n(t)_{\text{plasma}} = -\frac{\omega_0^2(t)}{2\omega_0^2} = -\frac{e^2 n_e(t)}{2m_e\varepsilon_0\omega_0^2}
\]  
\hspace{1cm} (2.7.21)

In solids, the generation of electron-hole pairs leads to a change of the refractive index that weightens up the Kerr effect. A plasma generation in a condensed media would lead to a permanent damage and needs to be avoided. The self-focusing and super continuum generation shows a strong dependence on the band gap of the medium where the MPE transition rate \(W\) follows the power law \(W \propto I^K\) with \(K = E_{\text{gap}}/h\nu\) [26]. The larger the band gap of a semiconductor, the higher is the clamped intensity at which self-focusing and plasma defocusing level off. Furthermore, the crystals have to sustain the high intensity so that sapphire \((E_{\text{gap}} = 9.9 \text{ eV})\) and YAG \((E_{\text{gap}} = 6.3 \text{ eV})\) which are proven host materials for laser crystals are the first choice for WLC generation. For the generation of a WLC that extends to the IR in order to get amplified by an OPA presented in section 4.3 we investigate the spectra of YAG and sapphire shown in figure 2.16. They are generated from a 1020nm pump pulse and span up to 2.5\(\mu\)m where the detection of our spectrometer limits. The strong pump region is apparent in the trace for sapphire where it is filtered out with a dichroic mirror for trace of YAG.
Figure 2.16: White light continua generated from a 1020nm pump. The WL of sapphire spans up to 2.5$\mu$m where YAG shows a considerable intensity only up to 2$\mu$m. A dichroic mirror is used to filter out the region of the pump for the case of YAG.
3 Pulse Characterization

A pulse characterization is a measurement of light using detectors that transform the energy of photons into an electronic signal. One might wish to track the temporal shape of the pulse directly using photo diodes and oscilloscopes but the experimental limits with fast photo detectors are in the picosecond range. Optical pulse measurements on shorter time scales have to be performed in the frequency domain, where slow detectors are sufficient to capture the spectrum and phase with the help of a (nonlinear) optical setup.

While it is uncomplicated to capture the spectral intensity of a pulse with a commercial spectrometer, the temporal shape and phase of the electric field needs more effort. Precise pulse measurement techniques gained attention around the 1970th when pulse durations as short as a few picoseconds were generated. The topic spans from field autocorrelators to today’s state of the art technique implying nonlinear processes, active piezo stages and a computer controlled acquisition and retrieval of the laser pulse. In order to be able to compare the methods to the work of this thesis, this chapter includes autocorrelation methods, the popular FROG method, the SPIDER setup and their derivatives before we finally arrive at the two-dimensional spectral shearing method (2DSI) and the experimental work of this thesis.

3.1 Correlation Methods

Interferometric techniques have a long history in the detection schemes of coherent light. Linear interferometric technique were used by Michelson and Morley in 1881 respectively 1887 to answer the theory of an ether acting as a medium for EM wave propagation [27]. Also modern gravitational wave detectors like GEO600 and LIGO use (huge) interferometers. In the case of ultrashort optical pulses which are among the shortest man-made events, the interferometric technique is used to sample the pulse with itself. Not only but part of the 2DSI setup also contains a Michelson interferometer (MI) as sketched in figure
The relation between the input electric field and the output electric field is given by the superposition of the electric field after being split by the beam splitter (BS) and delayed in one of the interferometer arms. The field at the output reads

$$E_{\text{out}}(t) = \frac{1}{2} \Re\{A(t-t_1)e^{i\omega_k(t-t_1)} + A(t-t_2)e^{i\omega_k(t-t_2)}\}$$ (3.1.1)

The intensity measured by the photo diode is given by the squared electric field. To account for the slow response of the photo detector, the signal out of the MI is integrated over time, losing the time dependence $P(t)$ but keeping the dependence on the relative delay $\tau$. The absolute size and delay of a symmetric MI is of no interest and we introduce the time delay between the arms $\tau = t_1 - t_2 = 2l_1/c - 2l_2/c$.

$$\text{SIG}_{\text{PD}} = \int dt (E_{\text{out}}(t))^2$$ (3.1.2)

It is shown in [12] that the linear interferometer is not capable of extracting the phase of the signal. We cannot distinguish between an optical pulse or a broadband signal of other origin like noise.

It turns out that using a nonlinear element in which the splitted pulse under test is recombined generates a signal that depends on the duration of the pulse under test [28]. The optical setup is shown in figure 3.2. The nonlinear crystal is phase matched for a SFG process of the pulses with identical spectra.
Figure 3.2: Intensity autocorrelation setup. The pulse is split by the beam splitter (BS) and send to the delay stage (framed) before launched into the second order ($\chi^{(2)}$) nonlinear crystal. An iris can be used to block the remaining fundamental and a photo diode is acquiring the signal.

Similar to the linear interferometer, the pulses are delayed with respect to each other and the nonlinearity of the crystal is giving a signal at twice the frequency that depends on the overlap of the incident pulses in time. The second order polarization is proportional to the delayed electric fields $E(t)$ and $E(t - \tau)$

$$P^{(2)}(t) \propto E(t) \cdot E(t - \tau). \quad (3.1.3)$$

In a noncollinear fashion, the second harmonic (SH) of the individual waves does not enter the detector and there is no background signal when the pulses do not overlap. The polarization is again proportional to the second harmonic field that is recorded as a time integrated intensity by the photo detector according to

$$I_{SH}(\tau) \propto \int I(t) I(t - \tau) dt. \quad (3.1.4)$$

The phase is again lost in this measurement but the recorded signal over the delay $\tau$ gives a FWHM pulse duration of the SH signal, that can be compared with calculated FWHM durations from common pulse shapes and their FWHM in time domain. The technique is rather insensitive to the shape of a pulse and only gives a reasonable result when the pulse shape is known a priori.

A method called interferometric autocorrelation (IAC) introduced by Diels [29] in 1985 combines the interferometric setup with a nonlinearity and gives additional coherence terms in the correlation function. The setup is shown in figure 3.3.
Figure 3.3: Interferometric autocorrelation (IAC) where both pulse copies undergo the same amount of dispersion before the second harmonic signal is recorded with a photo diode.

The electric field at the output of the MI is again 3.1.1 and the polarization and electric field after the nonlinear crystal is proportional to the squared electric field of the superimposed pulse copies

\[ E_{SH} \propto \left( A(t)e^{i\omega_t t} + A(t - \tau)e^{i\omega_t(t-\tau)} \right)^2. \]  

(3.1.5)

The photo detector integrates the intensity over time where the single terms are

\[ I(\tau) \propto \int dt \quad |A(t)|^4 + |A(t - \tau)|^4 \]

\[ + 4 |A(t)A(t - \tau)|^2 \]

\[ + 2A(t)|A(t - \tau)|^2 A^*(t - \tau)e^{i\omega_t \tau} + c.c. \]

\[ + 2A(t - \tau)|A(t)|^2 A^*(t)e^{-i\omega_t \tau} + c.c. \]

\[ + A(t)^2 A^*(t - \tau)^2 e^{2i\omega_t \tau} + c.c. \]

\[ = 2 \int I^2(t) dt \]

\[ + 4 \int I(t) \cdot I(t - \tau) \]

\[ + 4 \int \Re \left( (I(t - \tau) + I(t))A^*(t - \tau)A(t)e^{i\omega_t \tau} \right) \]

\[ + 2 \int \Re \left( (A(t - \tau))^2 (A^*(t))^2 e^{2i\omega_t \tau} \right) \]

(3.1.6)

and give a background signal \( I_{\text{back}} \), an autocorrelation term \( I_{\text{auto}} \), an coherence term swinging at \( \omega_c \) and one at the SH frequency \( 2\omega_c \).

The last two terms give additional information to the intensity autocorrelation signal which is present in the second term. The interference terms contain the phase information, so that the interferometric AC trace more sensitive to the spectral phase but the pure spectral phase can not be extracted directly. The calculated IAC trace of an assumed pulse is fitted to measured data in order to find the pulse duration, where one starts with a flat phase e.g. bandwidth limited pulse. Various IAC traces for different orders of dispersion are plotted in figure 3.4. The dispersion of a pulse can be distinguished best in the wings of the IAC signal with lifted spectral wings for second order dispersion and a modulation in the wings for third order dispersion.

Generally, an inverse algorithm need sufficient high signal to noise ratio (SNR) to converge
Figure 3.4: Calculation of IAC signals (a) hyperbolic secant pulse shape of 10 fs FWHM duration (b) Same pulse passed through 3 mm BK7 glass. (c) Solely 200fs$^2$ of GDD and (d) solely 1000fs$^3$ of TOD.

to a solution and additionally the single trace is the only data set without the possibility for consistency checks. So far, no single method presented was capable to extract the phase on its own and only relied on assumed pulse shapes. Those pulse shapes can be predicted for some sort of pulse generations (for example mode locked lasers in [30]) but in the sub 10 fs regime several authors have shown problems of methods like IAC. Figure 3.5 shows the comparison of a pulse measurement by IAC compared to the more sophisticated SPIDER method. The resolution of the pedestals is poor in the IAC trace and generally leads to an estimation of a shorter pulse duration.
In the ultrashort pulse regime, further limitations from every single optical element, as for example dispersion from beam splitters, lenses or bandwidth limitations from nonlinear crystals arise and give the motivation for the following two classes of sophisticated pulse characterization techniques: FROG and SPIDER.

### 3.2 FROG

The technique of frequency-resolved optical gating (FROG) [32] is based on autocorrelation methods but with the photodiode replaced by a spectrometer in order to capture a frequency resolved intensity autocorrelation trace. Spectrally resolved gating techniques in different versions opened up a whole family of FROG pulse characterization techniques. Rick Trebino’s book, dedicated to the FROG family [33] serves as the main reference here. The general FROG signal is constructed by an electric field $E$ and a gate pulse $g$ which is usually a copy of $E$. Since the signal is recorded spectrally resolved, it also depends on the angular frequency $\omega$ in addition to the delay $\tau$. The convolution is recorded in forms of an intensity as

$$S(\omega, \tau) = \left| \int E(t)g(t - \tau)\exp(i\omega t)dt \right|^2$$  

(3.2.1)
Since the gate pulse is most probably a copy of $E(t)$, it is not known a priori. When using other pulses to gate, it can be estimated by equation 3.2.1 that an infinitely long gate, let us assume a cw beam, would not change the signal $S(\omega, \tau)$ by delay and ruin the temporal resolution of the pulse. An infinitely short gate would solely result in a temporal dependence $S(\tau)$ and ruin the phase information. A gate at the duration of the pulse itself results in a set of spectra connected by the delay $\tau$. The pulse and its delayed copy generate a signal that is treated in a general way as $E_{SIG}(t, \tau)$ and integrated subsequently by the slow response spectrometer.

$$S(\omega, \tau) = \left| \int E_{SIG}(t, \tau) e^{i\omega t} dt \right|^2$$ (3.2.2)

The FROG signal $E_{SIG}$ depends on the geometry of the experimental setup and the chosen nonlinear order. Self-diffraction SD-FROG relies on the third order effect of self-diffraction. The two crossed beams as shown in figure 3.6 generate a modulation of the refractive index by the Kerr effect and build up a transient grating of the refractive index.

![Figure 3.6: Self-diffraction FROG](image)

Figure 3.6: Self-diffraction FROG: The pulse is split in a beam splitter (BS) and one part is delayed in a delay stage that scans the temporal overlap of the pulses in the nonlinear crystal. The third order susceptibility $\chi^{(3)}$ enables the effect of self-diffraction. The spectrometer records the spectral intensity.

Part of the light is diffracted by the grating. The mixed frequency terms are emitted under an angle to fulfill the phase matching $\tilde{k}_{SIG} = 2\tilde{k}_1 - \tilde{k}_2$ where the indices can be exchanged as the fields are copies of each other. An imprinted limitation is the self-diffraction itself, as it is not phase matched for crossed beams. Therefore sufficient thin materials have to be used, that results especially with the weak third order effect in a low efficiency.

Polarization gate PG-FROG uses again the Kerr effect in form of the induced birefringence according to $|n_o - n_e| \propto E^2$. Two polarizers in the signal arm as shown in figure 3.7 ensure that the beam is totally blocked when the other pulse copy, called probe beam, is out of delay.
Figure 3.7: Polarization Gate FROG: The pulse is split in a beam splitter (BS) and one part is delayed in a delay stage. Two perpendicular polarizers ensure a perfect attenuation of one pulse copy (here called signal) at first. A $\lambda/2$ wave plate (WP) rotates the one by $\pm 45^\circ$ respectively. When both pulses are temporarily overlapped, the Kerr effect acts like a wave plate and allows some light to be transmitted to the spectrometer.

When the probe pulse is overlapped in the crystal in time and space, the induced birefringence turns the polarization of the signal and an intensity after the second polarizer can be measured that depends on the temporal overlap of the signals. It has to be ensured, that the polarizers are of high quality to guarantee a low floor on the signal.

Second harmonic generation SHG-FROG [34] is widely used due to its higher efficiency that originates in the lower order non-linearity employed, compared to other FROG schemes. Furthermore the setup shown in figure 3.8 is background free and ensuring a high dynamic range. The optical setup and likewise the signal at the output is identical to the intensity autocorrelation with the photo diode replaced by a spectrometer.

Figure 3.8: Second harmonic FROG: The pulse is split in a beam splitter (BS) and one part is delayed in a delay stage to scan the overlap in the second order nonlinear crystal ($\chi^{(2)}$). The sum frequency signal is acquired with a spectrometer and the fundamental is blocked by an iris.

An integration over the spectrum gives the autocorrelation signal

$$M_{\text{time}}(\tau) \int d\omega \ S_{\text{SHG}}(\omega, \tau) \propto I_{\text{AC}}(\tau) \quad (3.2.3)$$

In contrast to third order FROG schemes the pulse serves directly as gate and the temporal symmetry yields symmetric spectrograms. With the loss of the direction in time, a cross check is needed to distinguish positive and negative dispersion. When the chirp of the pulse is not known a priori, a glass plate introduces (usually positive) dispersion and can
be used for a consistency check. A powerful consistency check is given by the marginals in time and frequency domain that can be calculated easily in the case of SHG FROG. Equation 3.2.3 is the marginal $M_{\text{time}}(\tau)$ in time domain and cross checks the FROG trace with the intensity autocorrelation. The frequency marginal

$$M_{\text{freq}}(\omega) = \int d\tau S_{\text{SHG}}(\omega, \tau) \propto |E_{\text{SHG}}(\omega)|^2$$  \hspace{1cm} (3.2.4)

can be computed simply from the spectrometer data. More than just a check, the frequency marginal is useful for a post correction of the spectral data in order to account for errors in the FROG trace [35]. It is a well known technique to multiply the FROG trace by the ratio with the frequency marginal but we want to emphasize that it should be done with care to force the FROG trace on the frequency marginal. Since the frequency marginal is time independent it should be clear that the systematic error to be corrected for must be also time independent.

All those FROG traces carry $N_{\text{time}} \times N_{\text{freq}}$ information about the pulse under test, where $N_{\text{time}}$ is the number of temporal delay steps and $N_{\text{freq}}$ is the number of pixels of the spectrometer. The final representation of the pulse in time domain is retrieved with an iterative algorithm based on the $N_{\text{time}} \times N_{\text{freq}}$ spectrogram. A number of algorithms were developed in order to reduce the computation time, converging to a single solution and improved SNR robustness. The common method of general projections [36] combines (1) the experimental FROG trace and (2) the FROG signal $E_{\text{SIG}}$. The description will be limited to the case of SHG-FROG. An initial guess of the electric field gives the input signal $E_{\text{SIG}}$ for the algorithm that approach a solution between the two constraints. Since the boundary conditions are settled in frequency and time domain, a Fourier transform is necessary for every half step when comparing the solution to one or the other. The calculation process follows the diagram in figure 3.9. Each round trip the iteration is guided by the error $Z$ between $E'_{\text{SIG}}$ and the current solution.

$$Z = \sum_{i,j}^{N} |E'_{\text{sig}}(t_i, \tau_j) - E(t_i)E(t_i - \tau_j)|^2.$$  \hspace{1cm} (3.2.5)

A widely accepted advantage of FROG is the FROG Error $G$ yielding a gauge for the reliability for the pulse reconstruction and derived pulse duration.

$$G = \sqrt{\frac{1}{N^2} \sum_{i,j}^{N} |I_{\text{FROG}}(\omega_i, \tau_i) - \mu I_{\text{measure}}(\omega_i, \tau_i)|^2}$$  \hspace{1cm} (3.2.6)

The parameter $\mu$ is adapted to obtain the lowest value for $G$. The error implies systematic errors in the optical setup, stray light and other experimental imperfections. The remaining error depends (1) on random noise that is not given in a calculated FROG trace. (2) The grid size $N$ of the FROG trace directly relates to $G$ and a cited FROG
Figure 3.9: The generalized projection algorithm to retrieve the spectral phase out of a FROG trace. The algorithm starts with an initial guess and is fed with the measured spectrogram. By "improving" the guess, the simulated spectrogram iteratively approaches the measured one.

error without at least the grid size is not very meaningful.
3.3 SPIDER

Based on spectral interferometry, Iaconis and Walmsley proposed the method of spectral phase interferometry for direct electric-field reconstruction (SPIDER) in 1998 [37]. An approach that captures the phase and amplitude of an electrical pulse independently and gives the possibility to optimize one or the other detection. From a measurement, the pulse reconstruction is performed in a direct way with no need for iterative algorithms. SPIDER is based on spectral interferometry where two beams are overlapped in space with a temporal delay on one pulse. Thinking of a monochromatic beam which would correspond to picking a single frequency in a broadband pulse, coherent and incoherent phase relation would lead to constructive and destructive interference. For broadband pulses, a spectrometer replaces the photo diode of figure 3.1 and captures all frequencies at once as sketched in figure 3.10.

![Figure 3.10: A Mach Zehnder interferometer enabling the detection of spectral fringes using a spectrometer at the output. The pulse is split with a beam splitter (BS) in signal field $E_S$ and reference field $E_R$ to distinguish them in the description but the names are interchangeable here. After recombination in a second BS, the signal is detected with a spectrometer.](image)

We term a reference field $E_R(t)$ and a signal field $E_S(t)$. Let us assume that the phase undergoes different dispersion and delay in the upper and lower interferometer arm. The spectrum at the output will show constructive and deconstructive interference after recombination at the second beam splitter and has a fringed shape. If the reference pulse would be known, the phase sensitive signal of the spectrometer could be used to retrieve the phase of the signal pulse. A longer optical path for the signal result in a delay $\tau$ and recombined with the reference in a beam splitter gives a signal

$$E_{IS}(t) = E_R(t) + E_S(t - \tau).$$

(3.3.1)

After detection by the spectrometer, the integration yields

$$I_{IS}(\omega, \tau) = \left| \int E_{SI}(t)e^{-i\omega t}dt \right|^2$$

$$= E_R(\omega)E_R^*(\omega) + E_S(\omega)E_S^*(\omega) + E_R(\omega)E_S^*(\omega)e^{i\omega \tau} + E_S(\omega)E_R^*(\omega)e^{-i\omega \tau}.$$  

(3.3.2)
The second, oscillating part contains the phase information of the electric field. The DC part and the AC part are isolated in the Fourier domain where we pick solely one side of the symmetric FFT without loss of information. In order to resolve the sideband, a suitable delay has to be adjusted to separate it from the DC part. An inverse FT of the sideband then contains the relative phase information in form of $E^*_R(\omega)E_S(\omega)e^{i\omega\tau}$. Taking the argument of the complex function

$$\Delta \phi(\omega) = \arg(E^*_R(\omega)E_S(\omega)e^{i\omega\tau}) = \phi_S(\omega) - \phi_R(\omega) + \omega\tau$$  \hspace{1cm} (3.3.3)

yields the relative phase between the signal and reference pulse up to a calibration $\omega\tau$. Note that the spectral phase is extracted from the interferogram without a full spectral measurement of the pulse. An independently taken spectrum completes the measurement. The only component missing to know about the phase of a pulse under test is the phase of the reference pulse that is usually not known a priori as it is split from the pulse under test itself. To overcome this problem, the spectral interferometry is extended by two almost single frequency pulses that upconvert the signal pulse in a nonlinear crystal to spectrally slightly sheared replicas of the signal to become the self-referencing SPIDER method.

In SPIDER, the signal is split in two copies with a variable delay to each other. A third pulse is generated from the signal or independently and synchronized with the pulse copies. The third pulse is strongly chirped in order to approach a narrowband cw beam for the temporal support of the pulse under test. Two frequency separated and independently generated narrowband pulses are possible as well. The pulses are focused in a nonlinear crystal where the broadband pulses are upconverted by almost single frequencies in an SFG process. When the shear frequency of the narrowband stretched pulses is chosen sufficiently small, both upconverted, still broadband pulses widely overlap in frequency and generate an interference pattern that depends on the initial delay $\tau$ between the fundamental pulse copies and their phase. The geometry is shown in figure 3.11.
The fields are given by the upconverted fundamental pulse copies, where we call the upconversion frequency $\omega_{up}$ and add a shear frequency $\Omega$ to the second slightly shifted one $\omega_{up} + \Omega$. The interferogram builds up similar to spectral interferometry as

$$
I_{SPIDER}(\omega - \omega_{up}) = |E_R(\omega - \omega_{up})|^2 + |E_K(\omega - \omega_{up} - \Omega)|^2 \\
+ |E(\omega - \omega_{up})|e^{i\phi(\omega - \omega_{up})}|E(\omega - \omega_{up} - \Omega)|e^{i\phi(\omega - \omega_{up} - \Omega)}e^{i\omega\tau} \quad (3.3.4)
$$

Where the argument of the sideband in the Fourier domain gives the phase $\Phi_{SPIDER}(\omega)$ of the SPIDER trace. It is assumed that the narrowband pulses add only a constant phase and do not create an additional chirp.

$$
\Phi_{SPIDER}(\omega) = \phi(\omega - \omega_{up} - \Omega) - \phi(\omega - \omega_{up}) - \omega\tau \quad (3.3.5)
$$

It is a crucial part of SPIDER to determine the delay $\tau$ in order to subtract the linear phase only and not the phase belonging to the pulse itself which is ensured when the sideband is clearly separated from the DC part in the Fourier domain. The phase $\Phi$ transforms into an absolute phase $\phi$ via the group delay $T_g = -\frac{d\phi}{d\omega}$ and the fundamental differential quotient.

$$
\frac{d\phi}{d\omega} = -\frac{\phi(\Delta \omega - \Omega) - \phi(\Delta \omega)}{\Omega} = \frac{d\Phi}{d\omega} \quad \leftrightarrow \quad \Phi(\omega) = -\Omega \frac{d\phi}{d\omega} \quad (3.3.6)
$$

Here, it became clear that the shear frequency $\Omega$ has to be known precisely and that it directly affects the phase, same as for the delay $\tau$. The final representation of the phase is obtained by integration

$$
\phi(\omega) = -\frac{1}{\Omega} \int_{\omega'}^{\omega} \Phi(\omega')d\omega' \quad (3.3.7)
$$
The conventional SPIDER apparatus has the ability to perform a full measurement on a single shot basis. Furthermore the phase retrieval is straightforward and based on highly efficient FFT. A crucial point in conventional SPIDER is the interferogram and the spectrometer itself. The Whittaker-Kotelnikow-Shannon (WKS) theorem tells us that a pulse of duration $\Delta T$ has to be sampled with a frequency of $\frac{2\pi}{\Delta T}$. The sampling frequency in a SPIDER setup is given by the spectral shear $\Omega$. In addition to the upper limit of the spectral shear, there is also a lower limit. The spectral shear is coupled via the second order dispersion to the time delay ($\Omega = \frac{\tau^2}{\phi(\tau)}$), where the time delay has to be kept sufficiently large to separate the DC from the AC part in equation 3.3.2 and the second order dispersion needs to stretch the pulse almost cw like for the duration of the test pulse. Numeric examples are given in [38] for a transform limit Gaussian pulse of $\Delta \tau \approx 50$ fs. In order to resolve the fringes with $5 - 10$ data points, the delay is fixed around $\tau \approx 1 - 2$ ps and a spectrometer resolution of 0.05 nm at $\lambda = 400$ nm is necessary. Scanning monochromators are able to deliver such a resolution but it is still demanding for today’s handy and easy-to-use spectrometers. The well established ocean optics spectrometers for example deliver a resolution of $\Delta \omega_{\text{pixel}} = \frac{900 \text{ nm}}{3648} \approx 0.25$ nm [39].

The generation of the stretched pulses can be done independently and gives the possibility to increase the power and therefore support the second order nonlinear process or set the center frequency to shift the interferogram to a wavelength range where an efficient detection is possible. Furthermore, the pulse under test can be left ”untouched” without the need of a beamsplitter as it is done in most FROG and the original SPIDER schemes. A variation called zero additional phase (ZAP) SPIDER is a techniques were no additional phase is added to the pulse under test are necessary to characterize ultrashort pulses below $\approx 10$ fs, particularly because the thinnest optics result in a non-negligible pulse broadening. In a ZAP configuration, there are two sheared copies of a strongly chirped or monochromatic pulse synchronized to the pulse under test instead of two replicas of the pulse under test synchronized with a chirped pulse. An overview of the most common SPIDER techniques is further given in [40].

A SPIDER derivate that overcomes the WKS theorem and also yields a two dimensional trace containing intuitive information about the phase of a pulse was introduced with spatially encoded arragement (SEA) SPIDER in 2005 [41]. The fringes of a SEA SPIDER trace are not generated in the frequency domain but with a noncollinear geometry in a spatial domain and recorded by a spatially resolving spectrometer using a 2D CCD camera as shown in figure 3.12.
Figure 3.12: SEA SPIDER: The Fresnel reflection from the wedge is transported to BBO only with mirrors. The transmission is used to prepare two massively in glass strechted and delayed pulses that are used for an upconversion in the BBO. The interfering SFG signals are recorded on an imaging spectrometer (CCD).

The angle between the spatially interfering sheared pulses and the pulse under test ensures the separation of the DC and AC part in the Fourier transform and an isolation of the phase dependent part. A filtering of the AC terms in the Fourier domain that contain the phase information describe the signal as [42]

$$I_{\text{SEA-SPIDER}}^{\text{filtered}} = |E(x, \omega - \omega_{up})|e^{i\phi(x, \omega - \omega_{up})}|E(x, \omega - \omega_{up} - \Omega)|e^{i\phi(x, \omega - \omega_{up} - \Omega)}e^{i\Delta k x}.$$.  

(3.3.8)

The difference between the transverse components of the propagation vectors of the upconverted pulses and the transversal coordinate $x$ make the difference to conventional SPIDER. The calibration of the phase $e^{i\Delta k x}$ in SEA SPIDER is done when zero shear is adjusted between the two stretched or filtered pulses. The dependence on the vertical axis is given via the convergence angle $\Delta k x$. The tilt between the upconverted pulses and the corresponding $\Delta k$ defines the appearance of the fringes on the 2D CCD sensor.
Two-dimensional spectral shearing interferometry (2DSI)

Two-dimensional spectral shearing interferometry (2DSI) \[5\] is a pulse characterization technique that follows the idea of spectrally sheared pulses from previous spectral interferometric techniques together with the second order nonlinearity. The measurement of the spectral phase by interference of two spectral sheared pulse replicas enables a direct phase retrieval based on Fast Fourier Transforms (FFT) and therefore avoids a computational intensive iterative retrieval that is inherent to the FROG techniques. The densely packed phase information of a SPIDER trace in the frequency domain and the need for a high performance spectrometer is circumvented in 2DSI by resolving the phase information in the pseudo temporal domain of a two-dimensional intensity diagram. Furthermore, the delicate calibration of the delay in a SPIDER setup is avoided within the 2DSI technique plus the raw two-dimensional diagram already gives a visually intuitive representation of the pulse. The 2DSI geometry preserves the pulse under test from additional distortions occurring in optics like beam splitters and solely uses mirrors to focus the pulse in a nonlinear crystal before the signal is recorded by a spectrometer.

It is within the scope of this thesis to set up a 2DSI measurement device, develop a code for data acquisition and pulse retrieval and test the device on broadband laser sources. Two narrowband spectrally sheared pulses are used for an upconversion of the pulse under test in a nonlinear crystal such as BBO, where one of the narrowband ancillae pulses is delayed by a variable delay stage. Identical frequency components interfere in dependence of the applied delay. We can already think of a perfectly compressed pulse when all frequencies have the same internal pulse position and constructive interference should happen at the same delay for all frequencies, vice versa for destructive interference. The generation of ancillae pulses is another degree of freedom in the 2DSI geometry. Originally they are produced from a split copy of the pulse under test by introducing a huge amount of dispersion (\(\approx 10 \text{ cm}\)) of fused silica and the strongly chirped pulse appears as a quasi cw pulse in the time window of the short pulse under test. Another method is presented in [43] using a 4-f pulse shaper geometry that filters the pulse spatially in the
Fourier domain. In principle, the ancillae out of the pulse shaper are completely free of chirp, while dispersed pulses will always show a minimal residual chirp that will pursue in a "chirped" upconversion.

The ancillae preparation can also be of independent origin, for example when the upconversion wavelength should be chosen to place the raw 2DSI data in a spectral range that is easily accessible to common spectrometer based on CCD’s. This is beneficial for infrared (IR) pulses where a wavelength of 3μm is mapped on 630 nm with an ancillae filtered from a Ti:Sa-laser around 800 nm.

The setup as we constructed it is shown in figure 4.1 and employs interference filters in each arm of a Michelson interferometer that ensures an easy experimental handling including a comfortable determination of the shear frequency and interferometric stability of the ancillae. The filtering approach was also proven in [44] to increase the performance of a spectral shearing interferometry setup.

![Diagram of the setup](image)

Figure 4.1: Setup of our Two-dimensional spectral shearing interferometry (2DSI) device. Input for the ancillae pulse that get spectrally filtered by interference filter (IF) and focuses with the pulse under test in the nonlinear crystal before the SFG signal is recorded by a spectrometer.

The interferogram that builds up from the interference of two slightly sheared pulses \( E(\omega) \) and \( E(\omega - \Omega) \) reads as

\[
I(\omega, \phi_{\text{delay}}) = |A(\omega) + A(\omega - \Omega)e^{i\phi_{\text{delay}}}|^2 \\
= |A(\omega)|^2 + |A(\omega - \Omega)|^2 + 2|A(\omega)A(\omega - \Omega)|\cos(\phi_{\text{delay}} + \Phi(\omega) - \Phi(\omega - \Omega)).
\]

(4.0.1)
Here, the frequency $\omega$ corresponds to the frequency range after upconversion and the delay introduced by the delay stage is implied in $\phi_{\text{delay}} = \omega \tau$. The phase information is hidden in the argument of the cosine function which expresses the differential of the group delay by implying the shear frequency $\Omega$

$$T_g = \frac{d\Phi}{d\omega} \approx \frac{\Delta \phi}{\Omega}.$$  \hspace{1cm} (4.0.2)

From this equation, the resolution of a measurement ultimately becomes clear to be the shear frequency. The shear frequency has to be chosen suitable to the pulse under test. Correspondingly, the phase is obtained from integrating the group delay. Following the Shannon Sampling Theorem, the shear frequency defines the maximum time window $\Delta T$ that can be captured correctly with a 2DSI measurement.

$$\Delta T \leq \frac{2\pi}{\Omega}$$ \hspace{1cm} (4.0.3)

Note that the temporal window needs to be much larger than a FWHM duration of the pulse. A time window that is too small aliases the remained energy excluded from the window into the calculation and leads to a false characterization [45]. Since the temporal shape is unknown a priori, a large time window should be used to start with. Measurements with decreasing shear frequencies that show convergence are a proof to sample the pulse correctly.

The filter for the ancillae generation are narrow bandwidth interference filter (Semrock) that can be tilted for a fine tuning of the filter wavelength. The transmission curves are shown in figure 4.2 A single filter shows a bandwidth at FWHM of 2.8 nm and two tuned filters with an angle difference of 3° transmit a pulse of 0.6 nm bandwidth maining a
transmission of 50 %. A single filter is chosen for low energy pulses, for instance from laser oscillators. High power sources can make use of two filters per ancilla to narrow down the bandwidth and possibly decrease the shear frequency.

4.1 MATLAB GUI

In order to make use of the fast pulse retrieval and online optimization of a laser source, an easy-to-use graphical user interface (GUI) is developed to take the data and automatically perform the pulse retrieval. We use MATLAB to control a piezo stage with a travel range of 100 μm and a resolution of 0.1 nm. The Physik Instrumente (PI) piezo controller exchanges the brand’s own CGS commands via a fast TCP connection with the computer. In MATLAB, the commands are translated using a library that can be downloaded from PI on request. At every start up, an auto calibration command is send. Furthermore we send positioning commands to the controller and receive high precision position read outs.

The chosen spectrometer is manufactured by Ocean Optics and the universal MATLAB instrument driver supports all necessary commands. During the work, both the USB2000 model with an optical resolution of 1.5 nm and the HR4000 with an optical resolution of < 1.0 nm worked with the GUI. The GUI is displayed in figure 4.3 The most important quantity for the retrieval is asked first in the upper right part of the GUI. The lower and upper ancillae wavelengths have to be typed in. They can be observed from any independent spectrum viewer or by using the small plot in the GUI. When the background is sufficiently low, a function is implemented to estimate the peak’s position automatically. The shear frequency defines the maximum time window that is captured correctly and displayed directly below. The fundamental spectrum of the pulse under test is usually taken independently and can be loaded in the lower axis while it is upconverted by the mean of the shear frequency. The upper figure displays the captured spectrum in real time. The integration time can be adjusted to the right side and the background is taken when pressing the button Background. As a reference spectrum the upconverted spectrum helps to ensure that the pulse is fully upconverted. The spectrometer data in the upper axis should show the same shape as the upconverted fundamental pulse. In figure 4.3 we see a measurement of a Ti:sapphire oscillator pulse that is nicely compressed and shows horizontal fringes in the 2DSI raw data. The measurement starts when the relevant entries to control the delay stage are done. To fine tune the temporal overlap to the fixed ancilla pulse, a starting point for the piezo stage can be set. The stage is moved freely by using the horizontal bar tap. A step size in μm translates to the temporal delay and the number of spectra per optical cycle. The number of steps together with the step size is giving the total amount of fringes. Empirically a number of <5 ensures a robust
Figure 4.3: The graphical user interface (GUI) written in MATLAB to control and evaluate the pulse characterization. The initial START button builds up the communication to an Ocean Optics spectrometer and the piezo stage. The wavelength range is set on the top and the ancillae are defined to the right where we call them Pump 1 and Pump 2. This enables the previously taken fundamental to be displayed over the upconverted frequency range (bottom axes) and to be compared to the measured upconverted spectrum (upper axes). Details for the measurement are set to the lower right with a bar to control the piezo position within 100 μm, number of spectra and integration time. The background has to be taken before a measurement will start by hitting perform 2DSI. All relevant data are packed and saved with the save command and the pulse retrieval starts with retrieval.

measurement and visually clear data. The measurement starts by pressing perform 2DSI. When the trace shows up in the middle, custom notes can be typed in the text window. The save command organises all information that are relevant for a full retrieval in a cell structure and saves it as .mat file with a time stamp. The file then includes the fundamental spectrum, the shear frequencies, custom notes, 2DSI data trace and spectral correction data for the spectrometers. The retrieval subsequently opens figures with the retrieved phase in the frequency domain and the pulse in time domain.

The phase retrieval is mainly following the equations 4.0.1 and 4.0.2 to calculate the group delay. The argument of the cosine is extracted using FFTs along the pseudo temporal domain. Since the measured data are real, the FFT is symmetric in the pseudo frequency domain. An isolation of the sidebands and extracting the phase at the frequency of the sideband reveals the desired phase information. The window of the FFT can be set manually, so that a signal and energy at the edges are avoided whereas a Hann filter is more convenient for an automatic retrieval. Figure 4.4 shows the FFTs from an artificial pulse retrieval where we perform the FFT from the raw data (a) and after the application of
a Hann filter (b). There are less high frequency components when using an appropriate windows and should improve the phase retrieval especially in the presence of noise. A

Figure 4.4: Comparison of Fourier transform without a window function (a) and with Hann window (b) giving less high frequency components.

spectrometer measures the intensity on a wavelength grid and has to be scaled to a constant frequency grid for ongoing calculations. It is especially important for the broadband fundamental spectrum to transform the intensity from a wavelength grid to a frequency grid correctly [47]

\[ I_\omega(\omega) = \frac{\lambda^2}{2\pi c} I_\lambda(\lambda)|_{\lambda = 2\pi/\omega}. \]

Since the spectrometer usually has a higher resolution than the sampling frequency given by the spectral shear, the Whittaker–Shannon interpolation formula which is basically a sinc-function that bandlimits a signal is applied on the 2DSI traces to avoid aliasing of higher frequency components in phase reconstruction. A concatenation of the group delay at the shear frequency returns the phase information. Finally the phase information is shifted back to the fundamental wavelength. Together with the fundamental spectrum, the electric field can be calculated in the time domain using a 1D FFT.

In order to cross check the retrieval, a typical 2DSI trace is simulated from a single pulse defined by an envelope, the carrier frequency and possibly a phase. Examples of such pulses are given in figure 4.5 with a comparison of the pulse by definition and the retrieved one. The simulation shows a perfect coincidence of both pulses, that proves the accuracy of the retrieval. Experimentally, the upconversion is up to the phase matching
Figure 4.5: 2DSI Simulations for a pulse (a) undergoing 3mm of BK7 glass, (b) purely -200fs\(^2\) of GDD and (c) 400fs\(^3\) of TOD. The initial defined pulse is plotted in black and the retrieved one in blue.

in the nonlinear crystal. The calculation for the right cut angle \(\theta\) is based on section and equation 2.7.9. The BBO shall match ancillae pulses around \(\lambda = 800\)nm and support pulses with wavelengths from 500 nm to 1000 nm in order to measure broadband oscillators and furthermore signals up to the IR(2100 nm) as part of the waveform synthesizer [4]. For a broadband phase matching a type II phase matching is beneficial as pointed out in [48] with the broadband signal polarized along the ordinary axis and the ancillae along the extraordinary axis of the crystal. The intensity of the sum frequency signal is given in figure 4.6. The plots confirm that a type II phase matching is advantageous and a noncollinear angle can extend the conversion efficiency from the visible to the near IR with constant efficiency.
Figure 4.6: Phase matching for BBO calculated for application in the 2DSI setup. All calculations are done for a thickness of 20 μm. (a) Type I phase matching in a collinear manner (b) Type II phase matching in a collinear manner (c) Type II phase matching in a noncollinear manner (d) Type II collinear for IR. The red line indicates the typical wavelength of 800 nm that is usually used for the ancillae generation from a Ti:sapphire laser and the cuts (e) show the conversion efficiency respectively.
4.2 Ti:sapphire Oscillator

Laser oscillators using broadband gain media and the mechanism of Kerr-lens mode locking (KLM) were first shown in [49]. An optimization of the cavity dispersion and the output coupler topped the pulse duration down to sub 5 fs pulses directly from a stable solid state laser device that corresponds to sub two cycle optical pulses [50].

To show the capabilities of the 2DSI technique, a Ti:sapphire laser oscillator serves as a source and its pulses are characterized. The oscillator is equipped with double chirped mirrors and BaF$_2$ wedges for the dispersion management. A phase matched output coupler (PMOC) enhances the output spectrum on the long wavelength side resulting in sub 4 fs Fourier transform limit pulses.

The oscillator is pumped by 5.1 W and supports 2.5 nJ pulses at 80 MHz repetition rate. The challenges for a pulse characterization are (1) phase matching the broad bandwidth and (2) generating a sufficiently large second harmonic signal that can be detected by a spectrometer. The setup shown in figure 4.7 is using a parabolic mirror with a focal length of 1 inch to focus the pulse under test and the ancillae in the BBO crystal.

Figure 4.7: 2DSI setup to measure the Ti:sapphire oscillator. The oscillator sits to the left side of the frame. Subsequently the external DCMs and BaF$_2$ compress the pulse. In order to match the type II phase matching the polarization is rotated by 90° in a periscope before the pulse is split with a wedge. The transmission is used to prepare the ancillae pulses after another rotation of the polarization in $\lambda/2$ wave plate.
The pulse is split by a wedge after turning the polarization in a periscope. Roughly 2% of the pulse are reflected as pulse under test and the transmission at the wedge is used for the creation of the ancillae pulses. We measured that $\approx 1\%$ of the oscillator output energy is left to each of the ancillae pulses after a double transmission of the interference filter. Therefore the thickness of the BBO should be chosen carefully in such a low intensity regime in order to weigh up bandwidth and efficiency of the second order process. The thickness of the BBO used for the measurements is 10 $\mu$m or 20 $\mu$m while measurements with the thin BBO take several minutes due to the long integration times of the spectrometer required for a sufficient SNR. We observe no bandwidth limitation even with the thicker crystal and use it for the following measurements. The cutting angle is $\theta = 44^\circ$. The 2DSI fringes in figure 4.8 show a good contrast and the calculated group delay matches the experimental trace. The measurement was taken with a shear frequency of $\Omega = 3.6$ THz and the phase was fine tuned under rotation of the BaF$_2$ plates to obtain an almost flat spectral phase and the shortest pulse duration. The phase and spectral intensity are shown in figure 4.9. The pulse duration at FWHM is calculated to 4.2 fs where the transform limit (TL) is 4 fs. The phase at the wings of the spectrum lift off when the sum frequency signal is too weak and the noise is detrimental to the phase detection. The phase at wavelength smaller than $\approx 600$ nm: greater than $\approx 1200$ nm is basically made up out of noise and not meaningful. Since the intensity of the fundamental goes down to zero in the same spectral region, the noisy part of the phase does not affect in the complex representation of the pulse.

Figure 4.8: 2DSI raw data trace with calculated group delay $\tau_G$. The group delay shows perfect coincidence with the small modulation of the fringes.
The phase and FWHM duration are proven by repeating measurements and multiple shear frequencies. With a limited frequency range of the interference filters it was not possible to exhaust the time window as given in equation 4.0.3 so that we basically obtained the same result for all measurements. A consistency check is done to prove that the idea of

![Figure 4.9: Pulse characterisation of a Ti:sapphire laser oscillator. (a) The spectral intensity and the spectral phase. (b) The pulse in time domain with a Fourier transform limit (TL) FWHM duration of 4fs. The final compressed pulse yields a FWHM duration of 4.2fs.](image)

2DSI and the retrieval code works properly. Therefore a material of known dispersion is put in the beam path. The measurement of the phase without and with 2 mm of fused silica is shown in figure 4.10. The frequencies out of the grey area are not covered by the fundamental pulse. The phase difference of the experimental traces matches the theoretical trace that is calculated from the Sellmeier equation.
Figure 4.10: Experimental verification of our 2DSI setup and retrieval code by a measurement of the dispersion in a 2mm fused silica plate. The difference matches the black trace that is calculated from the Sellmeier equation.

4.3 IR OPA

Apart from implication of the 2DSI technique, the work of this thesis contributed to the construction of a broadband optical parametric amplifier in the infrared regime (IR-OPA). The IR-OPA covers the wavelength range $\approx (1200-2100)$ nm and represents the long wavelength part in the waveform synthesizer project [4]. The amplification of broadband optical pulses employs the rules of nonlinear optics presented in former chapters. Those will be extended by the role of the group velocity that is a crucial factor to maintain the amplification over a long distance in the nonlinear crystal and a broadband amplification of a signal. We redefine the involved fields as $\vec{E}_P, \vec{E}_S, \vec{E}_I$, for pump, signal and idler waves and follow [24].

$$\vec{E}_{P,S,I}(z,t) = \frac{1}{2} \left[ E_{P,S,I}(z) e^{i(\omega_{P,S,I}t-k_{P,S,I}z)} + c.c. \right]$$  \hspace{1cm} (4.3.1)

The frequencies of those pulses have to maintain the energy conservation $\omega_P - \omega_S - \omega_I = 0$. From the nonlinear wave equation under the assumption of no free currents and charges in time domain

$$\left( \Delta - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \vec{E}(\vec{r},t) = \mu_0 \frac{\partial^2}{\partial t^2} \left( \vec{P}_{IN}(\vec{r},t) + \vec{P}_{NL}(\vec{r},t) \right)$$  \hspace{1cm} (4.3.2)
the three coupled equations for pump, signal and idler waves are derived. The nonlinear polarizations are at the frequencies of the incident waves according to equation 2.7.1:

\[
P_{NL,S}(z,t) = \frac{\varepsilon_0}{4} \chi^{(2)} A_P(z) A_I^*(z, t) e^{i(\omega_p - \omega_S) t - (k_p - k_I)z} \\
P_{NL,I}(z,t) = \frac{\varepsilon_0}{4} \chi^{(2)} A_P(z) A_S^*(z, t) e^{i(\omega_p - \omega_I) t - (k_p - k_S)z} \\
P_{NL,P}(z,t) = \frac{\varepsilon_0}{4} \chi^{(2)} A_I(z) A_S(z, t) e^{i(\omega_I + \omega_P) t - (k_I + k_P)z}
\]  

(4.3.3)

The second order temporal derivative of the envelope becomes negligible with the SVEA

\[
\left| \frac{\partial^2 A}{\partial z^2} \right| \ll \left| 2\omega \frac{\partial A}{\partial z} \right| \ll |\omega^2 A| 
\]  

(4.3.4)

and we obtain for the second order derivative

\[
\frac{\partial^2 P_{NL,S}}{\partial t^2} = -2\omega_S^2 \chi^{(2)} A_P(z, t) A_I^*(z, t) e^{i((\omega_p - \omega_S) t - (k_p - k_I)z)} \\
\frac{\partial^2 P_{NL,I}}{\partial t^2} = -2\omega_I^2 \chi^{(2)} A_P(z, t) A_S^*(z, t) e^{i((\omega_p - \omega_I) t - (k_p - k_S)z)} \\
\frac{\partial^2 P_{NL,P}}{\partial t^2} = -2\omega_P^2 \chi^{(2)} A_I(z, t) A_S(z, t) e^{i((\omega_p + \omega_P) t - (k_I + k_p)z)} 
\]  

(4.3.5)

Placing those equations in the nonlinear wave equation yields the coupled equations for envelopes of three interacting waves in a nonlinear medium of second order.

\[
\frac{\partial A_S}{\partial z} = -i \frac{\omega_S d_{eff}}{n_S c_0} A_I^* A_P e^{-i\Delta k z} \\
\frac{\partial A_I}{\partial z} = -i \frac{\omega_I d_{eff}}{n_I c_0} A_S^* A_P e^{-i\Delta k z} \\
\frac{\partial A_P}{\partial z} = -i \frac{\omega_P d_{eff}}{n_P c_0} A_I A_S e^{i\Delta k z} 
\]  

(4.3.6)

Here, the \(d_{eff}\) replaces the nonlinear susceptibility to reduce the quantities of nonlinear coefficients. Furthermore \(\Delta k = k_P - k_S - k_I\) is the phase mismatch between the interacting fields. In terms of cw beams, the temporal and therefore spatial overlap is given in any condition. This is different for optical pulses travelling with the group velocity of \(v_g = d\omega/dk\). Those optical pulses need to be overlapped precisely and the overlap will change due to different group velocities for different wavelength in the nonlinear crystal.
The propagation of the pulse envelope is included as

\[
\frac{\partial A_S}{\partial z} + \frac{1}{v_{g,S}} \frac{\partial A_S}{\partial t} = -i \frac{\omega_{Seff}}{n_{c_0}} A_S^* A e^{-i \Delta k z} \\
\frac{\partial A_I}{\partial z} + \frac{1}{v_{g,I}} \frac{\partial A_I}{\partial t} = -i \frac{\omega_{Ieff}}{n_{c_0}} A_S^* A e^{-i \Delta k z} \\
\frac{\partial A_P}{\partial z} + \frac{1}{v_{g,P}} \frac{\partial A_P}{\partial t} = -i \frac{\omega_{Peff}}{n_{c_0}} A_I A_S e^{i \Delta k z}
\]

and we define the retarded time with respect to the pump pulse \((t' = t - z/v_{g,P})\).

\[
\frac{\partial A_S}{\partial z} + \left( \frac{1}{v_{g,S}} - \frac{1}{v_{g,P}} \right) \frac{\partial A_S}{\partial t'} = -i \frac{\omega_{Seff}}{n_{c_0}} A_S^* A e^{-i \Delta k z} \\
\frac{\partial A_I}{\partial z} + \left( \frac{1}{v_{g,I}} - \frac{1}{v_{g,P}} \right) \frac{\partial A_I}{\partial t'} = -i \frac{\omega_{Ieff}}{n_{c_0}} A_S^* A e^{-i \Delta k z} \\
\frac{\partial A_P}{\partial z} = -i \frac{\omega_{Peff}}{n_{c_0}} A_I A_S e^{i \Delta k z}
\]

The coupled partial differential equations underlie approximations such as neglecting second and third order effects but nevertheless an analytical approach will reveal insight in the bandwidth limitation of the parametric amplification. We want to point out the role of the group velocity mismatch (GVM) between pump and signal, respectively pump and idler \((\delta_{(S,I)} p = 1/v_{g,S,I} - 1/v_{g,P})\) that is captured in these equations. The pulse splitting length as defined in [24]

\[
l_{(S,I)p} = \frac{\tau_p}{\delta_{(S,I)p}}
\]

gives the maximal length of interaction between pump and signal, respectively idler pulse. The shorter the duration of the pump pulse \(\tau_p\) the shorter is the range of interaction and amplification in general. The splitting length between pump and signal directly relates to the amplification. Despite the need of phase matching to fulfill the momentum conservation discussed in section 2.7 the group velocity mismatch limits the bandwidth of the signal to be amplified. In a common configuration, the pump is a narrowband pulse with a duration of \(\tau_p \approx 100\) fs whereas signal and idler pulses aim to be broadband. A higher frequency within the signal \(\omega_S + \Delta \omega\) corresponds to a lower frequency in the idler pulse \(\omega_I - \Delta \omega\) and vice versa with respect to their central frequencies due to energy conservation. The resulting phase mismatch

\[
\Delta k = k(\omega_p) - k(\omega_S + \Delta \omega) - k(\omega_I - \Delta \omega)
\]
writes in first order to
\[
\Delta k = k(\omega_p) - \left( k(\omega_I) + \frac{\partial k}{\partial \omega_I} \Delta \omega \right) - \left( k(\omega_S) - \frac{\partial k}{\partial \omega_S} \Delta \omega \right)
\]
\[= \left( \frac{1}{v_{g,I}} - \frac{1}{v_{g,S}} \right) \Delta \omega \]  \hfill (4.3.11)

which equals the GVM. To understand how the GVM acts on the amplification of the signal pulse, the pulse behavior in equation 4.3.8 again is neglected and a second derivation of the signal and inserting the expressions for idler and pump gives

\[
\frac{\partial^2 E_S}{\partial z^2} = \frac{\omega_S \omega_I d_{eff}^2}{n_S n_I c_0^2} |E_p|^2 E_I \Delta k \frac{\omega_I d_{eff}^2}{n_I c_0} E_S^* E_I e^{-i\Delta k z}
\]
\[= \Gamma^2 E_S - i\Delta k \frac{\partial E_S}{\partial z}. \]  \hfill (4.3.12)

A solution is given below when a signal seed is given \( E_S(0) = E_{seed} \) and there is no incident idler field \( E_I = 0 \) to

\[
I_S(L) = I_{seed} + I_{seed} \frac{\Gamma^2}{\Gamma^2 - \Delta k/2} \sinh^2(\sqrt{\Gamma^2 - \Delta k/2L})
\]
\[= I_{seed} \left[ 1 + \frac{\Gamma^2}{g^2} \sinh^2(gL) \right] \]  \hfill (4.3.13)

with the gain \( g \). Here, a phase mismatch translates to the intensity of the signal that builds up over the length \( L \) of the crystal. In the regime of large gain \((gL \gg 1)\) the intensity becomes

\[
I_S(L) = I_{seed} \left( \frac{\Gamma}{g} \right)^2 e^{2gL}
\]  \hfill (4.3.14)

and the total parametric gain \( G \) of the signal becomes

\[
G = \frac{I_S(L)}{I_{seed}} = \left( \frac{\Gamma}{g} \right)^2 e^{2gL}. \]  \hfill (4.3.15)

The parametric gain shows an exponential growth when perfectly phase matched and scales with the square of the pump intensity but this model in the undepleted pump regime has its limits when we want to describe high intensity and high efficiency. Looking a few steps back at the gain \( g \), the incorporation of the phase mismatch \( \Delta k \) in equation 4.3.13 subsequently gives a bandwidth limitation in the parametric gain \( G \) that started from a mismatch of the group velocities from signal and idler pulse. The limitation on the bandwidth tells us that matching the group velocities of signal and idler is necessary to keep a broad amplification. This can be done by frequency matching of signal and idler resulting in a degenerate OPA (DOPA) or a noncollinear (NOPA) geometry by introducing a longer path for the faster travelling pulse. The angle to match the group velocities.
is governed by a projection of the wave vectors with respect to the direction of the signal, similar to what we did for noncollinear birefringent phase matching in equation 2.7.15.

![Figure 4.11: Group velocity matching in a noncollinear OPA (NOPA) with the pump pulse (blue), the signal (green) and the idler (red).](image)

The projection of the noncollinear wave vectors on perpendicular and parallel direction using the trigonometric relations gives the phase mismatch to

\[
\Delta k_\parallel = k_P \cos(\alpha) - k_S - k_I \cos(\Omega) \\
\Delta k_\perp = k_P \sin(\alpha) - k_I \sin(\Omega)
\]

that should be matched to equal zero. For broadband pulses we introduce again the frequency dependence where we have to include it for the noncollinear angle \(\Omega(\omega)\), too as it changes with the frequency of the signal. A Taylor expansion and the small angle approximation for the angle \(\alpha\) leads to

\[
\Delta k_\parallel = -\frac{\partial k_S}{\partial \omega} \Delta \omega + k_I \cos(\Omega) \frac{\partial \Omega}{\partial \omega_I} \Delta \omega + k_I \cos(\Omega) \frac{\partial \Omega}{\partial \omega_I} \Delta \omega = 0 \\
\Delta k_\perp = \frac{\partial k_I}{\partial \omega_I} \sin(\Omega) \Delta \omega + k_I \cos(\Omega) \frac{\partial \Omega}{\partial \omega_I} \Delta \omega = 0.
\]

By multiplying the first equation with \(\cos(\Omega)\) and the second by \(\sin(\Omega)\), the equations are merged to

\[
v_{g,S} = v_{g,I} \cos(\Omega)
\]

where we used \(\frac{\partial k}{\partial \omega}\) being the group velocity.

This gives the condition for a broadband amplification when the group velocity of the signal matched the projected group velocity of the idler. The noncollinear angle will separate the beams slowly but negligible, especially because the strong pump does not need to be focussed tightly and the idler usually covers the signal completely.

The NOPA of interest in the following section of this thesis is part of a waveform synthesizer [4] and accounts for the long wavelength side of the overall spectrum. Broadband amplification in the IR regime was shown in [52] resulting in a pulse duration of only 8.5fs when compressed.

A Ti:sapphire laser generates pulses of \(\approx 130\) fs duration and a fraction of the energy \(0.3\)
mJ) is fed to the OPA as pump pulse. With another fraction of the Ti:sapphire laser, a CEP stable pulse is obtained as an idler from another OPA at λ=1020 nm that is used to generate a CEP stable white light continuum. We use a f=125 mm plano-convex lens to focus ≈ 6 µJ in a 3 mm thick sapphire plate. The scheme of the NOPA is drawn in figure 4.12 with a photograph of the WLC to the lower side and the amplified signal with remaining pump beam and idler in the upper right corner. Sapphire is chosen over YAG because of the enhanced bandwidth in the IR compared to YAG (see figure 2.16). The thickness of the sapphire is traded between a thin plate for a limited accumulation of spectral phase and a thicker plate for enhanced SPM and increased bandwidth. A 2 mm plate showed a sufficient bandwidth but broke down several times during operation of the NOPA, so that a thickness of 3mm is used in the final configuration. Since sapphire is slightly birefringent, it should be specified as ’z-cut’ or zero degree when the optical axis is perpendicular to the plane of the window. The recollimated WLC is send as a signal to the NOPA. The first stage is employing a 2 mm thick BBO matched for type I phase matching at θ = 20.1° to amplify the signal.

Tuning of the nonlinear crystal and the noncollinear angle between pump and signal brings an amplified spectrum that can be tuned to various shapes and bandwidths. Some exemplarily spectra are presented in figure 4.13. Not only the spectrum, but also the beam profile needs to maintain a good quality after amplification to re-amplify the pulse in the second stage and propagate the pulse to the stage of synthesis and compression. Therefore, the spectrum is tuned to a continuous shape with only moderate slopes and with a mild attenuation of the amplification in the long wavelength part. The IR part
will be emphasized more in the second stage of the OPA. In this way, we obtained a sufficiently broad spectrum and maintained a good beam profile.

![Graph](image)

Figure 4.13: Spectra of the IR OPA for different noncollinear angles and rotation tuning of the crystal. The angle differences between the spectra no. 1, 2, 3 are very small and not measured precisely. With larger angles ($\approx 1.5^\circ$) the spectrum tends to spectrum no. 2 and smaller noncollinear angles ($\approx 1^\circ$) results in less amplification of the redder part as shown for spectrum no. 3.

This IR OPA gave the possibility to test the 2DSI setup in the IR and to characterize the first stage of the NOPA. Another fraction of the Ti:sapphire laser is used to generate the ancillae pulses independently from the pulse under test. The fundamental spectrum is measured with an Ocean Optics NIR spectrometer. Since the 2DSI by principle is upconverting the light by the frequency of the ancillae, the 2DSI signal is accessible with a high resolution spectrometer in the visible.

The fundamental pump wavelengths of the WL generation and the amplification are attenuated with a dichroic mirror that low passes only the frequencies of interest (>1200nm). The raw data trace is given in figure 4.14 with the retrieved group delay on top. The measurement itself is working fast. With plenty of energy available from a big Ti:sapphire for the ancillae preparation, the integration time of the spectrometer can be kept to a few milliseconds and a full measurement and retrieval is done in half a minute. Furthermore the retrieved group delay coincides well with the raw 2DSI trace.
Figure 4.14: Calculated GD on the raw 2DSI trace from the NOPA. Again, the retrieved group delay perfectly matches the raw data fringes.

The retrieved spectral phase is plotted in figure 4.15. The measurement reveals some phase modulation on the short wavelength side but a smooth increasing positive phase to longer wavelength. The rather smooth phase behaviour show basically second and third

order dispersion and should be compressible straightforward. This manifests in the time domain given in figure 4.16 when we compare the shape to the theoretical curves in table 2.3. The FWHM duration is calculated to 8.2 fs in the Fourier limit and 15 fs for the uncompressed pulse. A compression of the pulse with DCM’s is planned together with
Figure 4.16: Retrieved pulse in time domain from the first stage IR NOPA. The Fourier transform limit (TL) FWHM duration is 8.2fs. The measured duration for the uncompressed pulse is 15fs.

investigation in the second stage in order to obtain a high energy, short IR pulse from this NOPA.
4.4 Discussion

The barrier of $\tau=10$ fs FWHM pulse duration is commonly cited as a next level for ultrashort pulse measurements. The broad bandwidth of sub 10 fs pulses encounters problems in the frequency mixing within the nonlinear crystal when the full spectrum of the pulse can not be phase matched perfectly with constant efficiency. Furthermore, such broadband pulses do not maintain a smooth and symmetric shape of secant hyperbolic or any other analytical nature and therefore introduce systematic errors in interferometric autocorrelation techniques. The article [53] discusses the problems of ultrashort pulses with respect to SHG FROG. Although systematic errors arise at ultrashort pulse durations, the powerful consistency checks of FROG give the possibility for a correction during the pulse retrieval. The signal of third harmonic FROG geometries show impurities of Raman ringing [54], a scattering effect where optical phonons give a non instantaneous response to the optical field. It is of experimental interest to maintain a high conversion efficiency from the pulse under test to the FROG signal for a robust and fast detection. Here, second order geometries show an advantage over third order FROG setups. Furthermore, the conversion to the second harmonic frequency range separates the signal from the fundamental and lowers the background in the measurement. The same argument applies for the technique of SPIDER and our 2DSI scheme. When dealing with pulses in the IR, the upconversion shifts the spectrum in a range where high efficiency and high resolution semiconductor detectors are available. This facilitates the measurement in the experiment. Especially in geometries like SEA SPIDER or ZAP SPIDER and in our 2DSI device, the frequency of the narrow bandwidth ancillae can be chosen freely to match the spectrum of the signal with the range of a spectrometer. Additionally the sum frequency generation of a broadband PUT with a narrowband ancilla has a reduced bandwidth compared to second harmonic generation of a broadband PUT. The constant intensity of an ancilla is beneficial, whereas maxima in a SHG are emphasized quadratically and exhaust the dynamic range of a spectrometer even more.

An advantage of FROG is the moderate demand on the spectrometer resolution. An oversampling as discussed in the section of SPIDER is not necessary for a correct retrieval of the temporal structure in a FROG apparatus. Here, the temporal delay is mapped on its own temporal axis that gives the 2d FROG spectrogram. The same applies for the 2DSI method where is phase information is encoded in a pseudo time domain and the spectrometer resolution does not need to be high in particular.

The integrated retrieval of spectral intensity and spectral phase of a pulse in the FROG algorithm suffers from the filtering behaviour of the nonlinear crystal when phase matching limitations arise for broadband pulses. The spectral intensities might be suppressed especially in the wings and without correction, the FROG algorithm ”sees” a pulse with narrower bandwidth. This results in an overestimation of the duration or convergence
problems of the retrieval algorithm and can be corrected by frequency marginals calculated for SHG FROG. The same limitation is less pronounced in SPIDER and 2DSI. There, the spectral intensity is captured independently and only the phase information is gained within the second order nonlinear process. For sure we still need to phase match the whole bandwidth as good as possible but a reduction in the wings is not detrimental as long as the contrast of the fringes is detectable by an FFT. You can also think about taking separate measurements in the case of ultrabroadband sources and stitching the phase numerically.

Noncollinear geometries in the majority of FROG and SPIDER measurements bring the separation of the remaining fundamental pulse copies and second harmonic signal by default. In the case of autocorrelation methods and FROG, with a gate pulse as short as the signal, the noncollinear geometry inconveniently lowers the temporal resolution of the measurement. This temporal smearing originates from a geometrical smearing in the nonlinear crystal, also called pancake effect and sketched in figure 4.17. The crossing angle and the finite beam width increase the measured pulse duration $\tau_{mess}$ due to an increased overlap resulting in a variation of $\delta t$.

$$\tau_{mess}^2 = \tau_{PUT}^2 + \delta t^2.$$  \hspace{1cm} (4.4.1)

The distance over which the crossed beams generate a second harmonic signal gives $\delta t$

$$\delta t = \frac{\sin(\gamma)d_{foc}n}{c}.$$  \hspace{1cm} (4.4.2)

with the beam diameter in the focus $d_{foc}$ and the beam crossing angle $\gamma$. The variables are described in figure 4.17a.

Figure 4.17: A visualization of the geometric smearing effect. The pulses are separated by $\Delta y$ and focused with an angle of $\gamma$. The beam diameter $d$ and the temporal duration $\tau_{PUT}$ are indicated by small arrows. The sum frequency signal (violet arrow) is sent for detection. (a) The noncollinear SHG FROG geometry with a short pulse and its copy serving as a gate pulse. The noncollinear overlap in the nonlinear crystal generates a second harmonic signal over a longer distance and for a longer time. (b) The noncollinear overlap in a 2DSI setup where the stretched ancillae are of almost single frequency for the temporal support of the PUT and no temporal variation occurs. Adapted from [55], [56]

In methods like SPIDER and 2DSI, the stretched pulse used to upconvert the PUT is of
almost single frequency with respect to the duration of the PUT and the retrieval algorithm does not depend on a gating characteristic of the second pulse so that geometric smearing does not occur in our 2DSI apparatus.

Ultrashort pulses are usually measured over many cycles. Depending on the pulse energy, the repetition rate and the bandwidth of a laser it is technically demanding to capture the spectrum of a single optical pulse and it is even more complicated to perform a full pulse characterization including the phase information. In principle the technique of SPIIDER is capable of single shot measurements but the required integration times are often too long for the majority of sources. The technique of FROG and 2DSI with multiple discrete delay steps are inherently multi-shot techniques. Pulses from mode locked lasers are supposed to be stable with negligible fluctuation from shot to shot. This is harder to state for sources like hollow fibres and OPAs where longer distances, pointing issues and intensity fluctuations are involved. Simulations have shown that a superposition of random pulses in a characterization technique like FROG or 2DSI yields in most cases a flat spectral phase [57] where the variations are averaged out. Concluding that the acquisition of a train of pulses ends in a shorter temporal estimation of the pulse compared to the one of a single pulse, most pulse characterization techniques yield too short pulse durations for an unstable train of pulses. This is called the coherent artifact, where only the coherent part of a pulse train is measured correctly. Up to now, the question of stability within a train of pulses can be addressed only in an indirect way with multi shot techniques. With pulses of random phase the contrast of the interference shrinks and for the technique of FROG, the FROG error $G$ increases. In a 2DSI trace the coherent artifact expresses in a reduced fringe visibility and a washed out deconstructive interference.

Nevertheless, these indicators are rather weak in a real experiment because a tiny misalignment of the beams shows similar symptoms. Up to now, we do not know about any technique that precisely displays the stability of a pulse train. Current techniques can only give an indirect hint on the pulse to pulse and therefore phase to phase stability.

Judging from the experience in the lab and the presented measurements, we find the technique of 2DSI to combine the interferometric accuracy of previous SPIIDER methods with lower demands on the spectrometer resolution and delay calibration. Solely the inherent multi shot requirement might be a limiting factor for very fast data acquisition but on the other hand enables a two dimensional raw data trace that is directly interpretable and indicates the validity of a measurement.
Outlook

The work in this thesis showed that our 2DSI geometry is capable to fully characterize ultrashort laser pulses as short as 4 fs from low energy in the visible spectrum to high energy in the IR. With the same set of spectrometers, a fully automated MATLAB GUI and corresponding retrieval algorithm we consider the setup as highly flexible.

It is planned to use this geometry for ongoing characterization in a synthesizer project conducted at CFEL. We assume that the broad bandwidth of very thin BBO crystals down to 5 μm together with high energy amplified pulses allows us to capture the whole synthesized pulse. The bandwidth will stretch over three broadband OPA’s from 500 nm to 2 μm. Finally a 2DSI setup might be attached permanently to monitor the pulses. An optimization towards a fast pulse retrieval results in update times less than a second. When some leakage of a mirror is used to feed the 2DSI device, experiments with the synthesized pulse on HHG or spectroscopy can be performed simultaneously and a correlation of driver pulse and its excitation can be interpreted.

For an even faster acquisition, the 2DSI scheme might be adapted to a geometry like ZAP SPIDER [58] with slight modification on the signal detection. There, the phase information is imprinted on a single spectrum that is modulated by interference as described in section 3.3. Due to the higher demands on the spectral resolution, the Ocean Optics spectrometer will have to be replaced by a more sophisticated spectrograph. Moreover an investigation on the pulse to pulse stability is possible when we approach single shot capabilities. Note that with an adaption to classical SPIDER geometries comes the drawback of that technique, first of all the precise delay stability and subsequent errors. Nevertheless we think that the application of more than one pulse characterisation technique is worthwhile for the benefits of comparing and ensuring a measurement when it incorporates limited effort.

The simulations in figure 4.5 are based on a code that already incorporates the possibility for an incident pulse free of choice, e.g. a measured spectrum or fully retrieved pulse.
It simulates the full process of upconversion from a fundamental pulse with narrowband ancillae. An extension of the simulation allows to study effects on the upconversion process from residual bandwidth and phase of the ancillae. Furthermore the effects of noise or the coherent artifact will be investigated. Finally the work on a 2DSI error as it is given within the technique of FROG is feasible by comparing the measured trace with a simulated trace from the retrieved pulse.

Spatio-temporal details are still hidden to experimental characterization techniques. The transverse coordinate of a pulse can not be separated in most detection schemes. In 2DSI, the method of the ancillae makes it possible to maintain the spatio-temporal details throughout the upconversion process in principle. Therefore it should be waived to focus the ancillae pulses in order to maintain flat phase fronts and use a collinear propagation of ancillae and PUT to maintain the transversal coordinates from the PUT in the sum frequency signal. Different to our current setup, a rigorous imaging geometry has to be satisfied in order to map the signal generated in the BBO on an imaging spectrometer. This will yield the 2D traces for single points in both transverse coordinates of the pulse.

Coming back to our very first motivation and the way how photo cameras evolved from elaborate, huge devices with long exposure times to small and high resolution features in a smartphone that capture the light, we seek the same path for ultrashort pulse characterization techniques to capture ultrashort laser pulses in a standard procedure with high resolution and validity. The reliability of the 2DSI device presented in this thesis makes it possible to run a pulse characterization in parallel with experiments as for example HHG to ensure the quality of the driving pulse. Additionally the flexibility given by the experimental geometry and the creation of the ancillae pulses serves as a basis for adaption and extension of the apparatus to reveal further insights in ultrashort optical pulses.
Bibliography


(Fabian Scheiba)
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