Next Generation
Attosecond Technology

A thesis submitted to attain the degree of
DOCTOR OF SCIENCES of ETH ZURICH (Dr. sc. ETH Zurich)

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2020
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<tr>
<td>$\hat{A}$</td>
<td>electric field envelope in frequency domain</td>
</tr>
<tr>
<td>$A$</td>
<td>vector potential</td>
</tr>
<tr>
<td>$B$</td>
<td>B-integral</td>
</tr>
<tr>
<td>$b$</td>
<td>confocal parameter</td>
</tr>
<tr>
<td>$c$</td>
<td>speed of light in vacuum</td>
</tr>
<tr>
<td>$d_{eff}$</td>
<td>effective nonlinear coefficient</td>
</tr>
<tr>
<td>$E$</td>
<td>electric field</td>
</tr>
<tr>
<td>$E_K$</td>
<td>kinetic energy</td>
</tr>
<tr>
<td>$e$</td>
<td>electron charge</td>
</tr>
<tr>
<td>$\varepsilon_0$</td>
<td>vacuum permittivity</td>
</tr>
<tr>
<td>$F_L$</td>
<td>electric field amplitude</td>
</tr>
<tr>
<td>$I$</td>
<td>intensity</td>
</tr>
<tr>
<td>$I_p$</td>
<td>ionization potential</td>
</tr>
<tr>
<td>$I_{peak}$</td>
<td>peak intensity</td>
</tr>
<tr>
<td>$k$</td>
<td>wave number</td>
</tr>
</tbody>
</table>
\( \Delta k \)  wavevector mismatch

\( \lambda \)  wavelength

\( \Lambda \)  periodic poling period

\( m_e \)  electron mass

\( n \)  refractive index

\( N_{atm} \)  particle density at 1 bar pressure

\( n_2 \)  nonlinear refractive index

\( P \)  polarization

\( P_{pk} \)  peak power

\( p_i \)  momentum

\( r_e \)  classical electron radius

\( q \)  harmonic number

\( \tau \)  pulse duration

\( U_p \)  pondermotive potential

\( w_0 \)  beam radius

\( \chi \)  susceptibility tensor

\( \mu_0 \)  magnetic constant

\( \omega \)  angular frequency

\( \omega_0 \)  central angular frequency

\( \phi(\omega) \)  spectral phase

\( \theta_s \)  noncollinear angle

\( \gamma \)  Keldysh parameter

\( \Psi \)  wavefunction

\( \eta \)  ionization fraction
List of Symbols and Acronyms

Acronyms

ADK    Ammosov-Delone-Krainov
APPLN  apperiodically poled lithium niobate
AR     anti-reflection
AOM    acousto-optic modulator
AOPDF  acousto-optic programmable dispersive filter

CEP    carrier envelope phase
CEO    carrier-envelope offset
CPA    chirped pulse amplification
cw     continuous wave
CA     cycle averaged

DFG    difference frequency generation

EUV    extreme ultraviolet

FWHM   full-width at half maximum
FROG   frequency resolved optical gating
FOD    fourth order dispersion
FOPA   frequency domain optical parametric amplification
FW     fundamental wave
GD     group delay
GDD    group delay dispersion
GVM    group velocity mismatch
GRIIRA green-induced infrared absorption

HHG    high-harmonic generation
HITRAN high-resolution transmission molecular absorption database

IAP    isolated attosecond pulse
IR     infrared
IDFG   intrapulse difference frequency generation
<table>
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<th>Full Form</th>
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<tr>
<td>LBO</td>
<td>lithium triborate</td>
</tr>
<tr>
<td>LN</td>
<td>lithium niobate</td>
</tr>
<tr>
<td>LCoS</td>
<td>liquid crystal on silicon</td>
</tr>
<tr>
<td>NOPA</td>
<td>noncollinear optical parametric amplification</td>
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<tr>
<td>OPA</td>
<td>optical parametric amplification</td>
</tr>
<tr>
<td>OPCPA</td>
<td>optical parametric chirped pulse amplification</td>
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<tr>
<td>OPG</td>
<td>optical parametric generation</td>
</tr>
<tr>
<td>OPO</td>
<td>optical parametric oscillator</td>
</tr>
<tr>
<td>PPLN</td>
<td>periodically poled lithium niobate</td>
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<td>PPT</td>
<td>Perelomov-Popov-Terentev</td>
</tr>
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<td>PSD</td>
<td>position sensitive detector</td>
</tr>
<tr>
<td>QPM</td>
<td>quasi-phase-matching</td>
</tr>
<tr>
<td>TDS</td>
<td>time-delay stabilization</td>
</tr>
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<td>TOD</td>
<td>third-order dispersion</td>
</tr>
<tr>
<td>THG</td>
<td>third-harmonic generation</td>
</tr>
<tr>
<td>TDSE</td>
<td>time-dependent Schrödinger equation</td>
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<tr>
<td>UVFS</td>
<td>ultraviolet-grade fused silica</td>
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<tr>
<td>VMIS</td>
<td>velocity-map imaging spectrometer</td>
</tr>
<tr>
<td>WC</td>
<td>walk-off compensating</td>
</tr>
<tr>
<td>WNC</td>
<td>walk-off non-compensating</td>
</tr>
<tr>
<td>XPM</td>
<td>cross-phase modulation</td>
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Publications

Parts of this doctoral thesis are published in the following journal papers and conference proceedings.

Journal Papers


Conference Papers

Conference on Lasers and Electro-Optics (CLEO), Munich, Germany (2019). (Poster)


Abstract

Attosecond pulses allow probing matter at its fundamental time scales. However, generation of the shortest pulses is closely linked to the laser technology. In particular, due to the high-harmonic generation scaling, the mid-infrared (mid-IR) laser sources hold the key in producing the shortest attosecond pulses and probing the fastest atomic and molecular dynamics.

The main obstacle in creating the shortest attosecond pulses and using them for science lies in the highly demanding requirements for mid-IR laser technology. In particular, energetic, few-optical-cycle pulses at high-repetition rates and high-average powers are needed. Up until now, such mid-IR pulses were demonstrated and used only at low repetition rates where possible experiments are limited. In this thesis, we show the first mid-IR laser system exceeding 14 GW peak power at 100 kHz repetition rate. We lay the foundations for the third-generation femtosecond technology at the mid-IR spectral range and we demonstrate a laser source capable of producing few-cycle pulses at greater than 25 W average power. Multiple innovative steps needed to achieve the high-repetition rate and sufficiently high-peak power pulses are unveiled in this thesis.

Finally, in this thesis we show, for the first time, a table-top high-repetition rate coherent soft x-ray continuum source spanning the complete water window range and extending beyond it. Based on this source we demonstrate the complete attosecond beamline design for use of such pulses in the next generation attosecond experiments.


Schliesslich zeigen wir in dieser Arbeit zum ersten Mal eine kohärente, weiche Röntgenkontinuumsquelle mit hoher Repetitionsrate und überschaubaren Platzbedarf, die den gesamten Bereich der Wasserfenster und darüber hinaus abdeckt. Basierend auf dieser Quelle zeigen wir das komplette Attosekunden-Beamline-Design für die Verwendung solcher Impulse in Experimenten mit hoher Wiederholungsrate.


Chapter 1
Motivation

The observation and study of the fastest events truly kicked off with the invention of the laser [1]. Very quickly we went from flash photolysis [2] to the use of the first nanosecond (ns) pulsed lasers [3]. With a demonstration of passive mode-locking, the first picosecond (ps) pulsed sources became available [4]. Colliding pulse mode-locking enabled sub-100 femtosecond pulse generation [5] leading to emergence of femtosecond chemistry [6]. The quest for the shortest pulses shortly followed with a nonlinear pulse compression already with 6 femtosecond (fs) pulses [7].

The ultrashort pulse producing lasers took their modern shape with the invention of Kerr-lens mode-locking [8] and semiconductor saturable absorber mirrors (SESAM) [9]. Shortly after, compressed white-light continuum pulses as short as 5-fs were demonstrated [10,11]. The ultrashort laser pulse technology was approaching the Fourier-limit of the visible light.

Meanwhile, in the background, another revolution was taking place. High-intensity laser pulses were used to drive matter beyond the perturbative regime with the first observation of coherent, laser-generated extreme ultraviolet (EUV) radiation [12,13]. The generation of the EUV light was explained by a three-step model [14,15]. According to this model, a sufficiently strong laser electric field is used to bend the potential of atoms in a way that electrons can tunnel out through the potential barrier. The freed electrons are accelerated with the laser field to high-energies and once they recombine with the parent ion, the EUV radiation is emitted. This emission takes place within every half-cycle of the carrier wave, thus leading to a harmonic structure in the frequency domain due to multiple emission bursts within a single laser pulse. Since emission occurs twice within a single oscillation, the generated radiation has harmonic structure in the frequency domain with spacing twice the carrier frequency. Due to this periodicity of the generated radiation, the process was named as a high-harmonic generation (HHG).

Yet a strong prerequisite for the HHG process to take place is achieving very high light intensities at the focus (on the order of 100 TW/cm²). At the same time as the first HHG experiments were performed, a new and very powerful concept was demonstrated: chirped pulse amplification (CPA) [16]. This technique allowed circumventing material damage threshold and nonlinear self-action limitations while allowing to amplify ultrashort pulses to very high energies. Since the first demonstration of the CPA technique, the frontier of the highest peak power laser pulses was continuously pushed forward.

The first HHG experiments were performed at few Hertz pulse repetition rates. It took nearly another decade until the same peak power in the 10-100-gigawatt (GW) range became available for the experiments with 1 kHz repetition rate. Such relatively high-repetition rate, high-peak power pulses were used for pioneering work where time-resolved spectroscopy frontiers were explored. At first, a
generated EUV pulse train of 250 attosecond (as) pulses was characterized. This experiment was shortly followed by an isolated attosecond pulse measurements and their use for attosecond time-resolved spectroscopy.

This pioneering spectroscopy work was performed with Ti:sapphire CPA systems generating probe (or pump, depending on experiment) pulses in the 100-as range. The bandwidth of the attosecond pulses is determined by the electron energy acquired during the free acceleration in the continuum. The generated high-frequency cut-off of the HHG process scales with the intensity of the laser $I$ and wavelength $\lambda$ of the carrier wave $\sim I\lambda^2$ [15]. However, the intensity of pulses cannot be increased arbitrarily. Too high ionization of the gas target will lead to a high-density plasma generation and impossible phase-matching of the HHG process [21]. Thus, to obtain a high-energy cut-off without excessive ionization, the mid-infrared (mid-IR) driving lasers are required.

Mid-IR laser light generation is a difficult task due to the lack of suitable laser gain materials. However, an alternative path to the mid-IR laser light generation is by transferring visible/near-infrared (near-IR) light into the mid-IR spectral range. This can be achieved with the use optical parametric amplification (OPA) techniques [22,23]. The OPA process can take place in a large variety of nonlinear optical materials, many of which can support mid-IR light generation. When OPA was first demonstrated together with chirping of the seed followed by its compression after the amplification (OPCPA) [24,25] it opened the doors for a wavelength flexible, highly scalable laser pulse generation. Up to this day, the OPCPA technique shows no scalability limits and we will discuss it in great detail.

Today, the OPCPA technique is revolutionizing attosecond science. Using a mid-IR OPCPA, up to 1.6 keV soft x-ray (SXR) continuum corresponding to 5000's order harmonic generation was demonstrated [26]. If such pulses were to be perfectly compressed, it would yield 2.5 as pulses. However, these SXR pulses were generated only at 20 Hz repetition rate, limiting the applicability of such source for time-resolved attosecond spectroscopy experiments.

Although the HHG cut-off scales favorably with the driving laser wavelength, the HHG yield from a single atom drops rapidly with wavelength. The single-atom response for a fixed energy interval at fixed intensity is expected to scale with $\sim\lambda^{-5.5}$ [27], meanwhile if the cut-off is tracked and intensity is adjusted for phase-matching to be possible, the single-atom yield around the cut-off scales with $\sim\lambda^{-8.4-9.0}$ [21]. This flux loss can be partly mitigated by phase-matching and extending the pressure-length product [21], but cannot be fully overcome. Therefore, high-average power mid-IR laser sources with pulses providing sufficient peak power are necessary to preserve the flux and enable SXR use for the next generation attosecond experiments.

A significant step forward in the recent years was made by using 1 kHz Ti:sapphire CPA output pulses, generating the mid-IR light via the OPA process and driving the HHG with it [28–30]. With this approach pulses as short as 43 as [31] and a HHG cut-off spanning the complete water window (SXR spectral range between the K-absorption edges of carbon at 284 eV and oxygen at 543 eV) up to 570 eV [28]
were demonstrated. However, this technique is effectively limited by the used laser technology. Due to the average power limits in the Ti:sapphire CPA only up to a few-Watt average power in the mid-IR can be achieved and thus it cannot be further scaled in the repetition rate needed to overcome the loss of flux. The repetition rate scaling is especially important for experiments where few photo-ionization events per shot are required to avoid space-charge effects [32] or to enable coincidence detection [33,34]. High-repetition rate attosecond sources are necessary to increase the signal-to-noise ratio of experiments, decrease data acquisition time and scale the SXR flux while keeping the experimental setups compact and table-top.

The first generation attosecond experiments were performed with low average power Ti:sapphire CPA systems. By employing the OPA technology, the center wavelength of the HHG driving pulses could be shifted towards the mid-IR and SXR generation with sub-100 as pulse duration was achieved. In this thesis I will focus on developing the next generation attosecond technology. In particular, I will address a general laser technology shift towards the high-power picosecond laser pumped few-cycle mid-IR OPCPA techniques. Furthermore, I will focus on scaling the average power of mid-IR lasers so that a sufficient peak power at high-repetition rates can be achieved for mid-IR driven HHG with cut-off extending beyond the water window.

Figure 1.1 shows so far demonstrated HHG cut-off versus repetition rate of the laser (for ≥1 kHz case). Blue diamonds indicate the demonstrated results before the start of the thesis and green diamonds show the results announced during the time of the thesis. To bring the attosecond technology forward we concentrated on achieved phase-matched HHG at 100 kHz repetition rates and with cut-off greater than the oxygen K-edge to be able to span the complete water window.
1. Motivation

Figure 1.1. Demonstrated HHG cut-off versus repetition rate. The overview was compiled during the thesis writing time. It is not an exhaustive list but represent most of the key results reporting the largest phase-matched cut-offs at the time. Blue diamonds correspond to the results published before the start of the thesis (<2016). Green diamonds belong to the results published during the thesis related work. Red diamond corresponds the achieved and targeted result. This result is described in the thesis. Corresponding references are: 1 [35], 2 [28], 3 [29], 4 [36], 5 [37], 6 [38], 7 [39], 8 [40], 9 [41], 10 [42], 11 [43], 12 [44], 13 [45], 14 [46], 15 [47], 16 [48].

To be able to achieve this goal, at first, we had to innovate the laser technology so that a high-repetition rate mid-IR laser system with sufficient peak power would be possible. Figure 1.2 shows state-of-the-art mid-IR systems with the same color code as for the previous figure. During the time of this thesis work multiple laser systems delivering greater than 1 GW peak power and higher than 10 W average power became available. We were able, for the first time, to demonstrate a 14 GW peak power mid-IR laser system delivering 25 W of average power at 100 kHz repetition rate.
Figure 1.2. Demonstrated peak power at high-repetition rate (>10 kHz) for systems with central wavelength above 1.5 μm. The overview was compiled during the thesis writing time. Blue diamonds correspond to the results published before the start of the thesis (<2016). Green diamonds belong to the results published during the thesis related work. Red diamonds correspond to the achieved and targeted results. These results are described in the thesis. Corresponding references from 1 to 8 are: [49–56]. The peak-power was tabulated by using the claimed pulse energy $E$ and the normalised temporal intensity function $I(t)$: $P_{pk} = E / \int I(t)dt$.

The layout of this thesis is as follows. In Chapter 2 I will discuss the theoretical foundations including perturbative nonlinear optics considerations relevant for OPCPA design, non-perturbative HHG aspects and its scaling. In Chapter 3 I will discuss general and practical high-repetition rate laser system design aspects relevant for attosecond science. I will show a powerful pulse shaping technique which allows spectral aberration-free amplitude and phase shaping of ultra-broadband pulses at arbitrary repetition rates. In Chapter 4 I will discuss in detail our new high-peak-power mid-IR OPCPA system and will discuss its performance and possible improvements in the future. In this chapter, I will show how periodically poled lithium niobate (PPLN) crystals can be used for high-power amplification with more than 100 W average power pump beams. In Chapter 5 I will unveil the general HHG scaling lows. Following these scaling laws an appropriate experimental apparatus for high-repetition rate mid-IR HHG is demonstrated. Then, I will show the first mid-IR HHG spanning the complete water window at 100 kHz repetition rate. Finally, I will discuss the design and implementation of the new attosecond pump-probe beamline which will enable the next generation attosecond experiments. In Chapter 6, I will discuss our auxiliary near-IR OPCPA system and its applications for the next generation of attosecond technology.
Chapter 2
Theoretical Foundations

In this chapter, I will discuss the theoretical framework which forms the basis for the work of this thesis. I will particularly address the aspects of a general theory which are relevant to understand the work presented in this thesis.

2.1 Electromagnetic Wave Propagation

Starting from the Maxwell equations one can show that an electromagnetic wave $E$ propagating in $z$ coordinate in space and $t$ coordinate in time can be described by the following equation:

$$\frac{\partial^2 E(z, t)}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E(z, t)}{\partial t^2} = \mu_0 \frac{\partial^2 P(z, t)}{\partial t^2},$$  \hspace{1cm} (2.1)

where $c$ is the speed of light, $\mu_0$ is the magnetic constant and $P$ is the polarization of the medium. In case of propagation in vacuum, the polarization of the medium would be null and the wave equation would reduce to the classical wave equation. However, the reason light is interesting for applications lies exactly in the interaction term with the propagation medium. Given the assumption that the light-matter interaction is perturbative, i.e. light is not strong enough to disturb the potential energy surface of the material it interacts with, we can apply a perturbative expansion of the induced polarization:

$$P(z, t) = \varepsilon_0 \sum_{n=1}^{\infty} \chi^{(n)}(z, t)E^n(z, t).$$  \hspace{1cm} (2.2)

Here $\varepsilon_0$ is the electric constant and $\chi^{(n)}$ is the susceptibility tensor. Since the interaction is perturbative, the sum is highly convergent where even the second term is negligible for usually encountered applications of light. In the first section, I will focus on the effects of wave propagation when the linear term dominates. Since the wave equation contains only linear terms in $E$, the linear system theory applies. This means that the output electric waveform is a convolution of the medium response function and the input electric field. From a mathematical standpoint, this can be simplified by transforming the description into the Fourier domain. In the Fourier domain, linear light-matter interaction can be described as a product between the input electric field and the spectral response function. The refractive index of the medium can be related to the linear spectral susceptibility via $n(\omega)^2 = 1 + \chi^{(1)}(\omega)$ and the wavenumber to be $k(\omega) = \frac{\omega}{c} n(\omega)$. Using this relation, we can write down the Helmholtz equation describing linear propagation of an electromagnetic wave in the frequency domain:

$$\frac{\partial^2 \tilde{E}(z, \omega)}{\partial z^2} + k(\omega)^2 \tilde{E}(z, \omega) = 0.$$  \hspace{1cm} (2.3)
The equation can readily be solved with a plane wave Ansatz \( \tilde{E}(z, \omega) = \tilde{A}(z, \omega) e^{-ik(\omega)z} \) and using a slowly varying envelope approximation (SVEA). SVEA assumes that the envelope \( \tilde{A}(z, \omega) \) of the pulse evolves in space and time much slower than the carrier-wave, thus allowing to drop the second order derivates of the envelope. As long as this approximation is true, the output electric waveform of a linear system can be retrieved by multiplying the input with \( e^{-ik(\omega)z} \). This result has multiple implications.

### 2.1.1 Linear Absorption

If the refractive index contains an imaginary part, the amplitude of the envelope can evolve in size. This can be related to absorption or gain. In the case of absorption, we directly recover the Beer-Lambert's law. When developing a laser system, it is crucial to consider the absorption bands of materials. Many materials transparent in the visible range start absorbing in the mid-IR wavelengths. For example, commonly used UV-grade fused silica (UVFS) normally has relatively broad absorption features at 2.2, 2.7 and above 3 µm and thus is not suitable for mid-IR systems. Molecular interaction in gas phase is much weaker and the absorption lines are very narrow. For example, water vapor is the main mid-IR absorbing gas in air. By using the absorption data available from HITRAN database and Voigt line broadening mechanism I calculate the transmission spectrum through 1 meter of air at 23 °C and 50% relative humidity as shown in Figure 2.1a. Very strong absorption at 2.6–2.8 µm is visible which can be attributed to O-H bond stretch vibrations. CO\(_2\) absorption is present as well, but it is much weaker than water in this spectral range. If one would be developing a mid-IR system around 2.5 µm spectral wavelength, water absorption would certainly be a significant obstacle. However, it could be avoided by immersing the laser system into N\(_2\).

![Figure 2.1. Linear absorption features. (a) Water transmission after 1 meter of propagation at atmospheric pressure, 23 °C and 50% relative humidity. Blue line shows a high-resolution and orange line shows a low-resolution picture in order to simulate a typical spectrometer response. (b) 100 nm thin film absorption at soft x-ray spectral range.](image)

The absorption situation is also very interesting when considered at EUV/SXR range. Here the photon energy is so high, that it can ionize electrons from core shells producing very sharp absorption features in spectrum as shown in Figure
2. Theoretical Foundations

2.1b for carbon, oxygen and iron. Having an x-ray continuum extending through these absorption features would allow to unambiguously map dynamics associated with those elements. Carbon and oxygen are particularly interesting due to their presence in biological molecules [57], whereas iron is interesting for study of ultrafast magnetization dynamics [58].

2.1.2 Linear Dispersion

Real values of the refractive index will contribute only to the change of the spectral phase, as given by the solution of the Helmholtz equation \( \tilde{E}(z, \omega) = \tilde{A}(z, \omega)e^{-ik(\omega)z} = \tilde{A}(z, \omega)e^{-i\phi(\omega)} \). When building an ultrafast system, it is crucial to manage this phase, since it relates to the pulse duration via the Fourier transform relation. If the spectral phase is constant (i.e. flat), the pulse duration will be Fourier-limited by the spectral width of the envelope. However, if the phase is not flat, the pulse will be chirped. Correct chirp management is crucial for achieving the desired pulse duration at the output of a laser system. Only if the pulse is compressed, the high-peak power can be achieved. In this sub-section, I will briefly describe the fundamental differences associated with chirp management at different spectral regions of relevance to the work of the thesis.

The spectral phase can be conveniently expanded by the use of the Taylor series around the center frequency \( \omega_0 \) of interest:

\[
\phi(\omega) = \phi(\omega_0) + GD_{\omega_0}(\omega - \omega_0) + \frac{1}{2!}GDD_{\omega_0}(\omega - \omega_0)^2 + \\
+ \frac{1}{3!}TOD_{\omega_0}(\omega - \omega_0)^3 + \frac{1}{4!}FOD_{\omega_0}(\omega - \omega_0)^4 + \cdots \tag{2.4}
\]

The absolute phase offset \( \phi(\omega_0) \) matters only for phase sensitive cases (such as interferometric experiments or attosecond pulse generation). The group delay (GD) term is relevant if one wants to calculate how much the pulse is delayed when propagating through the medium, but it does not affect the pulse shape. However, the group delay dispersion (GDD) term is responsible for a linear chirp of a pulse, whereas third-order dispersion (TOD), fourth-order dispersion (FOD) and higher-order terms contribute to higher-order pulse shape distortions. The higher-order dispersion terms (higher than GDD) become relevant only when bandwidth is approaching an octave or when there are non-material related sources of dispersion (such as a prism pair) which can introduce a very large TOD, with a negligible GDD. Which dispersion term dominates can be evaluated by comparing GD spread related to that dispersion given the bandwidth \( \Delta \omega \) of the pulse: \( GDD \cdot \Delta \omega, TOD \cdot \Delta \omega^2 \) etc. Furthermore, the higher-order-odd dispersion terms such as TOD are highly undesired for CPA systems, since it leads to an overlap of spectral components in the time domain. This therefore, creates interference fringes and causes gain competition between the spectral components. As a general rule of thumb, the desired pulse stretching for amplifiers should be such that \( \frac{TOD}{GDD} \cdot \Delta \omega < 1 \). This condition implies that the GD spectrum has no turning point within the bandwidth of the pulse due to TOD (under the assumption of no higher-order dispersion).
Following the considerations in the previous paragraph, it is most meaningful to consider dispersion management in terms of GDD and TOD values. Higher-order dispersion values can also matter for few-cycle pulse compression, but usually, are controlled by programmable pulse shapers, which will be discussed later. In a visible and near-IR spectral range all materials contribute with positive GDD and TOD values. Therefore, to manage dispersion at visible/near-IR spectral range, usually chirped mirrors, prism or grating stretchers/compressors are employed. In the mid-IR the situation becomes more interesting. Some materials which have their bandgap at the ultraviolet range (most of the transparent glasses in the visible range, such as CaF$_2$, Sapphire etc.) start exhibiting negative GDD at the mid-IR wavelengths. Whereas, materials which have their bandgap at the visible or near-IR will have positive GDD in the mid-IR (such as ZnSe, Si or KRS-5). Therefore, mid-IR CPA systems offer the unique opportunity of pulse compression in bulk, i.e. stretching by one and compressing by another material. Such type of compressors can exhibit especially high throughput. However, TOD still remains positive and thus it has to be managed separately. When it comes to minimizing TOD, an important criterion is GDD/TOD. A material survey is summarized in Table 2.1. From the table it can be seen that ZnSe or Si have favorable GDD/TOD ratio, however, this comes at the cost of larger nonlinear refractive index $n_2$.

<table>
<thead>
<tr>
<th>Material</th>
<th>Bandgap (nm)</th>
<th>GVD, fs$^2$/mm</th>
<th>GDD/TOD, fs$^{-1}$</th>
<th>$n_2$, cm$^2$/GW</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>730 nm</td>
<td>2.5 μm</td>
<td>730 nm</td>
</tr>
<tr>
<td>CaF$_2$</td>
<td>100</td>
<td>31</td>
<td>-55</td>
<td>1.99</td>
</tr>
<tr>
<td>Al$_2$O$_3$ (o)</td>
<td>124</td>
<td>67</td>
<td>-285</td>
<td>1.70</td>
</tr>
<tr>
<td>Si</td>
<td>1100</td>
<td>-</td>
<td>632</td>
<td>-</td>
</tr>
<tr>
<td>ZnSe</td>
<td>460</td>
<td>1317</td>
<td>211</td>
<td>1.00</td>
</tr>
<tr>
<td>MgO LiNbO$_3$ (e)</td>
<td>310</td>
<td>424</td>
<td>-216</td>
<td>1.66</td>
</tr>
<tr>
<td>β-BBO (e)</td>
<td>192</td>
<td>87</td>
<td>-381</td>
<td>1.76</td>
</tr>
</tbody>
</table>

Table 2.1 Dispersion properties of various materials. The values were calculated from Sellmeier relations [59–62] and $n_2$ was taken from literature [63–65].

Finally, we also need to consider material response in the EUV/SXR. In the EUV spectral range, it is possible to find material which exhibits negative GDD, thus allowing for pulse compression to sub-100 attosecond duration [66,67]. However, at SXR photon energies, the interaction cross-sections are smaller leading to complicated pulse compression options for photon energies above 500 eV [68]. An alternative approach was recently proposed to compress pulses in plasma [69]. During the writing time of this thesis, no successful attosecond pulse compression above 200 eV range was demonstrated in the community and it still remains the frontier.
2. Theoretical Foundations

2.2 Nonlinear Optical Interaction

In the previous chapter we have discussed pulse propagation aspects associated with linear response of matter. However, if the driving electric field strength is increased, the polarization response can become nonlinear. The microscopic explanation to this lies in the potential energy curve shape of the medium. There are no materials which exhibit a purely quadratic shape of the potential. Thus, if driven strong enough, they will start to respond anharmonically leading to the emergence of nonlinear terms in the polarization:

\[ P_{NL}(z, t) = \varepsilon_0 \sum_{n=2}^{\infty} \chi^{(n)}(z, t)E^n(z, t). \] (2.5)

A linear spring-ball system will have a harmonic potential (quadratic shape). Hence, a higher-order correction of this is potential would include a cubic term which breaks the inversion symmetry allowing for response at twice the driving frequency. There are plenty of materials which break this inversion symmetry, such as commonly used nonlinear crystals like beta-barium-triborate (BBO), lithium triborate (LBO), etc. There are, however, materials which have the inversion symmetry (such as silicon or fused silica) and even-order polarization response is not possible and therefore \( \chi^{(2)}, \chi^{(4)} \) and etc. cannot exist. In such materials the first nonlinear response will contain a \( \chi^{(3)} \) term.

If the driving field strength is further increased until it becomes comparable to the material internal field strengths, the actual potential becomes modified by the driving field non-perturbatively. Therefore, this leads to a highly transient response allowing the generation of attosecond light bursts. In such regime, the higher-order nonlinear response was found to stop decreasing exponentially, as it is common for perturbative processes, but rather to plateau in response strength [70]. More in-depth discussion into this topic is given in the HHG section of the chapter.

2.2.1 Second Harmonic Generation

As the first example of a nonlinear interaction let us consider the second harmonic generation of the driving field. This kind of nonlinear interaction was used in some of the nonlinear stages and pulse characterization measurements. To understand this nonlinear interaction, first let us consider a general three-wave mixing process expressed as a superposition of two waves with frequencies \( \omega_1 \) and \( \omega_2 \):

\[ E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c. \] (2.6)

Then the second-order polarization response will be

\[ P_{NL}(t) = \varepsilon_0 \chi^{(2)}(E_1^* E_2 e^{-i(\omega_1 + \omega_2) t} + 2E_1 e^{-i(\omega_1 - \omega_2) t} + 2E_2 e^{-i(\omega_1 + \omega_2) t} + \varepsilon_0 \chi^{(2)}). \] (2.7)

The nonlinear polarization term contains various three-wave frequency mixing processes, including the second harmonic generation \( (2\omega_1 \text{ and } 2\omega_2) \), sum-frequency generation \( (\omega_1 + \omega_2) \), difference frequency generation and optical
parametric amplification \((\omega_1 - \omega_2)\) and rectification (independent of the frequencies \(\omega_1\) and \(\omega_2\)).

Starting from the Helmholtz equation, assuming that the fundamental wave at the driving frequency does not get depleted and by using the SVEA it possible to show that the SHG output intensity \(I_2\) follows [71]:

\[
I_2(z) = I_1(0)(\Gamma z)^2 \text{sinc}\left(\frac{\Delta k z}{2}\right),
\]

(2.8)

where \(\Gamma = \kappa|E_1(0)|, \kappa = \frac{\omega_1 d_{\text{eff}}}{n_1 c}, \Delta k = 2k_1 - k_2\) is the phase-matching term and the \(d_{\text{eff}}\) is the effective nonlinear coefficient proportional to \(\chi^{(2)}\). From the equation, one can see that the second harmonic power will grow quadratically with length assuming a perfect phase-matching between the waves. In this case, power of the second harmonic will grow to the comparable magnitude as the driving field. This will lead to depletion of the driving field and overall saturation of the conversion efficiency. When considering the depletion of the driving field, the SHG intensity follows:

\[
I_2(z) = I_1(0) \tanh(\Gamma z)^2
\]

(2.9)

Given a sufficient length of the crystal or strong enough input intensity this function would converge to 100\% conversion efficiency. However, in practice, this is mostly not reached. As discussed in [71], the phase-matching width \(\Delta k L\) will rapidly decrease with increasing \(\Gamma z\) leading to enhanced sensitivity to phase-matching at high-conversion efficiencies. A phase-mismatch can appear when using ultrashort pulses due to self-action related effects. Because of this and other effects, usually we achieved around 70-80\% conversion efficiencies.

If SHG is operated at low conversion efficiencies, such as 25-40\%, the remaining fundamental beam peak will be significantly depleted as for example is shown in Figure 2.2. In the figure one can see the temporal profile of the depleted pulse and the generated second harmonic. Similar depletion also occurs in the spatial domain. This kind of beam flattening was employed in our setup and is discussed in section 4.2.
2. Theoretical Foundations

Figure 2.2. Temporal pulse flattening via SHG. Calculated depletion of our amplifier output (discussed in section 3.2.1) at 37% SHG efficiency.

2.2.2 Optical Parametric Amplification

Another three-wave mixing process of paramount importance for this thesis is the optical parametric amplification (OPA). It is described by the $(\omega_1 - \omega_2)$ difference frequency term discussed in the previous section. A careful treatment of the process is well described by [22,23] and here I will briefly summarize the key parameters. The highest frequency term in the interaction is always referred to as the pump, the frequency term which is amplified and is seeding the process is denoted as signal and the generated difference frequency term is the idler. Throughout the thesis these components will be denoted with corresponding suffixes $p$, $s$ and $i$. The frequency of the idler is always such that it satisfies the energy conservation relation:

$$\omega_p = \omega_s + \omega_i. \quad (2.10)$$

The phase-matching condition does not have to be rigorously satisfied, however, given the interaction length $L$, the phase-mismatch should be less than $\frac{\pi}{L}$ for the process to be efficient:

$$|\Delta \vec{k}| \equiv |\vec{k}_p - \vec{k}_s - \vec{k}_i| < \frac{\pi}{L}. \quad (2.11)$$

Following a rather involved coupled-wave derivation, assuming the SVEA, flat-top spatial and temporal profiles and no pump depletion the intensity gain for the signal frequency can be expressed as [22]:

$$G_s = 1 + \left(\frac{gL}{\alpha}\right)^2 \sinh^2(\alpha) \quad (2.12)$$
with the following coefficients:

\[ g^2 = \frac{d_{eff}^2 \omega_s \omega_i I_{\text{peak}}}{2 \varepsilon_0 n_s n_i n_p c^2}, \]  
\[ \alpha^2 = (gL)^2 - \left( \frac{\Delta k L}{2} \right)^2. \]  

(2.13)  
(2.14)

Here, \( I_{\text{peak}} \) is the intensity of the pump, \( n_x \) is the refractive index of the signal, pump and idler, \( \varepsilon_0 \) is the vacuum permittivity and \( c \) is the speed of light in vacuum.

Assuming a perfect phase-matching, i.e. \( \Delta k = 0 \), the gain is proportional to \( \sinh^2 gL \). Hence, the gain will stay constant as long as the \( gL \) product is constant. Since \( g \propto \sqrt{I_{\text{peak}}} \), halving the crystal length would require increasing the intensity by a factor of four to reach the same gain. Furthermore, if a broadband signal is amplified, the gain will not be spectrally uniform due to the product \( \omega_s \omega_i \). In such a case, the gain will maximize at the degeneracy point (i.e. \( \omega_s = \omega_p / 2 \)).

In case of an imperfect phase-matching at low pump intensities, the gain bandwidth will be limited by the phase-matching bandwidth. The phase-matching bandwidth can be studied by performing the Taylor expansion of the phase-mismatch term around the central frequency of the signal:

\[ \Delta k(\omega_0 + \Delta \omega) = k_p - k_s(\omega_0) - k_i(\omega_0) - \left( \frac{\partial k_s}{\partial \omega} \right)_{\omega_0} \Delta \omega - \frac{1}{2} \left( \frac{\partial^2 k_s}{\partial \omega^2} \right)_{\omega_0} \Delta \omega^2 + \ldots \]  

(2.15)

Noticing that the inverse of \( \frac{\partial k}{\partial \omega}_{\omega_0} \) corresponds to the group velocity \( v_g(\omega_0) \), \( \frac{\partial^2 k}{\partial \omega^2}_{\omega_0} \) corresponds to the group velocity dispersion (GVD) and assuming a perfect phase-matching at the central frequency we can approximate the phase-mismatch for a given bandwidth with the following equation:

\[ \Delta k(\omega_0 + \Delta \omega) \approx \left( \frac{1}{v_{gi}(\omega_0)} - \frac{1}{v_{gs}(\omega_0)} \right) \Delta \omega - \frac{1}{2} \left( \text{GVD}_s(\omega_0) + \text{GVD}_i(\omega_0) \right) \Delta \omega^2 \]  

(2.16)

From this equation, it is clear that the OPA bandwidth decreases as the group velocity mismatch between the signal and idler is increased. Therefore, to achieve a broadband OPA, first the group velocities need to be matched. This can happen around the degeneracy (in case signal and idler share the same polarization), if material dispersion allows for it or by introducing a noncollinear angle between the signal and the idler [72]. Once the group velocities are matched, the phase-matching bandwidth will be limited by the GVD between the idler and the signal which ideally should hold the opposite signs.
One important aspect which often remains unmentioned is the risk of amplifying quantum noise. If the gain is high enough, the OPA can amplify the quantum noise to the level comparable to the signal. The quantum noise can be modeled as an omnipresent seed which seeds the OPA at all angles, positions, frequencies and times. Therefore, if the actual seed level is comparably low and it is not well matched to the gain the pump creates a significant quantum noise amplification can occur. This process is usually referred to as an optical parametric generation (OPG) and is highly undesired in OPCPA systems since it reduces the useful energy in the main pulse and increases its shot-to-shot noise level. Throughout the work of the thesis various measures were taken to reduce the impact of the quantum noise.

2.2.2.1 Collinear BBO OPA Example

One of the best-suited materials for OPA is BBO. It has a large transparency range and was demonstrated to be suitable for nonlinear optics from 200 nm to 3200 nm and recently even up to 5 µm in case of a very short-length interactions [61]. Furthermore, it has a high-damage threshold and can have relatively large $d_{\text{eff}}$. Because of these properties, we have used this material for various experiments. Figure 2.3 shows a simulated undepleted OPA gain for the case of a 1.5-mm long BBO crystal pumped by 515 nm 50 GW/cm$^2$ pulses. In this case, a type-I birefringent phase-matching is considered (i.e. the signal and idler share the same and the pump has orthogonal polarizations). From the figure, one can see that the phase-matching is mostly broad at the degeneracy, where signal and idler group velocities are matched. Furthermore, there is one more turning point at 630 nm due to the group velocity matching. Since in this case there are two group velocity turning points, that means that GVD mismatch also crosses a zero-point somewhere (actually at 715 nm in this case). However, the pulse amplification in the 650–900 nm range is relatively narrowband supporting 50–100 fs pulses. If somehow it would be possible to bring the group-velocity matching point close to the GVD matching it would be possible to achieve an ultra-broadband phase-matching.
Nonlinear Optical Interaction

Figure 2.3. Collinear BBO OPA gain versus phase-matching angle. The gain curves were calculated assuming 50 GW/cm² pump intensity at 515 nm. The crystal length used for the calculation was 1.5 mm.

2.2.2.2 Noncollinear Optical Parametric Amplification

In the previous section, we found that to achieve a broadband phase-matching, a group velocity matching between the signal and the idler is needed. It turns out that by setting the signal noncollinear to the pump, one can achieve the group velocity matching in certain materials for the idler. Of course, this comes at the cost of an angularly dispersed idler beam. To better understand let us study the geometry in more detail. We assume a noncollinear angle $\theta_s$ between the pump and the signal, then the angle between the idler and the pump will be $\theta_i(\omega)$. Furthermore, we can decompose the vectors in two planes, one perpendicular and one parallel to the pump:

\[
\Delta k_\perp(\omega) = k_s(\omega) \sin \theta_s - k_i(\omega) \sin \theta_i(\omega), \tag{2.17}
\]

\[
\Delta k_\parallel(\omega) = k_p - k_s(\omega) \cos \theta_s - k_i(\omega) \cos \theta_i(\omega). \tag{2.18}
\]

Assuming a perfect phase-matching, the first equation can be used to find the idler noncollinear angle which will be frequency-dependent:

\[
\theta_i(\omega) = \arcsin \frac{k_s(\omega) \sin \theta_s}{k_i(\omega)}. \tag{2.19}
\]

To estimate the phase-matching bandwidth in the noncollinear geometry the same Taylor expansion as used in the previous section can be performed. When going through the calculation one will find that the group-velocity related expansion terms will cancel if the group velocities $v_{gs}$ and $v_{gi}$ are matched through the following relation:

\[
v_{gs} = v_{gi} \cos (\theta_s + \theta_i(\omega)). \tag{2.20}
\]

This, therefore, suggests that provided a right pump wavelength and crystal choice an OPA can be configured to support an ultra-broadband bandwidth by choosing a right noncollinearity angle.
2.2.2.3 Noncollinear BBO OPA Example

To compare with the collinear case, we study again the gain bandwidth of a BBO crystal however this time in a noncollinear optical parametric amplification (NOPA) configuration. If the internal noncollinearity angle between the pump and the signal is set to 2.4°, the group velocities will match at 740 nm. Furthermore, the GVD between the signal and idler will also be matched at 790 nm wavelength (for the case of 515 nm pump). Thus, as a consequence of this matching, the NOPA will exhibit an ultra-broadband gain bandwidth as it is shown in Figure 2.4a. On top of that, as we saw before, the influence of phase-mismatch is less if the pump intensity is larger. Figure 2.4b illustrates this gain bandwidth scaling with intensity. Therefore, for amplifying ultrashort pulses, a high-pump intensity is desired.

![Figure 2.4. BBO NOPA gain. (a) NOPA gain tuning for 1.5 mm long BBO crystal when pumped with 515 nm at 50 GW/cm² intensity and seeded with a noncollinear angle of 2.4°. (b) Calculated NOPA gain at three different intensities for a fixed crystal angle of θ = 24.5°.](image)

Finally, another very important aspect is spatial walk-off. Because of the birefringent phase-matching, the pump k-vector will not point in the same direction as the energy flow. For the BBO NOPA, this spatial walk-off angle is around 3.3°. Therefore, if the beam sizes are very small or crystals are very long, the interaction will be limited by the spatial overlap length between the signal and the pump. Furthermore, the gain will not be spatially and spectrally uniform and the spatial walk-off can lead to a pulse wavefront tilt [73,74]. To reduce the wavefront tilt effect, the NOPA can be configured in a walk-off compensating direction, where the signal noncollinearity angle matches the walk-off of the pump.

2.2.2.4 Quasi-Phase-Matching

It is not always possible to find a nonlinear crystal which will exhibit the desired dispersion, strong effective nonlinear coefficient and will allow phase-matching at the targeted wavelength range. To overcome this issue, a quasi-phase-matching technique can be used. By periodically poling the crystal (i.e. by inverting the sign of the nonlinearity every Λ μm) the nominally nonphasematched process can keep the phase-synchronization with the driving wave.

An advantage of such a technique is that it allows choosing the crystal axis such that the \( d_{eff} \) is the largest. For lithium niobate (LN) this is the \( d_{33} \) coefficient, which
is nearly 10 times larger for a 1030 nm pumped OPA process compared to the optimum in BBO. Because of the periodic c-axis inversion, the effective coefficient strength will be slightly reduced (by a factor $\pi/2$ in case of 50% duty cycle [75]), however, it still allows for very strong coupling between the different waves.

The periodic c-axis inversion for the phase is equivalent to a grating with a grating vector in the k-space of $K_g = 2\pi/\Lambda$. Therefore, this effectively will contribute to the phase-mismatch term:

$$\Delta k = k_p - k_s - k_i - K_g.$$  \hfill (2.21)

By setting the right periodicity of the poling, a desired nonlinear process can be phase-matched. On top of that, the polling could be performed aperiodically, i.e. $K_g$ can be a function of propagation distance $z$ or it can even have a transversal component as we will discuss later. This can allow for extremely broadband amplification or switching on and off various nonlinear processes during the propagation [76].

### 2.2.2.5 Periodically Poled Lithium Niobate NOPA example

The achievable poling periodicity depends on the choice of the crystal. For LN the poling periodicity can be in the range of to 5–50 µm allowing to correct for 100–1200 mm⁻¹ k-vector mismatch. For example, a 1030 nm pumped collinear periodically poled LN (PPLN) for 2.5 µm amplification will exhibit a phase-mismatch of 200 mm⁻¹. Hence 31 µm polling period would lead to a phase-matched amplification. Furthermore, because of the QPM geometry, the idler and signal will share the same polarization, thus leading to the group velocity matching at the degeneracy (2 µm) and broadband gain. Through the noncollinear operation, the GVM point can be shifted to the longer wavelength. Finally, the GVD mismatch between the signal and the idler is relatively small in PPLN due to low dispersion of the material at the mid-IR range. Since the $d_{eff}$ is very large for the QPM geometry, the effective crystal length can be very short and thus broadband amplification in the mid-IR can be achieved. The gain-bandwidth in 1030 nm pumped noncollinear PPLN is shown in Figure 2.5. One can see that octave-spanning amplification can be achieved.
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Figure 2.5. PPLN small-signal gain. (a) PPLN gain at different noncollinear angles. The 1030 nm pump intensity was assumed to be 13 GW/cm$^2$ and crystal thickness was set to 0.5 mm. (b) Small-signal gain at internal noncollinearity angle of 1.6°.

One of the main issues with the PPLN is the generation of free-carriers. The free-carriers are usually generated by the pump or its undesired second harmonic. The charges accumulate in the crystal and create a strong local electric field, which alters the properties of the material leading to photorefractive distortions of the beam. Over time it was found that doping PPLN with MgO can drastically reduce the photorefractive effect. By today this doping to 5%-level became the industry standard. Because of this, throughout this thesis the PPLN and LN are always considered to be doped with MgO unless otherwise specified.

2.2.3 Third-Order Nonlinear Processes

Third-order nonlinear processes, such as third-harmonic generation (THG), rarely play an important role in nonlinear optics due to very different frequencies involved (and thus limited phase-matching bandwidth) and much weaker coupling coefficient. However, some processes, such as self-phase modulation (SPM) or cross-phase modulation (XPM) are always phase-matched. Furthermore, many materials do not have the second-order nonlinear coefficient due to the inversion symmetry, however, all materials, including air will exhibit third-order nonlinear coupling which can accumulate dramatically.

One of such processes is the SPM. Its strength can be estimated by considering the B-integral:

$$B = \int_0^L \frac{2\pi}{\lambda} n_2(x) I_{\text{peak}}(x) dx.$$  \hspace{1cm} (2.22)

Here $\lambda$ is the center wavelength of the pulse, $I_{\text{peak}}$ is the peak intensity of the pulse and $L$ is a physical length of the material. In the time-domain, the nonlinear phase combined with the dispersion of the material can strongly distort the pulse shape. This kind of nonlinear intensity coupling is often desired in oscillators helping to form soliton-like pulses [8] or to nonlinearly compress the output pulses [77]. Meanwhile, in the spatial-domain, this can act as a focusing lens leading to a self-focusing action of the beam. It is desired for the Kerr-lens mode-locked oscillators
but it is highly undesired for high-power lasers used to pump nonlinear systems since it can lead to a self-focusing and damage of the optical components or distortion of the beam.

Using modal analysis performed by my colleague we can estimate the focusing strength (inverse of the focal length) of the Kerr-lens:

\[ F_{\text{Kerr}} = \frac{1}{w_0^2} n_2 LI_{\text{peak}} = \frac{\lambda}{2\pi w_0^2} B = B/b \]  

Here, \( b \) is the confocal parameter of the beam with waist \( w_0 \). A similar consideration applies in the time-domain and the temporal phase can be modified differently for different parts of the beam, leading to a not perfectly compressible pulse due to spatially varying temporal phase. If the laser intensity is fluctuating, this will couple to phase, temporal and spatial shapes of the beam. For these reasons the B-integral has to be kept as low as possible.

Often overlooked, but very important is the XPM between the pump and the signal waves in an OPA process. Usually, the intensity of the pump beam is rather large and the intensity of the signal is quite low. Although the SPM of the signal itself is low, the pump can modulate the phase of the signal via the third-order nonlinear coupling. The XPM coupling is much stronger if the pump and the signal share the same polarization, leading to the coupling coefficient \( \gamma_{ps} = 2 \) (for type-I phase-matching with different polarizations \( \gamma_{ps} = 2/3 \)) [78]. The B-integral due to the XPM effect (ignoring the idler contribution) is:

\[ B_{\text{XPM}} = \int_0^L \frac{2\pi}{\lambda_s} \gamma_{ps} n_2(x) I_{\text{peak}}(x) dx \]  

For example, for a BBO-based 2 mm long OPA stage pumped at 100 GW/cm\(^2\), the XPM contribution to the B-integral can be around 1 radian, already. Of course, usually, such high-intensity is used only at the pre-amplification stages where the signal is normally much smaller than the pump and thus is sampling less of the non-uniform phase. PPLN based nonlinear amplification stages can have stronger XPM due to larger \( n_2 \) and stronger coupling coefficient. Therefore, this kind of coupling is one of the important problems for high-energy, broadband OPCPA systems. Flat-top pump beams can decrease the non-uniformity of this added phase.

### 2.3 High-Harmonic Generation

#### 2.3.1 Classical Model

At the heart of this thesis lies the generation of attosecond light pulses through the HHG process. The HHG was first observed experimentally by focusing high-energy pulses into gas targets [13,79]. The experiments have found that the very high order harmonics did not follow anymore logarithmic decrease of intensity predicted by perturbative interaction models. Rather the intensity of the harmonics reached a plateau before dropping again at a certain cut-off. Because of this scaling, it
became possible to generate coherent EUV/SXR pulses with laser coherence properties.

One of the most intuitive and useful explanations to the HHG is the semiclassical three-step model first proposed in [14]. The description includes three steps: strong-field ionization, propagation in the continuum and finally recombination of the emitted electron with the parent ion leading to the emission of a high-energy photon or scattering.

The first step in the model is the tunnel ionization of the atom which we will more carefully discuss in the following section. For simplicity we can assume that the electron tunnels out with its initial momentum and position being zero. The second step in the model is the unbound electron classical propagation in the laser field. In the following derivation, I will ignore the magnetic field contribution, however, if very high-intensities or very long wavelength drivers are considered, the magnetic field of the laser starts to impact the propagation [80]. Furthermore, as the first approximation, the electric field of the parent ion can be ignored. This approximation can be justified by a significant propagation distance outside the atom, extending to a nanometer scale [81]. On such scales, the electric field of the laser strongly dominates. Considering the driving field $F_L$ being monochromatic and linearly polarized one can write down a classical free-electron equation of motion:

$$\dot{x}(t) = -\frac{e}{m_e} F_L \cos \omega_0 t.$$  \hspace{1cm} (2.25)

In this equation, $e$ is the electric charge, $m_e$ is mass of the electron and $\omega_0$ is the driving field frequency. Assuming the electron tunnels out at time $t_0$ with no initial velocity, the equation can be solved for the following electron trajectory function:

$$\dot{x}(t) = \frac{eF_L}{m_e \omega_0} [\sin \omega_0 t_0 - \sin \omega_0 t],$$  \hspace{1cm} (2.26)

$$x(t) = \frac{eF_L}{m_e \omega_0^2} [\cos \omega_0 t - \cos \omega_0 t_0 + \omega_0 (t - t_0) \sin \omega_0 t_0].$$  \hspace{1cm} (2.27)

This electron trajectory is graphically visualized in Figure 2.6. In this figure, only a single burst of electrons emitted during the first half-cycle of the driving field is considered. From Figure 2.6a one can see that not all emitted electrons will return to the origin. However, those which return will interact with the parent ion.
The last step of the HHG process is the electron recombination with the parent ion. At the return time $t_R$, the electron can recombine and emit an EUV/SXR photon or scatter. This can happen only if $x(t_R) \approx 0$. The equation can be numerically solved and the solution is shown in the Figure 2.7a. From the numerical calculation, it can be seen that an electron must be emitted at a phase between $0$ and $\pi/2$. If it is emitted earlier or later, it does not return to the residual ion. The recombination phase defines the return velocity. Thus, the final recombination momentum will be:

$$p_{inc} = \frac{eF_L}{\omega_0} [\sin \omega_0 t_0 - \sin \omega_0 t_R].$$  \hspace{1cm} (2.28)

Inserting the numerical solution of the recombination phase into the momentum equation one can find the final energy of the electron for different emission phases as it is shown in Figure 2.7b. From the figure one can see that the returning electron will have the highest kinetic energy if the emission phase is around 0.3129 radians. Then the maximum returning electron kinetic energy is:

$$E_{K,max} = \frac{p_{inc}(0.3129)^2}{2m_e} \approx 3.2 \frac{e^2 F_L^2}{4m_e \omega_0^2} = 3.2U_p,$$  \hspace{1cm} (2.29)

where $U_p$ is the electron ponderomotive energy. From this equation it follows that the maximum attainable HHG photon energy would be the ionization potential $I_p + 3.2U_p$ and it scales proportional to $\lambda^2I$. Therefore, the highest energy photons can be generated with mid-IR driving fields or very intense pulses. This kind of scaling motives development of intense long-wavelength sources.

Figure 2.6. Calculated electron trajectories during HHG. (a) Calculated trajectories with different emission times (30 randomly selected trajectories following the emission probability curve shown in part b. (b) The electric field cycle of 2.2 µm driving field with 300 TW/cm² intensity (blue). The ionization rate (red) was calculated for argon atoms using Ammosov-Delone-Krainov (ADK) rates (see the following section).
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Figure 2.7. Electron recombination and HHG emission. (a) Numerical solution to the recombination phase for a valid set of emission phases. Here phase represents timing with respect to the carrier wave as shown in Figure 2.6b. (b) The returning electron energy versus the emission phase (left axis). The right axis is GDD of high-harmonic radiation calculated for 2.2 µm driving wavelength.

From the Figure 2.7b it can also be seen that for the electron energies below 3.2\(U_p\) there can be two different emission phases. The electrons emitted later would recombine sooner and are usually referred to as “short trajectory” electrons. The electrons emitted earliest will recombine latest and these are usually referred to as “long trajectory electrons”. Because of this, the chirp of the harmonics will be opposite and if those harmonics would overlap in time domain, it will cause interferences. Later we will see that the trajectories can be selected via phase-matching. It is important to note that the chirp of the short trajectories is positive and it can be compensated by adding material in the path which introduces negative chirp.

2.3.2 Strong-Field Ionization

Understanding of the atomic ionization mechanism in an intense harmonic electric field is the key for a quantitative understanding of the HHG process. It is well known that if atoms are exposed to light which photon energy is larger than the ionization potential, photoelectrons are emitted [82]. However, if the photon energy is smaller than the ionization potential there is still a finite probability for the ionization. Generally, in this case, the ionization is explained using the perturbation theory, where the perturbation order corresponds to the number of photons absorbed with exponentially decreasing probability over the photon number needed. However, in the case of very strong fields, the perturbation theory does not hold anymore and an ionization plateau is observed [83]. This happens when the ionization enters the tunnelling ionization regime. This is the first step of the HHG process and it occurs when the laser electric field strength is comparable to the atomic electric field strength.

The pioneering work in understanding tunneling ionization was done by Keldysh [84]. According to the Keldysh theory, the tunneling ionization dominates over multi-photon ionization if the Keldysh parameter \(\gamma\) is less than unity:
\[ \gamma = \sqrt{\frac{I_p}{2U_p}} \]

with \( I_p \) being the ionization potential of an atom. As a general rule of thumb, all harmonics with photon energies above the ionization potential of the target will lie in the non-perturbative ionization regime.

For the exact calculation of the ionization rates, time-dependent Schrödinger equation (TDSE) for the atom with the external driving field needs to be solved. This normally can be done numerically, however even by using a single-active electron approximation, the calculation is extremely involved and is computationally highly demanding. One of the most successful models in simplifying this complex task was developed by Perelomov-Popov-Terent’ev (PPT) [85]. Even this treatment is quite involved. Because of this often a simplified version of it is used, which was derived by Ammosov-Delone-Krainov (ADK) [86]. Although, the ADK model works well in the tunneling regime, for HHG calculations one first needs to reach the intensity necessary for the tunneling regime. Therefore, a large number of electrons below the tunneling regime are ionized as well. Because of this, the ADK model will give a wrong absolute estimate for the ionization fraction of the medium, which is crucial for HHG phase-matching calculations (described later). Because of this reason, we resort only to the PPT model which we checked to match well the available TDSE calculation results in terms of predicted ionization rates. The PPT model was implemented following the equations from [87] and included the generalized Coulomb correction term [88]. Figure 2.8 shows ionization probability prediction of a helium gas target with 800 TW/cm\(^2\) intensity and 2200 nm central wavelength pulses. The PPT ionization rates are usually calculated for cycle averaged (CA) pulses (no sub-cycle resolution). However, to have a rough estimate for the sub-cycle ionization rates, the cycle averaging can be omitted. For comparison, on the same plot the ADK model prediction is plotted. It is worth noting, that these models are valid only for linearly polarized electric fields. The PPT ionization rate calculation for a single point can normally take up to a few seconds on a modern computer. For this reason, the code was used to generate a look-up table for a very rapid ionization rates estimates used in the high-intensity beam propagation simulations and HHG phase-matching calculations.
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Figure 2.8. Strong-field ionization with a few-cycle pulse. Left axis: integrated on-axis ionization probability predicted by PPT, cycle-averaged PPT and ADK models for helium atoms at 800 TW/cm² intensity and 2200 nm central wavelength. Right axis: the electric field waveform used for the calculations.

2.3.3 Strong-Field Approximation

The three-step model is well-suited for developing an intuitive understanding of the HHG process. However, it is not sufficient for obtaining the necessary atomic response function which is needed for making quantitative HHG predictions. Solving the TDSE could allow for this function, however it is numerically too demanding. Hence a simplified theory is needed which could include the quantum effects, such as wavepacket diffusion and quantum interferences of different trajectories. Lewenstein et al. [70] has developed such quantum treatment of the HHG process under the so-called the strong-field approximation (SFA).

Considering only a single-active electron and assuming a linearly polarized electric field, the TDSE in atomic units and in length gauge for an electron interacting with a laser field $F(t)$ and an ion with a potential $V_a(x)$ can be written:

$$i \frac{\partial |\Psi\rangle}{\partial t} = \left[ -\frac{1}{2} \nabla^2 + V_a(x) - x \cdot F(t) \right] |\Psi\rangle. \quad (2.30)$$

In case of no external laser field, the ground state $|0\rangle$ would evolve with a propagator $U_0 = e^{iI_p t}$, where, the $-I_p$ is its energy. In case of no atomic field being present, the $|p\rangle$ unbound electron would evolve in the external electric field with a Volkov propagator $U_V = e^{-i\Phi(p,t)}$, where

$$\Phi(p,t) = \int_{t_i}^t E_K(t')dt' = \frac{1}{2} \int_{t_i}^t p(t')^2 dt' = \int_{t_i}^t [A(t') - A(t_i) + p_i]^2 dt'.$$ \quad (2.31)
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Here $A(t)$ is the vector potential, $p_i$ is the initial momentum of the electron. This solution can be understood as a plane-wave of energy $E_K(t)$ propagating in a continuum.

Given these two propagators an ansatz for the TDSE can be constructed:

$$|\Psi\rangle = a(t)|0\rangle e^{ip\cdot t} + \int d^3pb(\vec{p},t) |\vec{p}\rangle,$$

(2.32)

where $a(t)$ is the population of the ground state and $b(\vec{p},t)$ describes a wave-packet. This kind of ansatz assumes that other bound states can be ignored and that the electron in the continuum can be propagated without the influence of the atomic potential (hence the SFA). Furthermore, the $a(t)$ can be calculated using PPT ionization rates discussed before, however in case of undepleted HHG (such that only a small fraction of the atoms is ionized) $a(t)$ can be assumed to be unity.

This ansatz can be back substituted into the TDSE. By noting the orthogonality between the ground state and the continuum state, calculating the inner product with $|\vec{p}\rangle$, remembering that the position operator in the momentum space acts like a derivate and denoting the dipole matrix element as $d_x(\vec{p}) = \langle \vec{p}|x|0\rangle$, the equation describing the $b(\vec{p},t)$ can be acquired. All the information about the atom remains in the dipole matrix element which indicates the bound-continuum transition. The $b(\vec{p},t)$ can further be analytically solved to acquire a closed expression for the $b(\vec{p},t)$. Then finally the $b(\vec{p},t)$ expression can be used in the ansatz to calculate the time-dependent dipole term $\langle \Psi|x(t)|\Psi\rangle$. The calculation is quite involved and more details are given in [70]. However, the main solution is the time-dependent dipole response function which describes a single-atom response to a strong laser field. Here I would like to present a version from [89], which I find to be more intuitive than in the original paper:

$$x(t) = 2Re \left\{ i \int^t_{-\infty} dt' \left( \frac{\pi}{\epsilon + i(t-t')} \right)^{3/2} \times \right.$$

$$\left. \times d^\ast(p_{st}(t',t) - A(t))d(p_{st}(t',t) + A(t'))F(t')e^{-iS_{st}(t',t)} \right\}$$

(2.33)

Here, $t'$ is the tunnel ionization time, $\epsilon$ is a small regularization constant to avoid singularity in the integral for numerical calculations. The $p_{st}(t',t) = \frac{1}{t'-t} \int^t_{t'} A(t'') dt''$ and finally the quasi-classical action of the trajectory is described by:

$$S_{st}(t',t) = (t-t') \left( I_p - \frac{p_{st}^2}{2} \right) + \frac{1}{2} \int^t_{t'} A^2(t'')dt''.$$  

(2.34)

For hydrogen-like atoms, the dipole matrix element can be written as:

$$d(p) = \frac{i2\pi(2I_p)^{3/2}}{\pi} \frac{p}{(p^2 + 2I_p)^{3/2}}.$$  

(2.35)
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In Figure 2.9, the dipole response of helium atoms is plotted for different driving wavelengths. The cut-off is found to scale proportional to $\lambda^2$. However, one can observe a dramatic decrease in dipole response strength if mid-IR driving sources are used. Furthermore, if gases with higher $I_p$ are used, such as helium, the dipole response decreases. However, for achieving an observable flux of high-harmonic radiation, it is necessary to phase-match the generation process. This imposes constraints on the possible ionization fraction of the medium. Therefore, the cut-off scales partly with intensity and thus the highest cut-off harmonics are achievable with the highest $I_p$ targets but at the trade-off of a lower flux.

![Figure 2.9. Dipole response in frequency domain versus driving wavelength. Intensity and pulse duration were fixed at 500 TW/cm$^2$ and 15 fs, respectively. Calculations were performed for the helium atom ($I_p = 24.6$ eV).](image)

2.3.4 Phase-Matching of High-harmonic Generation

Having understood the single-atom response via the SFA model, we can apply it to practical conditions. In practice, the high-harmonic generation occurs in a macroscopic ensemble of atoms, which therefore leads to a coherent addition of the emitted waves. To keep the phase-synchronicity of the generated harmonic radiation within the interaction volume one has to consider the full macroscopic picture of the harmonic generation. If the phase-matching condition is satisfied, the high-harmonic generation flux will grow proportional to the target pressure squared, i.e. $\propto P_n^2$. This follows from the fact that $P_{n}(t) \propto N_{\text{atoms}} \chi(t)$, and since the intensity scales as the square of the electric field, thus leading to flux growing $\propto N_{\text{atoms}}^2$, therefore the quadratic pressure scaling relation. Given a very weak dipole response around the cut-off frequencies, the phase-matching is essential for generating sufficiently intense attosecond pulses, which could be used for experiments with a high signal-to-noise ratio.

Non-phase-matched HHG was demonstrated multiple times, but either it involved very sensitive detection schemes [90] or ultraviolet driving lasers, where the dipole response is much stronger [91]. For cut-off scaling and isolated attosecond pulse generation associated reasons, we will focus only on the phase-matched HHG.
Starting from the Helmholtz equation with a nonlinear source term \( \hat{P}_{nl}(\omega) = FT[|N_{\text{atoms}}x(t)|] \), under the plane-wave Ansatz and SVEA, we can derive the \( q \)-th harmonic complex amplitude \( \hat{A}_q \) equation [81]:

\[
\hat{A}_q(z) = -\frac{i\mu_0\omega_q^2}{2k_q} \int_0^z e^{i\Delta k_q z} e^{-\alpha_q z} dz .
\]  (2.36)

Here \( \Delta k_q \) is the phase-mismatch term between the driving field \( k_1 \) and the \( q \)-th harmonic: \( \Delta k_q = k_q - qk_1 \). \( \alpha_q = \frac{\omega_q}{c} n_f(\omega_q) \) is the absorption coefficient defined by the imaginary part of the refractive index \( n_f(\omega_q) \). From the equation it can be seen that the harmonic signal will build up if \( \Delta k_q \approx 0 \) and absorption is low. The phase-matching of the HHG can be evaluated by considering the contributing terms to the phase-mismatch between the driving field and the generated high-harmonic radiation:

\[
\Delta k_q(z,t,p) = \Delta k_{\text{atomic}}(z,t,p) + \Delta k_{\text{SPM}}(z,t,p) +
+ \Delta k_{\text{plasma}}(z,t,p) + \Delta k_{\text{geo}}(z) + \Delta k_{\text{dipole}}(z).
\]  (2.37)

The first phase-mismatch term \( \Delta k_{\text{atomic}} \) is caused due to dispersion of the neutral atoms because the phase velocities between the high harmonic and driving radiation are different:

\[
\Delta k_{\text{atomic}}(z,t) = q\omega_q \left( 1 - \eta(z,t) \right) \times \\
\times \left( n_{\omega_q}(z,t,p) - n_{\omega_1}(z,t,p) \right) \approx -q\omega_q p\delta n \left( 1 - \eta(z,t) \right),
\]  (2.38)

where \( \eta(z,t) \) is the ionization fraction of the medium (effectively reducing the number of atoms), \( n_{\omega_q} \) is the refractive index of the medium seen by the EUV/SXR radiation. \( n_{\omega_1} \) is the refractive index experienced by the driving field and \( \delta n = n_{\omega_1} - n_{\omega_q} \) is the refractive index difference between optical and EUV/SXR frequencies.

The second order correction to the atomic phase term is the Kerr nonlinear refractive index contribution:

\[
\Delta k_{\text{SPM}}(z,t) = -q\omega_q pm_2 I(z,t) \left( 1 - \eta(z,t) \right).
\]  (2.39)

Here, \( n_2 \) is the nonlinear index of the target gas and \( I(z,t) \) is the intensity of the driving field. Normally, the SPM contribution for mid-IR driven HHG is relatively small, but it can be important for shorter wavelength driven harmonics.

The third term in the phase-mismatch equation is associated to the free electrons generated during the HHG process. The plasma dispersion can be estimated using the Drude model, approximating EUV/SXR refractive index to 1 and performing the expansion leads to a phase-mismatch contribution:

\[
\Delta k_{\text{plasma}}(z,t) = q\eta(z,t)r_e N_{\text{atm}} P \lambda.
\]  (2.40)

Here \( r_e \) is the classical electron radius, \( N_{\text{atm}} \) is the particle density and \( \lambda \) is the driving wavelength. Although, the atomic and plasma terms are opposite in sign, it
is not possible to achieve the phase-matching with these terms alone. Since phase-mismatch due to plasma and neutral gas scales the same way, the terms can be combined into a single dispersion term:

\[
\Delta k_{\text{atomic}}(z, t) + \Delta k_{\text{plasma}}(z, t) \equiv \\
\Delta k_{\text{dispersion}}(z, t) = -q_p \frac{2\pi}{\lambda} \delta n \left( 1 - \frac{\eta(z, t)}{\eta_{\text{crit}}} \right), \text{with} \ (2.41)
\]

\[
\eta_{\text{crit}} = \left( 1 + \frac{\eta N_{\text{atm}} \lambda^2}{2\pi \delta n} \right)^{-1}. \ (2.42)
\]

Here \(\eta_{\text{crit}}\) is the critical ionization, which meaning will soon become apparent. The next term in the balance equation is the geometrical mismatch term \(\Delta k_{\text{geo}}(z)\). For a free-focusing configuration, it is due to the Gouy-phase shift \([92]\):

\[
\Delta k_{\text{geo-free-focus}}(z) = (q - 1) \frac{2}{b \left( 1 + \frac{(2z)^2}{b^2} \right)^2}. \ (2.43)
\]

Here \(b\) is the confocal parameter and \(z\) is the propagation axis. This equation is valid only on-axis. In case instead of a free-focus a waveguide is used, then the geometrical term is \([93]\):

\[
\Delta k_{\text{geo-waveguide}} = \frac{q u_{11} \lambda}{4\pi a^2}. \ (2.44)
\]

Here \(u_{11}\) is mode factor, and \(a\) is the waveguide inner radius. Although hollow-core waveguide appears to be a very appealing choice for the HHG due to the possibility to extend the interaction length and possibly implement quasi-phase-matching, the high-repetition rate mid-IR driven HHG experiment execution would be very challenging. The waveguide attenuation coefficient \([94]\) scales as \(\lambda^2/\alpha^3\) and due to long wavelength and relatively low pulse energies a small radius would be needed. This leads to attenuation coefficients on the order of 1 mm thus basically no guiding at all.

The last term in the balance equation is the dipole phase term. It is an intrinsic phase to the HHG process and it is associated to the phase electron acquired in the continuum. In the previous sections we saw a distinction between short and long trajectories. This difference is reflected in the dipole phase coefficient \(\alpha\) which is first computed numerically. According to calculations presented at \([92]\) for 800 nm driving wavelength for short trajectories \(\alpha \approx 1.0 \times 10^{-14} \text{ rad cm}^2 \text{ W}^{-1}\), for the cut-off where trajectories merge \(\alpha \approx 13.7 \times 10^{-14} \text{ rad cm}^2 \text{ W}^{-1}\) and for long trajectories \(\alpha \approx 24 \times 10^{-14} \text{ rad cm}^2 \text{ W}^{-1}\). The dipole phase is proportional to \(\lambda^3\) due to longer trajectory \([95]\). The on-axis phase-mismatch term for a Gaussian beam then can be written as:

\[
\Delta k_{\text{dipole}}(z, t) = -\alpha \frac{\partial l}{\partial z} = \frac{8\pi a l_{\text{peak}}}{b^2 \left( 1 + \left( \frac{2z}{b} \right)^2 \right)^2}. \ (2.45)
\]
Here $I_{\text{peak}}$ is the peak intensity. The dipole phase is the only term which is asymmetric with respect to the focus position thus it allows to choose which trajectories to prefer in the phase-matching. It is worth noting, that the dipole phase is 0 at the focus. This allows to derive an intuitive equation for phase-matching pressure $p$ at the focus:

$$0 = -qp \frac{2\pi}{\lambda} \delta n \left( 1 - \frac{\eta(z, t)}{\eta_{\text{crit}}} \right) + (q - 1) \frac{2}{b},$$

leading to

$$p = \frac{q}{q - 1} \frac{\lambda^2}{2\pi^2 w_0^2 \delta n \left( 1 - \frac{\eta(z, t)}{\eta_{\text{crit}}} \right)} \approx \frac{\lambda^2}{2\pi^2 w_0^2 \delta n \left( 1 - \frac{\eta(z, t)}{\eta_{\text{crit}}} \right)}.$$  \hspace{1cm} (2.46)

This equation suggests that the phase-matching pressure for HHG scales as $\lambda^2$ and $1/w_0^2$. Therefore, this indicates that high-repetition rate mid-IR source driven HHG will require very high pressures to achieve phase-matching. It is also worth noting that if the $\eta(z, t) \geq \eta_{\text{crit}}$, then phase-matching pressure becomes infinitely large and thus no more phase-matching is possible. This is a very important observation since it suggests that if the laser intensity is too high, the HHG process will not be efficiently phase-matched. Thus, the efficient HHG requires a very intricate balance between ionization rate, focal position, confocal parameter and target gas pressure. Figure 2.10a shows the phase-matching terms for mid-IR driven HHG in spatial on-axis coordinate at $t=0$ at the peak of the pulse. In this case, the laser is calculated to be focused to 35 $\mu$m radius (3.5 mm confocal parameter) and 485 TW/cm$^2$ intensity. This yields a maximum of 0.05% ionization, whereas 0.06% is critical ionization. Such conditions should lead to cut-off extending to 720 eV. To phase-match such harmonics 39 bar pressure would be required. Figure 2.10b shows the temporal picture of phase-matching terms at $z=0$ position. In this case, a 15 fs pulse was used for the calculation.
2. Theoretical Foundations

Figure 2.10. HHG phase-matching terms. (a) contribution from different phase-matching terms in spatial on-axis coordinate at t=0. (b) contribution from the phase-matching terms in time-domain at z=0. The calculation details are explained in the text.

HHG phase-matching is strongly varying both in space and time and thus a complete spatiotemporal picture is more informative. Figure 2.11a shows the spatiotemporal phase-matching window for the discussed HHG case. The picture suggests that the HHG would be efficiently phase-matched over 1 mm propagation distance. However, this calculation does not consider plasma defocusing effects. In section 5.1.3 I will address the plasma associated limitations in more detail.

Figure 2.11. HHG in helium calculation. (a) Spatiotemporal HHG phase-matching picture. (b) Absorption length for high-harmonic radiation at 39.7 bar pressure of helium.

Furthermore, the absorption of the generated EUV/SXR radiation can be the limiting factor in flux. Figure 2.11b shows that for the discussed case the absorption length is around 6-8 mm around the cut-off. This absorption of the generated light will limit the achievable HHG flux and is further discussed in section 5.1.4.

2.3.5 Scaling of High-Harmonic Generation
From the SFA and phase-matching considerations, we can also infer some general scaling laws for the HHG. Figure 2.12a shows calculated cut-off scaling where at every wavelength the intensity is adjusted to have the ionization fraction close to 85% of the critical ionization. Since the dispersion is less for the mid-IR wavelengths, the critical ionization also decreases. This leads to lower possible intensities while still preserving phase-matching possibility. Because of this, the actual
Cut-off scaling is less than \( \propto \lambda^2 \) and more precisely it is \( \propto \lambda^{1.6} \). Similarly, we can also compare the expected HHG yield scaling. Figure 2.12b shows the calculated relative yield scaling where the cut-off was tracked and for every driving wavelength point 5\% of fractional bandwidth around the cut-off was integrated. This leads to the single-atom response scaling around the cut-off \( \propto \lambda^{-9.3} \). Similar single-atom response scaling around cut-off was reported by [21,96]. In contrast, authors from [27] predicted \( \propto \lambda^{-5.5} \) single-atom yield scaling for a fixed energy interval at a fixed intensity. Since for mid-IR HHG the phase-matching pressure grows close to \( \propto \lambda^2 \), the phase-matched flux then will grow with a square of the pressure, thus close to \( \propto \lambda^4 \). With a more refined phase-matching and reabsorption estimates discussed in 5.1.5, the phase-matched harmonic at cut-off yield for helium gas target can be estimated to scale with \( \propto \lambda^{-3.6} \).

Figure 2.12. Scaling of high-harmonic generation. (a) Phase-matched HHG cut-off scaling for a fixed ionization fraction. (b) Expected HHG flux scaling derived from SFA, phase-matching considerations and by integrating around the corresponding cut-off. Red points are calculated using SFA.
Chapter 3

High-Repetition-Rate Lasers for Attosecond Science

The next generation attosecond technology will heavily rely on high repetition rate laser systems providing attosecond pulses via the HHG process. If we consider that driving pulse energies of at least 100 $\mu$J are needed to reach relatively efficient HHG for high-photon energies, then at 100 kHz repetition rate, at least 10 W average power (GW-class peak power pulses at few-cycle pulse duration) mid-IR systems are needed for driving the beamlines. Such high average power mid-IR systems with few-cycle pulse duration were not demonstrated when I started my PhD work and when writing the thesis - only a few were available world-wide (one being ours). I expect that many more similar systems will emerge in the near future, especially because various companies started to offer similar systems commercially. These systems are particularly interesting for strong-field science and attosecond technology due to the extremely large pondermotive energies that the electrons can acquire during the strong-field light-matter interaction. Furthermore, as motivated in the introduction, due to a very large repetition rate, such systems could allow accessing much better SNR needed for precision measurements and fundamental studies.

In this chapter, I will begin by focusing on high-repetition-rate laser technology which enabled the development of our mid-IR OPCPA and near-IR OPCPA systems. I will discuss, in particular, the general aspects of mid-IR OPCPA system design. This was the most challenging and the least trivial part of the development which required multiple innovations before we could reach the required output parameters.

3.1 Key-Enabling Technology

As mentioned before, for efficient HHG with a large cut-off (> 100 eV) the pulse energies on the order of 100 $\mu$J – 1 mJ are desired. To achieve single isolated attosecond pulses at high-photon energies, a few-cycle driving pulse duration is needed. Hence, 10-GW-level peak powers are desired (this requirement is more clearly justified in section 5.1.2). Although HHG has been achieved with lower peak-power pulses, the cut-off was very low [97]. To scale the cut-off beyond 100 eV, a mid-IR driving laser is generally required, while to compensate for the decreasing flux in the mid-IR, a high average power is required. Hence here we have a set of requirements for the next generation attosecond beamline driving laser: GW-level peak power, high-repetition rate and few-cycle-pulse duration in the mid-IR wavelength regime.

It turns out that achieving all these requirements simultaneously is far from a straightforward task. We could try to consider starting from Ti:sapphire CPA systems. The state-of-the-art is a cryo-cooled multi-pass Ti:sapphire amplifier that can deliver more than 20 W average power 30 fs pulses [98,99]. Converting to the mid-
IR (>2 µm) from such amplifier pulses is possible only by pumping an OPCPA system. Such system could at best allow for 6 W output powers (quantum efficiency limited and assuming that the amplifier will run at 100% of its capacity). Power scaling with Ti:sapphire technology is very challenging due to expensive pulsed pump lasers, poor thermal lens performance of Ti:sapphire crystals and a relatively short upper-state lifetime of the ions in the crystal requiring pulsed pumping. Although a direct diode pumping was recently demonstrated with blue diodes [100], the blue diode output power is still comparably low. Hence, today, the state of the art attosecond beamlines are usually driven with 1 kHz cryogenically cooled Ti:sapphire amplifier which is pumping an OPA system. After a nonlinear pulse compression 200 GW peak power, pulses with 2.1 W average power can be achieved [28].

Another alternative is to use direct laser amplification in the mid-IR. The so-called "Ti:sapphire of the mid-IR", the Cr:ZnSe/Cr:ZnS are very promising gain materials. Few-cycle pulse durations in the mid-IR were achieved from mode-locked Cr:ZnSe oscillators and amplifiers [101]. Hence, by power-scaling this technology, it would be possible to achieve GW-level peak powers at high-repetition rates. However, so far, GW-peak powers have only been demonstrated at 1 kHz [102]. At least an order of magnitude in power scaling is still missing. The thermal properties of the Cr:ZnSe/Cr:ZnS gain material are not better than Ti:sapphire, but there is a fast emergence of suitable high-power pump lasers for these crystals such as Ho:YLF, Tm:YAG etc. So, although this technology could be useful in the future, during the course of my PhD studies, this technology was only developing.

The only remaining possible technology for this application is the Yb- and Nd-based amplifiers delivering sub-1 to 100 ps pulses at 1 µm with unprecedented powers which could be transferred to the mid-IR by using the OPCPA technique. At the start of my PhD work, one of the most powerful commercially available amplifiers was 300 W, 1 ps Yb:YAG Innoslab amplifier. A few years later, one can already buy multi-kW class Yb-based picosecond amplifiers (Trumpf GmbH, Amophos GmbH, Active Fiber Systems GmbH). The rapid progress of the technology is fueled by the demands for highly parallel micromachining technology and challenging scientific applications such as HHG and free-electron laser seeding. Furthermore, the Yb:YAG laser gain material is probably one of the most developed gain materials, allowing excellent power handling and supporting sub-ps pulses. If we consider 300 W as the pump power for an OPCPA pumping, then, considering the quantum defect, we could expect up to 100 W-class mid-IR systems to be feasible, which is certainly enough for bringing the attosecond science to the next level.

Here we very quickly drifted towards OPCPA as the only possible solution for the high-power, few-cycle mid-IR source. But it is worth to mention a few trade-offs associated with it. Although the OPCPA technology was the only solution for us, it still turns out to be an excellent technique when it comes to transferring high pump laser energies into any desired wavelength range. The amplification bandwidth is defined by phase-matching of the crystals, whose choice is broad and can also be engineered for any desired mid-IR wavelength range by using quasi-phase-matching (QPM) techniques. Furthermore, OPCPA works by exciting the atoms to virtual levels, hence no actual energy is stored in the crystals, making this technology a
great match for high-power pumping and high-repetition-rate applications. Finally, since some nonlinear crystals can be grown to large dimensions this technology is also an excellent choice for Joule-class, multi-PW peak power systems, holding records for the world’s most powerful laser systems [103]. The key drawback of the OPCPA technology is in its complexity due to the required perfect temporal synchronization between the pump and the seed.

3.2 The Pump Laser Choice

In the previous chapter, I motivated our choice to bring attoscience to the next level by using an industrial-grade laser at 1 µm and converting that power to the mid-IR spectral range. When it comes to choosing the right pump laser, there are few trade-offs to consider. Regarding the gain medium, Yb:YAG and Nd:YAG are the best developed and most advanced gain materials. However, the Nd:YAG provide narrower gain bandwidth, which could support pulses only down to 10 ps duration. Strictly speaking, this is not a problem for OPCPA, and actually, there are world-record OPCPA systems which are using Nd:YAG pump such as [104]. However, there is a trade-off: the damage threshold of materials in the ps regime scales as $\tau^{-1/2}$ [105], which will limit the possible intensities on the crystals. Higher intensity is desired for larger phase-matching bandwidth, as discussed in the theory chapter, and hence it is better to have a pump which is shorter than 10 ps. Following this reasoning, we chose to use a Yb:YAG based system, whose bandwidth can support down to sub-ps pulse durations.

When it comes to Yb:YAG systems, at the start of my PhD work three Yb:YAG amplifier technologies were competing in an industrial setting: thin-disk, Innoslab and fiber. All of these amplification geometries share a large surface area to volume ratio, allowing efficient 1-D heat flow and thus permitting high-power operation. Thin-disk regenerative amplifiers are challenging due to the required cavity switching and hence the maximum power available at the time was 200 W. Whereas with recent developments this obstacle seems to have been overcome and the maximum output power was increased further to 2.45 kW [106]. On the other hand, the thin-disk regenerative amplifier output is defined by the cavity, hence it allows for more reliable beam shape than the Innoslab geometry.

Meanwhile, the Innoslab-type amplifiers benefit from a high gain in multi-pass configurations and do not require Pockel's cells. However, the mode is highly dependent on the thermal lens from the pump light and can easily change with changes in the environment. Furthermore, the in-coupling and out-coupling at the gain crystal are geometrically constrained, leading to a clipping on the beam. This distorts the beam shape and requires beam cleaning later to recover an acceptable $M^2$ value in the so-called “bad axis”. Because of the geometry, the same “bad axis” corresponds to the lower brightness axis of the pump diode bars which are imaged to the crystal. For example, recently a 1.5 kW Innoslab amplifier was demonstrated, however after the beam cleaning and compression only 0.661 kW were remaining [107].
Finally, the fiber amplifiers were struggling in supporting large pulse energies due to high B-integrals. However, recently an efficient workaround was developed: a coherent combining of many fiber amplifiers allowing excellent power scaling to 3.5 kW level at high repetition rates [108]. This technique also enabled pulse energy scaling to 10 mJ pulse energy level with greater than 1 kW average powers (Active Fiber Systems GmbH). In recent years, also thulium fiber laser systems emerged delivering kW-level output powers at 80 MHz repetition rates [109] which will lead to exciting HHG results in the near future.

3.2.1 The Pump Laser Performance

For pumping our OPCPA system, we use an A400 Innoslab amplifier from Amphos GmbH. The system is seeded by taking part of a Ti:sapphire few-cycle oscillator (described in section 3.3) bandwidth around 1030 nm and coupling it to a fiber pre-amplifier which pre-amplifies the 1030 nm seed to 1 nJ pulses (Venteon Preamp) and then is used to seed the A400 amplification system. The A400 amplifier is actually a chain of amplifiers, including fiber amplifiers, a pair of AOM based pulse pickers to control the repetition rate, a fiber-Bragg-grating stretcher to stretch the seed pulses to around 100 ps duration, and a pair of bulk amplifiers to amplify the seed to a Watt-level. This pre-amplified seed is used to directly seed the Innoslab Yb:YAG gain medium. The A400 amplifier is capable of amplifying pulses to 440 W level.

The A400 output is compressed in an external compressor designed by Amphos GmbH. The transmission through the compressor (including the beam cleaning) is up to 68% allowing up to 300 W output. Using the grating compressor, the pulses are compressible below 1 ps duration, nearly to the transform limit.

The amplifier system can deliver up to 300 W of output power with excellent stability during a working day. When the amplifier is in a good condition one could expect well below 1% peak-to-peak power drifts during the day. This is outstanding power stability considering the power level of the system. However, the long-term output power stability (over many days) was one of the major issues during my PhD. We have found that normally the amplifier day-to-day output was always decreasing at an accelerating rate. This led to on average 2-4 service visits every year needed to recover the performance of the system and so in turn severely slowing down the progress of our system development. It is important to note that the amplifier requires 2-6 hours of thermalization time, because of which we keep it on all the time. After it thermalizes, the output stability is very good (<1% peak-to-peak drifts). Figure 3.1 shows the output power variation over 10 hours.
3. High-Repetition-Rate Lasers for Attosecond Science

As discussed in the introductory part of the chapter, due to the Innoslab amplification geometry, beam shape control is very challenging. Figure 3.2 below shows several output beam shapes we have managed to achieve and were working with. The beam shape could be improved by placing a “tail killer” in the Fourier plane of the amplifier plane. However, due to strongly evolving beam pointing of the amplifier output, the tail killing was not feasible. Due to this, for most of the time we worked without clipping the tail. Consequently, the desired intensity for the nonlinear stages was achieved by telescoping the pump, rather than focusing it down.

Figure 3.2. Output beam shapes achieved from the A400 amplifier. (a) Tail-clipped beam with visible fringes on the beam. (b) Best case example. (c) Typical output beam.

This kind of configuration proved to not be robust over the long term. After every service, the beam shape and divergence would be different and since we had a highly aberrated beam, the recovery of the same intensities on every nonlinear stage was not feasible without a significant beam reshaping. Because of this reason we have since reconfigured the compressor/tail killing of the system in such a way that the A400 output is directly stabilized on the tail killer (stabilization was done with a 4-D Aligna system from TEM Messtechnik GmbH). The tail killing is performed by focusing the beam in the “bad axis” and collimating back again into the compressor. After the compressor, the beam is actively stabilized again. This configuration is illustrated in Figure 3.3. A significant care was needed to minimize and control pump beam shape changes due to thermal and Kerr lensing effects.
The final amplifier configuration used to pump our OPCPA systems.

A significant effort from our side was invested in optimizing and characterizing the output pulse shapes. We have implemented both an SHG-based frequency resolved optical gating (FROG) and cross-correlation (with Ti:Sapphire) measurement setups. From these measurements, we could observe pre-pulses to the main pulse mainly originating from a large negative TOD remaining on the pulse. The TOD sign was verified by performing a cross-correlation between the Ti:sapphire output pulses and the A400 compressed output. This limited the extractable energy during the OPCPA process.

Such large TOD could only be coming from a mismatch between the stretcher and the compressor pair. To address the pre-pulse problem, we carefully analyzed the compressor. After complete realignment, changing of any damaged gratings and optimizing the angle of incidence (AOI) on the gratings we finally were able to achieve much better pulse shape. The TOD on the pulse could be reduced by decreasing the AOI and changing the separation between the gratings. However, due to the diffraction efficiency of the grating rapidly dropping at AOI other than 44-45 degrees, operation at lower AOI came along with a trade-off in the compression efficiency. Finally, after optimizing AOI the remaining TOD of -0.7 ps$^3$ could be achieved. The pulses could be compressed near the Fourier-limit of 950 fs. However, as discussed before, the shorter pulse is not necessarily more optimal. Hence, we have decided to stretch the pulses to 2 ps duration shown in Figure 3.4 with the estimated peak power factor being 0.88 and the corresponding peak power of 1.2 GW.
3.3 The Seed Laser

We have discussed at the beginning that the new generation attosecond beamline driving laser should produce few-cycle pulses in the mid-IR. For example, 2.5 μm central wavelength would correspond to 120 THz frequency and single cycle duration would be 8.4 fs. A mid-IR bandwidth of 1.7–3.2 μm (83 THz) would support 12 fs pulses (1.4 cycle duration with the carrier frequency at 120 THz). There are multiple possible solutions which could deliver such bandwidth.

The most commonly employed option is to seed the OPCPA with a white light generated by the pump laser. Such seeding scheme is the most often employed in the cases where the pump pulses are of 30-500 fs duration [110]. If ps-long pump pulses are used, then the generated white-light can consist of multiple pulses [111], thus complicating the OPCPA seeding possibilities. This approach is cost-effective, since it does not require a broadband oscillator and it also ensures perfect synchronization between the seed and the pump. However, it is also a risky solution given our pump laser instabilities. If the pointing, beam size or power slightly evolve it can easily change the filament formation in the crystal and lead to evolving spectrum and phase. Furthermore, the bulk material would see a relatively high intensity and would be likely to damage due to the high repetition rate. One of the recently published mid-IR OPCPA systems prevents damage by continuously rotating the material [112]. However, such a solution is not particularly straightforward.

Another option would be a direct seeding from another oscillator. This, for example, could be a few-cycle Cr:ZnSe oscillator. However, the technology of Cr:ZnSe oscillators was not yet mature at the start of my PhD, and only recently few-cycle oscillators were reported [101].

A third option could be to make use of an OPO based seed generation. This could be a master oscillator at 1030 nm delivering 100 – 200 fs pulses at 80 MHz repetition rate and 10–20 W average power. A small fraction of the power would be used to seed the A400 and the remaining power would pump an OPO. With proper
engineering of the OPO, it is possible to have better than 10-50% power conversion to mid-IR offering freely tunable ultra-broadband bandwidth nJ seed for the mid-IR OPCPA [113]. Although this approach seems very attractive, no OPCPA similar to that was demonstrated so far. Furthermore, a few-cycle OPO is very sensitive to pump-cavity synchronization and could be challenging to maintain and develop. Finally, such an approach would not guarantee a CEP stability of the system and would require for additional fast locking loops.

Finally, an alternative to the OPO seeding is using the well mature and established Ti:sapphire few-cycle technology. Ti:sapphire oscillators are excellent at producing few-cycle pulses at low powers and are readily available from commercial vendors. Hence, a few-cycle Ti:sapphire oscillator could produce a broad enough bandwidth to seed the A400 and the OPCPA simultaneously. The Ti:sapphire oscillator center wavelength is usually around 800 nm (375 THz) and the few-cycle operation bandwidth can exceed 150 THz, which, when transferred to mid-IR via DFG processes can support sub-cycle pulses (since the cycles are longer). Furthermore, such solution takes care of seed/pump synchronization, while passive CEP stabilization is possible via the DFG process. Finally, the Ti:sapphire oscillators can be directly CEP stabilized and thus another CEP stabilized system at the near-IR spectral region could be developed in parallel. Therefore, due to these considerations we chose to develop a Ti:sapphire oscillator seeded OPCPA.

It is worth mentioning that a direct full bandwidth seeding is not always necessary since nonlinear compression in the mid-IR is possible. This could be done by filamentation in gases, where the mid-IR beam is guided in a hollow-core waveguide. The waveguide can be a simple hollow glass tube or a microstructured fiber. Such nonlinear compression techniques were demonstrated for high energy pulses at low repetition rates [28,114,115], as well as at high-repetition rates for relatively low pulse energies [52]. Although this technique could yield ultrashort pulses, the overall peak power will be limited due to the high-order dispersion distorting the pulse and raising the temporal pedestal. Furthermore, such pulses become hard to route to different experimental setups, since any additional dispersion would need to be compensated.

3.3.1 Ti:sapphire Oscillator Performance
For our purposes, we have acquired a commercially available Ti:sapphire oscillator (Venteon Pulse One, Laser Quantum GmbH) delivering pulses shorter than 6 fs with 210 mW average power at 82 MHz repetition rate. The oscillator pulse bandwidth extends up to 1100 nm. Using a partially reflective mirror and an interference filter a 1030 nm part of the spectrum was selected and used for seeding a fiber pre-amplifier (Venteon Pre-amp, Laser Quantum GmbH). The output beam of the oscillator had small but visible wings, which, when focused interfered with the main beam. To avoid this issue, we inserted an aperture removing these unwanted features from the beam.

The laser is actively carrier-envelope offset (CEO) frequency stabilized by locking the CEO frequency [116] to a ¼ of the repetition rate. This is done by using an
3. High-Repetition-Rate Lasers for Attosecond Science

f-to-2f nonlinear interferometer and providing feedback to the oscillator. Furthermore, the pulse picker at the A400 amplifier picks every \( n^{th} \) pulse such that \( n \text{ mod } 4 = 0 \), ensuring that the OPCPA amplified pulses at 100 kHz have the CEO frequency of 0. This was needed for the near-IR OPCPA system to be carrier-envelope phase (CEP) stable.

Finally, to prevent possible damage to the A400 system if the laser would go out of cw mode-locking, we have installed a CEO stabilization interlock. If the stabilization would be interrupted, the A400 pump diodes would switch-off.

3.4 The Final Architecture of the Setup

In the previous sections, I have discussed the lasers used to pump and seed the mid-IR OPCPA. In this section, I will discuss the general architecture of the set-up. The full conceptual layout of our laboratory is shown in Figure 3.5. The seed laser pointing was actively stabilized (TEM Messtechnik GmbH) and small fraction of the seed was sent to an active time-delay stabilization (TDS) unit to stabilize any temporal drifts between the seed and the pump (discussed in section 3.4.1). The remaining energy of the seed was sent into a pulse shaper which shaping aspects are discussed in section 3.5. The output of the pulse shaper was split with a beam splitter into two beams to seed two OPCPA systems. The first system was the mid-IR OPCPA described in detail in Chapter 4. The second OPCPA system we have developed in parallel was a high-power near-IR OPCPA described in Chapter 6. Having two systems developed in parallel allowed to address various uncertainties in advance and also opened much more possibilities for experiments. The near-IR OPCPA works at the usual and well-researched wavelength range but is delivering very-high-power pulses (>30 W), which were not available from OPCPA systems before I started my PhD. Working on the two high-power OPCPA systems allowed to troubleshoot high-power aspects and cross-check the physics at different wavelengths. The output of these systems was sent into our home-made beam routing system, where beams were transported near the ceiling to our experimental setups in another laboratory. Both, the near-IR and mid-IR OPCPA, systems were intended to be used for the HHG-based attosecond beamline as well as for strong-field ionization experiments with a high-resolution VMI spectrometer. Furthermore, the VMI experiments were planned to be done with targets which are prepared via deep-UV ionization. Thus, also a world-record UV (5th harmonic of Yb:YAG) source was developed by my colleagues [117].
3.4.1 Time-delay Stabilization

A crucial degree of freedom for the OPCPA systems is the timing between the seed and the pump. Although this flexibility is powerful in shaping the output pulses, at the same time, any pump-seed drifts are highly undesired. Since the Ti:sapphire laser is seeding the laser amplifier and the OPCPA, this already took care of all fast timing-jitter [118]. However, slow time-delay synchronization drifts remain. These drifts occur mostly due to small changes in the temperature of the fiber amplifiers. Even a small fraction of a degree of temperature change can change the effective path length by a few micrometers over the amplification chain containing tens of meters of optical fiber. During normal operation, we observe delay drifts of 500 fs, which are sufficient to significantly change the output of the system. To prevent this, I have implemented an active time-delay stabilization system (TDS).

There are various ways to stabilize the time-delay drifts such as balanced cross-correlation [119], two cross-polarized OPAs [120], a stretched seeding of a noncollinear OPA (NOPA) [121], idler position of a NOPA stage [44] and various other. For the delay synchronization of our systems, we had multiple constraints: (1) – independent stabilization for the near-IR and mid-IR OPCPA systems so that the systems can be developed without worrying about the delay changes, (2) – stabilization independent of pump intensity changes, (3) – very low (< 10 pJ) seed pulse energy requirement. Constraint (1) requires that the TDS system must pick-up the seed before the pulse shaper. Constraint (2) rules out any classical cross-correlation schemes. Constraint (3) rules out SFG based schemes due to very low seed pulse energies. Following these constraints, we chose to implement a system similar to [121], except that we have used a collinear OPA.
In our TDS system, the 1030 nm pump is frequency-doubled and a part of the Ti:sapphire seed is amplified in an OPA. Additionally, the seed is significantly stretched, such that a delay drift would be directly translated into a shift of the amplified spectrum. This effectively decouples the delay measurement from any changes to the pump power. The OPA output was sent on a grating and then the spectrum was observed on a position sensitive detector (PSD), such that when the delay shifts, the PSD centroid moves. The PSD centroid position was actively stabilized by providing feedback to a delay stage by using a home-built computer software. The TDS was implemented with a user-friendly format shown in Figure 3.6. The sensitivity of the system was estimated to be <20 fs. The stabilization loop was optimized to control only the slow delay drifts (on a scale of a second).

![Figure 3.6. Time-delay stabilization (TDS) implementation. (a) User interface of the TDS software. X position curve shows the system response function where a linear response to the delay scan is visible. This corresponds to the maximum of the Sum Voltage reading (total signal). (b) The actual amplified spectrum in the TDS system.](image)

**3.4.1.1 Detailed Description of the TDS**

The TDS is pumped with 1 W of 515 nm (SHG of 2.5 W 1030nm light in a 10 mm long LBO), and the OPA itself is in a 5 mm long BBO. The seed is derived from the Ti:sapphire output by picking-up part of the beam with a flat window. It is then stretched in 20 mm of propagation in ZnSe oriented at Brewster angle. Finally, the seed is combined with the pump using a dichroic mirror. The collinear OPA phase-matching bandwidth is 16 nm FWHM. Since only a small fraction of the bandwidth is available for the amplification, the effective seed energy can be estimated to be 4 pJ. This seed is amplified above the 82 MHz background with an estimated gain of 45 dB. Since the gain is very high, the effective amplification window in the time domain is shorter than 400 fs. Thus, effectively, the pump is much shorter than the seed. To use this OPA output for the time-delay stabilization, the amplified beam is spectrally dispersed on a grating and then a fraction of the relevant part of the spectrum is transferred on a position-sensitive detector. When the delay drifts, the detector records the change of the centroid. Figure 3.7a shows a small-signal gain calculation when the pump-seed delay is scanned. Figure 3.7b shows the TDS feedback during the thermalization of the pump laser.
The actual optimal TDS configuration parameters were not clear at the start of the system development. To find the optimal configuration I have performed numerical simulations where the centroid position of the spectrum was calculated versus delay. From the centroid slope, the sensitivity to the delay could be inferred. Figure 3.8a shows the sensitivity parameter for a collinear OPA based TDS. In contrast, Figure 3.8b shows the sensitivity changes in a NOPA configuration. The NOPA sensitivity to delay peaks around 1100 fs\(^2\) of the seed GDD and is more sensitive compared to the collinear OPA. Because of this, the final TDS for experiments was performed using NOPA configuration.

**3.5 Pulse-Shaping**

As any CPA systems, the OPCPA systems require often complex dispersion management schemes. To optimize the pump energy extraction, the seed is stretched to match the pump pulse duration, in every stage of the OPCPA chain. After the amplification, the pulses are compressed to reach high-peak powers. For few-cycle systems, even small amount of dispersion is enough to stretch the pulses significantly. Therefore, usually a programmable pulse shaper is employed at the beginning of the system so that dispersion of the output high-energy pulses could be fine-tuned.
without any significant losses. For high-repetition-rate systems which operate above 100 kHz, the main employed pulse shaper technology is based on spatial light modulators (SLMs). However, the main issue with SLM-based pulse shapers lies in the pixelated nature of the device which, if not used right, could cause distortions of the pulses. Normally, these distortions are tolerable if the applied phase is below $2\pi$. However, if larger phase is needed, such as in a typical OPCPA application, then the phase profile has to include phase-wrapping points which the pulse shaper will not be able to follow and thus will distort the output waveform phase. During my PhD, we have overcome this limitation of the pulse shaping device and our findings were published in a peer-reviewed journal. The publication is reprinted in the thesis and the reader is invited to read the publication to understand how the pulse shaper is used in our system.

3.5.1 Programmable Time-gated Pulse Shaping

We present our study on programmable time-gated pulse shaping in the following Optics Express publication [122] (© 2019 Optical Society of America).
Programmable pulse shaping for time-gated amplifiers


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Abstract: We experimentally demonstrate a novel use of a spatial light modulator (SLM) for shaping ultrashort pulses in time-gated amplification systems. We show that spectral aberrations due to the pixelated nature of the device can be avoided by introducing a group delay offset to the pulse via the SLM, followed by a time-gated amplification. Because of phase wrapping, a large delay offset yields a nearly-periodic grating-like phase function (or a phase grating). We show that in this regime the periodicity of the phase grating defines the group delay spectrum applied to the pulse, while the amplitude of the grating defines the fraction of light that is delayed. We therefore demonstrate that a one-dimensional (1D) SLM pixel array is sufficient to control both the spectral amplitude and the phase of the amplified pulses.

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1. Introduction

Femtosecond pulse shaping is nowadays an integral part of ultrafast optics technology. Pulse shaper applications are extremely broad and include time-resolved spectroscopy, laser-matter interaction experiments, metrology, machining of materials, optical communication and many others [123]. In complex high-power laser systems, the pulse shapers are typically an integral part of the front-end of the systems and play a major role in managing dispersion [44,49,124].

Spatial light modulator (SLM) based pulse shapers are ubiquitous, despite the fact that the pixelated nature of the device leads to various kinds of waveform aberrations [125]. The discrete phase sampling nature and the presence of gaps between the pixels leads to a time-domain comb of sampling replica pulses. Furthermore, another class of waveform distortion appears when a large phase is applied. Because SLM-based pulse shapers are usually capable of applying only up to $2\pi$ radians of phase, the effective large phase is applied by phase wrapping, i.e. $\phi_{\text{wrapped}} = \text{mod}(\phi_{\text{ideal}}, 2\pi)$. Phase wrapping leads to abrupt phase jumps, which some SLMs cannot exactly reproduce due to pixel crosstalk or limited spectral resolution. In the presence of such crosstalk, wrapping causes the phase to smoothly evolve from $2\pi$ to 0 over a finite region of the SLM, as illustrated in Fig. 1(a). This transition corresponds to a phase error and introduces aberrations on the shaped pulse. If a large phase slope versus frequency is applied, the wrapped phase can effectively be
considered as a periodic phase grating in the frequency domain. In time-domain this leads to an appearance of a series of “diffraction” orders, known as modulator replicas [125].

Which kind of waveform distortions dominate depends strongly on the type of the SLM pulse shaper design. Liquid crystal (LC) SLMs typically have a low number of pixels separated by large gaps leading to significant sampling replicas and weaker modulator replicas, as it was well studied in [125]. On the other hand, liquid crystal on silicon (LCoS) SLMs have smaller gaps leading to a relatively strong inter-pixel coupling [126]. Because of this property, sampling replicas are typically well suppressed while stronger modulator replicas emerge.

Modulator replicas can be avoided using spatial diffraction-based pulse shaping with a 2D-SLM [127]. Although this method additionally allows for amplitude shaping, it requires a 2D pixel array. It has also been demonstrated that a 1D-LCoS SLM can be used for both phase and amplitude control by oversampling the Fourier plane [128]. However, spectral distortions due to phase wrapping are still present in this technique. An alternative to the SLM-based pulse shaper is an acousto-optic programmable dispersive filter (AOPDF) [129]. AOPDFs are used in chirped pulse amplification (CPA) systems because they do not generate pulse replicas [130], and they can apply a large spectral phase while also controlling the spectral amplitude [50, 76, 131]. However, AOPDFs can be operated at a limited repetition rate and diffraction efficiency due to the need to continually generate new acoustic waves to modulate the acousto-optic crystal at the laser repetition rate. Currently, the highest repetition rate AOPDF-controlled system was demonstrated at 100 kHz [50].

In this paper, we experimentally demonstrate an alternative use of a one-dimensional (1D) SLM which allows for a simultaneous amplitude and phase control without spectral interferences from the replica pulses. Our approach applies to time-gated amplifiers, which include optical parametric chirped pulse amplifiers (OPCPAs) [25, 132] or frequency-domain optical parametric amplifiers (FOPAs) [133, 134]. In these systems amplification only occurs within the temporal duration of the short pump pulse, providing a time-gating effect. Here we show that by adding a large group delay (GD) offset on a phase-only 1D-SLM followed by a time-gated amplification, we can achieve a full amplitude and phase shaping of ultrashort pulses without aberrations or modulator replica pulses.

In section 2 we explain the scheme conceptually, while, in section 3 we demonstrate it experimentally using an OPCPA system. Finally, in section 4 we experimentally and theoretically demonstrate the amplitude shaping capability.

2. Time-Gated Filtering Scheme
The time-gated filtering scheme is conceptually illustrated in Fig. 1, where we sketch a pulse compression scheme using a pulse shaper followed by a time-gated amplifier. As discussed in the introduction, applying a phase larger than 2π radians on a pulse using a pulse shaper can lead to waveform distortions. Because the phase smoothly evolves from 2π to 0 due to crosstalk, as shown in Fig. 1(a), the spectral components around the 2π to 0 transition will experience a different phase than
originally applied, which will distort the waveform around the zero-delay. Here we overcome this problem by applying a sufficiently large GD to the SLM, such that the delayed light can avoid these temporal aberrations.

Fig. 1. a) Phase-wrapped group delay (GD) of 2000 fs. For illustrative purposes, the pixel crosstalk was simulated using a moving average filter with a width of 8 pixels. The snippet shows a zoomed in phase where deviation from the ideal wrapped phase (dashed black line) is clearly visible. b) Conceptual illustration of how we can increase the engineered waveform quality by adding a GD on the pulse with the pulse shaper and amplifying the delayed pulse using a time-gated amplifier.

As an example, suppose we wish to impose a spectral phase profile $\phi_{\text{target}}(\omega)$, onto a pulse in order to optimize its compression. Phase wrapping and pixel crosstalk will lead to aberrations, and these aberrations will distort the shaped pulse (see Fig. 1(b), usual pulse shaping). However, by applying a large GD offset, the corresponding ideal phase $\phi_{\text{ideal}}(\omega)=\phi_{\text{target}}(\omega)+GD \cdot (\omega-\omega_0)$ will be a nearly-linear function (here $\omega_0$ is the center frequency). The subsequent phase wrapping operation will yield a nearly periodic $\phi_{\text{wrapped}}(\omega)$ phase. This wrapped phase resembles a phase grating in the frequency domain. However, due to the pixel crosstalk, the amplitude of the phase grating $\phi_{\text{wrapped}}(\omega)$ will be reduced and smoothed as we show in Fig. 1(a). This periodic deviation from an ideal linear spectral phase will lead to the appearance of the modulator replicas in the time domain. From the Fourier theory, these replicas are temporally delayed by integer multiples of the applied GD offset, where the GD offset is the inverse of the period in frequency. The modulator replicas are illustrated in Fig. 1(b) (“shaped” waveform). Although the GD offset leads to replicas, the waveform can effectively be cleaned using a temporal filter. For example, a time-gated amplifier can be controlled to only amplify the first replica, which is free of the temporal aberrations appearing around the zero-delay, as shown in Fig. 1(b) (“output” waveform).

3. Experimental Setup

Fig. 2 shows our experimental implementation. Here we use a part of our already reported OPCPA chain [49,135] and we focus on the pulse shaping aspect in more detail. A reflective liquid crystal on silicon (LCoS) SLM is placed in a Fourier plane of a 4-f pulse shaper setup. The SLM is a one-dimensional array of 12288 pixels (BNS-Linear-12288, Meadowlark Optics Inc.). The pixel dimension is 1 μm horizontally and 19.66 mm vertically with 0.6 μm gaps between the electrodes. The
pulse shaper is used to control the spectral phase and amplitude of a Ti:sapphire oscillator output (Venteon Pulse: One < 6 fs, Laser Quantum Ltd.) operating at a repetition rate of 82 MHz. To increase the spectral resolution of the Fourier plane and thereby leverage the large number of pixels available, we expanded the Ti:sapphire output beam in the horizontal axis to obtain a radius of 1.4 mm ($1/e^2$) while the vertical axis has a radius of 0.7 mm before entering the pulse shaper. The beam is spectrally dispersed using a reflection grating with 600 grooves/mm (10RG600-800-1, Newport Corp.) and spectrally focused in the horizontal axis on the SLM with a cylindrically curved mirror of 100 mm focal length. The SLM is slightly vertically tilted so that the returning beam is reflected with a D-shaped mirror.

![Image of experimental setup]

**Fig. 2.** Our experimental setup. A reflective 4-f pulse shaper is used to control the phase of the seed for a noncollinear optical parametric amplifier (NOPA). The geometry of the pulse shaper setup (dashed box) is as follows. The incident beam is first reflected from the diffraction grating, then a D-shaped mirror (D1) directs the angularly chirped beam towards the cylindrical mirror. The cylindrical mirror is tilted upwards so that the reflected light reaches the spatial light modulator (SLM). The SLM is tilted downwards so that the returning beam is reflected by a second D-shaped mirror (D2). For time-gated amplification, a narrow-band pump pulse (wavelength 1030 nm) is frequency doubled in a second harmonic generation (SHG) stage and is used to amplify the seed in the NOPA.

Part of the Ti:sapphire oscillator bandwidth around 1030 nm is used to seed an Yb:YAG based amplifier chain (A400, Amphos GmbH) which yields high-power 1.8 ps pulses at 100 kHz. A fraction of the amplifier output power is frequency doubled, and this 515-nm pump beam is used to amplify the shaped Ti:sapphire pulses in a noncollinear optical parametric amplifier (NOPA). In this paper we use the NOPA to perform a time-frequency analysis of the shaped near-IR pulses by varying the
pump-signal delay. The NOPA, which is operated in a non-saturated regime, is based on a 1.8-mm-thick BBO crystal configured for type-I phase-matching. We characterize the amplified signal pulses with a spectrometer and a second harmonic frequency resolved optical gating (SH-FROG) [136]. Because the NOPA was optimized for generating the mid-infrared seed pulses for the above-mentioned OPCPA chain, only spectral components between 650-800 nm are amplified. However, the implemented pulse shaper supports a larger bandwidth of 640-930 nm.

The pulse shaper acts as a main dispersion control element for our OPCPA system. Such OPCPA systems usually require a relatively large phase tuning range. For example, to compensate for a third-order dispersion (TOD) of 50000 fs$^3$ over the 650-800 nm spectral range for a central wavelength of 745 nm, 160 radians of phase need to be applied as it is illustrated in Fig. 3(a). However, as explained in section 2, this requires phase wrapping and leads to modulator replica pulses and corresponding spectral interferences. To illustrate this issue, in Fig. 3(c) we show the NOPA amplified spectrum versus delay between the pump and the seed for an example configuration. Positive delay values correspond to the pump pulses arriving after the seed pulses. In the example shown, we have applied -50000 fs$^3$ TOD at a central wavelength of 745 nm. From the Fig. 3(c), it can be clearly seen that some spectral components are temporally spread out around the main pulse. Fig. 3(a) shows the phase profile which we wrapped and sent to the SLM device. The rapid change in phase derivative due to phase wrapping corresponds to the spread-out temporal features in the measurement. These waveform distortions can be attributed to the smoothed phase wrapping due to pixel crosstalk as discussed in section 2. We validated this by simulating the influence of a wrapped and smoothed spectral phase profile (Fig. 3(b)) by performing a time-frequency analysis on a simulated time-domain representation of the waveform (Fig. 3(d)).

![Fig. 3. (a) Ideal phase of -50000 fs$^3$ third-order dispersion (TOD) at a center wavelength of 745 nm. (b) Wrapped and smoothed phase from (a). (c) Noncollinear optical parametric amplifier (NOPA) amplified spectrum versus pump delay for a pulse with -50000 fs$^3$ TOD applied using the pulse shaper. For better contrast, only spectral amplitudes between 0.005 and 1 are plotted. (d) Simulated time-frequency analysis of a pulse on which the phase from (b) is applied.](image-url)
Since we observe that the phase errors due to the pixel crosstalk mostly manifest ahead of the pulse, we chose to impose a negative GD offset of -2000 fs. In Fig. 4(a), we show the NOPA signal versus the pump delay when -2000 fs GD and -50000 fs^3 TOD was applied. From the Fig. 4(a), we can identify 3 pulses: the zero-delayed pulse, which did not acquire the imposed phase; the pulse delayed by -2000 fs with the expected imposed phase and free from aberrations; and the replica advanced by 2000 fs due to phase smoothing errors with an opposite phase, i.e. positive TOD. The spectral amplitudes of the replica pulses depend strongly on how well the SLM phase response is calibrated. In this particular case, the SLM calibration is excellent over most of the bandwidth and hence only a very small part of the replica is visible. In Fig. 4(c), we contrast the acquired NOPA output spectrum for the case of 0 fs GD as it was shown in Fig. 3(c), and for the case when -2000 fs GD was imposed. Typically, in OPCPA systems the first nonlinear stage has a very high gain which naturally acts as an excellent temporal filter. By this temporal filtering, the effect of replica pulses is suppressed and a spectrally smooth amplification is achieved, as shown with the blue curve in Fig. 4(c). Finally, the seed pulse energy loss to replicas does not influence the overall system design, since the gain in the first amplification stage (which is usually very high gain for OPCPA systems) can be increased accordingly.

![Fig. 4. (a) Noncollinear optical parametric amplifier (NOPA) amplified spectrum versus pump delay for a pulse with -2000 fs group-delay (GD) and -50000 fs^3 third-order dispersion (TOD) applied using the pulse shaper. For better contrast, only spectral amplitudes between 0.005 and 1 are plotted. (b) Simulated time-frequency analysis of a pulse on which a wrapped and smoothed phase of -2000 fs GD and -50000 fs^3 TOD was applied. (c) The measured NOPA output spectrum for the 0 fs GD case [as in Fig. 3(c)], and for the -2000 fs GD case [shown in (a)].](image)

To establish the utility of amplifying the pulse with a GD, we demonstrate that the amplified pulses can be compressed to the Fourier limit. By manually optimizing the phase \( \phi_{\text{target}}(\omega) \) at fixed GD offset of -2000 fs, we could obtain a compressed pulse after amplification which we characterized with SH-FROG. The required phase \( \phi_{\text{target}}(\omega) \) varied by over 30 radians across the spectrum. Without the GD offset, this profile would cause 6 phase-wrapping points over the bandwidth, which in turn would distort the shaped waveform. In Fig. 5(a), we show the retrieved and measured spectra, as well as the remaining phase. Note that the phase...
over the energetic part of the spectrum is virtually flat, while at the wings of the spectrum the SH-FROG retrieval errors usually dominate. For comparison, the measured and retrieved traces are shown in Fig. 5(b). Furthermore, using a similar pulse shaper configuration, we have also demonstrated excellent pulse compression in the mid-infrared down-converted spectral range: our system yielded 1.7-cycle pulses at a center wavelength of 2500 nm with 12.6 W average power at 100 kHz, which is a record-short pulse duration for a >10 W mid-IR OPCPA [49].

Fig. 5. (a) Retrieved spectrum and phase of the compressed pulse. The measured spectrum is overlaid for comparison. (b) Measured and reconstructed traces.

4. Spectral Amplitude Shaping

A powerful feature of using the pulse shaper with a large GD is the possibility to control the spectral intensity of the delayed components. This control can be achieved by scaling the amplitude of the wrapped phase profile. Intuitively, this approach can be understood by considering a spatial phase grating analogy. When a diffraction grating modulates the spatial phase by a modulation amplitude of $2\pi$, then the diffraction to the 1st order is the most efficient. However, when the modulation amplitude is reduced, the optical power is redistributed into other diffraction orders. The same effectively happens in our pulse shaper in the time domain when a frequency grating is applied, as conceptually illustrated in Fig. 1.

In the pulse shaping case, when a large ideal phase profile is wrapped, it effectively acts like a phase grating applied in the frequency domain. Consider a large spectral phase $\phi_{\text{ideal}}(\omega)$ applied on a pulse with spectral intensity distribution defined by $A(\omega)$. We apply an amplitude scaling factor $\xi$ on a wrapped phase such that the modulation amplitude is scaled symmetrically over $\pi$:

$$\phi_{\text{wrapped}} = \xi \cdot \text{mod}(\phi_{\text{ideal}}, 2\pi) - \pi (\xi - 1), \text{ for } \xi \in \mathbb{R}[0,1].$$

It is important to stress that this is not equivalent to multiplying the unwrapped phase with the amplitude scaling factor since the modulo is not scale-invariant. As an example, consider applying a large GD which is phase-wrapped with a period of $\omega_p$ such that
This periodic phase function can be conveniently decomposed using Fourier series which leads to the output electric field:

\[
E(\omega) = A(\omega) \sum_{n=-\infty}^{\infty} e^{-i\pi(n-1)} \text{sinc}(\pi(\xi - n)) \exp\left( i \frac{2\pi\xi}{\omega_p} n \right).
\]  

(3)

In this form, Eq. (3) can be interpreted as a frequency domain phase grating which leads to an infinite number of possible diffraction orders in the time domain separated by the applied GD. For the case when there is no phase wrapping amplitude modulation, i.e. \( \xi = 1 \), we find that all Fourier coefficients are zero, except the \( n=1 \), which represents a linearly delayed pulse by \( 2\pi/\omega_p \). For the case when \( \xi \neq 1 \), all of the Fourier orders are contributing. If, in this case, we choose to time-filter only a chosen \( n \)th diffraction order, we find that the amplitude of the spectral component, \( \omega \), in the chosen diffraction order is defined by \( \text{sinc}(\pi(\xi-n)) \). It is worth noting that the applied phase is also scaled by the diffraction order \( n \) and the even terms acquire additional \( \pi \) phase shift. In contrast to the amplitude shaping described in [128], this scheme does not rely on a spatial diffraction-based filtering. The spectral components remain present in the beam, but are not delayed by the applied GD.

The crosstalk between the SLM pixels will lead to an effective smoothing of the wrapped-phase (as illustrated in Fig. 1), which in turn will reduce the grating modulation amplitude. Hence due to the crosstalk \( \xi \) will not be equal 1 and the modulator replicas will be present.

Because the GD is determined by the periodicity of the phase grating (i.e. \( \text{GD}=2\pi/\omega_p \)), the Eq. (3) can be extended to a more general phase which contains a large GD component. Any additional phase added will slightly change the local (i.e. in the vicinity of a spectral component \( \omega \)) phase-wrapping periodicity. This will lead to an effectively different GD for this spectral component. Due to the varying period of the phase grating, this additional phase will be mapped to a GD spectrum.

Finally, because the phase grating has a spatial extent, it also acts as a weak spatial grating placed in the Fourier plane of the pulse shaper. This leads to a small, but noticeable lateral beam shift when a large GD is applied. This connection between delay and displacement (i.e. a form of spatiotemporal coupling) is a general property of pulse shapers based on SLMs [137]. This property can also lead to a spatial chirp if the applied additional phase significantly modifies the GD offset [138].

An SLM mask designed for a frequency-dependent amplitude shaping effect can be most easily evaluated numerically. We assume a compressed input pulse with a Gaussian spectral energy distribution. On this pulse, we apply a GD of -2000 fs and group delay dispersion (GDD) of 500 fs\(^2\), and we additionally apply a flat-top amplitude mask of \( \xi=0.5 \) for a part of the bandwidth as shown in Fig. 6(a). We model the pixel crosstalk by smoothing the wrapped phase using a moving average filter.
with a span of 8 pixels. Due to this pixel crosstalk and the amplitude mask, the modulator replicas appear in the time domain, as can be seen in Fig. 6(b). Furthermore, from the figure we can see that: (i) the undelayed pulse did not acquire any GDD; (ii) the \( n=1 \) diffraction order has acquired the designed phase and amplitude modulation; (iii) the \( n=2 \) and \( n=-2 \) replicas acquired a phase which is twice larger than the designed phase, as expected from the Eq. 3. By temporally filtering the output, we can extract the spectral amplitude (Fig. 6(c)) of the delayed pulse, which indicates that the targeted spectral components were successfully suppressed.

Fig. 6. (a) Simulation of an applied amplitude-scaled wrapped phase on a simulated Gaussian transform-limited pulse. The phase was -2000 fs group delay (GD) and 500 fs\(^2\) group delay dispersion (GDD) for a central wavelength of 745 nm. (b) The time-domain representation of the simulated waveform in (a). The peak of the -2000-fs delayed pulse is suppressed as it would be expected for a linearly chirped and spectrally shaped pulse. (c) The spectral contents of the input and shaped pulses indicating spectral intensity shaping. (d) An experimental example of amplitude shaping where a nearly rectangular spectral intensity distribution was achieved after the noncollinear optical parametric amplifier (NOPA).

Next, we demonstrate this shaping experimentally by achieving a nearly flat-top amplified spectrum after the NOPA using a suitable phase profile at the SLM, as shown in Fig. 6(d). Note that because of the pixel crosstalk in the SLM and spectral gain coupling in the NOPA, an iterative algorithm would be required to achieve a perfectly flat-top shape.

In line with [76,127], we further demonstrate the amplitude and phase control by engineering a double-pulse waveform which is within the temporal gating window of our NOPA. We apply an additional GD of 400 fs for part of the bandwidth.
and, using amplitude filtering as shown in Fig. 7(a), we experimentally obtain the double-pulse profile at the output of the NOPA, as shown in Fig. 7(b). The relative amplitude of the pulses can be most easily controlled by adjusting the seed delay with respect to the pump.

Fig. 7. (a) Experimentally applied amplitude control on the phase grating. The phase-grating contains the phase used to compress the pulse (as in Fig. 5.), a -2000 fs group delay (GD) over the complete bandwidth, as well as an additional 400 fs GD offset added for the spectral components with wavelength longer than 720 nm. (b) Reconstruction of the double-pulse from a second harmonic frequency resolved optical gating (SH-FROG) characterization.

5. Conclusion
We experimentally demonstrate a new SLM-based pulse shaping technique with which a large spectral phase can be imposed on the pulse without reducing the waveform quality. We achieve this operation by applying a large GD and time-gating the delayed pulse. Due to phase wrapping of this large GD, we create a phase grating in the frequency domain, which corresponds to a series of pulse replicas in time. We show that temporal filtering of a specific replica via time-gated amplification allows removing the temporal aberrations created by the pixelated nature of the SLM. Furthermore, we demonstrate that we can control the spectral amplitude of the shaped pulse by tuning the amplitude of the phase grating.

This is a powerful technique which allows shaping the amplitude and phase of ultrafast pulses by using only a 1D-SLM in conjunction with a time-gated amplifier. Implemented within the front-end of an OPCPA system, this pulse shaping technique can be used to compensate for a large dispersion without adding temporal aberrations to the pulses, hence avoiding the need for additional highly dispersive stretchers. Furthermore, the amplitude shaping function can be used to compensate for a spectrally dependent gain in broadband systems, enabling engineering of the most optimal waveforms for various applications. Particularly, gain narrowing in OPCPA systems can be mitigated and very high peak power waveforms can be achieved for strong-field applications, such as high-harmonic generation and attosecond science.
3.6 Conclusions

In this chapter I have presented the general high-power mid-IR and as well as near-IR OPCPA considerations. I have also presented our laboratory infrastructure and some key aspects of the general system management necessary to be able to develop a successful OPCPA system. I have shown time-delay stabilization approach to have a good long-term stability between the OPCPA pump and seed, and I have explained our novel time-gated pulse shaping technique. Significant improvements in the mid-IR and near-IR OPCPA performance is possible to obtain after the pump laser performance is improved. Given the availability of the new kW-class sub-ps-cosecond pump sources I expect many new high-power OPCPA systems to emerge. Our work stands as a pioneering work in using such industrial-grade pump laser systems and we expect that our learnings described in this chapter will further the high-power OPCPA development.
Chapter 4

High-power Mid-IR OPCPA

In the previous chapter, we have discussed the general design considerations for the high-power OPCPA systems. I have shown, the seed and pump laser configurations we have used, time-delay stabilization and time-gated pulse shaping schemes. In this chapter, I will describe in detail our mid-IR OPCPA system. The first version of the system development was led by my colleague and was published in [49]. The developed system reached up to 6.3 GW peak power pulses with an ultrabroad-band configuration spanning from 1.7 to 3.2 µm. The second development version of the system was led by me and during the development, we have focused on achieving higher peak power pulses with an increased power extraction efficiency and stability of the system. Multiple innovative steps were taken to reach the 14.2 GW peak power result and they are outlined in this chapter. The complete mid-IR OPCPA layout is shown in Figure 4.1 and each aspect of the setup will be discussed in the following sections.

![Figure 4.1. High-power mid-IR OPCPA conceptual layout. SHG - second harmonic generation, NOPA - noncollinear optical parametric amplifier, DFG - difference frequency generation, BBO - beta barium borate, PPLN - periodically poled lithium niobate, LBO - lithium triborate, ZnSe - zinc selenide.](image)

4.1 BBO-Based NOPA

By using the previously presented time-gated pulse shaping technique a large negative GDD of -2400 fs² and TOD of -10400 fs³ is applied on the input waveform. Furthermore, the input waveform is spectrally flattened by using the programmable amplitude shaping capability in order to have no back-conversion in mid-IR pre-amplifier (to be discussed in section 4.6). This shaped electrical waveform is time-gated and amplified in a NOPA stage based on a BBO operated in a type-I nonlinear interaction geometry. First, we tried a 1.5-mm long crystal and were able to obtain a broader amplification bandwidth. However, we ultimately used a 2-mm long crystal since it allowed for higher power while still providing a sufficient bandwidth. The NOPA is pumped with 13.9 W of 515 nm radiation, focused to 432 x 363 µm² 1/e² diameter, reaching intensities up to 85 GW/cm². The pump pulses are derived from a low-efficiency SHG stage (37 % depletion...
efficiency). From the depletion efficiency, the SHG pulse duration was estimated to be 1.4 ps. The SHG depletion calculations are presented in more detail in the following section. The seed is focused on the BBO crystal in such a way that it is at the image plane of the pulse-shaper SLM and the spot is round of 440 x 448 µm² 1/e² diameter. The image plane placement on the time-gating position is very important because as the GD imposed by the pulse shaper is tuned, it will cause beam position change. However, at the image plane of the SLM, the seed will not move. The seed power is measured to be 14 mW at 82 MHz pulse repetition rate, thus without considering diffraction efficiency of the time-gated pulse shaping, the seed energy is estimated to be 173 pJ over the complete bandwidth. Taking this into account and that only fraction of the bandwidth is amplified, the true seed energy can be estimated to be below 30 pJ. In case of no spectral amplitude suppression, the seed is amplified to 380 nJ level. This is estimated to be 41 dB gain and the amplified spectrum is shown in Figure 4.2.

![Figure 4.2](image.png)

Figure 4.2. The seed spectrum vs the amplified NOPA spectrum.

The noncollinear angle of the NOPA was estimated to be 2.1 deg. Phase-matching was optimized for 650–780 nm spectral components amplification. The larger bandwidth of the NOPA can be obtained by choosing a thinner, 1.5-mm long crystal and increasing the pump intensity up to 120-160 GW/cm². At higher intensities, the crystal is damaged.

The NOPA can be operated in multiple orientations between the seed and the pump w.r.t. to the c-axis of the crystal. In our case of θ_s = 2.1° noncollinear angle, the pump beam has θ = 23.8° angle with respect to the c-axis of the crystal. Also, the crystal is cut at φ = π/2, so the pump beam lies in the z-y plane. This positioning of the pump defines the phase-matching condition. As long as the noncollinearity is preserved, the seed does not have to be in the z-y plane as the pump. However, the pump energy will flow at a 3.3° angle from the wave vector (spatial walk-off phenomena) (calculated using Sellmeier relation from [139]). Thus, in case of longer crystals or smaller pump beams, it is advantageous to have the signal as well in the z-y plane with noncollinearity oriented in such a way that the walk-off is partly compensated by it. In the case of θ_s = 2.1° noncollinearity, the
walk-off between the pump and the seed could be 1.2° (walk-off compensating case (WC)) or at the extreme case where the walk-off and noncollinearity are opposite, it would be 5.4° (walk-off non-compensating case (WNC)). The WC case could help to ensure spatial overlap between the seed and the pump for up to 10 mm interaction lengths, whereas WNC would allow only for 2.3 mm of overlapping interaction. Since we are using crystals shorter than this length, it does not really matter which orientation is used. However, in case WC is used, a parasitic SHG of the signal can occur. However, this occurs only for wavelengths longer than 860 nm and thus it does not concern this NOPA. For near-IR OPCPA we have built, the choice between WC and WNC was more critical. There are subtle differences between these configurations and they were studied in depth by [73,74,140]. In this NOPA we have used a WNC case since we observed it to provide a slightly greater gain.

4.2 SHG-Based Flattening

The SHG-based beam flattening plays a pivotal role in this OPCPA system. The flat-top beams help to delay the onset of back conversion at OPA stages and thus allow for higher pump energy extraction. Furthermore, a flat pump pulse in the time domain allows avoiding gain narrowing effects during CPA. Because our Innoslab amplifier system was not designed to deliver flat-top beams which are needed for OPCPA systems, we did implement pump flattening separately. One of the simplest and effective beam-flattening technique is based on depletion of the fundamental wave (FW) to the second harmonic (SH) via SHG process [141]. Following the equations described in section 2.2.1 we can calculate spatiotemporal flattening effects on the pump beam. Figure 4.3 shows the temporal profile of our pump pulse after 37% depletion. The calculated profile is for on-axis intensity, meaning that the wings of the beam will have less depleted temporal profile. To check the calculation experimentally and measure the depleted pump pulse shape we have performed a second harmonic frequency resolved optical gating (SH-FROG) measurement. For the measurement, the wings of the depleted beam were cut with an iris. Both, the FROG measurement and the theoretical expectation, suggest that the input pulse was strongly depleted so that its FWHM duration became 3.2–3.8 ps long.
Similar depletion effect occurs in the spatial domain. Figure 4.4a shows the spatial gaussian beam profile as an input for calculations and Figure 4.4b shows the beam profile after depletion. These profiles were calculated for $t=0$ of the pulse, i.e. for the most intense part of the pulse. The calculated intensity factor ($I = \frac{I_{factor}}{P_{pk}} \frac{P_{pk}}{Area}$) is 1 (2 for gaussian beams), which is equivalent to a flat-top beam. Note that in this equation $Area = \pi w_0^2$, with $w_0$ being the beam radius $1/e^2$ at the $t=0$. Since the depleted beam is spatiotemporally varying in size and any beam profiler camera would integrate the beam in time, the actually recorded profile will not seem so much flattened. Thus the peak intensity estimate from the directly measured beam radius becomes inaccurate. Furthermore, the actual energy in the pump pulse is not clear since there could a significant amount of amplified spontaneous emission and as well as pre- and post-pulses not captured within the range of the FROG measurement. Because of these reasons, the peak intensity estimate of the flattened beam is stated as a range of intensities.

Figure 4.4. Simulated beam profile at $t=0$. (a) Undepleted case. (b) Depleted via 37% efficient SHG.
If one considers that useful energy for OPCPA is only above FWHM of the beam (both space and time), the energy density increase due to the SHG flattening can be calculated. By considering the loss of power to the SH, I have calculated the beam energy above the spatiotemporal FWHM. Figure 4.5 shows the results. From the figure, one can see that even if some energy is lost to the SH, the actual energy density can be increased by about 30%. Finally, if the generated SH radiation can actually be useful in the system, then it does not have to be considered as a loss all. In our case, the SH beam from the SHG-1 (see Figure 4.1) is used to perform time-gating on the seed waveform. In the case of the SHG-2, the beam is simply discarded.

Figure 4.5. Energy above the spatiotemporal FWHM of the pump versus the SHG loss. The loss is considered in the energy above the FWHM estimate.

Finally, to be able to actually make use of these flattened beam profiles, the beams have to be imaged onto the nonlinear crystals. Without the image transfer, the spatial flat-top features would be lost and the flattening effect would be useful only because of the reduced gain narrowing effect. Because of this reason, the image from the SHG stages is transferred onto the nonlinear crystals. Furthermore, since the pump beam was continuously evolving due to drifts in the commercial pump laser, some special arrangements were necessary to be able to control the beam shape. This was achieved by having an adjustable pair of telescopes to control the beam size and divergence on the SHG crystal and from the SHG crystal, the beam image was just simply transferred with an appropriate magnification factor. This not only helped us to improve the long-term stability of the system but also could have a positive impact on the noise of the system. First of all, the transferred image size is not determined by the divergence/input angles on the SHG crystal and thus any beam pointing effects are reduced by the magnification factor. Secondly, the SHG removes the most intense parts of the beam and if the beam intensity fluctuates it depletes more or less depending on the change. Thus, the SHG stages work as a nonlinear noise filter reducing the pump energy fluctuations.
4.3 Prism Pair

Because of the previously described SHG-flattening, the NOPA is pumped by 1.4 ps SH pulses and the first mid-IR stages, such as DFG or PPLN-based NOPA are pumped by 3.2 ps long FW. This means that the pulses after the BBO-based NOPA have to be stretched even more than the time-gated pulse shaper can allow to. This is done by introducing a prism pair-based stretcher in the beam path so that a sufficient negative GDD and TOD is accumulated. We found that prism pair description by [142] is without unjustified assumptions and is well reproducing our experimental observations. We are using a SF10 prism pair separated by 380 mm distance and with a minimal possible glass insertion to avoid clipping parts of the spectrum that are used. This allows us to have -1400 fs² GDD and -17200 fs³ TOD for 730 nm center wavelength. Dispersion prediction was verified with a FROG measurement performed on the pulse at the DFG position.

4.4 Difference Frequency Generation

The stretched BBO-based NOPA output pulses (near-IR) are then sent on to our DFG stage. These pulses serve as the pump pulses for this nonlinear BBO-based DFG stage ($\theta = 20.2^\circ, \varphi = 90^\circ, L = 300 \text{ mm}$), i.e. they are the highest-frequency wave in the three-wave mixing process. The stage is collinearly seeded with a 2 W 1030 nm beam. The near-IR beam is depleted and amplifies the 1030 nm beam. During the process, it generates idler photons at the mid-IR spectral range. Since the stage is collinear, the idler beam is free from angular chirp. Any misalignment at this stage will lead to a strong angular chirp in the system. The seed beam for this stage is imaged from the SHG-1 stage to 240 x 227 $\mu$m² 1/e² diameter leading to an estimated intensity in the range of 12-25 GW/cm². The pump is effectively imaged from the BBO-based NOPA position (which is at the image plane of the pulse shaper) to a 93 x 103 $\mu$m² 1/e² diameter leading to an estimated intensity of 100 GW/cm². For this intensity estimate, the total dispersion up to the DFG crystal was accounted for. This stage outputs around 4 mW of mid-IR light. This power can vary a lot depending on the NOPA configuration and the desired spectral bandwidth. With an amplitude shaped pump pulse, the DFG provided 2 mW output. In an alternative configuration when the BBO-based NOPA is driven stronger, the DFG could deliver more than 50 mW of mid-IR. However, the following mid-IR amplification stage is saturated and its output power does not change if the DFG output is increased. In the case of the amplitude shaped NOPA, the depletion of the NOPA output was measured and is shown in Figure 4.6a. From the depletion measurement, the DFG spectral shape was predicted and measured as well. The measured DFG was found to fit the expected DFG (up to the noise floor of the measurement device (Mozza, Fastlite)) as shown in Figure 4.6b.
4. High-power Mid-IR OPCPA

Figure 4.6. Near-IR depletion and mid-IR generation. (a) Measured depletion of the near-IR pump in the DFG stage. (b) Measured mid-IR DFG spectrum (smoothed to reduce the noise due to its low power) and the expected DFG spectrum from the depletion of the near-IR.

Figure 4.7a shows BBO-based DFG phase-matching curves versus the crystal angle $\theta$. From the curves, we can see that the crystal could support ultra-broadband DFG spanning up to 3.2 $\mu$m. However, for going deeper into mid-IR it is limited by its absorption. An alternative crystal for BBO is BiBO ($\text{BiB}_3\text{O}_6$) which we also had in our laboratory ($\theta = 10.7^\circ$, $\varphi = 0^\circ$, L = 300 $\mu$m), but we did not use it due to the laser beam time shortage. The BiBO-based DFG phase-matching curves are plotted in Figure 4.7b. From the phase-matching curves we can see that an octave spanning mid-IR bandwidth could be obtained with this crystal.

Figure 4.7. DFG phase-matching. (a) DFG between near-IR and 1030 nm phase-matching in a 0.3-mm long BBO. (b) DFG between near-IR and 1030 nm phase-matching in a 0.3-mm long BiBO.

4.4.1 Difference Frequency Generation in APPLN

As an alternative to bulk DFG, we have also investigated using aperiodically poled lithium niobate (APPLN) for energy transfer from the near-IR to the mid-IR. We have used a custom design crystal of 1.2x0.5x45 mm$^3$ dimension manufactured by Gooch and Housego PLC. The crystal was designed to have a transversal grating component of $K_{g,x} = 105$ $mm^{-1}$. Because of this transversal component it was possible to obtain an angular chirp-free idler when seeded with 1030 nm narrowband source with a small noncollinear angle of 1 degree (external) to the
Difference Frequency Generation

near-IR pump. Along the propagation direction of the pump, a chirped poling was added. The poling period was varied from \( K_{g,z} = 502 \text{ mm}^{-1} \) to \( K_{g,z} = 274 \text{ mm}^{-1} \) in such a way that adiabatic energy transfer from the pump to the signal could take place \([143]\). Furthermore, the higher-order slope of the \( K_{g,z} \) evolution allowed for control of the group delay of the generated mid-IR light. Therefore, with appropriate design, such crystal can be used for obtaining a sufficiently stretched mid-IR pulse with the desired TOD. This way the prism stretcher would not be needed in the system.

This crystal was tested by using the pre-amplifier output from the near-IR OPCPA system (described in Chapter 6). For the experiment, we have used 100 mW near-IR broadband pump focused up to 2 GW/cm\(^2\) intensity and the seed was narrowband 1030 nm light focused to the same intensity but had 433 mW average power. The mid-IR light (after a 1500 nm long-pass filter) was measured to be 6 mW (after accounting for Fresnel losses). At the same time, the mid-IR and near-IR spectra were characterized. The measured depletion of the near-IR is shown in Figure 4.8a. Meanwhile, the expected mid-IR spectrum from the depletion in contrast to the actually measured mid-IR spectrum is shown in Figure 4.8b. From the measured depletion of the near-IR one could expect 56 mW of mid-IR to be generated (56% depletion). However, only 6% conversion efficiency is observed (around 24% quantum conversion efficiency).

![Figure 4.8. APPLN DFG. (a) Measured near-IR spectrum after APPLN crystal when not overlapped (undepleted) and when overlapped (depleted) with 1030 nm seed. (b) Measured mid-IR spectrum from DFG at the APPLN. The dashed line indicates the expected spectral shape if the depleted light was directly transferred to mid-IR.](image)

Such low conversion efficiency was also obtained in another recently published work \([76]\). Therefore, this suggests that a significant fraction of the mid-IR light is actually lost to some other nonlinear parasitic process taking place in the APPLN. This hypothesis is further backed-up with the fact that the measured mid-IR spectrum does not match the expected mid-IR spectral shape shown in Figure 4.8b. Another possible loss mechanism is related to the excitation of polarons in the APPLN \([144]\). The excited polarons can dramatically accumulate due high-repetition rate of our laser and due to their long lifetime in millisecond time scale can lead to a strong pump and signal absorption. Figure 4.9 shows the measured transmission of 1030 nm light as a function of the input power. From the plot, we
can see that the seed transmission decreases with the input power suggesting that some energy is lost in the crystal. Because of this low conversion efficiency, we have decided to mitigate risks and not use this DFG output to seed our mid-IR amplification chain.

Figure 4.9. Seed (1030 nm) transmission through APPLN crystal versus the input power with no pump (near-IR) present. Co-propagating parasitic SHG was also included in the power measurement.

### 4.5 Passive CEP Stability

For attosecond experiments involving isolated attosecond pulses (IAPs), often carrier-envelope phase (CEP) stability is a major requirement. The CEP determines the emission time of the attosecond pulses under the driving pulse envelope. Thus, if the measurement is to have attosecond pump-probe precision, the emission time of the burst has to be controlled precisely. For example, for 800 nm driving pulse, 100 mrad CEP jitter would directly translate to 42 as timing jitter. This is usually not a problem, because such short pulses are normally not acquired directly from the 800 nm driven HHG. However, in the case of mid-IR drivers, the CEP stability becomes much more demanding in terms of specification. For the same 100 mrad CEP jitter of 2200 nm driving wavelength would lead to 117 as timing jitter of the attosecond pulse. Furthermore, with mid-IR driving HHG shorter than 50 as pulses can be achieved [31]. Thus, to benefit from the short attosecond pulses, the CEP stability has to be better than 50 mrad.

In our system, the DFG is generated between the 1030 nm signal and the amplified Ti:sapphire frequency comb. Since the 1030 nm signal is also derived from the Ti:sapphire laser, the carrier-envelope offset frequency ($f_{\text{CEO}}$) [116] of the idler from the DFG should be zero [145]. $f_{\text{CEO}} = 0$ means that the CEP between every pulse is the same. However, in the Innsolab amplifier system, the pulse picking is performed by an acousto-optic modulator (AOM). The AOM performs pulse picking by diffracting the seed with an acoustic wave with carrier frequency $f_{\text{AOM}}$. The diffracted pulses will effectively experience a Doppler shift equal to $f_{\text{AOM}}$. 
Hence, our frequency comb from the DFG will have $f_{CEO} = f_{AOM} \mod \frac{f_{rep}}{n}$, where $f_{rep}$ is the Ti:sapphire repetition rate and $n \in \mathbb{N}$ is the pulse picking ratio such that we have $100 \, \text{kHz}$ pulse train. This, therefore, means that the pulse-to-pulse CEP will continuously evolve.

This can be avoided by locking the pulse picker electronics to the Ti:sapphire repetition rate in such a way that $f_{AOM} = \frac{f_{rep}}{n} \cdot m \in \mathbb{N}$ [146]. However, even if this would be taken care of, the overall CEP stability of the system would be not ideal due to the extreme phase change imposed by the grating compressor. In the compressor, the pulses from the A400 are compressed from a few-hundred-ps duration to 2 ps, which gives a very large phase change. Thus, any pointing jitter of the beam would translate to a large phase fluctuation. Because of this reason, we have decided not to pursue a modification of the AOM pulse picking system, but rather use a different concept to ensure passive CEP stability of the system.

![Diagram](image)

Figure 4.10. High-power mid-IR OPCPA conceptual layout with passive CEP stability. The DFG seed in this case is directly derived from Ti:sapphire oscillator and amplified by a high-gain OPA with remaining power from NOPA.

Figure 4.10 shows our planned mid-IR OPCPA modification to ensure passive CEP stability in the system. The 1030-nm DFG pump is replaced with a 1064-nm pump derived directly from the Ti:sapphire oscillator, time-gated and amplified via a single-stage OPA. In case the amplifier not deliver sufficient output power, an additional OPA stage could be added to have a higher CEP-OPA output power.

The feasibility of this scheme was evaluated in a few steps. First of all, the Ti:sapphire spectrum was measured to extend beyond $1100 \, \text{nm}$. Secondly, the time-delay stabilization (TDS) was built and it was shown with it that with less than 3 W of 1030 nm of the pump, it is possible generate a sufficient amount of SHG and have more than $45 \, \text{dB}$ gain from a single OPA stage. Thus, with careful optimization and optimal seed stretching, it should be possible to reach higher gain. In an additional project (not reported here), we have also tried to fiber-couple 1 μm components of the Ti:sapphire output and amplify in a narrowband NOPA. The NOPA was based on a LBO ($\theta = 90^\circ, \varphi = 11.6^\circ, L = 15 \, \text{mm}$) and could provide
more than 60 dB of gain in a single stage while being driven by a few-Watt level 515-nm light.

Another alternative to the implementation of the CEP-OPA is changing the architecture of the DFG in such a way that DFG would be performed from the same time-gated NOPA output pulse. This technique is often called as intrapulse DFG (IDFG) and is highly attractive for generating CEP stable seed for the system. However, the problem with this scheme is that it does not permit for linear phase transfer to the mid-IR. Of course, a numerical algorithm could be developed to solve the phase relation between the mid-IR and the near-IR, but so far, we did not manage convergence on an efficient algorithm. In principle, this problem could be overcome by the use of our pulse-shaper and imposing a large group delay to one edge of our spectrum so that we could have one long pulse and one short pulse within the same time-gated window. It is possible to identify a scheme where the two pulses have such phase that the generated mid-IR phase is well predictable and it has a strong negative TOD component imposed so that no prism pair is needed. The main issue with such IDFG is that our NOPA bandwidth is limited by the pulse-shaper and thus we could not generate mid-IR components below 2 µm via IDFG. Thus, we would need to shift the center of the bandwidth more into the mid-IR. This could be done in the future, but it would come with a reduced HHG flux.

4.6 Mid-IR Pre-Amplification

In our team we have studied various amplification techniques to parametrically amplify the generated DFG idler components. For example, we have explored using PPLN amplifiers with a transversal grating component. This allowed decoupling the interaction geometry from phase-matching considerations. The transversal grating component can be used to shift the group velocity matching point to a collinear configuration. Another use of the engineered transversal grating component is a possibility to use two tandem QPM structures so that multi-octave spanning spectrum is amplified. All these results we have been reported in a publication [135] and the reader is invited to familiarize oneself with it. For the result described in this thesis, the gratings with a transversal component were not used.

In the final scheme, the mid-IR beam after the collinear DFG is selected by a dichroic beam splitter and is used to seed a 1-mm long PPLN-based NOPA. For this and later stages we have used high-aspect-ratio, 2x10.9 mm² aperture samples with 29.3 µm poling period (HC Photonics Corp.). The pre-amplification crystal was not temperature controlled. The noncollinear angle between the seed and the pump was varied between 1.7 and 2.2 degrees and it was optimized for a larger bandwidth. The seed is delivered directly by a single 150 mm lens close to the imaging condition from the DFG with a tunable magnification factor around x5. Meanwhile, the 20.6-W 1030-nm pump is imaged from the SHG-1 stage to 570 x 543 µm² 1/e² diameter leading to an estimated intensity in a range of 23-40 GW/cm².
Figure 4.11. Pump beam propagation in the pre-amplifiers. (a) Pump beam propagation from the entrance into the setup to the DFG crystal. The solid blue line indicates the horizontal calculated beam size (in case of a Gaussian beam propagation). The dashed blue line indicates the calculated vertical axis of the beam. The solid orange line indicates the B value from the ABCD matrix with respect to the SHG crystal position. B=0 indicates imaging condition. If B=0, then A value (not shown here) determines the beam size accurately, even for non-gaussian beams. (b) Pump beam propagation from the entrance of the setup to the NOPA pre-amplifier.

The complete pump path from the input of the setup up to the crystals was fully traced with our ABCD propagation code. Figure 4.11 shows an example of how the pump beam was imaged from the SHG crystal to the DFG and NOPA crystals. Imaging of the pump path was essential for obtaining flat-top beam profiles on the crystals as well as reducing system sensitivity to the changes in the pump beam size and divergence. Moreover, since the imaged beam is angle insensitive and position is de-magnified, the overall stability of the beam on the crystal is dramatically improved compared to a direct beam delivery without satisfying the B=0 condition.
4. High-power Mid-IR OPCPA

Figure 4.12. Spectra delivered by the pre-amplifier. The amplifier can deliver ultra-broadband bandwidth with 1.6 cycle transform limit with 1.07 W output power. The final configuration was achieved after the complete system optimization, including the power amplifiers leading to 2.2 cycle transform limit with 1.2 W output power.

The pre-amplifier can deliver ultra-broadband sub-2 cycle transform-limit output spectra such as shown in Figure 4.12 with more than 1 W of average power. During the complete system optimization (including the power amplifiers) we have decided to make compromise on bandwidth to reduce water absorption related effects and have higher output power possible. This led to the final output spectrum shown in Figure 4.12. The corresponding power slope is shown in Figure 4.13a. From the slope, we can see that the pre-amplifier was saturated. The spectra taken with increasing power show no signs of back-conversion (Figure 4.12b). By changing the BBO-based DFG stage into a BiBO-based DFG stage it should be possible to achieve sub-1.5 cycle transform limit output spectra from this single pre-amplifier.

Figure 4.13. The pre-amplifier slope. (a) Power slope of the pre-amplifier. (b) The corresponding spectral evolution at different pump powers.
4.7 High-Power Amplification of Mid-IR in PPLN

The pre-amplified mid-IR already had 12 µJ pulse energies at 100 kHz repetition rate. However, this output power was achieved with a relatively low pump energy extraction and only 12% quantum conversion efficiency. As it was nicely illustrated by H. Fattahi in [147], to have an overall OPA-based amplification efficient, one has to amplify in a few stages. The first stage, the so-called pre-amplifier, is used to increase the seed energy to a level comparable to the pump via high-gain (30 dB in our case). Then the following amplification stages can be run with a low gain but high efficiency. Following this trade-off, we targeted having 10 dB gain in the second amplification stage and 5 dB in the last amplification stage.

However, achieving high-power amplification in the mid-IR is challenging due to the lack of suitable amplification crystals. For example, the most commonly used nonlinear material, BBO, cannot be used for high-power mid-IR amplification due to its absorption above 2.3 µm. Neuhaus et al. ([51]) reported crystals to break during high-power amplification. A BBO alternative is BiBO crystal which starts absorbing above 2.5 µm. Thus, this crystal will also dramatically limit power scaling possibilities in the mid-IR. Other alternative options are using potassium titanyle arsenate (KTA) or potassium niobate (KN) used for the OPCPA systems reported in Refs. [52,53]. Although these crystals have excellent power handling capabilities, they require type-II phase-matching which limits the amplification bandwidth. A good alternative to these crystals is lithium niobate (LN). As discussed in the introductory chapter, here in this thesis we discuss only MgO doped LN crystals. LN is susceptible to photorefractive effect and thus has limited power scaling possibilities. However, the birefringent phase-matching bandwidth can support few-cycle pulses.

Another option is to use periodically poled LN (PPLN) crystals. The strong advantage of PPLN is that it allows accessing d_{33} nonlinear coefficient via type 0 nonlinear interaction geometry, thus offering much higher effective nonlinearity. Because of this reason, the PPLN crystals can be very short and be designed for ultra-broadband amplification supporting single-cycle pulses. Small signal gain from different nonlinear crystals, assuming realistic intensities for each crystal, is shown in Figure 4.14. Motivated by the possible amplification bandwidth and thus achieving highest peak power pulses we have decided to focus on using PPLN for the high-power amplification.
Figure 4.14. NOPA small signal gain for various nonlinear crystals. The parameters were adjusted for realistic conditions which could be used for the final amplification stage. The BiBO crystal parameters were: $L = 0.7 \text{ mm}$ length, $I = 80 \frac{\text{GW}}{\text{cm}^2}$ pump intensity, $\theta = 6.6^\circ$ crystal angle, $\theta_s = 1.5^\circ$ noncollinear angle, $d_{\text{eff}} = 2.6 \frac{\text{pm}}{\text{V}}$. The LN crystal parameters were: $L = 0.8 \text{ mm}$, $I = 40 \frac{\text{GW}}{\text{cm}^2}$, $\theta = 52.7^\circ$ crystal angle, $\theta_s = 2.2^\circ$, $d_{\text{eff}} = 4.4 \frac{\text{pm}}{\text{V}}$. The PPLN crystal parameters were: $L = 0.5 \text{ mm}$, $I = 13 \frac{\text{GW}}{\text{cm}^2}$, $\theta_s = 1.6^\circ$, $d_{\text{eff}} = 12.1 \frac{\text{pm}}{\text{V}}$ with QPM grating periodicity of 29.3 $\mu$m. Note that in practice due to gain saturation effects, to achieve this gain, higher pump intensities are needed.

The key limitations of PPLN are its power handling capability, susceptibility to photorefractive effects and aperture restriction. Furthermore, the phase-matching of various parasitic nonlinear processes, often through higher-order QPM periods, cannot be fully mitigated. Phase-matching of parasitic processes can also occur due to various random duty cycle errors from the targeted poling periodicity [75,148]. Some of these parasitic processes include SHG of the pump, SHG of the signal or idler, and sum-frequency generation between the pump and signal or idler. Thus, this effectively creates photons which are absorbed by the medium. Although LN bandgap is at 310 nm its absorption starts increasing already below 1 $\mu$m [149–151]. Due to the absorption of the pump and parasitic light, the crystal temperature both locally and globally increases. Moreover, photorefractive and pyroelectric effects can occur which modify the refractive index of the medium. Part of this effect can be mitigated by short-circuiting the c-facets. Good electrical contact for mounting such crystals is important. However, even if the global current can be short-circuitied in the crystal, the local heating associated with the pump intensity profile remains. This will cause photorefractive distortion effects even in MgO doped LN crystals [152].

From the simulations of Ref. [152] one can see that effective negative lensing can appear along the c-axis and positive lensing perpendicular to the c-axis. We
measured the transmitted pump under various conditions in order to better understand how to avoid these effects. It should be noted that in our experiments high-intensity, high repetition rate ultrashort pulses are used, which can introduce various additional effects not present in the cw interactions considered in [152]. Figure 4.15a shows measured transmitted beam profile after 0.5 mm thick PPLN sample under 1030 nm, 2 ps, 100 kHz, 15 W, 23 GW/cm² illumination. The beam is not distorted. However, if the intensity is further increased to 45 GW/cm² and 30 W of average power, the beam fans-out as shown in Figure 4.15b. Along the c-axis (vertical in the picture), the beam spreads out and focuses perpendicular to the axis in a similar way to the predictions from Ref. [152]. Since the introduced lensing is non-parabolic, the beam quality degradation is visible as well. To get an additional information, also one has to look into the image of the pump beam on the crystal (i.e. the near field). The measured image is shown in Figure 4.15c. From the figure, one can see that the pump amplitude (which was originally a Gaussian) becomes strongly distorted and the highest intensity part of the beam is lost (possibly due to absorption).

![Figure 4.15. Photorefractive beam fan-out. (a) 15W, 23 GW/cm² beam transmitted through a 0.5 mm PPLN sample. (b) 45 GW/cm², 30W beam transmitted through the same sample. Clear photorefractive fan-out occurs on the pump beam. The vertical axis corresponds to the c-axis of the crystal. The beam was measured 90 cm away from the crystal (far field). (c) The pump beam image on the crystal corresponding to the (b) measurement (near field).](image)

Light absorption, including multi-photon absorption, can excite long-lived free carriers [153]. These charge carriers will diffuse in the crystal effectively forming quasi-particles called polarons. The excited polarons will have their own absorption spectra with different lifetimes depending on the MgO doping in the crystal [154]. Another effect which can occur is Green-Induced Infrared Absorption (GRIIRA) [155], in which absorption of green light leads to an increase in the absorption coefficient of infrared light. In our experiments, such green light is generated at 515 nm by parasitic pump SHG. Although this SHG process is far from phase-matching, its efficiency is not negligible due to random duty cycle errors in the QPM crystal, which lead to a phase-matching pedestal [148]. Therefore, there is a chance for a run-away mechanism causing severe distortion effects of the beam and possibly leading to a damage of the crystal. Still, in the context of high-power picosecond pumped PPLN crystals, a detailed study of the complicated interplay between the mentioned effects remains a topic for future work.

It is also worth mentioning, that the damage threshold measurements performed with 100 kHz, 1-ps pump laser [156] indicate that the damage threshold
in PPLN and LN is similar and around 174 mJ/cm$^2$ and 183 GW/cm$^2$. In our system, we have operated the crystals with 3-10 times lower fluences and we have not observed laser-induced damage of the crystals. During the operation of the crystals, we set the pump intensity to onset of the photorefractive fan-out of the beam (reversible damage). Therefore, the photorefractive fan-out of the beam and not the irreversible damage threshold determines the maximum practical intensity. Finally, it is also worthwhile to remark that PPLN has some absorption features in the mid-IR as well, as it was studied in [157].

4.7.1 Implementation of High-Power PPLN Amplifiers
Following the discussion in the previous section, we can define certain guidelines in the design of PPLN-based high-power parametric amplifiers. First of all, the intensity and fluence on the crystals have to be limited, thus large apertures are necessary for high-power operation. However, the poling of LN over large aperture is very challenging. Often, commercially available PPLN crystals have 1 mm thickness. Because the electrical poling does not propagate easily through the material, at larger thickness crystals wedge-like poling structures develop [158]. At the start of our project, up to 10 mm poling thickness for PPLN was demonstrated [158]. Meanwhile, commercial vendor (HC Photonics Corp.) could offer custom 2-mm-thick PPLN samples with good poling quality or 3-mm-thick samples with reduced poling quality. At the time of writing this thesis, 5 mm thick PPLN samples were already commercially available [159]. To try the wide-aperture PPLN with wedge-like poling structure, we have acquired 5 mm thick PPLN samples with 30.6 µm poling periodicity from our collaborators. In principle, the large aperture could be obtained with different topology in the crystal growth [160], however this approach is costly and difficult to implement and is not pursued by the crystal growers.

The crystals had 5x16 mm$^2$ aperture and were grown along the beam propagation direction using Czochralski process. Due to larger poling aperture higher poling errors were expected, leading to a reduced effective nonlinear coefficient, and thus the crystals were diced to have greater lengths than one would normally use in a near-ideal QPM structure. For example, for our 9.1 W output power result [161], we have used 2.3 mm long sample with 190 W of pump average power. Although the large-aperture samples permitted high-power operation, we also have observed severe pump beam distortion effects, sometimes appearing a short time after the power is put on the crystal. To better understand these observations, we systematically studied the behavior of the crystal at different positions. In this study, we have observed strongly varying parasitic second harmonic generation efficiency throughout the crystal. On the other hand, the small signal OPA gain was mostly centered on the side of the crystal. The measurements performed on the crystal are shown in Figure 4.16. The spatially non-uniform gain can in principle be partly compensated by saturating the nonlinear process. However, the crystal could not withstand sufficient intensity needed to achieve strong enough saturation. Since we have also observed the pump beam to dramatically fan-out after a short time when a high-power is applied, we have
concluded that thermal effects in the crystal and thus the photorefractive crystal modification limits the high-power use of our sample.

Figure 4.16. Large-aperture PPLN samples. (a) Parasitic SHG in a 5 mm thick, 1.3 mm long PPLN sample. The crystal was pumped by 6.6 W 1030 nm pump, focused to 5 GW/cm$^2$ intensity. The crystal was elevated to 110 °C temperature. (b) Small signal OPA gain in the same sample when seeded with 10 mW mid-IR. The vertical and horizontal axis are relative to an arbitrary reference. The horizontal is the c-axis.

Following the conclusions from the experiments with the large-aperture PPLN samples we have decided: 1) to use the largest thickness samples for which high-poling quality could be maintained (i.e. smaller poling thickness); 2) to work with a very high aspect ratio samples and fill the aperture with the pump beam so that a quasi-1D heat flow could take place. For this purpose, we have acquired 2 mm thick PPLN samples from HC Photonics with 29.3 µm poling periodicity, 10.9 mm height (2x10.9 mm$^2$ aperture). The samples had 0.5 mm and 1 mm lengths and were AR/AR coated for 515 nm, 1030 nm and 1700-4500 nm. The mounted crystal during the high-power operation was photographed and is shown in Figure 4.17.
PPLN crystals have traditionally been operated at elevated temperatures, since this increases mobility of the free carriers and thus decreases the photorefractive effect. However, after performing a systematic temperature scan of the crystals and monitoring the pump beam shape, we could find the minimal fan-out when the crystal was at the room temperature or below. Table 4.1 shows our measured transmitted beam profile shapes and image profiles on the 0.5 mm thick PPLN sample at 100 W of pump power. From the table, we can see that water-cooled case (21 °C) and 90 °C case were showing the least distorted pump beam shapes. The same test was performed with a 1-mm-long crystal where we have observed the smallest beam distortions for the smallest temperatures. Since cooled crystals are beneficial for the overall stability of the enclosed setup (less heat load when the amplification compartment is sealed) we have decided to water-cool the crystals. During the high-power operation, we still observe the crystal mount to heat-up by a few degrees, partly due to the amplified signal and the generated parasitic light absorption and partly due to the scattering of the idler from a water-cooled beam dump nearby.
High-Power Amplification of Mid-IR in PPLN

Table 4.1. Pump profile after PPLN crystal at different crystal temperatures. Far field profile (measured 90 cm away from the crystal) and imaged pump beam profiles after 0.5 mm long PPLN at 100 W pump power (weakly depleted via a low-efficiency SHG).

For the above-mentioned experiment and also for the following experiments, the pump beam was partly depleted via a variable low-efficiency SHG [141]. From the previous discussions, we concluded that significant beam distortions in PPLN emerge from non-uniform heat load on the crystal. This can be partly overcome with flat-top beams. Since we could not use the full available pump power on the high-power stages, we have decided instead of attenuating the pump power via waveplate and polarizer pair, attenuate it via variable efficiency SHG. This proved to be very convenient and sensitive means on controlling the final intensity of the crystals. For pump depletion, we have started with 1-mm long LBO crystal leading to an estimated maximum 22% depletion. However, during the experiments, we could maximize the output power of the system while still keeping a good output beam shape with only 8% beam depletion by detuning the SHG phase-matching.

To make efficient use of the beam flattening effect, the pump beam from the SHG crystal had to be imaged on the PPLN crystals by using cylindrical telescopes. For the first high-power amplifier we imaged only one axis (along the c-axis of the crystal) since for another axis we could find the correct beam size with a flat-top profile already at the desired position (for this particular position the aberrated Innoslab amplifier beam happened to be a flat-top). Figure 4.18a shows the beam propagation calculations from the entrance of the setup up to the PPLN crystal. Notice that the high-power beam was focused in the air up to an estimated 200 GW/cm² for imaging to be possible. Figure 4.18b shows the measured beam profile at 65 W power level and with an estimated intensity of 18-23 GW/cm² (4122 x 762 µm² 1/e² diameter). This pump beam was used to pump 1-mm long PPLN crystal. The crystal was seeded with the pre-amplifier output. The transmitted pump profile is shown in Figure 4.18c and the output power slope is shown in Figure 4.18d. In this amplification stage, the seed could be amplified from 1-Watt level up to 12 W (gain of 12) corresponding to 36% quantum conversion efficiency. The seed was also shaped with cylindrical lenses to maximise the pump-seed spatial overlap needed for the high extraction efficiency.
Figure 4.18. The first high-power amplifier characteristics. (a) High-power pump beam propagation calculation from the entrance into the setup up to the first high-power amplifier. The solid line is for the horizontal axis and the dashed line is for the vertical axis. (b) Measured pump profile at the crystal position. (c) The transmitted pump profile through a 1 mm PPLN crystal (measured 90 cm after the crystal) with seed on. (d) The first high-power amplifier output slope vs pump power.

Similarly, to the first high-power amplification stage, the pump beam from the entrance into the setup was imaged onto the second high-power amplification stage with imaging scheme shown in Figure 4.19a. The imaging yielded a beam profile shown in Figure 4.19b. At 105 W of pump level, the beam had 3700 x 1000 μm² 1/e² diameter corresponding to estimated 25-34 GW/cm² intensity. For the last amplification stage, we have used 0.5 mm thick PPLN crystal and the transmitted pump beam was measured 63 cm away from the crystal is shown in Figure 4.19c. The seed from the first high-power amplification stage was again shaped independently on both axes to match the pump beam shape on the crystal. In this stage, we could achieve up to 30 W of output power with power slope shown in Figure 4.19d. This corresponds to a gain of 3 and 41% quantum conversion efficiency. Best to my knowledge, this is one
of the highest ever reported single-stage quantum conversion efficiency for a high-power, few-cycle OPCPA system.

Figure 4.19. The second high-power amplifier characteristics. (a) High-power pump beam propagation calculation from the entrance into the setup up to the second high-power amplifier. The solid line is for the horizontal axis and the dashed line is for the vertical axis. (b) Measured pump profile at the crystal position at 105 W power level. (c) The transmitted pump profile through a 0.5 mm PPLN crystal (measured 63 cm after the crystal) with seed on. (d) The second high-power amplifier output slope vs pump power.

The output beam from the second high-power amplification stage was collimated and shaped with a set of cylindrical telescopes. For beam shaping, we have used D-coated lenses (Thorlabs) which reflected a large fraction of the parasitic second harmonic of the signal generated in the last amplification stage. The output beam was then further reflected by ultra-broadband high-reflectivity Si/SiO$_2$ mirrors (Optoman) which completely filtered away any remaining parasitic light. Then the beam was transmitted through a compressor (discussed in section 4.8) where the final output power of the system was measured to be 25 W. The system was optimized for long-term stability by removing all possible stray reflections, capturing idler components. The long-term power stability measurement is shown in Figure 4.20a suggesting no power drifts in the system.
Part of the beam was reflected with a CaF$_2$ wedge where it was telescopied (to have it x3 times smaller) into a diagnostic area for spectrum, pulse duration and beam shape measurements. Figure 4.24b shows the measured beam profile when the last high-power amplification stage is not on (only the first one). Figure 4.20c shows the final measured beam profile at the full output power of the system. The beam shape is excellent considering how the pump beam is distorted. The small thermal distortions around the beam are due to the fan-out in the last amplification stage. Finally, Figure 4.20d shows the spectral evolution in the system after every amplification stage.

![Graph](image)

**Figure 4.20.** High-power output beam characteristics. (a) Long-term output power stability of the system measured after compressor. (b) Output beam shape when only the first high-power stage is on. (c) Output beam shape when only the second high-power stage is on. (d) Spectral evolution in the system after each amplification stage. OPA1 stands for the pre-amplifier and OPA2 and OPA3 stand for the high-power amplification stages.

### 4.8 Dispersion Management

The final and one of the most important aspects of the system left to discuss is the dispersion management in the CPA scheme. As discussed in Chapter 2, the mid-IR has an advantage for compression possibility in bulk materials since there are
materials exhibiting both negative and positive GDD. However, all standard transparent optical materials exhibit positive TOD in this spectral range. Thus, a careful dispersion management has to be implemented to minimize the TOD and material in the system. The conceptual dispersion management layout in our system is shown in Figure 4.21. We start from a Ti:sapphire oscillator which effectively yields compressed pulses. Then via the time-gated pulse shaping technique, we have a freedom to impose an arbitrary large phase. However, the pulse shaping window is shorter than the mid-IR amplification window. This is because the temporal extent supported by the pulse shaping is limited by the 515-nm OPA pump derived via SHG, while the subsequent NOPA stages are pumped by 1030-nm pulses which have undergone low-efficiency SHG flattening as discussed before. Hence, we use a prism stretcher to further stretch the pulses. The stretcher is configured to provide a large negative TOD. Depending on the prism insertion, the prism pair can either provide a large negative or positive GDD. Our OPCPA chain is based on PPLN, it uses many lenses and windows made out of CaF\(_2\) and thus the system inherently provides large negative GDD and positive TOD for the mid-IR pulses. Finally, we target to compress the pulses in bulk since bulk compressors (using anti-reflection coated material or uncoated material at Brewster incidence) exhibit very low loss, are very simple to add into the beam path, and do not involve any multi-pass arrangements or the introduction of angular chirp. These features are especially important in the challenging mid-IR spectral region. Furthermore, in contrast to chirped mirror-based compressors, bulk compressors provide smooth phase, which is then easy to compensate.

![Diagram of dispersion management scheme](image)

Figure 4.21. Conceptual dispersion management scheme in our mid-IR OPCPA system.

Given these constraints, we could compress the pulses either by using negative or positive GDD materials. However, because the OPCPA chain has a negative GDD, it makes the most sense to compress with a positive GDD material. This way dispersion in the system acts as a natural stretcher increasing the pulse duration between the amplification stages as the gain between the stages becomes smaller and extraction efficiency higher. For our first version of the system [49], we have tried to use a negative GDD based compression scheme (in sapphire), but we found that this scheme yielded a significant cross-talk between the spectral components in the pre-amplifiers. Therefore, for this version of the system, I chose to use a positive GDD compressor. This choice of the phase sign allowed to reduce the TOD contribution nearly by half. A lens-free system was an attractive option, which could highly simplify dispersion management. However, the need for cylindrical tunable optics would have dramatically complicated the optimization of the amplification stages and would have required much more space than it was available for the system.
4. High-power Mid-IR OPCPA

Figure 4.22. Dispersion propagation in our system. (a) Group delay dispersion (GDD) as a function of dispersive elements in the system. (b) Third order dispersion (TOD) as a function of dispersive elements in the system.

Figure 4.22 shows how GDD and TOD evolves as a function of dispersive elements in our system. We start from a large negative GDD and TOD imposed by the pulse shaper. Then after the time-gating, the pulses are further stretched in a prism pair. Then the pulses are further stretched due to the dispersion of the amplification chain and finally are compressed in a 25 mm thick ZnSe compressor oriented at Brewster angle. We chose to use ZnSe as our compression material since it offered favorable GDD over TOD ratio. Furthermore, compared to silicon (Si) it also had a smaller nonlinear refractive index and thus smaller B-integral. We have attempted using Si samples to compress the pulses, but we have observed excessive heating of Si. This could be due to a very strong two-photon absorption in the material.

A significant disadvantage of using ZnSe or Si as compressor materials is that if negative GDD materials are used before the experiment (such as CaF$_2$ windows) before incoupling into a vacuum chamber, the pulses will be compressed twice: in the bulk of positive GDD material and at the output facet of a negative GDD material. To minimize this problem, the beam has to be maximally expanded on ZnSe or Si. Also, since the refractive index of these materials is very large, Brewster angle is very large, thus leading to a beam projection over a large area of the material and thus minimizing the intensity inside. Nevertheless, for this purpose, I have prepared a propagation calculation where the B-integral can be estimated. Consider the beam expanded to 3 mm radius $1/e^2$ (beam size needed for routing the mid-IR beam to another lab via our periscope system discussed in section 5.2.4). Figure 4.23 shows the calculated intensity evolution during such double pulse compression configuration before the beam would enter a vacuum chamber for the HHG experiment. From this calculation, the accumulated B-integral up to the HHG target is found to be 2.2 rad. Such nonlinear phase is tolerable. In case it would prove to deterioe the pulse shape, the beam could be further expanded.
To characterize the output pulses from our system, we have used a frequency-resolved optical gating (FROG) technique [136]. Since the pulses were centered near 2 \( \mu \)m, the second harmonic FROG signal was crossing 1 \( \mu \)m, where instead of standard Si-based spectrometers one must use noisier InGaAs spectrometers. To leverage good signal-to-noise ratio from Si-based spectrometers, we used the third harmonic FROG to characterize the pulses. For this purpose, we have built a FROG setup and recorded the spectrograms. Figure 4.24 show the recorded (a) and reconstructed (b) spectrograms. Figure 4.24 (c) shows the reconstructed pulse temporal shape with 16.5 fs FWHM duration. Figure 4.24 (d) shows the reconstructed, measured spectrum and reconstructed phase. The measured spectrum matched well the reconstructed spectrum indicating a reliable FROG reconstruction. The reconstructed phase is virtually flat, meaning that the output pulses are transform-limited. The center wavelength of the measured spectrum was 2.2 \( \mu \)m and the pulse duration was 16.5 fs FWHM, which corresponds to only 2.2 carrier-wave cycles. From this FROG measurement, we could estimate the peak power of the pulses to be 14.2 GW.
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Figure 4.24. Output pulse duration characterization. (a) Recorded FROG trace. (b) Reconstructed FROG trace. (c) Reconstructed pulse shape with 16.5 fs FWHM duration. (d) Solid line indicated the reconstructed spectrum. Dashed red line indicates the independently measured spectrum. Dashed orange line shows the reconstructed phase.

4.9 Conclusions

In this chapter, I have described in detail our new mid-IR OPCPA system delivering world-record parameters. The system excels in its peak power, which is more than twice higher compared to our previous record-system [49] (published in 2018 and not described in this thesis). The system also delivers very high-average power needed to overcome the low HHG flux issue. Furthermore, in this system, we systematically demonstrated how PPLN crystals can be used at high average powers. We also have shown more than 41% quantum efficiency from a single ultra-broadband amplification stage, which is among one of the highest efficiency parametric amplification stages reported for such class systems. Although the system in this state is already suitable for strong-field ionization and HHG experiments, it still has room for improvements. First of all, the bandwidth of the system can be optimized to generate even shorter, sub-two cycle pulses. The output power of the system can be increased only if the A400 amplifier problems in the grating compressor are solved (the current gratings are very lossy). Furthermore, the spatiotemporal flattening of the pump pulses for the last amplification stages can be further optimized to be able to extract more power from the pump while improving the output beam shape. Finally, the CEP stabilization of the system can be implemented by using the CEP-OPA concept. In the next chapter, I will describe how this system was used for the first proof-of-principle HHG experiments.
Chapter 5
Attosecond Technology

In the motivation chapter, I have explained the state-of-the-art attosecond technology and the requirements for the next generation attosecond measurements. This particularly includes generation of soft x-ray radiation which could be used to generate sub-10 as pulses with sufficient flux for experiments. Motivated by the versatility of high-repetition rate sources, we have developed a mid-IR laser system operating at 100 kHz repetition rate, yet providing sufficient peak power for HHG in noble gases. This laser system was described in the previous chapters. In this chapter, I will demonstrate its use for the HHG experiments, design considerations associated with the experimental apparatus and I will present the complete attosecond pump-probe beamline design.

5.1 HHG Scaling Considerations

5.1.1 Dipole Response Scaling
To be able to design a complete attosecond pump-probe beamline, a good understanding of the HHG process and its phase-matching is needed. Starting from the SFA described in the theoretical foundation chapter we can derive general dipole response strength scaling laws. First of all, as shown in the Figure 5.1a, the dipole strength scales as $\propto I^{9.5}$ when considering an integrated response above 30 eV. This scaling effectively holds until a significant ionization is reached (on the order of 1% ionization probability). At higher intensities, the dipole response starts to saturate. Another very important and general scaling is associated to the decreasing dipole response strength as the ionization potential is increased as shown in Figure 5.1b. From the figure it can be seen that if helium noble gas is used compared to xenon, the dipole response strength will be $2 \cdot 10^5$ times weaker. However, as we have seen in the theoretical foundations chapter, the intensity on the target has to be limited to avoid excessive ionization. As we will see later, the optimal intensity for xenon is 7 times lower than for helium, thus effectively leading to a weaker dipole response strength possible for xenon than for helium. Therefore, from these considerations, the highest ionization potential gases at the maximum allowed intensity should be used.
Figure 5.1. Dipole response strength scaling. (a) The dipole response strength calculated for helium gas driven by two-cycle 1030 nm pulses. The response strength was evaluated by integrating from 30 eV up to the cut-off. The fit was $\propto (I/I_0)^{9.5}$, $I_0 = 1$ TW/cm$^2$. (b) The dipole response strength as a function of the ionization potential normalised to xenon response. For the calculations two-cycle 1030 nm pulses at 200 TW/cm$^2$ intensity were assumed. The fit was $\propto e^{-0.98I}\rho$.

In the theoretical foundations chapter, using the SFA I have demonstrated that the single-atom response strength around the cut-off at the intensity where phase-matching is still feasible will scale as $\propto \lambda^{-9.3}$. Combined with the dramatic dipole response strength scaling to the intensity it can be concluded that for a given targeted photon energy, the highest flux can be obtained by using the shortest possible driving wavelength with the highest possible ionization potential target at the maximum allowed intensity. For example, let us consider the industrially relevant 13.5 nm (92 eV) wavelength. It is possible to generate this wavelength via phase-matched HHG from 515 nm driven HHG in helium ($I_p = 24.6$ eV), 1030 nm HHG driven in argon ($I_p = 15.8$ eV) or helium and also via 2200 nm driven HHG in xenon ($I_p = 12.1$ eV). When targeting similar ionization fraction and considering only two-cycle pulses, the intensities can be estimated as shown in the legend of the Figure 5.2. Here, and elsewhere in the thesis, the ionization fraction means ionization probability normalized to the critical ionization level (defined in 5.1.2). The mentioned figure shows the calculated dipole response strength for various possible approaches to generate the EUV radiation. From the figure it is clear that the dipole response is strongest if the shortest possible driving wavelength with the highest possible ionization potential is used. However, the dipole response can be slightly different than predicted by the used SFA model due to many-electron interaction effects [162] as well as Coulomb interaction terms and other effects not included under the SFA formalism [163].
HHG Scaling Considerations

Figure 5.2. Single-atom response $|x_n|^2$ calculated using SFA for different types of gases and driving wavelengths. The intensities were chosen so that phase-matching of HHG would still be possible.

This calculation does not consider the phase-matching related effects due to the pressure and absorption length scaling for different generation conditions [93]. These aspects will be discussed later. However, the general trend will remain the same. Actually, the highest HHG conversion efficiency in this spectral range was demonstrated by using UV driving laser and multiple ionization of gases (i.e. the HHG from ions with even higher ionization potentials than available from helium gas) [91]. Unfortunately, this approach has limited cut-off scaling possibilities and it would be very challenging to span the complete water window and be able to generate sub-10 as isolated attosecond pulses (IAPs) at the same time. Because of this reason, we have decided to focus on scaling the phase-matched HHG cut-off with mid-IR lasers and using helium as the gas target. On top of this, since the cut-off is generated from the most intense cycle of the waveform, it can directly contain only a single attosecond burst and thus in this thesis I will focus on HHG target design considerations always following the cut-off and allowing for IAP generation with few-cycle pulses.

### 5.1.2 Scaling of HHG Phase-matching

The general HHG phase-matching terms I have already discussed in the theoretical foundations chapter. In that chapter we have derived the equation $p = \frac{\lambda^2}{2\pi^2 w_0^2 \delta n \left(1 - \frac{\eta(x,t)}{\eta_{\text{crit}}}ight)}$, which can be used to estimate the phase-matching pressure at the focus of a beam. Note that throughout the thesis, the pressure is used interchangeably with density. The equation is parametrized with the critical ionization level $\eta_{\text{crit}} = \left(1 + \frac{r_e N_{\text{atm}} A^2}{2\pi \delta n}ight)^{-1}$ parameter with $\delta n = n_{\omega_1} - n_{\omega_q}$. If the actual ionization...
level $\eta(z, t)$ is too large, no phase-matching is possible. Thus, it is crucial to understand the impact of this parameter.

The critical ionization parameter depends on the generated EUV/SXR cut-off via the refractive index $n_{\omega_q}$. However, a priori, it is not possible to know what EUV/SXR energy to use for calculations, since it is coupled to the laser intensity and thus to the ionization fraction $\frac{\eta(z, t)}{\eta_{\text{crit}}}$. To solve this coupled nonlinear problem, I have used the following algorithm:

1) $n_{\omega_q}$ is approximated to be 1 and the critical ionization level for the desired driving wavelength and gas type is estimated. This is a relatively good approximation for SXR.

2) The needed peak intensity to reach a predefined targeted ionization fraction $\frac{\eta(z, t)}{\eta_{\text{crit}}}$ for a given input pulse duration at the peak of the pulse is calculated by using PPT-CA ionization rates.

3) From the determined peak intensity, the corresponding cut-off photon energy is calculated.

4) The calculated cut-off photon energy is used to estimate new $\eta_{\text{crit}}$ value.

5) The newly estimated $\eta_{\text{crit}}$ is used to refine the intensity estimate and the new cut-off estimate can be found.

Since $\eta_{\text{crit}}$ is slowly varying with respect to the EUV/SXR photon energy, the algorithm converges to the targeted ionization fraction $\eta(z_{\text{target}}, t_{\text{target}})/\eta_{\text{crit}}$ after 3-4 iterations. As a consequence of this, the needed intensity for the targeted ionization fraction can be determined precisely. This is a crucial step to be able to study the HHG phase-matching properties at different conditions, such as different types of gases, different driving wavelengths or pulse durations.

Using the previously described algorithm, the phase-matching pressure was calculated as a function of the ionization fraction at the peak of the pulse and at the focus, i.e. $\frac{\eta(t=0, r=0)}{\eta_{\text{crit}}} / \frac{\eta}{\eta_{\text{crit}}}$. Since the peak intensity is determined by the ionization fraction, the beam radius can be parametrized via the peak power of the pulses. Hence, the phase-matching pressure curves for different peak powers and ionization fractions can be obtained and are shown in Figure 5.3a. The calculation was performed for a helium gas target and 2200 nm driving wavelength. From the figure it can be seen that in order to achieve mid-IR driven HHG phase-matching with a significant ionization fraction $\eta/\eta_{\text{crit}}$, and without excessive pressures, greater than 10 GW peak power pulses are necessary. Furthermore, from Figure 5.3b it can be seen that the 10 GW peak power pulses need to be focused to around 35 μm with confocal parameter of 3.5 mm.
Figure 5.3. Helium phase-matching conditions at different peak powers and ionization fractions. The calculation was performed for helium gas target and 2200 nm driving wavelength. The peak power was parametrized in such a way that the beam size is adjusted to achieve the targeted ionization fraction $\eta/\eta_{\text{crit}}$. (a) The acquired phase-matching pressure curves. (b) The beam waist needed to achieve the targeted ionization fraction at different peak powers.

From the Figure 5.3 it is also possible to infer a certain targeted ionization fraction. Since we are intending to use >10 GW peak power pulses for ionizing the helium target, the expected ionization fraction without excessively high phase-matching pressure would then be $\eta/\eta_{\text{crit}} = 0.85$ leading to around 40 bar expected phase-matching pressure for helium. For this fixed ionization fraction, I have performed a set of calculations to compare different driving wavelengths and different types of gases. Figure 5.4a shows the calculated intensity yielding 85% ionization fraction to the critical ionization shown in Figure 5.4b. This corresponds to the phase-matchable classical cut-off shown in Figure 5.4c. The corresponding pressure needed to achieve the phase-matching condition considering 10 GW peak power pulses is shown in Figure 5.4d. For other peak powers the pressure can be calculated by the $p \propto 1/P_{\text{pk}}$ scaling relation. It is worth noticing that for mid-IR wavelengths ($> 2 \mu$m), the critical ionization level is around 0.01%. This therefore requires to use high-purity helium for experiments, with impurities less than $10^{-5}$. Another useful observation is that the phase-matching pressure for HHG in helium scales as $\propto \lambda^{1.6}$. The same scaling power is found to apply to the cut-off as well.
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Figure 5.4. Phase-matched HHG scaling vs wavelength and gas type. (a) Shows the peak intensity needed to reach 85% ionization fraction \( \eta/\eta_{\text{crit}} \). (b) Critical ionization level around the cut-off with peak intensity from (a). (c) Phase-matched cut-off calculated with intensity from (a). (d) Phase-matching pressure scaling for 10 GW peak power pulses.

5.1.3 Scaling of Plasma Effects in HHG

The calculations presented in the previous section demonstrated the general HHG phase-matching considerations. However, the actual scaling will be modified due to the self-action effects effectively limiting the interaction length. For example, due to the ionization, the beam will experience strong plasma defocusing [95] and spectral blueshifts [164]. These effects will dramatically limit the achievable peak intensity, flux and cut-off.

To be able to estimate scaling of the self-action effects, we can first estimate the B-integral related to the plasma generation with \( B_{\text{plasma}} = \frac{2\pi}{\lambda} \Delta n_{\text{plasma}} L \) (assuming that the beam is not significantly modified by the nonlinearity). Here the refractive index change \( \Delta n_{\text{plasma}} \) is related to the ionization level, which I assumed to be 85% of the critical ionization at the peak of the pulse. \( L \) is the target length, which I take to be 1 mm (later it will be rescaled). The B-integral evolution depending on the driving wavelength and target gas type for phase-matched cut-off HHG is shown in Figure 5.5a. Although the presented B-integral values do not seem very large, they are acquired within a single-cycle duration and thus are of major importance as we will see later. From the figure we can see that the nonlinear self-action effects will get stronger for mid-IR driven HHG conditions and that they are especially strong for helium gas targets. These B-integral curves scale with \( 1/P_{pk} \) and suggest that
for a constant interaction length, the B-integral for higher peak power pulses will be smaller.

The B-integral calculations can also be used to estimate the plasma defocusing strength. We can approximate the plasma lens to scale similar to a Kerr lens and thus the description from the theoretical foundations chapter can be used: \( F_{\text{plasma}} = \frac{\lambda}{2\pi w_0^2} B_{\text{plasma}} \). However, to compare the defocusing to the interaction length, it is convenient to multiply \( F_{\text{plasma}} \) with a confocal parameter \( b \), which leads to \( F_{\text{plasma}} b = B_{\text{plasma}} \) expression. Therefore, the plasma defocusing effects will scale the same way as the B-integral.

From the B-integral, the blueshift can readily be estimated with \( \Delta E_{\text{blueshift}} = \frac{\hbar B_{\text{plasma}}}{(\tau/2)} \). Here \( \tau \) is the pulse duration and since we target to achieve 85% ionization fraction at the peak of the pulse, the effective time for the associated phase change is halved. Note that for all calculations presented here I have assumed two-cycle pulses. For a given B-integral, the severity of the pulse distortion effects will become less pronounced for longer pulses with scaling \( \Delta E/E_{\text{photon}} = B/(\pi N_{\text{cycle}}) \), where \( N_{\text{cycle}} \) is the number of optical cycles in the pulse.

Figure 5.5b shows the blueshift calculation results for different driving wavelengths and various target gases used at pressures where the cut-off is phase-matched. For the calculation I have assumed 10 GW peak power pulses. From this figure it can be seen that the blueshift from helium will be significantly worse compared to the lower ionization potential gases. Thus, it is expected that the helium HHG will be strongly limited by the nonlinear phase-shifts associated to the plasma generation. Note that this calculation assumes the same peak intensity, and since the blueshift will distort the pulse, the achieved intensity and thus the actual blueshift will be smaller. The curves in the Figure 5.5b can be linearly scaled with the peak power of the laser pulses, i.e. \( \Delta E_{\text{blueshift}}/E_{\text{photon}} \propto 1/P_{\text{pk}} \) under the assumption of a constant interaction length \( L \).

![Figure 5.5. Self-action effects scaling for the phase-matched HHG with 10 GW peak power pulses. (a) B-integral accumulated up to the peak of the pulse during HHG in a gas target. (b) Blueshift of the carrier wave normalised to the carrier wave photon energy.](image-url)

The calculation above addressed self-action effects scaling for a fixed interaction length. However, the acquired scaling can be used to estimate the optimal
interaction length for the HHG before the nonlinear effects distort the pulse. To be able to estimate the optimal interaction length, first the amount of tolerable blueshift needs to be determined. By considering the experimental results (to be presented later), \( \Delta E_{\text{blueshift}}/E_{\text{photon}} = \exp(-2) = 13.5\% \) could be a good estimate. The particular choice of this parameter will not affect the determined scaling laws but will produce a constant offset. This parameter also determines the maximum tolerable B-integral depending on the number of cycles in the pulse with relation \( B = \pi N_{\text{cycle}}/e^2 \), leading to 0.85 rad B-integral criterion for a two-cycle pulse.

Figure 5.6a shows the calculated corresponding interaction length for different driving wavelengths and different gases considering phase-matched HHG at cut-off with ionization fraction of 85\% and blueshift up to 13.5\% for two-cycle and 10 GW peak power pulses. Note that the demonstrated interaction length scales linearly with the peak power and pulse duration. This figure suggests that the interaction length decreases unfavorably for mid-IR driven HHG.

![Figure 5.6](image)

**Figure 5.6.** Phase-matched HHG interaction length scaling. (a) Calculated interaction lengths for 13.5\% blueshift of the carrier wave photon energy and assuming 10 GW peak power pulses. The interaction length for other peak powers can be scaled linearly. (b) Interaction length normalised to the corresponding confocal parameter needed to reach the targeted ionization fraction. This parameter is independent of the peak power.

The interaction length is compared with the corresponding confocal parameter in Figure 5.6b. Note that in all cases the interaction length is much shorter than the confocal parameter of the laser beam and is equal to around 7.5\% of it. This parameter is invariant to laser peak power but can be linearly extended with number of cycles in the pulse. Thus, the phase-matched HHG by few-cycle pulses is always limited by the plasma associated dephasing and not by the diffraction effects. Thus, the coherence length of HHG is determined in Figure 5.6a.

This therefore suggests that waveguide geometries with extended interaction lengths are not well suited for IAP generation schemes, since the extended interaction length will only lead to in decreased temporal coherence of the generated radiation.

It is also worth mentioning that some authors hinted at a possible nonadiabatic self-phase-matching [165,166]. It is expected to occur on the sub-cycle time scales
during very rapid ionization with few-cycle pulses and when strong self-action effects are present. This regime is not yet fully understood and more future work is required.

5.1.4 Reabsorption of the EUV/SXR

The generated EUV/SXR radiation will be directly reabsorbed by the same medium it is generated in because the generated photon energies are above the ionization potential of the medium. Therefore, a careful choice of the target length as well as a good differential pumping design is needed to remove any residual gas from the generated beam path.

Using HHG phase-matching calculations as described in the previous section, the absorption length for the achievable cut-off with 85% ionization fraction $\eta/\eta_{\text{crit}}$ and 10 GW peak power assumption used to calculate the phase-matching pressure is estimated in Figure 5.7. From the figure it can be seen that HHG driven with a 10 GW peak power at 2200 nm will have an absorption length of 9.5 mm. This is significantly more than the confocal parameter or the interaction length and thus the absorption-limited HHG will not be achieved. Therefore, in order to be able to achieve absorption-limited HHG, the waveguide interaction geometry should be used [93]. However, as discussed in section 2.3.4, the hollow-capillary waveguide loss is too high for guiding high-repetition rate mid-IR pulses focused to small areas suitable for helium ionization. Recently, an antiresonant-reflection photonic crystal fiber was demonstrated for mid-IR guiding with low loss at 88 μm core size [52], which could hold the key for achieving absorption-limited mid-IR HHG in helium with high-repetition rate lasers. However, in case when long HHG targets are used, the group velocity matching between EUV/SXR and the driving field needs to be considered since it will limit the flux scaling and IAP generation possibilities [167]. Furthermore, in the last section we have seen that the effective interaction length before nonlinear effects dramatically distort the pulse are around 7.5% of the confocal parameter, thus the waveguide geometries will effectively reduce the temporal coherence of the generated SXR radiation.
5.1.5 HHG Flux Scaling

Finally, in this section I will address the HHG flux scaling. Constant et al. [168] suggested that to maximise HHG output, absorption-limited conditions should be reached. To be able to reach absorption-limited HHG, the medium length has to be at least three times larger than the absorption length $L_{abs}$ and the HHG coherence length $L_{coh}$ has to be at least five times longer than the absorption length. From the last sections we have seen that the interaction length of the HHG is limited by the self-action effects. On top of that, the generated SXR radiation will be reabsorbed. Thus, in this section I will combine the discussed parameters and also consider the SPA predictions.

Starting from the equation (2.36) discussed in the theoretical foundations chapter for the $q$-th harmonic complex amplitude $\tilde{A}_q$, the flux scaling $|\tilde{A}_q|^2$ can be estimated. After solving the integral, the HHG intensity scaling at the cut-off can be estimated:

$$
|\tilde{A}_{E_{cut-off}}|^2 (L) \propto x_{nl}^2 p^2 (1 - \eta)^2 E_{cut-off}^2 \frac{L_{abs}^2}{1 + 4\pi^2 \left(\frac{L_{abs}}{L_{coh}}\right)^2} \times
$$

$$
\times [1 + e^{-\frac{L}{L_{abs}}} - 2 \cos \left(\frac{\pi L}{L_{coh}}\right) e^{-\frac{2L}{2L_{abs}}}]
$$

with $x_{nl}$ being the dipole response strength, $p$ pressure, $\eta$ ionization level, $E_{cut-off}$ harmonic energy of interest. For further calculation I have used the pressure needed to phase-match at the cut-off and the ionization level was set at $\eta = 0.85\eta_{crit}$. $L_{coh}$ is taken to be the interaction length determined in Figure 5.6a.
optimum medium length $L$ is determined by numerically solving the equation $[1 + e^{-\frac{L}{L_{\text{abs}}}} - 2 \cos \left( \frac{\pi L}{L_{\text{coh}}} \right) e^{-\frac{L}{2L_{\text{abs}}}}]$ and choosing the length maximizing its value. Depending on the $L_{\text{abs}}$ optimum interaction length $L_{\text{opt}}$ is found to be close to $L_{\text{coh}}$. If the absorption is strong, the optimal target length is slightly less than $L_{\text{coh}}$. For mid-IR driven HHG in helium, the function maximises at $L_{\text{coh}}$. Note that $|A_{\text{cut-off}}|^2$ is invariant to the peak power.

To estimate the effect of phase-matching and absorption for HHG conversion efficiency, it is convenient to define an effective length:

$$L_{\text{eff}}^2 = \frac{L_{\text{abs}}^2}{1 + 4\pi^2 \left( \frac{L_{\text{abs}}}{L_{\text{coh}}} \right)^2 \left[ 1 + e^{-\frac{L}{L_{\text{abs}}}} - 2 \cos \left( \frac{\pi L}{L_{\text{coh}}} \right) e^{-\frac{L}{2L_{\text{abs}}}} \right]_{L=L_{\text{opt}}}}, \quad (5.2)$$

so that the complex amplitude scales with

$$|A_{\text{cut-off}}|^2 \propto x_{\text{nl}}^2 p^2 L_{\text{eff}}^2 (1 - \eta)^2 E_{\text{cut-off}}^2. \quad (5.3)$$

This therefore suggests that the pressure-effective length product $pL_{\text{eff}}$ is useful to determine the influence of phase-matching in HHG. Figure 5.8 shows the pressure-effective length product scaling for different gas types and different driving wavelengths at cut-off. These pressure-effective length product curves are invariant under the peak power of the pulses. The curves suggest that helium and neon gases offer favorable flux scaling possibilities at mid-IR and thus possibly could help to offset the weaker dipole response.

![Figure 5.8. HHG phase-matching pressure and absorption length product scaling.](image)

To estimate the flux, $|A_{\text{cut-off}}|^2$ is multiplied with the interaction area parametrized with the peak power. Finally, to have the complete picture, I have included the dipole response strength estimated from the SFA model. In optimal conditions,
the HHG flux in helium gas is predicted to scale as shown in Figure 5.9. The figure shows that although the dipole response strength scales highly unfavourably with $\lambda^{-9.3}$, the phase-matching can partly compensate this scaling to $\lambda^{-3.6}$. Each term contributing to this final scaling is shown in Table 5.1. Note that here the flux is estimated in normalized energy units for a constant peak power pulses with two-optical-cycle pulse duration.

<table>
<thead>
<tr>
<th>Term</th>
<th>$x_{nl}^2$</th>
<th>$p^2$</th>
<th>Area</th>
<th>$I_{\text{eff}}^2$</th>
<th>$E_{\text{cut-off}}^2 (1 - \eta)^2$</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scaling</td>
<td>$\lambda^{-9.3}$</td>
<td>$\lambda^{3.15}$</td>
<td>$\lambda^{0.45}$</td>
<td>$\lambda^{-0.8}$</td>
<td>$\lambda^{2.89}$</td>
<td>$\lambda^{-3.6}$</td>
</tr>
</tbody>
</table>

Table 5.1. HHG flux scaling with wavelength: the contributing terms.

These HHG flux scaling considerations are useful as guidelines for experimental apparatus design. However, they cannot predict the exact spatiotemporal structure of the generated attosecond pulses. To be able to do so, we have developed a full 3D beam propagation code using SFA [169] and PPT ionization rates. This numerical simulation code includes full macroscopic propagation of the laser and EUV/SXR fields. The code still remains to be used for systematic studies with mid-IR driving wavelengths, but its usability was already established in studies at 800 nm driving wavelength range. In one study we have investigated laser noise coupling to the HHG process via the blueshift. The generated EUV spectra at 1 kHz were correlated with laser power fluctuations. We have shown that laser fluctuations can be used to predict EUV spectral noise and thus allows for effectively balanced noise cancellation. The experimental results were supported by the nonlinear propagation simulations. The study was published in [170] and is not discussed in this thesis. The reader is invited to read the paper to get better understanding of the scheme. The scheme is expected to be implemented at the high-repetition rate beamline.
5.2 HHG Experiment Assembly

In the last two sections I have discussed some general scaling rules for the HHG experiment design which could allow us to reach the targeted soft x-ray continuum spanning the complete water window with our mid-IR laser system. We have found that a helium gas target should be used since it provides the best cut-off scaling possibilities and minimum reabsorption for the generated photons. We have also identified that the beam focus size has to be around 35 μm and that the phase-matching pressure will be around 40 bars. This is an extreme target pressure and no to date pump-probe beamline is demonstrated with such high target pressure. In this section I will detail the HHG experiment’s practical implementation.

5.2.1 HHG Module Design

To be able to design a high-pressure HHG module a good understanding of the gas flow dynamics is needed. At the interface between high-pressure and vacuum, gas will expand into vacuum without any resistance and thus the expansion will be supersonic. In case a gas nozzle is used, the pressure just outside the nozzle will be dramatically less than the backing pressure due to the expansion. This kind of gas target is well suited for experiments limited to reabsorption, since the laser propagation direction can be orthogonal to the gas propagation direction. However, this also means that the backing pressure needs to be much higher than what is achieved just near the nozzle. In case 40 bar target pressures are needed, such
design becomes impractical due to the limited gas pressures and gas capacity available from standard gas bottles. Because of these reasons, I chose to generate HHG inside a gas needle, which the laser drills itself through. This way, the HHG is generated in the gas before it is expanded and thus high-gas density targets can be achieved.

Our high-pressure HHG module is illustrated in Figure 5.10a. The laser light is focused into a gas needle (blue) which it drills through. The HHG occurs inside the needle and the generated EUV/SXR radiation then exits the module through two apertures. The first aperture is used to create the first differential pumping compartment (marked in dashed red). The second aperture is used not only to define the second differential pumping stage (marked in dashed yellow), but also it serves as a diffractive mid-IR beam filter. This way most of the mid-IR power after HHG can be already separated.

![Figure 5.10](image.png)

Figure 5.10. High-pressure HHG module. (a) Cross-section parallel to the beam propagation. The red dashed-line indicates the first differential pumping chamber. The yellow dashed-line indicates second differential pumping chamber. (b) High-pressure HHG module side-view. (c) Photograph of the needle during mid-IR driven HHG.

To explain the HHG module in more detail, first I will provide some key dimensions. The entrance window is 39 mm away from the HHG needle. We have used 3 mm and 5 mm thick CaF₂ windows. We also had mechanical spacers to be able to
place the entrance window further away from the HHG needle in case the B-integral in the window would be too large. For the first HHG experiments we have used needles made out of steel which had an inner diameter of 1 mm. Since steel is easy to ablate, the drilled hole size was expected to be larger than the beam radius. In case this would have led to too-high mass flows, we also had HHG target needles made out of quartz available. Since quartz is very hard to drill, the hole size was estimated to be around the beam diameter size. From the HHG needle to the first differential aperture there was 7 mm distance. This distance was chosen to be small enough so that the generated radiation would travel in a region of relatively high-pressure only for a very small distance and that the pressure leak to the next differential pumping stage would be small enough. The aperture diameter was machined to be 0.5 mm. After this aperture, the beam travels 91.5 mm distance up to a 45° gold coated copper mirror with a 0.7-mm-diameter hole oriented along the beam propagation direction. Most of the mid-IR beam is reflected upwards and then collimated with a lens-window placed 40.5 mm away from the 45° mirror. Meanwhile, the SXR beam exits to the experiment or diagnostics via the hole in the mirror. In case the SXR/driving field separation would prove to be not sufficient to avoid damage of filters, annular driving beam shapes could be used for the HHG [171,172].

The whole HHG module is welded onto a flange as shown in Figure 5.10b. Therefore, it can be directly plugged into a vacuum chamber with an attosecond interferometer and used to generate SXR radiation in one of the interferometer arms.

Finally, Figure 5.10c shows a photograph of the HHG taking place in the gas needle. In the figure it can be seen that the entrance hole size in the steel needle is larger than the exit hole since a wider supersonic gas expansion with a Mach diamond is visible at the entrance side. At the same time, it can be seen that the zone of silence at the exit side is interrupted by the reflection from the skimmer with the first aperture. The skimmer was placed on purpose into the zone of silence to avoid turbulence related noise transfer to the attosecond pulse propagation. The disruption of the zone of silence could possibly be partly mitigated by having a sharper skimmer construction.

### 5.2.2 Differential Pumping Scheme

To understand the differential pumping scheme performance and to appropriately dimension the apertures and vacuum pumps, first a good mass flow into the vacuum system estimate is need. At the high-pressure gas expansion to vacuum conditions, the gas flow becomes choked and its mass flow $Q$ is predicted by the following equation [173]:

$$ Q = A_{\text{min}} \sqrt{\frac{\pi}{4} \bar{c} \psi p} $$

(5.4)

where $A_{\text{min}}$ is the aperture area (in case of a well-defined holes $A_{\text{min}} = 0.86 \cdot A_{\text{geometric}}$), $\bar{c}$ is the average thermal velocity of gases, $\psi$ is the flow function, which
is clamped at a value of 0.513 for critical flow of helium and \( p \) is the gas supply pressure. Once the mass flow is determined, the pressure in the vacuum chamber can be found by knowing the effective pumping speed \( S_{\text{eff}} \): \( p = \frac{Q}{S_{\text{eff}}} \).

For evacuating the first differential pumping stage I have chosen a A100L (Pfeiffer) pre-vacuum pump with the specified pumping speed of 27.8 L/s at 2-5 mbar range. This is a high-duty vacuum pump designed to handle high gas flows. However, in order to avoid the noise and vibrations generated from this and other pre-vacuum pumps, we have located the pumps 10 meters away in another room with a special construction helping to minimize vibration transfer to the experimental apparatus. When the pump is placed so far, it is crucial to design appropriately dimensioned vacuum pipes so that the pump is not strangled. For this purpose, we have installed large diameter vacuum pipes near the ceiling which allowed to reach better than 24 L/s effective pumping speed at the first differential pumping stage.

The second differential pumping stage was already supported by a turbo-molecular pump HiPace 300 (Pfeiffer) with an estimated effective pumping speed of 224 L/s. To minimize noise transfer from the turbo-pump, we have used our home-build vibration-dampening mechanism, which is visible in Figure 5.10b. The pump is separated from the vacuum chamber via a bellow having the same aperture as the pump. Compared to commercially available solutions, this approach has larger footprint, but does not strangle the pump.

In order to be able to verify the differential pumping capability and the model used to predict the mass flow, mechanically pre-drilled gas needles were used. First, a needle with two holes of 150 \( \mu \text{m} \) was used and a pressure slope was measured as shown in Figure 5.11. From the slope of the fit, the effective pumping speed could be calculated to be 24 L/s, very well fitting the expected speed. Since this verified the model, a pressure slope can be used to calculate the effective hole size in the needle drilled by the laser.
In order to verify the differential pumping capability of the setup, a needle with two 60 μm holes was used. The recorded pressure in the first and the second differential pumping stage is shown in the Figure 5.12. The pressure in the first stage rises up to 8 mbar with 70 bar pressure in the target. This approximately corresponds to a pressure differential of 8750. From the pressure slope, the hole size could be estimated to be 56 μm, close to the expected value. The pressure in the second stage was increasing quadratically to the input pressure, possibly due to a decreasing turbo-molecular pump capacity at higher pressures and increasing number of atoms traveling directly from the needle hole as a jet. However, even at the highest pressures, the pump electrical current was below 5 A (out of 10 possible), thus indicating that there is a significant room for supporting larger diameter holes in the needle. The HHG module was connected to yet another vacuum stage supported by a HiPace 300 pump. The pressure in that stage was 4.7 \cdot 10^{-7} \text{ mbar} indicating excellent differential pumping performance of the system with a pressure gradient greater than 11 orders of magnitude over 16 cm distance. Therefore, this concludes the design of the high-pressure HHG module.
5.2.3 Soft X-ray Spectrometer

To characterize the generated SXR radiation a commercial spectrometer 251MX from McPherson was used. The spectrometer offers the possibility to switch between two diffraction gratings without the need to break the vacuum environment. The grating design was optimized for a flat-field with focal plane length of 25-20 mm. To be able to cover the spectral range from 15 eV up to keV we have used three different diffraction gratings: 300 grooves/mm covering a range from 15 to 65 eV, a 1200 grooves/mm grating covering the 62-248 eV range and a 2400 grooves/mm grating covering 248-1240 eV.

To detect the spectrum, we have used an Andor Newton 940 charge-coupled device (CCD). The camera is thermo-electrically (TE) cooled down to -85 °C for the best signal-to-noise performance. A signal recorded on a CCD does not directly correspond to spectral intensity, since effectively the units of the measured data are analog-to-digital (A/D) counts per pixel per second. Therefore, to extract the true spectral shape, our measured CCD data are processed to represent the spectrum in units of spectral power density (i.e. W/eV). This is done in the following steps:

1) The A/D counts are related to electron counts via the calibration data provided by the CCD manufacturer. In our case it was 4 electrons per A/D count.

2) The number of detected electrons is related to the number of incident photons by considering that a single photon of energy $E$ will create $E/3.65$ eV electron-hole pairs [174].

3) The probability for a photon to interact with the CCD is corrected using the manufacturer specified quantum efficiency (QE) curve.

4) The number of photons per pixel per second is converted to average power per pixel by multiplying each corresponding pixel by the energy of the photon at the respective pixel.
5) To convert average power per pixel to average power per eV a Jacobian transformation function is applied.

6) Finally, each grating diffraction efficiency is compensated using data available from our manufacturer.

Figure 5.13 shows the impact of these amplitude correction terms combined for each of the used gratings in the later described experiments. From the curves it can be seen that at least an order of magnitude change in the spectrometer sensitivity range occurs. Thus, it is crucial to take this response function into account to show the true spectrum.

![Figure 5.13. Spectrometer raw data conversion curves to power spectral density. The curves were calculated for the gratings we have used. The inverse of these curves can be considered as the spectrometer sensitivity curves.](image)

Finally, the pixel axis of the spectrometer is calibrated by fitting the wavelength to pixel mapping expected from the diffraction equation. For this purpose, filters with absorption edges in the range of interest are introduced in the beam. Also, since the gratings have low diffraction efficiency, the higher diffraction orders are well visible and can be used to improve the calibration accuracy. Furthermore, if peaks at odd-order harmonics of the driving laser energy are visible, they can be used to help improve the fit. Finally, the spectrometer had a strong carbon-based contamination and thus the carbon K-edge was strongly present in the measured spectra. This is present in all SXR spectra reported in the community, such as [28–30].

5.2.4 Beam Delivery
The beams from our laser systems were delivered to the neighbouring laboratory which accommodates the high-energy SXR beamline via our home-built beam routing system. With this system, the beams are elevated to the height of the ceiling, transported to another laboratory and then lowered down to the usual beam height on another optical table. To avoid coupling of vibrations between the optical tables, we have used bellows to isolate vibrations between the tables. Since the beams were transported by more than 12-meter distance, a significant pointing noise is
added on the beam. Therefore, to stabilize the beams on another optical table we have used active 4-D beam stabilization systems (TEM Messtechnik GmbH). To transport the beams, we had three different channels: mid-IR, near-IR and 1030 nm. The final beam delivery architecture is shown in Figure 5.14. For the beam transport of the mid-IR radiation we have used custom, ultrabroadband Si/SiO$_2$ mirrors (Optoman). These mirrors have better phase and reflectivity properties if used for s-polarization. Because of this reason, the mirrors were oriented for s-polarized reflection. Finally, the complete beam path was immersed in nitrogen to avoid any water or carbon dioxide related absorption issues.

![Beam routing implementation. Mid-IR, near-IR and 1030 nm beams were transported from the right side mounted on an optical table in our laser laboratory to the left side mounted on an optical table in our experiment laboratory.](image)

**Figure 5.14.** Beam routing implementation. Mid-IR, near-IR and 1030 nm beams were transported from the right side mounted on an optical table in our laser laboratory to the left side mounted on an optical table in our experiment laboratory.

### 5.3 Water Window Soft X-ray Generation

In this section I will describe the first water window SXR generation at 100 kHz repetition rate. For the experiment, we have used our previously described mid-IR OPCPA output. The output beam was transported more than 15 meters away from the last amplification stage in a nitrogen purged path. The beam was actively stabilized, compressed in a ZnSe bulk compressor and focused into our high-pressure HHG module with a 75 mm long focal length plano-convex CaF$_2$ lens.

The HHG module output was directly characterized with the previously described 251 MX SXR spectrometer. The spectra were measured after blocking the residual pump light with a 100 nm thick aluminium filter. For the first measurements, an argon gas target was chosen. The laser intensity was too high for efficient HHG phase-matching; hence the input beam was clipped with an iris. The recorded spectra have been corrected for the spectrometer response and filter transmission. Figure 5.15a show the argon spectrum measured with the 1200 grooves/mm grating at different gas pressures. For a direct comparison, the recorded spectra with the highest overall flux and with the highest cut-off are shown in Figure 5.15b.
the figure, it can be seen that the measured cut-off dramatically decreases with the increased pressure, although the flux improves a lot. This strongly suggests plasma related limitations on the achievable intensity in the target as well as strong blueshifts of the carrier wave.

![Graph showing pressure scan in log scale and two measured spectra in the linear scale.](image)

Figure 5.15. HHG in argon. (a) Pressure scan in log scale. (b) Two measured spectra in the linear scale. The spectrum measured at 2.4 bar had the highest estimated flux. The spectrum measured at 0.4 bar had the highest measured cut-off, but the integrated energy was 38 times smaller over the same range (60-220 eV).

In order to quantify the plasma related blueshift, we have used a 300 grooves/mm grating, which allowed to resolve well the harmonic structure at the lower end of the spectrum. By fitting the shifting harmonic spacing, the blueshift could be estimated to be around 0.08 eV/bar for the case of HHG in argon. A central wavelength of 2200 nm corresponds to 0.56 eV, thus a 300-500 nm blueshift in 1-2 pressure range is observed. This blueshift matches very well with theoretical expectations shown in Figure 5.5.

Although, the argon gas target already allowed to extend the HHG spectrum beyond what is possible with Ti:sapphire driven beamlines, the spectrum did not extend to the water window. Therefore, as the next gas target, we have used neon to generate SXR radiation. Figure 5.16a shows the spectra measured from neon gas target at different gas pressures. The flux from neon peaked at 10 bar pressure and is shown in Figure 5.16b. The spectrum clearly extends well into the water window and up to 420 eV. Strong carbon contamination associated K-edge absorption is visible at 282 eV.
Since the HHG from neon did not cover the full water window, we finally used a helium gas target to generate the SXR radiation. Figure 5.17 shows the acquired results. The SXR HHG was well phase-matched in the expected pressure range of 30-50 bar. The HHG cut-off extended up to 620 eV, spanning the complete water window and extending beyond it. This recorded spectrum demonstrates the world's first coherent SXR laser source spanning the complete water window at 100 kHz repetition rate. The shown spectrum can support down to 9 as pulses.

Due to the lack of suitable calibrated instruments, the flux of the recorded spectra was not calibrated. However, from the CCD response, a lower bound estimate can be made. Without considering the beam clipping on the slit, the flux from argon could be estimated to be 5.5 nW (60-220 eV), 300 pW from neon (220-420 eV) and 32 pW (250-620 eV) from helium.

This data stands as the first demonstration of a water window SXR source at 100 kHz. However, there is a lot of room for improvements in the measurement and generation of the SXR radiation. First of all, the beam focusing has to be improved to have aberration-free focusing condition. Secondly, the absolute flux has to be...
characterized with a calibrated photodiode. Third, the focusing conditions need to be optimized based on SXR beam profiles recorded with CCD so that the optimal trajectories are phase-matched. Unfortunately, this was not possible to be done during the available time because of an unexpected pump laser downtime.

5.4 Attosecond Beamline Design

In the last section I have demonstrated the world’s first 100 kHz coherent SXR laser source. In this section I will show an attosecond beamline design suitable for characterizing the pulses originating from SXR source and using these pulses for attosecond pump-probe measurements.

5.4.1 General Design Aspects

The conceptual beamline design is shown in Figure 5.18. The beamline contains six vacuum chambers. The input beam is split into two beams: one used for interacting with an experimental target and the other used for HHG. The HHG point is imaged with a toroidal mirror at grazing incidence so that 1:1 imaging is achieved. The generated high-harmonic radiation is recombined with the split beam in the experiment chamber where dynamics are probed in gas or solid targets. The electrons emitted during the interaction are recorded with an electron spectrometer and the transmitted high-harmonic radiation is refocused into an SXR spectrometer.

The planned implementation of the conceptual design is shown in the Figure 5.19. This beamline design allows for a minimal footprint of 3 x 1.6 x 0.9 (L x W x H) m³ which was necessary due to the lack of available experimental space. The high-pressure HHG module described before is installed into the generation chamber as can be seen in the figure. All of the turbo-molecular pumps are decoupled from the chamber by using our vibration dampeners.
To avoid vibration coupling from the pumps and alignment changes when evacuated, the optical setups inside the vacuum chambers are built on separated optical breadboards. However, due to the relatively small size of the breadboards, solid steel breadboard vibration resonances would be close to the turbo-molecular pump rotation frequencies (500-1500 Hz depending on the pump). Thus, effectively allowing noise to couple to the breadboard modes. We avoid this problem by using high-stiffness breadboards.

This can be achieved with honeycomb structures or using high-stiffness materials, such as carbon fiber reinforced polymer (CFRP). A drawback of honeycomb structures is that they require a lot of epoxy glue and to be vacuum compatible, the inside of the breadboard has to be evacuated simultaneously as the full vacuum chamber is evacuated by a separate pump. For experiments were vacuum better than $10^{-7}$ mbar is not needed, CFRP breadboards can be used. For this purpose, we have acquired CFRP breadboards from CabonVision offering the highest possible stiffness solution for our application. These breadboards have outgassing rate of $5.4 \times 10^{-7}$ mbar L/s/cm$^2$, which can be reduced when thermally cycling up to 100 °C. At UHV, the breadboard outgasses water, nitrogen, oxygen and carbon dioxide. With a custom design of the breadboards, the resonances could be achieved to lie outside the pump excitation range (1626, 2502 and 2698 Hz). Finally, it is worth mentioning that the thermal expansion of these breadboards is 5-10 times smaller than steel and it helps to mitigate heating induced changes inside the interferometer.

Figure 5.19. Attosecond beamline design rendering.

The attosecond interferometer is planned to be actively stabilized by measuring the phase of a co-propagating cw laser light and actuating on a nanopositioning
stage PZ 38 2 VAC (piezosystem jena GmbH) optimized for faster than 20 ms response time. This concept was demonstrated and implemented in one of the beamlines in our laboratory [34]. Such stabilization scheme was recently used to reach better than 10 as rms interferometer stability reported by another group and demonstrating extremely fast feedback in the range of up to 300 Hz [175].

5.4.2 SXR Imaging

The generated SXR radiation needs to be transferred to the target area. However, at the SXR spectral range there are no ultra-broadband mirrors. Thus, a grazing incidence optics has to be used. Since initially the beamline was designed to support greater than 1 keV photon energies and the space was limited, the imaging optic design was highly constrained.

To be able to reflect the SXR radiation around 1 keV, a grazing incidence of $3^\circ$ must be used [176]. It was shown by [177] that toroidal mirrors can be used to focus attosecond pulses, however it is crucial that the imaged HHG generation point to the interaction point lies in a perfect 1:1 imaging plane. This is achieved if the imaged point and the object are on the so-called Rowland circle defined by

$$p = q = R \cos \alpha = \frac{\rho}{\cos \alpha},$$

with variables defined graphically in Figure 5.20. $R$ is the tangential radius of curvature and $\rho$ is the sagittal radius of curvature. Because of the targeted $3^\circ$ grazing angle of incidence, the ratio between the curvatures was necessary to be 365. However, due to the lack of space, the smallest possible to manufacture sagittal radius of curvature had to be used. Thus, for imaging 1 keV photons over the shortest possible distance, which was for 114 cm image transfer, we have acquired a gold coated toroidal mirror with 30 mm sagittal radius of curvature and 10953 mm tangential radius of curvature (Winlight).

![Figure 5.20. SXR imaging with a toroidal mirror. $p$ – distance from the object to the toroid, $q$ – distance from the toroid to the image point, $R$ - the tangential radius of curvature, $\rho$ - the sagittal radius of curvature, $\alpha$ – angle of incidence.](image)

After the interaction point, the SXR photons are imaged in the SXR spectrometer only in the horizontal axis using a spherical mirror with a curvature of 11032 mm. This mirror was planned to be used at $2^\circ$ angle of incidence and for
transferring the image over 77 cm distance. The SXR imaging system is expected to have better than 20% transmission for 1 keV photons from the HHG position up to the SXR spectrometer.

5.4.3 Photoelectron Spectrometer

One of the most important instruments in the beamline is the electron detector, which allows to measure electron kinetic energy spectra. These electrons are emitted during the interaction between SXR photons and the material of interest. If during the interaction a sufficiently intense laser field is present, then the emitted electron trajectories will be modified by the laser field. A pump-probe delay scan can be performed and a spectrogram containing the electron emission and the SXR phase is recorded. If an attosecond pulse train is used a reconstruction of attosecond beating by interference of two-photon transitions (RABBITT) can be performed. Alternatively, if an IAP is used, an attosecond streaking measurement is performed. These techniques were contrasted and discussed in [178] and both can yield valuable phase information for the attosecond pulse or photoemission phase measurement.

However, an important challenge for implementation of these techniques in the SXR spectral range is due to low resolving power in a typical time-of-flight electron spectrometer. For instance, if the beamline is driven by a 3 µm laser source which generates 1 keV photons, then the electron spectrometer resolution must be better than 0.41 eV, which corresponds to the resolving power of 2500. In terms of temporal resolution, over 1 meter of free electron propagation (which is large for a time-of-flight spectrometer) this translates into a better than 10 ps time-resolution requirement, which is not feasible with usual electronics. At the same time, the spectrometer must be suited to be used with relatively high background pressures in the range of 10^{-4} mbar necessary for gas targets. On top of that, the photoemission from gas targets is not well localized, due to the extended nature of the interaction. To achieve this demanding goal under such conditions, we have worked with SPECS GmbH who provided us with a modified version of a high-resolution time-of-flight spectrometer Themis 600 suitable for elevated pressures. Now this spectrometer is available as a commercial product under the name of Themis 600 EP. The high-resolving power is achieved by using special electron lens mode settings, scanning over a relatively small energy window which leads to a quasi-time-of-flight/velocity map image recorded on a 2D delay-line-detector (DLD). This image can be transformed back to the electron kinetic energy space with a pre-calculated electron trajectory transformation matrix embedded in the spectrometer software. The actual performance of the device for broadband, high-energy electron spectrum measurements remains to be investigated.

The spectrometer is intended to be used inside a vacuum chamber which has a separate compartment built from μ-metal to shield the interaction area (the spectrometer has 40 mm working distance) from external magnetic fields, such as from the earth. The time-of-flight spectrometer is installed into the interaction vacuum chamber as shown in Figure 5.21 and it has an ultra-high-vacuum compatible valve to be able to exchange the samples without removing vacuum from the
spectrometer. Furthermore, the spectrometer has two differential pumping stages to receive an excellent vacuum at the 2D-DLD position. Moreover, the turbo-pump mounted on top of the chamber is coupled with a gas catch to be able to reduce the background pressure in the chamber and allow for high gas density targets.

Figure 5.21. Interaction chamber rendering.

5.4.4 Vacuum System
An attosecond beamline is an intricate instrument involving high-density gas targets, regions of ultra-high vacuum, and photons which are easily absorbed by any molecules. Thus, a careful vacuum control of the instrument is needed. To achieve a suitable pressure for each vacuum chamber, the vacuum system scheme containing eight turbo-molecular pumps and three pre-vacuum pumps was designed and is shown in Figure 5.22. The system is supported by three pre-vacuum pumps: the A100L (Pfeiffer) pump is used to remove high-gas load from the HHG stage; two more pre-vacuum pumps, nXDS15i (Edwards), are used to support the remaining turbo-pumps. One of the nXDS15i pumps is devoted to higher gas load pre-vacuum and another one is for used for high-vacuum support. The first HHG differential pumping stage pressure sensor is connected to a pneumatic interlock system which allows to interrupt the HHG gas supply in case of a sudden overpressure event.
6. Attosecond Technology

Figure 5.22. High-photon energy attosecond beamline vacuum scheme.

5.5 Conclusions

In this chapter I have demonstrated HHG phase-matching considerations tracking the cut-off at optimal generation conditions. This allowed to infer general phase-matched HHG scaling. The HHG cut-off was found to scale with $\lambda^{1.6}$ and the phase-matched HHG flux is expected to decrease with $\lambda^{-3.6}$. The considerations also infer the optimal HHG experimental conditions, particularly suggesting helium utilized at high gas pressures and very short interaction lengths as the most suited gas target for cut-off scaling with mid-IR lasers.

Lead by this general HHG scaling considerations, I have demonstrated a high-gas-load HHG module capable of being operated at as high as 70 bar target pressures and well suited for mid-IR HHG in a tight-focusing configuration. This HHG module was used to demonstrate the first proof-of-principle mid-IR HHG at 100 kHz repetition rate spanning the complete water window and extending beyond it. This experimental demonstration is expected to enable next generation attosecond technology experiments where high-photon energies are combined with high-repetition rates.
Conclusions

As the first step of the employing the mid-IR HHG for experimental studies, a compact attosecond beamline suitable for operation at greater than 1 keV photon energies was designed and prepared for assembly. This experimental apparatus is expected to enable first attosecond experiments at 100 kHz repetition rates.
Chapter 6
Near-IR OPCPA

To complement the mid-IR OPCPA, in parallel a near-IR OPCPA system was developed as well. Having two OPCPA systems operating at different wavelength ranges allows to cross-checking various experiments with different driving wavelengths. At the same time, the near-IR OPCPA operates in a spectral range where high-quality crystals and optical coatings exist. Thus, this system allowed to decouple risks.

The layout of the system is shown in Figure 6.1. The pre-amplification part delivered Watt-level 100 kHz pulses with 690-920 nm bandwidth. The pre-amplified pulses were further boosted up to 35 W average power level in high-power amplification part of the system. The pre-amplification part of the system contains two amplification stages sharing the same pump beam. The high-power amplification part of the system uses a SHG cascade scheme [141] to have spatiotemporally shaped flat-top beam for the last high-power amplification stage.

During my PhD time, I have worked on the first version of the pre-amplifiers which delivered up to 2.2 W of average power and later were improved to deliver up to 2.5 W average power. The pre-amplifiers has enabled the first angular streaking experiments at 100 kHz [179] as well as HHG I will show in section 6.2. The development of the high-power amplification part of the system is led by my colleague and the system is not yet finalised. However, we were already able to report greater than 21 GW peak power pulses at 100 kHz repetition rate [180]. The system is expected to deliver greater than 30 GW peak power pulses soon. In this chapter I will describe only the pre-amplification part of the system.

![Conceptual layout of near-IR OPCPA system.](image)

6.1 Near-IR OPCPA Pre-Amplification

The pump pulses for the pre-amplification part of the system are derived from 1030 nm A400 output SHG. For the initial version of the system I have used 22 W of 1030 nm which was converted to second harmonic (515 nm) in a 5 mm long LBO crystal with 68% conversion efficiency leading to 15 W of pump power. These pump pulses were focused up to 82 GW/cm² peak intensity in a 1.5-mm long BBO crystal. The amplification took place in the same geometry as discussed in section
4.1 but the nonlinear angle was chosen to be 2.4° which offered the widest amplification bandwidth for the targeted spectral range.

The seed pulses were derived from the $f_{CEO} = f_{rep}/4$ stabilized Ti:sapphire output with at 82 MHz repetition rate. The A400 amplifier picked every 4th multiple pulse, thus the amplified pulses by the near-IR OPCPA were CEP stable. To control the coarse and fine phase, we have used the same time-gated pulse shaping scheme as it was used for the mid-IR OPCPA and thus the first amplification stage also served as the amplifier which gates the pulse shaper output waveform. The near-IR OPCPA was seeded only with 58 pJ pulse energy and in the first amplification stage the pulses were already amplified to 1.6 µJ level (160 mW at 100 kHz, 44 dB gain). The corresponding power slope and spectral evolution are shown in Figure 6.2. The first pre-amplification stage was configured in an unsaturated regime to mitigate the risk of excessive amplification of quantum noise in the system.

![Figure 6.2. Near-IR OPCPA first amplification stage slope. (a) Power slope. (b) Spectrally resolved power slope.](image)

The pump beam from the first amplification stage was directly 1:1 imaged onto the second amplification stage which had a 0.7-mm long BBO. The pre-amplified signal from the first stage was also delivered to the second amplification stage yielding greater than 1.4 W average output power level with 13.7 W of 515 nm pump. This output level was sufficient for seeding high-power amplification stages.

The pre-amplifier output was also used for strong-field ionization experiments. In this case, the intensity on the crystals was increased to more than 115 GW/cm² by sending more pump power (up to 19 W of 515 nm) and hence saturating the amplification. This yielded more than 2.2 W output average power with the spectrum shown in Figure 6.3. The transform-limit was calculated to be 8 fs. This corresponded to 2.5 GW output peak power at 100 kHz.
Dispersion in the system is managed in the following way: the OPCPA seed is chirped with a negative TOD via the programmed phase on the pulse-shaper and GDD is close to zero on the first amplification stage. Then the seed is stretched between each amplification stage with material dispersion providing positive GDD and TOD (lenses and windows). Finally, the pulses are compressed with a chirped mirror pair providing negative GDD of -120 fs$^2$ at 800 nm center wavelength. To compress the pre-amplifier output, 18 bounces were used. The phase on the SLM was fine-tuned until plasma generation in the air was observed.

### 6.2 HHG with Near-IR Pre-Amplifier Output

Since the pre-amplifier output pulses were able to ionize ambient air, it meant that the achievable peak intensity was sufficient for the first strong-field ionization experiments. The pre-amplifier output was routed to our experimental room with our beam routing system discussed in 5.2.4. The output pulses were focused into the HHG module discussed before in section 5.2. The beam was focused with a 75 mm focal length CaF$_2$ lens to a 66 x 40 µm$^2$ beam diameter leading to an estimated 240 TW/cm$^2$ peak intensity. Such peak intensity was sufficient for HHG from lower ionization potential gases, such as xenon, krypton and argon. The generated HHG spectrum in argon is shown in Figure 6.4. The near-IR HHG with pre-amplifiers was performed before the mid-IR system was available for experiments. Thus, it was used to verify and the HHG module operation.
6.3 Conclusions

Since the first demonstration of strong-field ionization at 100 kHz in our group, the output characteristics of the near-IR system were dramatically improved [180]. With the improved output characteristics, the near-IR OPCPA is well suited to replace the "workhorse" Ti:sapphire CPA systems operating at 1 kHz. The upgraded near-IR OPCPA output will be used for versatile HHG generation in any gas target spanning complete photon energy range up to the helium cut-off. Furthermore, the system output will be used to configure the designed attosecond beamline discussed in the previous chapter.

The unique advantage of our attosecond beamline design is that it is arranged for a tight focusing configuration. This means that the high-power near-IR OPCPA output can be used to explore HHG scaling beyond phase-matching. In the past, it was already demonstrated that keV photon energies can be achieved via HHG in near-IR [90]. Due to very high-average power of our laser system as well as high-gas pressures possible in our target, it is expected that a sufficient HHG signal for pump-probe experiments will be generated in this regime. Finally, the second harmonic or third harmonic of the near-IR OPCPA can be used for generation of extreme brightness HHG from multiply ionized plasmas [181]. In combination with the polarization gating technique, it is expected that this approach could offer an alternative path for generating IAPs at high-photon energies.
Chapter 7
Conclusions and Outlook

In this dissertation, I have described a journey starting from no high-repetition rate and GW-peak-power class mid-IR lasers available worldwide to the first demonstration with more than 14 GW peak power pulses in the mid-IR at 100 kHz repetition rate. The work in developing this system laid the foundations for the third-generation femtosecond technology in the mid-IR. The developed laser system is shown to be capable of driving the next generation attosecond experiments where high-repetition rates and high-photon energies are combined.

The developed laser system enabled the first high-harmonic generation (HHG) experiments generating soft x-ray (SXR) photons at this high-repetition rate. We were able to generate SXR continuum spanning complete water window. This work stands as a two-order of magnitude improvement compared to the previous SXR generation experiments conducted at a 1 kHz repetition rate [28,29].

Finally, a complete attosecond beamline design suited for pump-probe measurements at photon energies as high as 1 keV was unveiled. This beamline is designed to be used at the tight-focusing configuration, thus allowing for the first high-photon energy attosecond pump-probe experiments at this high-repetition rates. The experimental apparatus could support the generation of the shortest attosecond pulses to date.

None of these results would have been possible without the innovative achievements we have made in the process. One of the key-enabling innovation was the demonstration of the time-gated pulse shaping scheme [122]. This pulse shaping scheme enabled fully programmable phase and amplitude control just with a single 1-dimensional spatial light modulator (SLM). The technique allows avoiding spectral aberrations typically occurring if a too large phase is applied in a pixelated pulse shaping device. Thus, the time-gated pulse-shaping scheme not only to ensures excellent fine-phase control needed to achieve the highest peak power pulses but also allows to adjust phase in large scale and hence dramatically simplifying the optical parametric chirped-pulse amplifier (OPCPA) design aspects. This pulse shaping scheme is independent of the repetition rate of the laser and thus applies to any time-gated amplifier system provided a suitable SLM.

In this thesis, we have demonstrated a mid-IR OPCPA system based on periodically poled lithium niobite (PPLN) crystals. The PPLN crystals enable engineerable and flexible OPCPA bandwidth control supporting nearly single-cycle pulse amplification. However, these crystals are generally limited by photorefractive distortions. During the OPCPA system development, we were able to mitigate these PPLN associated distortions by choosing appropriate amplification conditions. Thus, we were able to demonstrate that the PPLN crystals can be used at greater than 100 W average powers with high-repetition rate picosecond pump pulses. The final amplification stage in our system used 105 W of pump power and had up to 41% of quantum conversion efficiency. Our work hints that PPLN crystal application at
high-average powers is limited by thermal effects. With the appropriate design of the crystal mounting, it is possible to mitigate the thermal effects and thus enable versatile high-power conversion into the mid-IR platform.

Since our mid-IR OPCPA system is based on PPLN crystals, the supported amplification bandwidth enables single-cycle pulse amplification. With amplification bandwidth optimization we were able to demonstrate high-power amplification with shorter than 1.7 cycle pulses [49]. The bandwidth was limited by our difference frequency generation (DFG) based on BBO crystal. In the near future the crystal will be exchanged with BiBO, which will allow for extending the achievable output bandwidth further into the mid-IR. This extension will enable not only the shorter mid-IR output pulses but also wavelength tuneable few-cycle output. Note that these few-cycle waveforms are achieved directly from the amplifier and involve no nonlinear post-compression and hence the full energy is contained in the pulse.

Recently, we were able to re-design the system in such a way that its efficiency could be improved and new world-record parameters could be reached [35]. The current system configuration delivers up to 25 W of average power with 14.2 GW peak power few-cycle pulses at 2.2 µm. The output parameters of this system were limited by the pump laser. With improvements in the pump laser configuration, higher output powers and even greater peak powers are within reach.

A significant time of my PhD work was invested in the development of our pump and seed laser systems. Our learnings described in the thesis lay foundations for high-power OPCPA pumping and management necessary to have stable long-term characteristics of the system at high-average powers.

Since our mid-IR system bandwidth was extending through the water-absorbing lines, we also encountered various challenges usually not encountered in other low power systems. Particularly, we have observed strong gas lensing effects in air, related to high-power mid-IR absorption during the propagation. To be able to transport the mid-IR output for experiments to another laboratory, we have designed and utilized our home-built beam-routing system. Future challenges will include optimization of mid-IR beam wavefront as well as ultra-broadband mirrors design improvements needed for high-power mid-IR pulses.

The laser pulses from our mid-IR system were used for HHG in gas targets. In this thesis, I have unveiled general phase-matched HHG scaling laws parametrized via peak power of the laser pulses. These scaling laws can be used as a general design guideline for any phase-matched HHG where short attosecond bursts are desired.

Using these scaling considerations, I have designed a high-pressure HHG module with high-efficiency differential pumping scheme. The HHG module contains a short gas cell which supports greater than 70 bar target pressures needed for SXR generation at high-repetition rates.

The module was used for the first proof-of-principle HHG experiments at 100 kHz repetition rates. The HHG experiments in helium gas target yielded SXR continuum extending beyond water window up to 620 eV. This is the first
demonstration were such SXR radiation was demonstrated with high-repetition rate laser source. This demonstration marks the tipping point in attosecond technology where high-photon energies are combined with high-repetition rates.

This laser source will enable space-charge free photoemission experiments from deep core electrons as well as high signal-to-noise ratio experiments in coincidence detection schemes. To be able to benefit from this laser source, I have designed and prepared for assembly a compact attosecond beamline which could support even up to 1 keV photons. This beamline will enable the first pump-probe experiments at such high repetition rates.

In the last chapter I have also demonstrated near-IR OPCPA design and its application for the strong-field ionization experiments. The system is well suited for scaling EUV flux used for attosecond experiments in the typical energy range up to 160 eV. Due to very high-power of the system, the near-IR OPCPA can be used to explore exotic HHG regimes discussed in the previous section.

In the last few years, we have witnessed dramatic progress in high-power OPCPA systems enabled by the emergence of high-power pump laser sources. Today, 1 µm and 2 µm ps pump laser sources already surpass kW-levels [108,109] and thus already enable the next level power scaling of OPCPA laser sources. It seems that there is no end in sight for power scaling of OPCPA. Enabled by the scalability of the parametric amplification scheme, the next generation attosecond experiments will become a routine.


Bibliography


Curriculum Vitæ

Removed for the online publishing
Acknowledgements

Isaac Newton once said that “If I have seen further it is by standing on the shoulders of Giants”. Although this dissertation rests on the past work of the community, my true personal Giants are those who tolerated, supported and helped during this thesis.

My greatest Giants are my family. I am deeply grateful for their continuous support, understanding, encouragement and patience during my thesis. I am especially grateful to my fiancée Dovilė who always inspired and helped me to find motivation.

I would like to say some special words of gratitude to some people I worked the closest:

Christopher Phillips, his contributions and insights in the work of this thesis are invaluable. He has contributed extensively in troubleshooting various experimental challenges, design of OPCPA aspects, particularly he is behind the time-gated pulse shaping idea and many simulations which helped to understand the underlying physics. Truly thank you for always finding time to discuss and teach and yet showing a path for independent work.

Nicolas Bigler, is another Giant on whose shoulders I am standing. When he started his PhD, there was no laboratory. He is behind the first mid-IR system version and countless hours of fixing the pump laser. He laid the foundations for the work I built upon.

Benjamin Willenberg, I am very glad I have had a chance to work together with him. His competence, energy and an unbreakable will to go an extra mile are truly fascinating. We teamed up with Benjamin for the modification of the laboratories to accommodate for our experiments. I truly learned a lot from you.

Stefan Hrishafov, who joined our small OPCPA team sometime later, helped a lot for maintaining and improving our setups and laboratory. He is behind the near-IR OPCPA development efforts. Thank you, a lot Stefan, for your patience and also that you always took effort and time to listen, understand and help.

Mikhail Volkov, with whom I shared so many weekends and had so many intellectually stimulating discussions in our office. He invited me to join to his HHG noise study, which was a truly refreshing and exciting work for me. Thank you for being the bests office mate.
Acknowledgements

Fabian Brunner, who is behind various important improvements in the laboratory necessary for high-efficiency work. Thank you, Fabian, for bringing energy and Powerwolf to E4.

Pierre-Alexis Chevreuil, who joined to work with me for the last year of my PhD. Together with him, we were able to reach the final results of this thesis. His desire for systematic work was truly enabling for the results. I think the group is really lucky to have you joined. Keep it up! Alone or not, I am sure you will reach outstanding results!

Ivan Graumann, with whom I had a chance to work on Yb:CALGO mode-locking project. He taught me a lot about ultrafast oscillators. I am looking forward to completing the GR20 and climbing Bristen with you!

I also would like to thank Lukas Gallmann who always took time to consider and discuss any questions I would pose. He laid the first ideas for the attosecond beamline concept.

Further, I would like to express my gratitude to other members of the team who supported my work by taking their time to advise and help when needed: Jochen Maurer, Fabian Schläpfer, Florian Emaury, Aline Mayer, Andreas Diebold, Sergej Neb, Matthias Golling, Francesco Saltarelli, Jannie Vos, Laura Cattaneo, Matteo Lucchini, André Ludwig and Cornelia Hofmann. Team is also not only about work but also about great atmosphere, for which I would like to thank as well: Arthur Niedermayr, Jaco Fuchs, Marco Gaulke, Nadja Hartmann (the best party minister!), Jonas Heindrich, Léonard Krüger, Lukas Lang, Jacob Nürnberg, Luca Pedrelli, Cesare Alfieri, Zeno Schumacher, Behçet Özgür Alaydin, Ajanta Barth, Dominik Waldburger, Lamia Kasmi and Sandro Link.

I also would like to acknowledge and thank Marcel Baer from Ingenieurbüro for his design and ideas of custom mechanical components needed to perform experiments. It is also hard to image the experiments to be possible without Physik-Werkstatt team lead by Andreas Stuken.

Finally, I would like to express my special thanks to Prof. Ursula Keller. PhD in her group was an excellent School of Life. The resources and freedom available at her group offered excellent conditions for exploration and growth.

Thank you all and I hope we keep in touch!

Zürich, 2019