Doctoral Thesis

Few-cycle mid-infrared OPCPA and applications in strong-field physics

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Few-cycle mid-infrared OPCPA and applications in strong-field physics

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

presented by
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2015
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<th>Description</th>
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</thead>
<tbody>
<tr>
<td>A</td>
<td>vector potential (Vs/m)</td>
</tr>
<tr>
<td>$\alpha_0$</td>
<td>excursion amplitude of a classical electron due to the electric field (a.u.)</td>
</tr>
<tr>
<td>B</td>
<td>magnetic field (T=Vs/m^2)</td>
</tr>
<tr>
<td>$\beta_0$</td>
<td>excursion amplitude of a classical electron due to the magnetic field (a.u.)</td>
</tr>
<tr>
<td>c</td>
<td>speed of light (m/s)</td>
</tr>
<tr>
<td>$\chi^{(1)}$</td>
<td>first-order susceptibility</td>
</tr>
<tr>
<td>$\chi^{(2)}$</td>
<td>second-order susceptibility (pm/V)</td>
</tr>
<tr>
<td>$\chi^{(3)}$</td>
<td>third-order susceptibility (pm^2/V^2)</td>
</tr>
<tr>
<td>D</td>
<td>electric displacement field (As/m^2)</td>
</tr>
<tr>
<td>$d_{\text{eff}}$</td>
<td>effective nonlinear coefficient (pm/V)</td>
</tr>
<tr>
<td>$\Delta k$</td>
<td>wave vector mismatch (mm^{-1})</td>
</tr>
<tr>
<td>$\Delta k_{\text{OPA}}$</td>
<td>gain bandwidth for unapodized linearly chirped grating (mm^{-1})</td>
</tr>
<tr>
<td>D(z)</td>
<td>local QPM duty-cycle</td>
</tr>
<tr>
<td>E</td>
<td>electric field (V/m)</td>
</tr>
<tr>
<td>e</td>
<td>electron charge ($1.602 \times 10^{-19}$ As)</td>
</tr>
<tr>
<td>$\varepsilon_0$</td>
<td>vacuum permittivity ($8.85 \times 10^{-12}$ As/Vm)</td>
</tr>
<tr>
<td>$\eta_{\text{max}}$</td>
<td>maximum plane-wave conversion efficiency</td>
</tr>
<tr>
<td>Symbol</td>
<td>Description</td>
</tr>
<tr>
<td>--------</td>
<td>-------------</td>
</tr>
<tr>
<td>$f$</td>
<td>focal length (mm)</td>
</tr>
<tr>
<td>$g$</td>
<td>coupling rate for $\Delta k \neq 0$ (mm$^{-1}$)</td>
</tr>
<tr>
<td>$G_{ss}$</td>
<td>small-signal gain</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>coupling rate (mm$^{-1}$)</td>
</tr>
<tr>
<td>$\gamma_0$</td>
<td>peak coupling rate (mm$^{-1}$)</td>
</tr>
<tr>
<td>$\gamma_K$</td>
<td>Keldysh parameter</td>
</tr>
<tr>
<td>$H$</td>
<td>magnetizing field (A/m)</td>
</tr>
<tr>
<td>$I_p$</td>
<td>atomic ionization potential (a.u. or eV)</td>
</tr>
<tr>
<td>$j$</td>
<td>free current density (A/m$^2$)</td>
</tr>
<tr>
<td>$K_g$</td>
<td>grating wave vector (mm$^{-1}$)</td>
</tr>
<tr>
<td>$k_i$</td>
<td>idler wave vector (mm$^{-1}$)</td>
</tr>
<tr>
<td>$k_p$</td>
<td>pump wave vector (mm$^{-1}$)</td>
</tr>
<tr>
<td>$k_s$</td>
<td>signal wave vector (mm$^{-1}$)</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>QPM chirp rate (mm$^{-2}$)</td>
</tr>
<tr>
<td>$L$</td>
<td>crystal length (mm)</td>
</tr>
<tr>
<td>$\lambda_i$</td>
<td>idler wavelength ($\mu$m)</td>
</tr>
<tr>
<td>$\lambda_p$</td>
<td>pump wavelength ($\mu$m)</td>
</tr>
<tr>
<td>$\lambda_s$</td>
<td>signal wavelength ($\mu$m)</td>
</tr>
<tr>
<td>$\Lambda(z)$</td>
<td>local QPM period ($\mu$m or mm)</td>
</tr>
<tr>
<td>$\Lambda$</td>
<td>signal gain coefficient</td>
</tr>
<tr>
<td>$L_{coh}$</td>
<td>coherence length ($\mu$m)</td>
</tr>
<tr>
<td>$m_e$</td>
<td>electron mass ($9.109 \times 10^{-31}$ kg)</td>
</tr>
<tr>
<td>$\mu_0$</td>
<td>vacuum permeability ($4\pi \times 10^{-7}$ Vs/Am)</td>
</tr>
<tr>
<td>$n_e$</td>
<td>extraordinary refractive index</td>
</tr>
<tr>
<td>$n_o$</td>
<td>ordinary refractive index</td>
</tr>
<tr>
<td>$n_i$</td>
<td>refractive index for the idler</td>
</tr>
<tr>
<td>$n_p$</td>
<td>refractive index for the pump</td>
</tr>
<tr>
<td>$n_s$</td>
<td>refractive index for the signal</td>
</tr>
<tr>
<td>$\omega_i$</td>
<td>idler angular frequency (s$^{-1}$)</td>
</tr>
<tr>
<td>$\omega_p$</td>
<td>pump angular frequency (s$^{-1}$)</td>
</tr>
<tr>
<td>$\omega_s$</td>
<td>signal angular frequency (s$^{-1}$)</td>
</tr>
<tr>
<td>$P$</td>
<td>induced polarization (As/m$^2$)</td>
</tr>
<tr>
<td>$P^{(1)}$</td>
<td>first-order polarization (As/m$^2$)</td>
</tr>
<tr>
<td>$P^{(2)}$</td>
<td>second-order polarization (As/m$^2$)</td>
</tr>
</tbody>
</table>
Symbols

- \( \mathbf{P}^{(3)} \) third-order polarization (As/m²)
- \( \mathbf{P}^{(NL)} \) nonlinear polarization (As/m²)
- \( P_{p,pk} \) pump peak power (MW)
- \( \rho \) free charge density (As/m³)
- \( \tau_{\text{eff}} \) effective seed pulse duration in Fourier plane (ps)
- \( \vartheta \) phase matching angle (° or rad)
- \( \Theta_p \) noncollinear pump angle (° or rad)
- \( U_p \) ponderomotive potential (a.u. or eV)
- \( w_p \) pump beam radius (µm)
- \( w_{wo} \) transverse spatial walk-off (µm)
# Acronyms

<table>
<thead>
<tr>
<th>Acronym</th>
<th>Definition</th>
</tr>
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<tbody>
<tr>
<td>a.u.</td>
<td>atomic units</td>
</tr>
<tr>
<td>ADK</td>
<td>Ammosov-Delone-Krainov</td>
</tr>
<tr>
<td>APPLN</td>
<td>aperiodically poled lithium niobate</td>
</tr>
<tr>
<td>APT</td>
<td>attosecond pulse train</td>
</tr>
<tr>
<td>AR</td>
<td>anti-reflection</td>
</tr>
<tr>
<td>BBO</td>
<td>beta barium borate</td>
</tr>
<tr>
<td>BPM</td>
<td>birefringent phase matching</td>
</tr>
<tr>
<td>CEP</td>
<td>carrier envelope phase</td>
</tr>
<tr>
<td>CPA</td>
<td>chirped-pulse amplification</td>
</tr>
<tr>
<td>CTMC</td>
<td>classical trajectory Monte Carlo</td>
</tr>
<tr>
<td>cw</td>
<td>continuous wave</td>
</tr>
<tr>
<td>DFG</td>
<td>difference-frequency generation</td>
</tr>
<tr>
<td>DM</td>
<td>dichroic mirror</td>
</tr>
<tr>
<td>DS</td>
<td>dispersion-shifted</td>
</tr>
<tr>
<td>EDFA</td>
<td>erbium-doped fiber amplifier</td>
</tr>
<tr>
<td>eV</td>
<td>electron Volt</td>
</tr>
<tr>
<td>far-IR</td>
<td>far-infrared</td>
</tr>
<tr>
<td>FOPA</td>
<td>frequency-domain optical parametric amplification</td>
</tr>
<tr>
<td>FROG</td>
<td>frequency resolved optical gating</td>
</tr>
<tr>
<td>FWHM</td>
<td>full-width at half maximum</td>
</tr>
<tr>
<td>GDD</td>
<td>group delay dispersion</td>
</tr>
<tr>
<td>GV</td>
<td>group-velocity</td>
</tr>
<tr>
<td>GVM</td>
<td>group-velocity mismatch</td>
</tr>
<tr>
<td>HH</td>
<td>higher-order harmonic</td>
</tr>
<tr>
<td>HHG</td>
<td>high-harmonic generation</td>
</tr>
<tr>
<td>HWP</td>
<td>half-wave plate</td>
</tr>
<tr>
<td>IAP</td>
<td>isolated attosecond pulse</td>
</tr>
<tr>
<td>keV</td>
<td>kilo-electron Volt</td>
</tr>
<tr>
<td>LBO</td>
<td>lithium triborate</td>
</tr>
<tr>
<td>MgOLN</td>
<td>magnesium-oxide doped lithium niobate (MgO:LiNbO₃)</td>
</tr>
<tr>
<td>mid-IR</td>
<td>mid-infrared</td>
</tr>
<tr>
<td>Acronyms</td>
<td>Definition</td>
</tr>
<tr>
<td>----------------</td>
<td>-------------------------------------------------------------</td>
</tr>
<tr>
<td>Nd:YAG</td>
<td>neodymium-doped yttrium aluminum garnet</td>
</tr>
<tr>
<td>NIR</td>
<td>near-infrared</td>
</tr>
<tr>
<td>OP-GaAs</td>
<td>orientation-patterned gallium-arsenide</td>
</tr>
<tr>
<td>OPA</td>
<td>optical parametric amplification</td>
</tr>
<tr>
<td>OPCPA</td>
<td>optical parametric chirped-pulse amplification</td>
</tr>
<tr>
<td>OPG</td>
<td>optical parametric generation</td>
</tr>
<tr>
<td>OR</td>
<td>optical rectification</td>
</tr>
<tr>
<td>PMD</td>
<td>photoelectron momentum distribution</td>
</tr>
<tr>
<td>PPLN</td>
<td>periodically poled lithium niobate</td>
</tr>
<tr>
<td>QPM</td>
<td>quasi phase matching</td>
</tr>
<tr>
<td>RDC</td>
<td>random duty-cycle</td>
</tr>
<tr>
<td>ROC</td>
<td>radius of curvature</td>
</tr>
<tr>
<td>SC</td>
<td>supercontinuum</td>
</tr>
<tr>
<td>SCG</td>
<td>supercontinuum generation</td>
</tr>
<tr>
<td>SFG</td>
<td>sum-frequency generation</td>
</tr>
<tr>
<td>SHG</td>
<td>second-harmonic generation</td>
</tr>
<tr>
<td>SNR</td>
<td>signal to noise ratio</td>
</tr>
<tr>
<td>SPM</td>
<td>self-phase modulation</td>
</tr>
<tr>
<td>SVAA</td>
<td>slowly-varying amplitude approximation</td>
</tr>
<tr>
<td>THG</td>
<td>third-harmonic generation</td>
</tr>
<tr>
<td>Ti:sapphire</td>
<td>titanium-doped sapphire</td>
</tr>
<tr>
<td>TL</td>
<td>transform-limit</td>
</tr>
<tr>
<td>TOD</td>
<td>third-order dispersion</td>
</tr>
<tr>
<td>UV</td>
<td>ultraviolet</td>
</tr>
<tr>
<td>VMIS</td>
<td>velocity-map imaging spectrometer</td>
</tr>
<tr>
<td>XPW</td>
<td>cross-polarized wave generation</td>
</tr>
<tr>
<td>XUV</td>
<td>extreme ultraviolet</td>
</tr>
<tr>
<td>Yb:YAG</td>
<td>ytterbium-doped yttrium aluminum garnet</td>
</tr>
</tbody>
</table>
Publications

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

Journal Papers


Conference Papers


Abstract

In the framework of this thesis the generation of few-cycle mid-infrared (mid-IR) pulses by means of optical parametric chirped-pulse amplification (OPCPA) is discussed in order to perform strong-field ionization experiments by the use of a long wavelength driving light source. Strong-field ionization processes require peak intensities in the order of $10^{13} \text{ W/cm}^2 - 10^{15} \text{ W/cm}^2$.

The peak intensity of an ultrashort laser pulse linearly and inversely scales with its pulse energy and pulses duration respectively. Accordingly, OPCPA schemes pushing the limits in terms of energy transfer to the mid-IR spectral range, e.g. pulse energy, and pulse duration are investigated. Thereby, the main focus is put on the investigation of the capabilities of conventional and chirped quasi phase matching (QPM) devices. These devices are also referred to as periodically poled lithium niobate (PPLN) and aperiodically poled lithium niobate (APPLN)-devices, respectively.

The output pulse duration of an OPCPA system is strongly related to the achievable phase matching and amplification bandwidth for a certain phase matching technique and nonlinear crystal. In contrast to the use of birefringent phase matching (BPM), QPM in lithium niobate ($\text{LiNbO}_3$) offers great flexibility in terms of phase matching bandwidth in the mid-IR spectral range and also reduces the experimental complexity by collinear
beam propagation. QPM devices with a constant grating wave vector $K_g$ throughout its spatial extent can phase match one single set of pump, signal and idler frequencies, which is determined by the QPM poling period. Such devices are referred to as PPLN crystals. By introducing a spatially varying grating wave vector $K_g(z)$, a wider range of frequencies can be phase matched at different positions inside of the QPM device. Such devices are referred to as APPLN devices. Relying on the bandwidth capabilities of APPLN devices, sub-four cycle pulse durations of 41.6 fs at a center wavelength of 3.4 µm were obtained from a three-stage, all-collinear OPCPA system. The compressed average output power was 600 mW, corresponding to a pulse energy of 12 µJ at 50 kHz repetition rate. The obtained pulse energy corresponds to an internal photon conversion efficiency of 24.5%. Furthermore, the optical parametric generation (OPG) background was negligible.

Although the 12 µJ-, sub-four-cycle pulses centered at 3.4 µm were sufficient for the demonstration of strong-field ionization in xenon, further amplification schemes were explored to enhance the pulse energy. Higher pulse energies in combination with sub-four-cycle pulse durations increase the electron yield obtained from strong-field ionization, e.g. increasing the electron count and thereby leading to higher statistics. Accordingly, an achromatic phase matching scheme based on a noncollinear interaction in a conventional PPLN was investigated. This type of amplifier was installed as the final power amplifier, while the preamplifiers remained the same as in the all-collinear OPCPA system. The adaption from an all-collinear to a hybrid system resulted in a compressed average power of 1.09 W, corresponding to 21.8 µJ at 50 kHz repetition rate. The final pulse duration after compression was 44.2 fs, corresponding to sub-four optical cycles at a center wavelength of 3.4 µm. The increase of pulse energy is more than 80% improvement in comparison to the all-collinear scheme, while keeping the OPG background comparably low.

For the demonstration of the capabilities of QPM devices, the great design flexibility was exploited to demonstrate a more sophisticated functionality, i.e. frequency-domain optical parametric amplification (FOPA).
In this arrangement, a two-dimensionally patterned QPM device was used for the purpose of optical parametric amplification (OPA) of a spatially chirped mid-IR idler seed in the Fourier plane of a 4f pulse shaper. A FOPA consists of a series of individual narrowband OPAs, which help to decouple the amplification of individual frequencies more robustly. At the output of the FOPA, an average power of 1.03 W was measured corresponding to 20.6 $\mu$J at a repetition rate of 50 kHz and a center wavelength of 3.4 $\mu$m. When accounting for the linear losses from the second diffraction grating and various beam shaping optics, a pulse energy of 33 $\mu$J in the Fourier plane was obtained, corresponding to a photon conversion efficiency of 32%. The pulse duration was measured to be 53 fs, with a 42 fs transform-limit (TL), corresponding to sub-four optical cycles.

The theoretical description of electron recollision processes, such as high-harmonic generation (HHG), strongly relies on the concept of the electric dipole approximation. By performing a strong-field ionization experiment with the use of the few-cycle pulses at a center wavelength of 3.4 $\mu$m emitted by the hybrid OPCPA system in combination with a velocity-map imaging spectrometer (VMIS), the breakdown of the dipole approximation in the long wavelength limit was observed. This breakdown manifested itself in an asymmetry in the photoelectron momentum distributions (PMDs) along the beam propagation direction. It was found that the asymmetry arises due to the onset of magnetic field effects. These observations pose new challenges for the theoretical description of strong-field processes in the long wavelength limit.
Kurzfassung


Die erzielbare Pulsdauer am Ausgang von OPCPA-Systemen hängt von der Seed-, Phasenanpassungs- und Verstärkungsbandbreite des verwendeten nichtlinearen Kristalls und der verwendeten Phasenanpassungstech-

von 50 kHz.


Theorien, die zur Beschreibung von Elektronenrückstreuprozessen in der Starkfeldphysik zum Einsatz kommen, beruhen auf der sogenannten elektrischen Dipolnäherung. Ein bekanntes Beispiel von Rückstreuprozessen ist die Erzeugung von Höheren Harmonischen (HHG). In einem Experiment zur Starkfeldionisation kamen die Laserpuls mit einer zentralen Wellenlänge von 3.4 µm in einem Elektronenimpulsspektrometer (VMIS) zum Einsatz. Dabei konnte eine Asymmetrie in den Photoelektronenimpulsspektren festgestellt werden. Es stellte sich heraus, dass die Asymmetrie durch Magnetfeldeffekte hervorgerufen worden ist. Diese Beobachtungen zeigen die Grenzen der Gültigkeit der elektrischen Dipolnäherung bei Starkfeldionisation mit langwelligen Lichtquellen auf, was eine neue Herausforderung bei der theoretischen Beschreibung von Starkfeldprozessen mit langwelligen Lichtquellen darstellt.
Chapter 1

Motivation

"Light is in short the most refined form of matter" - stated by Louis de Broglie as a way of formulating the light quantum hypothesis [1]. With this statement, in fact, he describes the nature of light either in terms of particles, particularly by means of photons or in terms of an electromagnetic wave. However, this hypothesis has already been postulated by Albert Einstein when he explained the photoelectric effect in 1905 [2]. The discovery of the photoelectric effect in combination with the preceding formulation of Planck’s radiation law in 1900 and the introduction of Planck’s constant opened up a new era in physics. In particular, it paved the way for the physical understanding of optical absorption, spontaneous- and stimulated emission. These observations and the understanding of the underlying physical effects represented an essential breakthrough in quantum mechanics and were the trigger for the discovery and the first demonstration of the laser by Theodore Maiman in 1960 [3]. LASER is an acronym and stands for Light Amplification by Stimulated Emission of Radiation.

Moreover, in the subsequent years since its first demonstration, laser technology has progressed rapidly. Nowadays, lasers have become extremely important in our daily life and are also inevitable in modern physics. Its applications range from optical communications to DVD players as part of our everyday’s life and from spectroscopy to microscopic imaging as part of science, to mention some prominent examples.

A unique feature of laser light is its deterministic propagation behav-
ior with respect to space and time which is characterized in terms of coherence. Coherence represents one of the key features why lasers have become as successful as they are nowadays in many spheres of our daily life.

Lasers can be operated in various modes. On the one hand the first lasers were operated in a “continuous wave (cw)”-mode, which means that the laser is continuously pumped and also continuously emits light. On the other hand, soon after the invention of the first laser, it was found out that lasers can also be operated in a pulsed mode. The first pulsed laser operation was achieved by so-called Q-switching which was reported in 1962 [4]. A further laser pulse generation technique is active modelocking which was theoretically proposed in 1964 [5], and was in fact also experimentally verified in the same year [6]. Complementary to active modelocking, the technique of passive modelocking, which employs a saturable absorber for starting and stabilizing modelocking, was introduced soon after [7, 8].

In the subsequent years, great efforts were put into the advancement of pulsed lasers, since they enable the generation of high peak powers and consequently higher focused intensities compared to conventional cw operation. Although dye lasers have demonstrated the first modelocked operation, these type of lasers went into strong competition with solid-state lasers, based on ion-doped laser gain materials such as neodymium-doped yttrium aluminum garnet (Nd:YAG) [9], ytterbium-doped yttrium aluminum garnet (Yb:YAG) [10] and titanium-doped sapphire (Ti:sapphire) lasers [11, 12]. Especially Ti:sapphire lasers turned out to be very promising in terms of reliability, operational simplicity and moreover in the broad emission cross section supporting few-cycle pulse durations [13].

In addition to the rapid evolution of laser oscillator technologies a strong focus was put simultaneously on laser amplifier technologies in order to enable the power amplification of oscillator-delivered few-cycle pulses to higher average powers. Accordingly, the first demonstration of chirped-pulse amplification (CPA) marked a breakthrough in ultrafast laser amplifier technology [14]. Here, the laser pulses are temporally stretched, then amplified and thereafter temporally compressed. With such a scheme one minimizes the problems caused by nonlinear phase
shifts (B-integral). At the output one can achieve high average powers in short pulse durations. The most common amplifier gain media employed in such CPA-schemes are Nd:YAG, Yb:YAG and also Ti:sapphire. Moreover, the continuous evolution of laser oscillator and amplifier technology was accompanied by the performance of experiments in nonlinear optics.

Surprisingly, the first experimental and theoretical demonstration of optical parametric amplification (OPA), i.e. a prominent nonlinear optical process, was done within five years of the first demonstration of the laser [15, 16, 17, 18, 19]. OPA has the advantage to enable energy transfer to a wavelength (frequency) range other than that present at the input. These wavelengths are then referred to as the idler wavelengths (frequencies). However, the generated idler wavelength range is constrained by energy conservation, phase matching and the transparency of the nonlinear gain media. That is the reason why this technique comes along with great wavelength (frequency) flexibility. Thus the next logical step was the combination of the advantages of OPA together with the ones of CPA, which was demonstrated for the first time in 1992 [20], resulting in optical parametric chirped-pulse amplification (OPCPA).

OPCPA consists of a modified CPA-system, where a nonlinear crystal (beta barium borate (BBO), lithium triborate (LBO), lithium niobate (LiNbO₃), etc.) acts as a parametric gain medium instead of a conventional laser gain medium (Yb:YAG, Ti:sapphire, etc.). The ultimate goal of OPCPA is to achieve broadband power amplification at a desired center wavelength of few-cycle pulses leading to focused intensities in the order of $10^{13} \text{ W/cm}^2 - 10^{15} \text{ W/cm}^2$, being used for studies of light-matter interaction. Moreover, the great wavelength flexibility makes OPCPA an ideal tool for the generation of few-cycle pulses at a desired center wavelength.

Due to the wide-spread deployment of Ti:sapphire laser systems, OPCPA was predominantly driven at laser wavelengths of 800 nm together with BBO as the nonlinear crystal. For these systems, predominantly collinear and noncollinear arrangements in birefringent crystals were the phase matching schemes of choice. However, for transferring energy from the near-infrared (NIR) to longer wavelengths in the (mid-infrared (mid-IR)) it turned out that LiNbO₃ accompanied by quasi phase matching (QPM) is a very promising combination. The investigation of the capabili-
ties of such QPM devices in connection with mid-IR-generation by means of OPCPA is a central part of this thesis. In contrast to broadband birefringent phase matching (BPM), QPM allows for broad phase matching bandwidths in a collinear fashion. QPM can be achieved by periodical or aperiodical poling, i.e. inverting the sign of the nonlinear coefficient $d_{\text{eff}}$ of the host crystal material (LiNbO$_3$ or LiTaO$_3$). These devices are then referred to as periodically poled lithium niobate (PPLN) or aperiodically poled lithium niobate (APPLN), respectively. Especially the latter has the potential for ultrabroadband phase matching bandwidths since the range of wave vectors $\Delta k$ can be set by the range of grating wave vectors $K_g$ supported by the grating design and hence enable shorter pulse durations than periodic QPM devices.

The development of a mid-IR light source was motivated by recent developments in attosecond and strong-field physics, showing a clear trend towards the use of long wavelength driving lasers. This trend was mainly triggered by the fact that high-harmonic generation (HHG) is a highly nonlinear process which has a strong nonlinear dependence on the driving laser wavelength. HHG was demonstrated for the first time in 1987 and 1988 [21, 22].

According to the first theoretical description in 1993 [23, 24, 25], HHG can be described by a semi-classical three-step model. The process steps are illustrated in Fig. 1.1 for different instances of time of the driving electric field. First, the electric field of a linearly polarized few-cycle pulse bends the atomic Coulomb potential of the target atom. Then the bound electron can escape into the continuum by tunneling through the potential barrier. This situation is illustrated in Fig. 1.1(a). Secondly, the highly modulated electric field of the few-cycle pulse exerts a force onto the liberated electron which drives the electron away from the parent ion and is thereafter accelerated back again (Fig. 1.1(b) and (c)). It is finally reabsorbed in a third step under the emission of an extreme ultraviolet (XUV) or a soft X-ray photon. This step is illustrated in Fig. 1.1(c). Furthermore the cut-off energy of the emitted photons can be expressed as:

$$E_{\text{cutoff}} = I_p + 3.17U_p \quad \text{with} \quad U_p = \frac{e^2E^2}{4m_e\omega^2}. \quad (1.1)$$

Here $I_p$ represents the first atomic ionization potential, $U_p$ the pondero-
Figure 1.1: Three-step model of HHG. (a): In the first quarter of the optical cycle of a few-cycle pulse, the Coulomb potential is bent, such that the bound electron can tunnel through the potential barrier being released into the continuum. (b): In the second quarter, the released electron is accelerated in the laser field thereby gaining kinetic energy. (c): When the electric field amplitude changes its sign, the free electron is accelerated back towards the residual ion. The electron can recombine with the residual ion under the emission of an energetic photon, corresponding to the HH-radiation.

motive potential, $E$ the electric field strength, $e$ the electron charge, $m_e$ the electron mass and $\omega$ the angular frequency of the driving electric field. According to Eq. (1.1), it is favorable to use long wavelength driving lasers to increase the energy cut-off of HH-radiation and possibly allow for energies exceeding the water window. Therefore in the framework of this thesis strong efforts were put into the investigation of mid-IR few-cycle pulse generation schemes by means of OPCPA.

Furthermore, we observed that the exploration of the wavelength scaling behavior of strong-field ionization processes itself revealed new insights and therefore could challenge theoretical models used to describe these processes. Accordingly, mid-IR few-cycle pulses delivered by our OPCPA-system in combination with a velocity-map imaging spectrometer (VMIS) were used for experiments to explore strong-field ionization in the momentum space. The outcome and insights gained by these experiments are discussed in the scope of this thesis.

This thesis is organized as follows: In chapter 2 the fundamentals of OPA/OPCPA are presented. It starts out with the introduction of the nonlinear polarization $\mathbf{P}^{(NL)}$, the derivation of the coupled wave equations
starting from Maxwell’s equations, illustration and discussion of parametric amplification and phase matching considerations. The chapter is closed by a discussion on the fundamentals of strong-field ionization. Chapter 3 outlines a mid-IR OPCPA system that exploits the capabilities of APPLN devices and delivers pulses with 42 fs pulse duration (sub-four-cycles) and 12 µJ of pulse energy at a repetition rate of 50 kHz. Chapter 4 covers the discussion of the amplification of mid-IR pulses by the help of an achromatic QPM power amplifier. Pulse energies of 21.8 µJ with pulse durations of 44 fs (sub-four-cycles) were achieved. In chapter 5 a new concept in nonlinear optics is introduced. The concept exploits the great potential of two-dimensionally-structured QPM devices as the amplification medium together with spatially-dispersed mid-IR idler seed pulses in a 4f pulse shaper arrangement. Pulse energies of more than 20 µJ with pulse durations of 53 fs (42 fs TL) were achieved. In chapter 6 a discussion is presented about the results from strong-field ionization experiments performed by the help of the OPCPA system, which is discussed in chapter 4, in combination with a VMIS. The analysis of the acquired data revealed new insights into strong-field ionization at long wavelengths. In fact, it describes the observation of the breakdown of the dipole approximation in strong-field ionization in the long-wavelength limit. In chapter 7 the thesis is concluded with a summary of the most important achievements. Furthermore a short outlook is presented regarding new energetic mid-IR pulse generation schemes potentially supporting single cycle pulse durations.
Since their theoretical description and experimental observation in the years 1962 to 1965 [16, 15, 17, 18, 19], OPA and related techniques, for example difference-frequency generation (DFG), have developed and progressed rapidly. Nowadays systems deploying these techniques generate light pulses covering a wide spectral range from the near-ultraviolet (UV) to the far-infrared (far-IR) [26, 27, 28, 29, 30, 31, 32, 33].

Besides its wavelength flexibility, OPA offers significant advantages over conventional laser amplifiers, such as CPA [14]. Those advantages range from broad phase matching bandwidths, the capability to support few-cycle pulse durations and high single-pass gain. Furthermore, OPA benefits from low thermal effects due to the fact that no population inversion is involved in the gain medium, high quantum efficiency, reduced amplified spontaneous emission and scalability to high pulse energies [34]. State-of-the-art OPA- and OPCPA-systems generate microjoules to millijoules of pulse energies and pulse durations down to a few femtoseconds. High pulse-energies in combination with few-cycle pulse durations result in high pulse peak powers on the order of MW and GW and lead to focused peak intensities ranging from $10^{13} \text{ W/cm}^2$ to $\geq 10^{15} \text{ W/cm}^2$. Such peak intensities are a necessary prerequisite for the ionization of noble gases and to perform strong-field experiments and the generation of HH [23, 35]. The purpose of most OPA- and OPCPA systems ranges from their application in conventional spectroscopy, fundamental investigations in strong-field ionization and time-resolved studies in attosecond science,
which heavily builds on the generation of attosecond pulse trains (APTs) or isolated attosecond pulses (IAPs). Particularly, attosecond science is an emerging branch of physics and physical chemistry, which makes use of the full potential of OPA- and OPCPA-systems [36, 37]. Therefore it is important to understand the nonlinear interaction during the OPA process to transfer energy from one wavelength to another, i.e. from the pump to signal and idler.

In this chapter we focus on the theoretical basics of nonlinear processes, e.g. OPA, which cover the description of the nonlinear susceptibility and induced polarization, the fundamental coupled wave equations and phase matching considerations. Furthermore we discuss the basic theory of the dipole approximation in strong-field ionization which is an essential part in chapter 6 when applying OPCPA to strong-field experiments and discussing our observations.

2.1 Theory of OPA

The OPA process is a three-wave mixing process and it is responsible for the transfer of energy from an energetic pump wave to a signal and idler wave. Energy transfer is obtained in the presence of an optical medium, which enables efficient coupling between the incident waves under the condition of energy- and wave vector conservation, the so-called phase matching. In fact the involved waves are coupled to each other via the induced nonlinear polarizations. Particularly, induced polarizations play an important role in any kind of nonlinear optical interaction. These relationships can be described by coupled wave equations, one for each wave, which can be derived from Maxwell’s equations. These equations build the foundations of OPA.

One of the great advantages of OPA is that the amplification process can be tailored by the use of suitable nonlinear crystals allowing for broad-band gain and broad phase matching bandwidths and consequently allow for the generation of few-cycle pulse durations. The following discussion is based on the mathematical treatment in Ref. [38].
2.1. Theory of OPA

2.1.1 Nonlinear susceptibility and polarization

When an electromagnetic wave is traveling through a material, the electric field $E(t)$ causes the occurrence of induced electric dipoles. The presence of electric dipoles in a material can be described by a macroscopic physical quantity called induced polarization $P(t)$ and can be written in terms of a function of the electric field strength $E(t)$. The following Taylor-series expansion describes the induced polarization by

$$P(t) = \varepsilon_0 \left\{ \chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \ldots \right\}$$

$$= \mathbf{P}^{(1)}(t) + \mathbf{P}^{(2)}(t) + \mathbf{P}^{(3)}(t) + \ldots$$

$$= \mathbf{P}^{(1)}(t) + \mathbf{P}^{(NL)}(t), \quad (2.1)$$

where $\chi^{(1)}$ represents the linear susceptibility, $\chi^{(2)}$ and $\chi^{(3)}$ represent the second- and third-order nonlinear susceptibilities. Three-wave mixing processes, such as second-harmonic generation (SHG), sum-frequency generation (SFG), DFG, OPA and optical rectification (OR) have in common to be induced by the second-order susceptibility $\chi^{(2)}$, whereas third-harmonic generation (THG), self-phase modulation (SPM) and cross-polarized wave generation (XPW) are four-wave mixing processes. Therefore the latter three processes are induced by the third-order susceptibility $\chi^{(3)}$.

For the case of OPA, which is the process of interest in the framework of this thesis, two waves, i.e. pump and signal at frequencies $\omega_1$ and $\omega_2$, are present at the input of the nonlinear medium. We can write the optical field as a superposition of the two waves as

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + \text{c.c.}, \quad (2.2)$$

with c.c. being the conjugate complex part of the superposition. When an intense optical field interacts with a nonlinear optical medium represented by the second-order susceptibility $\chi^{(2)}$, the induced second-order
polarization $P^{(2)}(t)$ can be written as

$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} E^2(t) = \varepsilon_0 \chi^{(2)} \left\{ E_1^2 e^{-i2\omega_1 t} + E_2^2 e^{-i2\omega_2 t} \right\}_{\text{SHG}}$$

$$+ 2E_1 E_2 e^{-i(\omega_1 + \omega_2) t} + 2E_1^* E_2 e^{-i(\omega_1 - \omega_2) t} \right\}_{\text{SFG}}$$

$$+ E_1 E_1^* + E_2 E_2^* + \text{c.c.} \right\}_{\text{OPA/DFG}} + \text{OR}. \right\}_{\text{OR}}$$

Equation (2.3)

According to Eq. (2.3), the interaction of the superimposed optical waves at frequencies $\omega_1$ and $\omega_2$ leads to the generation of additional frequency components. These additionally generated frequencies range from SHG at 2$\omega_1$ and 2$\omega_2$ of the fundamental frequencies $\omega_1$ and $\omega_2$, SFG at $\omega_1 + \omega_2$, DFG/OPA at $\omega_1 - \omega_2$ and OR representing a DC electric-field. In the case of OPA, the frequencies $\omega_1$ and $\omega_2$ can be renamed to $\omega_p$ and $\omega_s$ representing the frequency of the pump and the signal wave with the strict requirement that $\omega_p > \omega_s$.

Furthermore nonlinear mixing processes can also be described and understood in terms of virtual energy levels as shown in Fig. 2.1. In the virtual energy level picture the generation of new frequencies can be described by means of photon energies. According to the energy conservation condition $\hbar \omega_p = \hbar \omega_s + \hbar \omega_i$, for every photon created of frequency $\omega_i = \omega_p - \omega_s$, a photon of pump frequency $\omega_p$ is absorbed while an additional photon of signal frequency $\omega_s$ is created. The energy level diagram shown in Fig. 2.1 illustrates the generation of a photon at the difference

![Figure 2.1: (a): Nonlinear interaction of pump $\omega_p$ and signal wave $\omega_s$ represented by the $\chi^{(2)}$-susceptibility. The signal wave is amplified by the presence of the pump. However, the amplification process can only take place under energy conservation and consequently another wave at the difference frequency $\omega_i = \omega_p - \omega_s$ is generated. (b): Virtual energy-level diagram for the OPA/DFG-process. Adapted from [38].](image-url)
frequency $\omega_p - \omega_s$, when a pump photon of frequency $\omega_p$ and a signal photon of frequency $\omega_s$ are interacting within a $\chi^{(2)}$-medium, as it is the case for OPA and DFG. The optical field oscillating at $\omega_s$ is parametrically amplified under the presence of a pump optical field at $\omega_p$ during the DFG/OPA-process.

2.1.2 Coupled wave equations and parametric amplification

Any interaction of electromagnetic waves and matter can be macroscopically described by the Maxwell’s equations. In order to understand the dynamics of mixing processes and to derive the fundamental relationships between the pump, signal and idler wave for the process of OPA, we start out with the set of Maxwell’s equations and derive the set of coupled wave equations for the pump, signal and idler wave. For this procedure we assume that there are no free charges ($\rho = 0$) and no free currents ($j = 0$). The Maxwell’s equations can then be written as the following set of equations

\[
\begin{align*}
\nabla \cdot E &= 0 \quad (2.4) \\
\nabla \cdot B &= 0 \quad (2.5) \\
\nabla \times E &= -\frac{\partial B}{\partial t} \quad (2.6) \\
\nabla \times H &= \frac{\partial D}{\partial t} \quad (2.7)
\end{align*}
\]

with electric field $E$, electric displacement field $D$, magnetic field $B$ and magnetizing field $H$. Under the assumption of no free currents and a non-magnetic material the relation between $B$ and $H$ is given by $B = \mu_0 H$ and by $D = \varepsilon_0 E + P$ for the electric field $E$ and displacement field $D$. Here the quantity $P$ represents the induced polarization.

Under the assumption that nonlinear processes are allowed we can make use of the material relationship $D = \varepsilon_0 E + P$ and insert it into Eq. (2.7). Moreover, the curl-operator ($\nabla \times a$) can be applied to Eq. (2.6), and we furthermore assume that the order of the applied operators, i.e. temporal derivative $\partial / \partial t$ and the curl-operator ($\nabla \times a$), is interchangeable. Here, $a$ represents an arbitrary vector field, with $a \in \mathbb{R}^3$. Given that $B = \mu_0 H$, then Eq. (2.7) can be inserted into Eq. (2.6) and the set of
Maxwell’s equations has been decoupled and the following equation represents the wave equation in its most general form
\[ \nabla \times \nabla \times E + \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = -\frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P}{\partial t^2}, \tag{2.8} \]

with \( c = 1/\sqrt{\varepsilon_0 \mu_0} \), the speed of light. This rather complicated wave equation can be further simplified under certain assumptions and by making use of some mathematical identities from vector calculus. By applying the vector identity \( \nabla \times (\nabla \times E) = \nabla (\nabla \cdot E) - \nabla^2 E \) and the assumption of transverse, infinite plane waves we can make use of the vanishing divergence of the electric field, i.e. \( \nabla \cdot E = 0 \) [38]. In general, this assumption is not valid in nonlinear optics, even for isotropic media. However, for the most cases of interest it can be neglected. For the remainder of this chapter we assume that the contribution of \( \nabla \cdot E \) is negligible. Finally the vector identity simplifies to \( \nabla \times \nabla \times E = -\nabla^2 E \). Taking into account all these assumptions the wave equation (2.8) can be written in terms of the electric field \( E \) and polarization \( P \) as
\[ \Delta E = \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} + \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P}{\partial t^2}, \tag{2.9} \]

with \( \nabla^2 E = \Delta E \). When applying the following ansatz for the polarization \( P(r, t) = P^{(1)}(r, t) + P^{(NL)}(r, t) \) we can write Eq. (2.9) as
\[ \Delta E = \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} + \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 }{\partial t^2} \left\{ P^{(1)}(r, t) + P^{(NL)}(r, t) \right\}. \tag{2.10} \]

This equation can be further simplified by taking into account that \( P^{(1)}(r, t) = \varepsilon_0 \chi^{(1)} E(r, t) \). Then we can write the previous equation as
\[ \Delta E - \frac{1}{c^2} \left( 1 + \chi^{(1)} \right) \frac{\partial^2 E}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P^{(NL)}}{\partial t^2}, \]
\[ \Delta E - \varepsilon^{(1)} \frac{\partial^2 E}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P^{(NL)}}{\partial t^2}, \tag{2.11} \]

where \( \varepsilon^{(1)} = 1 + \chi^{(1)} \). This equation represents an inhomogeneous wave equation. The solutions of this equation are waves propagating at velocity \( c/n \) with \( n = \sqrt{\varepsilon^{(1)}} \). However, for the case of dispersive media we have to consider each spectral component of the electric field separately and
therefore consider the electric field $E(r, t)$ as the sum over its individual frequency components, i.e. $E(r, t) = \sum_n E_n(r, t)$, where the summation is only performed over positive frequency components since the c.c.-part of the individual electric fields $E_n(r, t)$ account for negative frequency components. For the following ansatz for the electric field and the polarization with its individual frequency components

$$E_n(r, t) = E_n(r)e^{-i\omega_n t} + \text{c.c.}$$  \hspace{1cm} (2.12)

$$P^{(NL)}(r, t) = \sum_n P^{(NL)}_n(r, t) = \sum_n \left\{ P^{(NL)}_n(r)e^{-i\omega_n t} + \text{c.c.} \right\},$$ \hspace{1cm} (2.13)

the inhomogeneous wave equation can be written for each individual spectral component as

$$\Delta E_n(r, t) + \frac{\omega_n^2}{c^2}\varepsilon^{(1)}(\omega_n)E_n(r, t) = -\frac{\omega_n^2}{\varepsilon_0 c^2}P^{(NL)}_n(r, t),$$ \hspace{1cm} (2.14)

where the temporal derivative $\partial^2/\partial t^2$ of the individual frequency components of the electric field $E_n(r, t)$ has been substituted by $-\omega_n^2$. When we assume plane waves and propagation only into the direction of the spatial coordinate $z$, then the following plane wave ansatz represents a solution of the wave equation (2.14)

$$E_n(z, t) = E_n(z)e^{-i\omega_n t} + \text{c.c.} = \frac{1}{2} \left\{ A_n e^{ik_n z}e^{-i\omega_n t} + \text{c.c.} \right\},$$ \hspace{1cm} (2.15)

where $A_n$ represents the field amplitude. For the sake of simplicity, we only consider the OPA/DFG-part of the second-order nonlinear polarization $P^{(NL)}_n(z, t) = P^{(2)}_{\text{OPA/DFG}}(z, t) = 2\varepsilon_0\chi^{(2)}E_1E_2^*e^{-i(\omega_1-\omega_2)t} + \text{c.c.}$. In the terminology of OPA the difference-frequency $\omega_1 - \omega_2$ can be interpreted as the idler frequency $\omega_i = \omega_p - \omega_s$ with $\omega_1 = \omega_p$ and $\omega_2 = \omega_s$. When we apply the ansatz of Eq. (2.15) to the nonlinear polarization oscillating at the idler frequency, we get

$$P^{(2)}_{\text{OPA/DFG}}(z, t) = P_i(z, t) = P_i(z)e^{-i\omega_i t} + \text{c.c.}$$

$$= 2\varepsilon_0\chi^{(2)}A_p A_s^* e^{i(k_p-k_s)z}e^{-i\omega_i t} + \text{c.c.}.$$ \hspace{1cm} (2.16)

Due to the effect of chromatic dispersion, the wave vectors of the involved waves behave differently than the associated frequencies, such that in general $k_i \neq k_p - k_s$. When we insert Eq. (2.15) and Eq. (2.16) into the wave
2. Theoretical foundations

Theoretical foundations (2.14), then the coupled wave equation for the idler wave can be written as

$$\frac{d^2 A_i}{dz^2} + 2ik_i \frac{dA_i}{dz} + \text{c.c.} = -\frac{d_{\text{eff}} \omega_i^2}{c^2} A_p A_s^* e^{i(k_p - k_s - k_i)z} + \text{c.c.}, \quad (2.17)$$

where the relation $2d_{\text{eff}} = \chi^{(2)}$ has been used to describe the connection between the second-order susceptibility and the nonlinear optical coefficient [38]. Furthermore, we can drop the c.c. - part of the coupled wave equation and still keep the validity of the equation.

The slowly-varying amplitude approximation (SVAA), which assumes that the fractional change of the amplitude in a distance of the order of an optical wavelength must be smaller than unity, can be applied to the coupled wave equation when the relation

$$\left| \frac{d^2 A_n}{dz^2} \right| \ll \left| k_n \frac{dA_n}{dz} \right|, \quad (2.18)$$

is fulfilled for $n \in \{i, p, s\}$. Moreover, the wave vector mismatch can be defined by $\Delta k = k_i + k_s - k_p$, where the individual wave vectors are given by $k_n = (n \omega_n / c)$ for $n \in \{i, p, s\}$. Under the assumption of the SVAA and the full permutation symmetry, we can write the set of coupled wave equations for the process of OPA as

$$\frac{dA_s}{dz} = i \frac{d_{\text{eff}} \omega_s}{n_s c} A_i^* A_p e^{-i\Delta k z}, \quad (2.19)$$

$$\frac{dA_i}{dz} = i \frac{d_{\text{eff}} \omega_i}{n_i c} A_p A_s^* e^{-i\Delta k z}, \quad (2.20)$$

$$\frac{dA_p}{dz} = i \frac{d_{\text{eff}} \omega_p}{n_p c} A_s A_i e^{i\Delta k z}. \quad (2.21)$$

Without further assumptions no analytical solutions exist for the set of coupled wave equations. In most cases the set of equations is solved numerically. In the remainder of this treatment we assume no pump depletion. Consequently the amplitude of the pump stays constant, i.e. $dA_p/dz = 0$. We solve the coupled wave equations for the case of a vanishing wave vector mismatch $\Delta k = 0$, or perfect phase matching. First we take the derivative of Eq. (2.20) with respect to the spatial coordinate $z$ and then insert the complex conjugate of Eq. (2.19) to eliminate $dA_s^*/dz$ from the
2.1. Theory of OPA

right-hand side, resulting in

\[
\frac{d^2 A_i}{dz^2} = \frac{d^2 \omega_s \omega_i}{n_s n_i c^2} A_p A_p^* A_i = \gamma^2 A_i,
\]

(2.22)

where the prefactor \( \gamma^2 \) is interpreted as the coupling rate given by

\[
\gamma^2 = \frac{d^2 \omega_s \omega_i}{n_s n_i c^2} |A_p|^2.
\]

(2.23)

When we assume for the boundary conditions that no idler wave is available at the input and the signal wave amplitude has an arbitrary value, i.e. \( A_i(0) = 0 \) and \( A_s(0) = A_{s,0} \), we find the solution for the set of the coupled wave equations to be

\[
A_s(z) = A_{s,0} \cosh(\gamma z)
\]

(2.24)

\[
A_i(z) = i \sqrt{\frac{n_s \omega_i}{n_i \omega_s}} \frac{A_p}{|A_p|} A_{s,0}^* (0) \sinh(\gamma z).
\]

(2.25)

The signal and idler wave experience monotonic growth as shown in Fig. 2.2, which is also suggested from the structure of the solutions presented in Eqs. (2.24) and (2.25). On the one hand the signal wave encounters an additional phase shift originating from the propagation in the medium. On the other hand the generated idler wave shows a dependence on the phase of the input signal and the pump, respectively.

The description of parametric interaction and amplification in the preceding section is based on the assumption of a vanishing wave vector

![Figure 2.2: Illustration of the exponential growth experienced by the signal and idler wave during the process of parametric amplification under the assumption of an undepleted pump and perfect phase matching \( \Delta \kappa = 0 \). For illustration purposes, arbitrary boundary conditions are used. Adapted from [38].](image)
mismatch $\Delta k = 0$, or under the assumption of perfect phase matching. However, in reality the condition $\Delta k = 0$ is not inherently fulfilled for all the involved frequencies. The following sections deal with important aspects of phase matching.

2.1.3 Phase matching considerations

The basic idea of phase matching is to prevent all the involved frequencies from temporal walk-off or in other words from dephasing during the propagation in an optical medium. This can be achieved in different ways. The most common technique for achieving phase matching in nonlinear optics is birefringent phase matching. This technique uses the birefringence of a nonlinear crystal to eliminate the phase mismatch, e.g. $\Delta k = 0$. However, the actual techniques used to achieve birefringent phase matching can slightly differ in their implementation. On the one hand, critical birefringent phase matching uses the angular adjustment of the crystal to tune the extraordinary refractive index experienced by an extraordinarily polarized wave to find the optimum point of operation leading to a vanishing wave-vector mismatch [39, 40]. On the other hand for non-critical phase matching, the polarization directions of all the involved waves are oriented along the principal crystal axes. This has the effect, that for this particular configuration the beams propagate along some principal axis of the birefringent crystal and therefore the interaction is less sensitive to the angular crystal adjustment. Due to the strong temperature dependence of the optical birefringence of some crystals, the phase mismatch can be minimized by adjusting the crystal temperature so that the phase velocities of the interacting waves are matched to each other [41, 42].

A special case for non-critical phase matching is quasi phase matching (QPM) [17, 43]. QPM can be achieved also in non-birefringent media and therefore in most cases the involved waves have equal polarization states.

Regardless of the phase matching technique, the wave vectors of the involved waves either can point into the same direction or different directions. These distinct configurations are referred to as collinear and noncollinear phase matching, respectively [44].

Under normal conditions, almost every optical medium exhibits a weak nonlinear behavior. However, second-order nonlinearities can only occur
in non-centrosymmetric crystals [45]. The applicable second-order nonlinearity can vary from one non-centrosymmetric crystal and phase matching technique to the other. When such a nonlinear optical medium is exposed to an incident signal (photons of energy $\hbar \omega_s$) and additionally to a high intensity pump wave (photons of energy $\hbar \omega_p$), then the nonlinear optical interaction can give rise to the emission of an additional wave, the so-called idler wave (photons of energy $\hbar \omega_i$). Here, the idler is referred to as the unseeded wave with the smallest oscillating frequency. The emission of an additional photon can either happen in some cases by spontaneous parametric emission and in some other cases by stimulated parametric emission. In fact, such processes can only occur under the constraint of energy conservation. The energy conservation can be formulated by means of photon energies by the following relationship

$$\hbar \omega_p = \hbar \omega_s + \hbar \omega_i.$$ (2.26)

However beyond this, efficient energy transfer from the pump to the signal and idler wave can only be achieved when an additional condition is met. That is the so called phase matching or wave vector conservation condition. It can be formulated in terms of wave vectors, frequencies, wavelengths and refractive indices by

$$\Delta k = k_i + k_s - k_p = 0 \quad \text{with} \quad k_j = n_j(\omega_j) \frac{\omega_j}{c} = n_j \frac{2\pi}{\lambda_j},$$ (2.27)

and $j \in \{i, p, s\}$. In contrast to energy conservation, wave vector conservation is not intrinsically fulfilled. Accordingly, the effect of phase matching has to come into play. In literature, the expressions of wave vector conservation and phase matching are used synonymously. For a non-zero wave vector mismatch the interacting waves are propagating out of phase and consequently one has to take care to compensate for the difference in phase.

Due to material dispersion, a minimum of $\Delta k$ can only be achieved for the propagation of distinctly polarized waves in birefringent optical media in the case of birefringent critical and non-critical phase matching. Specifically, critical phase matching, which was first demonstrated by Giordmaine [39] and Maker et al. [40] in 1962, can be achieved either in a collinear- or noncollinear fashion.
As mentioned earlier, a special case of a non-critical phase matching scheme is QPM. In contrast to BPM, the involved frequencies are not constantly locked in phase to each other, however, the sign of the nonlinear coefficient is periodically or aperiodically flipped so that the energy keeps flowing from the pump to signal and idler respectively [17, 46, 47]. This fact is illustrated in the section 2.1.3 about QPM.

Angular phase matching with collinear wave propagation

Most common anisotropic media used for parametric amplification are negative uniaxial birefringent crystals, such as BBO for example. The phase matching scheme applied to this type of crystal can either be of type-I or of type-II. Depending on the type of phase matching, there are some constraints regarding the polarizations of the interacting waves. For type I phase matching in a negative uniaxial crystal, the signal and idler waves (the lower frequency waves) must have both ordinary (o-) polarization whereas the pump wave must have extraordinary (e-) polarization. In contrast, for type II phase matching, the lower frequency waves must exhibit both orthogonal polarizations, i.e. signal wave exhibits ordinary, idler and pump wave exhibits extraordinary polarization [48]. Accordingly, the refractive indices for these waves are then given by $n_o$ for ordinary- and $n_e$ for extraordinary waves respectively. The refractive indices $n_o$ for ordinarily polarized waves are independent of the angular crystal orientation in a uniaxial crystal. In contrast, the refractive index $n_e$ experienced by an extraordinarily polarized wave, shows a dependence on crystal orientation. The value of it can be obtained by

$$n_e(\vartheta) = \frac{n_e n_o}{\sqrt{n_e^2 \cos^2 \vartheta + n_o^2 \sin^2 \vartheta}}, \quad (2.28)$$

where $n_e$ and $n_o$ represent the refractive indices along the principle axes of the birefringent uniaxial crystal [49]. By inserting the values for the refractive indices of the distinctly polarized waves into Eq. (2.27), one can calculate the wave vector mismatch for the chosen type of crystal and an angle $\vartheta$ with respect to the optical axis. In particular, the phase matching properties can then be tuned by rotating the crystal with respect to the incident beams. This simple tuning flexibility is one of the advantages
of this particular phase matching scheme. Due to the employment of distinctly polarized waves, the usable nonlinear optical coefficient $d_{\text{eff}}$ does not exhibit a maximum and therefore the achievable gain in such a configuration can be lower compared to the case of QPM. Furthermore, the group velocities are generally not matched to each other. Consequently, perfect phase matching can only be achieved for one certain set of frequencies $\{\omega_i, \omega_p, \omega_s\}$.

In the following section, the relation between phase matching bandwidth, group velocities and gain bandwidth is presented. This treatment is based on the analysis in [44]. When considering Eq. (2.27), the wave vector mismatch can be written as

$$\Delta k = k_p(\omega_p) - k_s(\omega_s) - k_i(\omega_i) = 0,$$

(2.29)

where $k_j(\omega_j)$ with $j \in \{i, p, s\}$ represent the wave vectors evaluated at the center idler, signal and pump frequencies. For illustration purposes, the pump is considered as monochromatic wave with frequency at $\omega_p$. Signal and idler exhibit a certain bandwidth according to $\omega_s + \Delta \omega$ and $\omega_i - \Delta \omega$ respectively. Then the wave vector mismatch can be written as

$$\Delta k = k_p - \{k_s(\omega_s) + \Delta k_s\} - \{k_i(\omega_i) + \Delta k_i\} = -\Delta k_s - \Delta k_i.$$

(2.30)

Furthermore we can approximate this expression by a first-order Taylor expansion

$$\Delta k \approx -\frac{\partial k_s}{\partial \omega_s} \Delta \omega + \frac{\partial k_i}{\partial \omega_i} \Delta \omega = \left\{ \frac{1}{v_{gi}} - \frac{1}{v_{gs}} \right\} \Delta \omega = \delta_{si} \Delta \omega,$$

(2.31)

where $v_{gs}$ and $v_{gi}$ are the group velocities of the signal and the idler wave. Now we can define the group-velocity mismatch (GVM) between signal and idler wave as $\delta_{si}$. Consequently, the wave vector mismatch $\Delta k$ is proportional to the GVM.

Another important parameter in connection with OPA is the full-width at half maximum (FWHM) gain bandwidth. Under the assumption of high gain and negligible pump depletion, the gain bandwidth can be written in terms of the crystal length $L$ and the GVM $\delta_{si}$ as

$$\Delta \nu \propto \frac{1}{\sqrt{L |\delta_{si}|}},$$

(2.32)
From this relation, we can conclude that the crystal length $L$ is also an important parameter to control the gain bandwidth. According to the relations given by Eqs. (2.31) and (2.32), we find that the achievable phase matching- and gain bandwidth for non-degenerate collinear OPA/OPCPA is ultimately limited by the GVM and the crystal length $L$. However, there is a trade-off between the achievable gain and the gain bandwidth: on the one hand, a shorter crystal leads to a broader gain bandwidth but on the other hand also leads to a lower gain compared to a longer crystal for the same pump parameters (peak power, spot size). Due to the effect of GVM, the phase matching condition can only be fulfilled for a narrowband spectral region.

In a collinear OPA/OPCPA the propagation direction inside of a crystal is chosen to meet the phase matching condition for a certain signal wavelength, i.e. $\Delta k = 0$. Then the group velocities of signal and idler are fixed and in general not matched. Group-velocity (GV)-matching can only be obtained when the OPA/OPCPA is operated at degeneracy, i.e. $\omega_s = \omega_i = \omega_p / 2$, provided that the signal and idler have the same polarization (type I phase matching in a birefringent crystal or QPM in a periodically or aperiodically poled crystal). Consequently such an operational configuration results in broadband phase matching and gain bandwidths. However, when the signal wavelength is tuned away from degeneracy then the condition $\delta_{si} = 0$ is lost in a collinear configuration leading to narrowband phase matching bandwidths [44, 50]. Accordingly, for the generation of few cycle pulse durations in non-degenerate OPA/OPCPA, the phase matching scheme has to be slightly adapted. There is the need for a more elaborate phase matching technique. The idea is to exploit the
2.1. Theory of OPA

capabilities of noncollinearly propagating waves in a nonlinear crystal. In the following section, the influence of noncollinearly propagating waves is discussed.

Angular phase matching with noncollinear wave propagation

In the previous section we have introduced a configuration which allows for broadband OPA/OPCPA in a collinear interaction geometry. However, broad bandwidths can only be achieved for a certain set of frequencies, e.g. when the OPA is operated at degeneracy. Here, the possibilities for broadband amplification for non-degenerate cases are discussed. The idea is to employ a noncollinear configuration by introducing another degree of freedom in addition to the phase matching angle $\vartheta$. The situation for such a geometry is shown in Fig. 2.3. Signal and pump propagate into different directions, such that the wave vectors form an angle $\Theta_p$, and overlap in space and time at the position of the birefringent crystal. The noncollinear angle $\Theta_p$ shows no dependence on frequency, however the signal and idler exhibit a frequency dependent angle $\Omega$ with respect to each other. The spectral dependence of $\Omega$ is necessary in order to fulfill the phase matching condition in a noncollinear configuration. A disadvantage of such a configuration is the inherent angular chirp and pulse front tilt among the generated spectral components of the idler which in turn is the result of the noncollinear nature of this technique. Different from collinear

![Diagram](image)

Figure 2.4: (a): Illustration of signal and idler wave propagation in a collinear geometry. The offset between the envelopes of signal and idler indicates a GVM. (b): The noncollinear propagation of signal and idler wave prevents temporal walk-off and hence there is no significant GVM. Adapted from [44].
phase matching, the signal and idler GVs are matched to each other. The relation of the GVs between signal and idler can be expressed by

\[ v_{gs} = v_{gi} \cos \Omega. \]  

\hspace{1cm} (2.33)

According to Eq. (2.33) broadband phase matching is obtained when the signal GV is equal to the projection of the idler GV on the propagation axis. This situation is illustrated in Fig. 2.4. For collinear geometry signal and idler quickly separate because of different GVs, whereas a noncollinear arrangement prevents signal and idler from temporal walk-off resulting into broader phase matching bandwidths [44, 50].

**Quasi phase matching (QPM)**

Although birefringent phase matching BPM is still the most common technique used for OPA/OPCPA, QPM has become more important, particularly for mid-IR generation [31, 32, 51, 52, 53]. Due to its capabilities, it relaxes some constraints inherent among birefringent phase matching techniques. The idea behind quasi phase matching is to spatially modulate the nonlinearity, i.e. flip the sign of the relevant effective nonlinear coefficient \( d_{\text{eff}} \). A nonlinear interaction in a QPM device operates in a nominally phase mismatched regime where a phase mismatch is acquired via the propagation over some distance, the so-called coherence length.

![Figure 2.5: (a): Illustration of the evolution of the SHG field amplitude of quasi phase matched and phase mismatched interaction in a QPM device. Moreover for comparison purposes a phase matched interaction for a non-QPM medium is also shown. (b): Spatial sign modulation of the nonlinear coefficient for the QPM.](image-url)
$L_{\text{coh}}$. However, at the positions where the direction of photon conversion is going into the wrong direction (energy flow from the signal back to the pump), the sign of the nonlinear coefficient is flipped. Consequently the energy continues flowing from the pump to the signal wave. This situation is illustrated in Fig. 2.5 for the case of SHG, where a quasi phase matched interaction is compared to a phase mismatched interaction in a QPM device and to a phase matched interaction in a non-QPM device. QPM in LiNbO$_3$ can be achieved by electrical poling, which is the technique for flipping the sign of the nonlinear coefficient and accordingly preventing energy from flowing back of the seeded- and generated wave to the pump wave. These QPM crystals are then called PPLN-devices.

It is not necessary to have different polarization states for the interacting waves and therefore in the most common cases the waves have equal polarization states. This enables to use a stronger element of the nonlinear tensor in comparison to conventional phase matching techniques [43]. As the conversion efficiency is proportional to $d_{\text{eff}}^2$, comparable or even higher conversion efficiencies can be achieved compared to birefringent phase matching techniques [54]. However, there are some limitations mainly in terms of fabrication. Until fairly recently, it was not possible to obtain PPLN crystals of good poling quality beyond 2 mm of thickness. It has been shown that electrical poling can also be applied to even thicker crystals, allowing for high quality large-aperture PPLN-devices [55]. The improvement in terms of fabrication, marks an important progress regarding the applicability of such crystals for high power OPCPA systems. One of the greatest advantages of QPM devices is its flexibility for various designs specified by the user for the intended application [43, 56].

According to the mathematical treatment in [56, 57], the spatially varying nonlinear coefficient for an arbitrary grating, can be described by

$$
\frac{\bar{d}(z)}{d_{33}} = \text{sgn} \left[ \cos \left( \varphi_g(z) \right) - \cos \left( \pi D(z) \right) \right] 
$$

(2.34)

$$
= \bar{d}_0(z) + \sum_{m=-\infty, m \neq 0}^{+\infty} \bar{d}_m(z)e^{im\varphi_g(z)},
$$

(2.35)
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with a grating phase given by

\[ \varphi_g(z) = \int_0^z K_g(z')dz'. \]  

Here \( m \) defines the QPM-order, \( K_g(z) \) is the local grating wave vector, \( D(z) \) the local grating duty-cycle and \( d_{33} \) the nonlinear coefficient. The Fourier coefficients are given by \( \bar{d}_m(z) = 2 \sin(\pi m D(z))/(\pi m) \) for \( m \neq 0 \) and \( \bar{d}_0(z) = (2D(z) - 1) \). For a standard QPM-grating with a constant duty-cycle of 50% along the spatial coordinate \( D(z) = 0.5 \). When only first-order QPM effects are considered, then the effective nonlinear coefficient is given by \( d_{\text{eff}} = d_{33} \bar{d}_1 \) with \( \bar{d}_1 = 2/\pi \). According to Eq. (2.29), the phase matching condition can be expressed for a general QPM-device as

\[ \Delta k(z) = k_p(\omega_p) - k_s(\omega_s) - k_i(\omega_i) - K_g(z), \]  

with \( K_g(z) = 2\pi/\Lambda(z) \) and \( \Lambda(z) \) as the local QPM poling period. Consequently, phase matching and the parametric interaction is tailored by the design in combination with the duty-cycle \( D(z) \) and the QPM poling period \( \Lambda \).

A grating with constant grating wave vector \( K_g \) throughout its spatial extension can properly phase match only one single set of frequencies. Similar to the situation in collinear BPM, this can lead to a limited gain bandwidth. However, when a spatially varying grating wave vector \( K_g(z) \) is introduced, a wider range of frequencies can be phase matched, which in turn can lead to broader gain bandwidths. Figure 2.6(a) shows

![Figure 2.6](image-url)
a schematic of an exemplary QPM-structure, whereas Fig. 2.6(b) shows a microscope image of a real QPM device, which is constituted by multiple linearly chirped grating.

Under the assumption of a strong undepleted pump and a zero input idler wave, the OPA signal power gain for the case of a linearly chirped grating can be expressed as

$$ G_{ss} = \exp \left( 2\pi \frac{\gamma^2}{|\kappa|} \right), \quad (2.38) $$

where \( \gamma^2 = \left( \omega_i \omega_s d_{33}^2 d_1^2 \right) / \left( n_i n_s c^2 \right) |A_p(0)|^2 \) is the square of the coupling rate between the signal and idler and \( \kappa = -dK_g / dz \) is the QPM chirp rate. The range of wave vectors supported by the grating, e.g. the phase matching bandwidth, is basically given by the product \( \Delta k_{BW} = |\kappa|L \). However in reality, the OPA gain bandwidth is not necessarily equal to the phase matching bandwidth, similar to the case of birefringent phase matching. The OPA bandwidth associated with a linearly chirped grating is approximately given by

$$ \Delta k_{OPA} \approx |\kappa|L - 4\gamma_0. \quad (2.39) $$

Here \( \gamma_0 \) is the peak coupling rate, which is related to the peak intensity of the pump, and to the pump beam profile characteristics.

Regarding experimental considerations, an initial system design is often based on the assumption for the required small-signal gain and phase matching bandwidth. According to these assumptions, the grating length \( L \) and the chirp rate \( \kappa \) can be determined by the help of Eqs. (2.38) and (2.39) for a certain set of pump parameters and coupling rate \( \gamma \).

Moreover, the importance of apodization has been studied [58, 54]. Apodization is a technique to adiabatically switch on and off the coupling among the involved waves. Accordingly, one should append apodization regions at the beginning and the end of a QPM grating in order to obtain useful gain spectra from such QPM devices. More details and a more thorough analysis on the physics and the effect of apodization can be found in [58, 54].

Due to its great flexibility, QPM is a very versatile and promising technique for the purpose of OPA/OPCPA. This technique is extensively applied to LiNbO$_3$. However, it can also be applied to other materials, for example and GaAs [59] and LiTaO$_3$ [60]. LiTaO$_3$ comes into play when
higher damage thresholds are required. On the one hand this material can sustain higher damage thresholds than LiNbO$_3$ but on the other hand it also shows a lower nonlinear optical coefficient [61], which in turn can be again a drawback. Due to the favorable mid-IR properties of LiNbO$_3$, QPM devices exclusively fabricated in LiNbO$_3$ are used in the remainder of this thesis. In chapter 3, 4 and 5, mid-IR OPCPA systems are discussed in more detail, which are all based either on conventional-, chirped- or even more sophisticated two-dimensional QPM structures.

### 2.2 Fundamentals of strong-field ionization

The interaction of intense laser pulses with gaseous and condensed media has been a very important topic in atomic, molecular and optical physics since the advent of ultrashort laser pulses. Light-matter interaction processes are also the key subjects of modern strong-field physics and attosecond science. HHG is a prominent example for one of those processes. It has been shown that HHs can either be generated in gaseous or solid media [21, 22, 23, 62, 63]. In recent years, HHG has become very important for the purpose of time resolved studies with attosecond temporal resolution [64]. There are several theoretical models describing the physical mechanism of generating HHs. A widely used and intuitive model is a semi-classical three-step model which was first introduced by Corkum [23], Schafer et al. [24] and Kulander et al. [25] in 1993 and has already been explained in the introduction of this thesis.

According to this model, the generation of HHs in gaseous media strongly relies on the ionization of the atoms of a particular gas target. Moreover, it was found that the energetic cutoff of the HH-radiation scales like

$$E_{\text{cutoff}} = I_p + 3.17U_p \quad \text{with} \quad U_p = \frac{e^2E^2}{4m_e\omega^2}. \quad (2.40)$$

Here $I_p$ represents a first atomic ionization potential, $U_p$ the ponderomotive potential, $E$ the electric field strength, $e$ the electron charge, $m_e$ the electron mass and $\omega$ the angular frequency of the driving electric field. There is a strong inherent dependence on the strength and the oscillating frequency of the driving field. As suggested by Eq. (2.40) the cutoff increases with decreasing field oscillating frequencies or longer driving
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wavelengths. In fact this scaling behavior is also one of the main motivations for the development of light sources operating at longer wavelengths, for example mid-IR-OPA/OPCPA systems, compared to common systems operating at 800 nm.

It has been shown that HHs driven at longer wavelengths can exhibit a cutoff beyond the water window and therefore enable the generation of coherent x-ray radiation [65, 66] which again can be applied for time resolved studies or XUV- and x-ray imaging [67, 68].

The HHG scaling behavior triggered further experimental investigations to get a deeper understanding of HHG, particularly for the strong-field ionization process itself and the subsequent propagation of the ionized electron in the continuum under the presence of an intense electromagnetic field. This includes the investigation of attosecond ionization and tunneling delays in strong-field ionization, which have been subject to various studies [69, 70, 71]. Furthermore, studies on the wavelength scaling behavior of strong-field ionization dynamics shed new light on this fundamental process by observing the emergence of low-energy structures in photoelectron spectra [72] and holographic structures formed by photoelectrons [73].

A so far experimentally lesser-explored aspect of strong-field ionization is discussed in chapter 6: It presents the experimental results obtained from a strong-field ionization experiment performed with few-cycle pulses emitted from a state-of-the-art mid-IR OPCPA. These results reveal new insights on non-dipole ionization dynamics in strong-field ionization in the long wavelength limit [36].

Strong-field ionization is governed by different mechanisms and regimes. The ionization regime strongly depends on the laser intensity, the oscillating frequency of the driving field and the ionization potential of the ionizing target. The ionization regime can be classified by a parameter $\gamma_K$, which was introduced by Keldysh in 1965 [74].

### 2.2.1 Ionization regime and Keldysh parameter

In strong-field ionization, intense laser light is focused into an atomic target and thereby certain mechanisms can lead to the release of a bound electron into the continuum. This can happen either by the absorption
2. Theoretical foundations

Figure 2.7: (a): Atomic Coulomb potential under the influence of an external laser field for the case of multiphoton ionization. (b): Coulomb potential under the influence of an external laser field for the case of tunneling ionization.

of multiple photons with a total energy exceeding the binding energy of the electron [75], i.e. defined by the atomic ionization potential $I_p$, by tunneling ionization [74] or by an intermediate mechanism, so-called non-adiabatic ionization [76]. In the tunneling regime the electric field strength is strong enough to bend the atomic potential such that there is certain probability for electrons to tunnel through the barrier and being released into the continuum. The situation for the regimes of multiphoton- and tunneling ionization is illustrated in Fig. 2.7.

The different ionization regimes can be classified by the so-called Keldysh parameter which is given by

$$\gamma_K = \omega \sqrt{\frac{2m_e I_p}{eE}} = \sqrt{\frac{I_p}{2U_p}} \quad \text{with} \quad U_p = \frac{e^2 E^2}{4m_e \omega^2}, \quad (2.41)$$

with a first atomic ionization potential $I_p$, the electric field strength $E$ and the angular frequency $\omega$ of the laser field [74]. Multiphoton ionization is the dominating ionization mechanism when $\gamma_K \gg 1$, whereas tunneling ionization is dominating when $\gamma_K \ll 1$. There is also an intermediate regime between those two, which is indicated by $\gamma_K \approx 1$. This is a typical regime used for HHG at laser wavelengths of 800 nm.

The Keldysh parameter drastically decreases when employing significant longer laser wavelengths. For laser intensities ranging from $1 \times 10^{13}$ W/cm$^2$ to $1 \times 10^{14}$ W/cm$^2$, a laser wavelength of 3.4 µm and the noble gas xenon with $I_p = 12.1$ eV as target, the Keldysh parameter varies in between $0.24 \leq \gamma_K \leq 0.75$. This indicates that the experimental results being discussed in chapter 6 were obtained from a strong-field ionization experiment dominated by the mechanism of tunneling ionization.
2.2.2 Dipole approximation in strong-field ionization

In this section we briefly explain the meaning of the dipole approximation and discuss its validity for strong-field ionization [36, 77].

We consider the interaction of an electromagnetic field with an atomic system and assume moderate intensities. Furthermore we assume a driving laser wavelength which is large in comparison to the size of the atomic target. When these two conditions are fulfilled the dipole approximation is assumed as valid because the spatial variation of the laser field across the atoms of the gas target is neglected. Then the laser field can be described by the vector potential so that $A(t) \equiv A(x, t)$, e.g. the dependence on the spatial coordinate $x$ is neglected. As a consequence, the magnetic field inevitably inherent in a laser pulse is zero, since $B = \nabla \times A(t) = 0$. Accordingly, magnetic field effects are neglected in all considerations under the assumption of the dipole approximation.

It can be expected that the dipole approximation breaks down for wavelengths which become comparable to the size of the target. This breakdown in the short wavelength limit is also known as the "upper dipole limit". Figure 2.8 illustrates the intensity-wavelength parameter space and

![Figure 2.8](image_url)

**Figure 2.8:** Intensity-wavelength parameter space in strong-field ionization. The green-dotted area indicates the parameter space where the dipole-approximation is considered as valid and is referred to as the "dipole oasis". The short wavelength limit of the dipole approximation is indicated by the violet line "upper dipole limit". The long wavelength limit arises when the onset of magnetic field effects become evident. This is indicated by the brown-dashed line "magnetic displacement". There are two other limits shown, i.e. the "radiation pressure limit" indicated by the green-dashed line and the "relativistic limit" when true relativistic effects start to occur indicated by the dashed blue line. The red triangles indicate the parameter space used for the experiment discussed in chapter 6. Adapted from [36].

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the breakdown in the short wavelength limit is indicated by the violet solid line labeled as “upper dipole limit”. In fact there is also a lesser-known limit towards longer wavelengths which was theoretically formulated in [78, 79, 80, 81]. In such a regime the magnetic field component of the Lorentz-force acting on the liberated electron linearly depends on the ratio \( \frac{v}{c} \) (expressed in the CGS-system of units) with \( v \) the electron’s velocity and \( c \) the speed of light. Hence, high-energy electrons are influenced by the magnetic field. Such high-energy electrons inevitably occur in strong-field ionization by the use of intense long wavelength driving lasers. Thus, it is expected that the dipole approximation breaks down in the long wavelength limit due to the onset of magnetic field effects. It is important to note that throughout the following classification of the different breakdown regimes atomic units (a.u.) are used.

A fully relativistic treatment is necessary when twice the ponderomotive potential \( U_p \) approaches the rest energy of the electron, i.e. \( 2U_p/c^2 = I/2\omega^2c^2 = 1 \), with the laser peak intensity \( I \) [78, 81]. This limit is indicated by the line labeled as “relativistic” in Fig. 2.8. However, the onset of the influence of the magnetic field can already be observed at significant lower peak intensities and higher laser oscillating frequencies; particularly the limit is reached when the magnetic field induced amplitude of a free electron’s motion in the frame where the electron is in average at rest exceeds 1 a.u., i.e. \( \beta_0 = \frac{U_p}{2\omega c} = 1 \text{ a.u.} \) [78, 82]. Then the free electron undergoes a “Figure-8”-motion due to the combined action of the electric and magnetic field of the laser pulse, which is illustrated in Fig. 2.9. This

![Diagram](image.png)

**Figure 2.9:** “Figure-8” motion of an electron under the presence of an optical field of a long wavelength driving laser. The electron excursion \( \alpha_0 \) is in the direction of the electric field and the excursion \( \beta_0 \) is in beam propagation direction caused by the action of the magnetic field [81, 82].
distinct breakdown is indicated as brown dashes line and labeled as “magnetic displacement” in Fig. 2.8. Moreover, another limit which is caused by radiation pressure becomes evident for $U_p^2 / 2c^2 = 0.5 \text{ a.u.}$ [81].

The investigation of the above mentioned non-dipole effects in strong-field ionization have been subject to a variety of works and their findings will become more important in the future for extending theoretical models, e.g. models used for HHG in attosecond science. These effects will probably also become more severe since there is a clear trend towards long wavelength driving lasers for the purpose of generating x-ray-HHs and attosecond pulses [66]. Furthermore, studies of non-dipole effects have often assumed a negligible influence of the Coulomb potential which has recently been shown in an experiment with circularly polarized laser pulses at 800 nm and 1.4 μm [83] and also in theoretical investigations [79, 80, 84].

In chapter 6 we present an experimental study on non dipole effects in strong-field ionization for the important case of linearly polarized few-cycle pulses. These pulses are delivered from the mid-IR-OPCPA system, which has been developed in the framework of this thesis [31, 32]. It was found that the combined action of the Coulomb potential of the residual ion and the magnetic field component of the laser pulse is of significant importance [36].
Chapter 3

All-collinear mid-infrared OPCPA

Recent developments in the generation of HH show a clear trend towards the application of coherent and bright light sources operating at long wavelengths, enabling the generation of XUV- or multi-hundred-electron Volt (eV) and even kilo-electron Volt (keV) soft x-ray pulses [85, 66]. Long-wavelength ultrashort laser pulses thus enable new capabilities for multidimensional spectroscopy and studies on atomic and molecular dynamics [86, 87, 88, 72] in both the mid-IR and XUV regions. To achieve a high signal to noise ratio (SNR) in sensitive detection schemes used for many applications, light sources with a high average photon flux are needed. In particular, higher repetition rates than those of conventional Ti:sapphire based systems (which mainly operate in the few-kHz regime) are strongly favored [89]. The development of suitable OPA- and OPCPA-based sources is progressing rapidly, with reported systems operating at central wavelengths near 1.8 µm [90, 91, 92], 2 µm [93, 94, 52], 3 µm [95, 53], 3.4 µm [96, 51] and 4 µm [97]. As indicated by the rich diversity of OPA- and OPCPA systems emitting over a wide range of wavelengths, nowadays the generation of few-cycle pulses is mainly achieved by the employment of broadband OPA or OPCPA. However, this technique requires very broad phase matching bandwidths to allow amplification of the full seed spectrum and to support few-cycle pulse durations. There are different ways and techniques to minimize the wave vector mismatch Δk and hence to achieve broadband phase matching. A noncollinear geometry among the interacting waves, which are spatially overlapped in a nonlinear bire-
fringent medium, guarantees a minimum in the GVM [44]. This is a necessary requirement for achieving broad phase matching bandwidths in a noncollinear phase matching scheme. Another technique for achieving broadband phase matching, is QPM in nonlinear dielectric media. QPM was observed for the first time in 1962 [17]. Here, ferroelectric domains are periodically flipped in order to avoid dephasing of the involved waves which are spatially overlapped in a collinear fashion. This scheme greatly reduces experimental complexity and the necessity for angular tuning to find the right operational point for broadband phase matching [43].

In this chapter a collinear, high-repetition-rate, mid-IR OPCPA system is presented. It is operated at a center wavelength of 3.4 µm and is based on APPLN OPA devices. In contrast to a noncollinear phase matching scheme in bulk crystals, the collinear interaction in such QPM devices gives rise to an idler output without an angular chirp. Compared to the previous system [51], the present system has been fully redesigned to account for recent developments in the theory of these devices [54, 57], and to suppress parasitic spatial effects [98]. The new system design overcomes the seed bandwidth limitations of the previous system [51], greatly reduces parasitic optical parametric generation (OPG) and maintains a good quantum conversion efficiency (∼24.5%) of the final OPA stage. Furthermore, the new system delivers sub-four-cycle pulses (41.6 fs FWHM without being limited by the damage threshold of the gain crystals, or by the bandwidth of the seed pulses provided by the primary fiber laser (in contrast to our previous result [51])). In particular, the bandwidth capabilities of the APPLN amplification devices are shown here for the first time by demonstrating 1000-nm-wide gain while maintaining compressible output idler pulses.

In such aperiodic or chirped QPM gratings, the grating wave vector \( K_g \) is swept smoothly and monotonically through phase matching for all spectral components of interest. Signal and idler spectral components experience gain in the vicinity of their local phase matching points. A unique aspect of this approach to OPCPA is that the gratings can support almost arbitrary phase matching bandwidths with the potential for customized gain profiles [43, 99], good conversion efficiencies [54, 100], and access to bandwidths beyond those possible with conventional birefringent phase
matching techniques without operating at the crystal damage threshold. Furthermore, the system complexity is kept minimal by the collinear arrangement of the OPCPA chain. The excellent properties of the present approach pave the way towards relatively compact systems offering intense few-cycle mid-IR pulses for attosecond science and strong-field physics.

The following sections present the individual parts of an all-collinear OPCPA system, which is based on parametric amplification in chirped QPM or APPLN devices. It covers the seed generation, starting with spectral broadening to overcome the seed-bandwidth limitations, temporal chirping, the two-stage pre-amplification, the power amplification and the final pulse compression. This chapter is based on a manuscript which has been published in [31].

3.1 Seed generation - Spectral broadening and chirping

The OPCPA system is shown schematically in Fig. 3.1. First a broadband seed source around 1.56 µm is generated and thereafter amplified in the OPCPA chain. The generated idler pulses are extracted after the final OPCPA stage. As primary seed source for the system, the pulses delivered from a mode-locked femtosecond fiber laser (Toptica FFS pro) operating

![Figure 3.1: Schematic overview of the mid-IR OPCPA setup. DM: dichroic mirror, DM1: HR 1064 nm / HT 1560 nm and 3400 nm; DM2: HR 1560 nm / HT 3400 nm. The pump beam at 1.064 µm is drawn as blue line, the seed at 1.56 µm in green and the idler at 3.4 µm in red respectively. The abbreviations “cyl.” and “Si” stand for “cylindrical lens” and “silicon lens” respectively. Adapted from [31].](image-url)
at $\lambda = 1.56 \mu m$ are used. The pulses exhibit a bandwidth corresponding to a transform-limit (TL) pulse duration of 65-fs and a pulse energy of 3.1 nJ at 80 MHz repetition rate (250 mW average power). For the generation of few-cycle mid-IR idler pulses, the required seed bandwidth in the near-infrared spectral range has to be generated before seeding the OPCPA system. For this purpose, the pulses are first compressed to their transform limit via a pair of silicon prisms in a double-pass configuration. Prior to spectral broadening, the pulses are collimated with a pair of standard lenses and then coupled into a 1-m-long dispersion-shifted (DS) telecom fiber (DCF3, Thorlabs Inc.) with a zero-dispersion wavelength of $\approx 1.6 \mu m$. This fiber operates in the normal dispersion regime, in contrast to the anomalous dispersion regime, which can be utilized for soliton pulse compression. The initially compressed input pulses are thus spectrally broadened while being temporally dispersed through the fiber. This configuration avoids the modulation instability regime while still utilizing convenient off-the-shelf telecom fiber technology. At the output of the spectral broadening fiber, the seed pulses support more than 250 nm of signal bandwidth, supporting sub-30 fs pulse durations. Furthermore,

![Figure 3.2: Pulse characterization after temporal compression and subsequent spectral broadening in dispersion-shifted fiber. (a): Measured spectrogram from SHG-FROG characterization. (b): Retrieved spectrogram. (c): Measured and retrieved spectra and spectral phase and (d) retrieved temporal intensity profile with temporal phase.](image)
the output of the fiber is characterized by near-IR SHG frequency resolved optical gating (FROG). The plots shown in Fig. 3.2 represent an exemplatory pulse characterization after initial temporal compression and subsequent temporal chirping. To provide a clean OPCPA seed, the polarization of the spectrally broadened pulse is optimized with a half-wave plate (HWP) and then stretched by a 4f pulse shaper in a double-pass configuration. The pulse shaper consists of two metallic plane folding mirrors, two plane-concave cylindrical mirrors with a radius of curvature (ROC) of -800 mm, and two holographic transmission gratings with 600 lines/mm. The gratings exhibit a single-pass efficiency of greater than 85% at the design wavelength of 1.55 µm. An exemplatory pulse characterization after the 4f pulse shaper is shown in Fig. 3.3. It is evident from the spectrograms and the reconstructed spectral phase that the pulse shaper mainly adds group delay dispersion (GDD) to the spectral phase. A double-pass configuration of a subsequently arranged pair of silicon prisms provides the third-order dispersion (TOD) required to compress the final output pulses. An exemplatory pulse characterization after the silicon prism pair is shown in Fig. 3.4. It is evident from the spectrograms as well the reconstructed spec-

![Figure 3.3: Pulse characterization after 4f pulse shaper in double-pass configuration. (a): Measured spectrogram from SHG-FROG characterization. (b): Retrieved spectrogram. (c): Measured and retrieved spectra and spectral phase and (d) retrieved temporal intensity profile with temporal phase.](image-url)
Figure 3.4: Pulse characterization after pair of silicon prisms. (a): Measured spectrogram from SHG-FROG characterization. (b): Retrieved spectrogram. (c): Measured and retrieved spectra and spectral phase and (d) retrieved temporal intensity profile with temporal phase.

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This is intended to pre-compensate before the mid-IR pulses are finally compressed via bulk propagation at the output of the OPCPA system. The spectrum after the 4f pulse shaper and the pair of silicon prisms in comparison to the spectrum of the primary seed is shown in Fig. 3.5 (“Broadened Seed” curve). It reveals that the spectrum has broadened by a factor of 1.5 to 2, which is necessary for the generation of shorter pulses than obtained from the previous system [51]. The pulse energy available to seed the first OPCPA stage (depicted as OPA1 in Fig. 3.1) is 92.5 pJ. Furthermore, the spectral ripples seen in Fig. 3.5 originate from the imperfect pulse profile used to seed the DS fiber: while spectral broadening in the positive dispersion regime can in principle provide smooth output spectra, in our case, the pulses are already partially distorted by the nonlinear broadening that also occurs in the erbium-doped fiber amplifiers (EDFAs) of the fiber laser system itself. This conclusion was confirmed via numerical modeling of the spectral broadening process in the DS fiber using standard techniques [101, 102], assuming the FROG reconstructed pulses emitted by the fiber laser as input to the model. During the OPA process, any modulations of the seed spectrum are transferred to the generated idler spectrum.
3.2 APPLN-based amplifier stages

The primary pump source for the OPCPA system is an industrial laser (Time-Bandwidth-Products Inc., “Duetto”) operating at 1.064 µm and delivering 12-ps pulses at a repetition rate of 50 kHz and average output power of up to 10 W. The output of this laser is split in two parts. One part is further amplified by a Nd:YVO₄ Innoslab-type amplifier [103], to provide a higher power pump for the final OPA stage. The remaining part is collinearly overlapped with the seed in OPA1 via a dichroic mirror (DM), denoted DM1 in Fig. 3.1 (transmissive (AR) for the seed at 1.560 µm and reflective (HR) for the pump at 1.064 µm). The average pump power reaching OPA1 is 5.7 W (113 µJ), and it is focused to a spot size of 315 µm (1/e² radius), corresponding to a peak intensity of 4.9 GW/cm². The pump output after the uncoated OPA1 crystal is split from the collinearly propagating signal and idler by another dichroic mirror DM1. In the new layout of the OPCPA chain, this pump power is then re-used to pump OPA2, providing a power of 3.8 W. The pump is refocused in OPA2 to an intensity of 6.7 GW/cm² (1/e² radius of 220 µm) and collinearly overlapped with the signal output of OPA1 (∼1.56 µm), which is split from the idler by dichroic mirror DM2 after the first amplification stage. The gain of the pre-amplification part of the system (i.e. between the OPA1 input and the OPA3 input) corresponds to 42.8 dB. The spectra of the seed and the amplified signals are shown in Fig. 3.6(a). The amplified spectra also contain the unamplified 80 MHz background from the seed source. With
Figure 3.6: (a) Comparison of seed and signal spectra. Spectrum of the primary seed (green line), spectrum of the spectrally broadened seed (black line) and spectra of the amplified signal after OPA1 and OPA2 (red and blue lines, respectively). An offset has been added to the seed and signal curves for clarity. (b) Normalized mid-IR spectra after OPA2 (red line) and OPA3 (blue line), respectively. Adapted from [31].

respect to these spectra, there is evidence of gain-narrowing, due to the non-flat-top pump temporal profile, reducing the amplification for signal spectral components below 1.475 µm and above 1.625 µm. The third and final amplification stage, denoted OPA3, is pumped by the output of an Innoslab amplifier [103]. The pump power reaching the OPA3 crystal is 15.2 W, corresponding to 304 µJ. The signal output of OPA2 is used as the seed for this stage, and the generated mid-IR idler around λ = 3.4 µm is extracted afterwards as the final output of the system. Dichroic mirrors DM1 and DM2 are used to combine and split up the waves. In OPA3, a moderate peak intensity of the pump is used not to drive the crystal too strongly. A consequence of pumping the crystal too strongly is the onset of beam-fanning, likely due to photorefractive effects driven by parasitic green light, which is generated primarily due to random duty cycle variations in the QPM grating [99]. To use all of the available pump power in the 1-mm-aperture APPLN devices, elliptical foci for the pump and signal beams are used. To obtain the required beam sizes, both spherical and cylindrical lenses are employed (indicated by “cyl.” in Fig. 3.1), forming one spherical and one cylindrical telescope. The pump beam is focused to an intensity of 4.4 GW/cm² (1/e² width of 700 µm in the horizontal dimension corresponding to the direction of the crystal c-axis, 1/e² width of 1700 µm in the vertical dimension). The resulting idler spectra of OPA2 and OPA3 are shown in Fig. 3.6(b). The spectra are measured with an
3.3 Pulse compression

For idler compression after OPA3, propagation through a bulk medium is utilized. The compressor consists of a 50-mm-long rod of anti-reflection (AR)-coated sapphire, with the length chosen as a tradeoff between efficiency and gain narrowing in the preamplifier OPCPA stages. Note that adjustment of the dispersion compensation primarily takes place on the seed pulse in the near-infrared spectral range. Due to the phase-reversal properties of OPA, the even-order spectral phase components of the idler inherit, approximately, the even-order components of the seed spectral phase with a flipped sign, whereas the sign of odd-order components of the idler phase are not flipped [43]. The combination of the applied seed phase, which utilizes the 4f pulse shaper and the pair of silicon prisms described above, and idler propagation through the bulk sapphire after the final OPA stage, enables compensation of the idler spectral phase up to fourth order. With this approach, the chirped 3.4-µm-pulses were compressed to 41.6 fs (sub-four-cycles), with 12 µJ of pulse energy after collimation and compression. The compressed pulses were characterized with mid-IR SHG-FROG, as shown in Fig. 3.7.

It is important to note that inside the APPLN grating, the generated idler power was approximately 948 mW ($\approx$ 19 µJ). This value was calculated when accounting for all linear losses encountered from the optical elements after OPA3, including Fresnel losses from the end-facet of the imaging grating spectrometer.

In all stages, newly designed uncoated magnesium-oxide doped lithium niobate (MgO:LiNbO$_3$) (MgOLN) crystals are used, each having an 11-mm-long, 1-mm by 3-mm-wide, apodized APPLN grating design with a chirp rate of $\kappa = -2.5 \text{mm}^{-2}$ (corresponding to a decreasing period versus position) in order to achieve a broad phase matching in a spectral window spanning the 3- to 4-µm spectral range. Concerning apodization, the importance of apodization profiles for such chirped QPM devices has been experimentally studied [58], and it has been recently shown how they can be implemented systematically for a wide range of parametric interactions [54].
uncoated APPLN-grating. This corresponds to an internal conversion efficiency (number of idler photons at the end of the QPM grating divided by the number of pump photons at the input) of 24.5%. With respect to the overall gain in the system, the net amplification between OPA3 input and compressor output corresponds to 11.8 dB.

In order to develop a working OPCPA system using APPLN devices, several important design constraints must be met which go beyond using QPM gratings to support a sufficient phase matching bandwidth. For example, with a given phase matching bandwidth and OPA gain, the non-collinear gain guided modes, discussed in [98], can be fully suppressed with a sufficient pump peak power, thereby suppressing excess OPG. By the design of the system layout and the APPLN gratings, the presence of OPG has been minimized. Specifically, by operating the system at 50 kHz and re-using the OPA1 transmitted pump for OPA2, it was possible to satisfy this pump peak power constraint in each stage, and also ensure sufficient seed power for OPA3 without excessively saturating OPA1 and OPA2. The remaining OPG background was characterized first by measuring the idler output power while operating the amplification chain in an
3.3. Pulse compression

unseeded configuration and also by a spectral characterization of the OPG. The spectra of the amplified idler (seeded case) and the OPG (unseeded case) are shown in Fig. 3.8. Note the different scales for the idler spectrum of OPA3 and the idler OPG spectrum. In both cases, the amount of amplified quantum noise accounts less than 2.5% of the total idler power, and this value represents an overestimate of the actual OPG in the seeded system. This value is a substantial improvement compared to the background in the previous OPCPA-system [51] and to reported OPG-levels of other comparable systems [93, 94, 52]. To the best of our knowledge, this is the lowest OPG background within output pulses delivered by a high-repetition rate, mid-IR OPCPA reported so far, and proves that the excess quantum noise amplification discussed in [98] for this type of gain medium can be fully suppressed by appropriate system design [56].

In addition to redesigning the OPCPA stages to minimize OPG while improving bandwidth and maintaining conversion efficiency, the sign of the pulse chirp was also flipped (now a sapphire compressor is used instead of a silicon compressor [51]). This change was made in order to optimize the OPCPA temporal dynamics. Furthermore, in addition to being more convenient experimentally, seeding the power amplifier stage OPA3 with the signal rather than the idler helps to suppress unwanted processes such as idler second-harmonic generation.

![Figure 3.8](image_url)

**Figure 3.8:** (a): OPG spectrum for the unseeded case of the amplifier chain in comparison to the idler spectrum of OPA3. Note the different scales for the idler and OPG spectrum. (b): Comparison of the idler and OPG spectrum on the same scale to emphasize the relation. The power contained by the area under the green OPG curve accounts for less than 2.5% of the total amount of idler power. Adapted from [31].
3. **Summary and conclusions**

In conclusion, a mid-IR light source operating at a 50-kHz repetition rate and delivering 41.6-fs record-short pulses directly from a high-repetition-rate OPCPA at 3.4 µm central wavelength was presented. The system provides a compressed pulse energy of 12 µJ. Additionally, an internal conversion efficiency of 24.5% was achieved and the OPG background was minimized by the re-design of the seeding and amplification scheme. The internal conversion efficiency is an indication that the power amplifier stage OPA3 is operating in a strongly saturated regime, with the potential to eventually access the fully saturated adiabatic frequency conversion limit [57]. So far, the system employs 1-mm by 3-mm-wide APPLN gratings. In recent years, wide-aperture samples have become available [55], allowing QPM technology to support very high energies. In combination with new developments in high power ultrafast amplifiers [104, 105], this technology should enable high power and high repetition rate few-cycle pulse generation in the mid-IR.
High-intensity light sources emitting in the mid-infrared (mid-IR) spectral range have received great attention in recent years for applications including spectroscopy and strong-field physics. As examples, mid-IR sources have been used for spectroscopic investigations of molecular dynamics, and for HHG beyond the water window [106, 66, 107]. Probing the wavelength-scaling behavior of strong-field photoionization is another key motivation for developing intense long-wavelength driving fields [72, 108, 36]. Ultrafast and high peak-power mid-IR light sources have also been used for accelerator applications, such as enhancing the electron peak current in x-ray free-electron-lasers [109]. Development of laser technologies combining high intensities and few-cycle pulse durations is necessary for many of these applications in order to access the relevant strong-field regimes. Moreover, high repetition rates (and hence high average powers) are important for obtaining a higher photon flux as well as a good signal to noise ratio in reasonable experimental time-frames. OPCPA is especially attractive in this respect that it allows us to combine the benefits of few-cycle pulse durations and high-power technologies (few-cycle for seeding; high-power for pumping). Moreover, new wavelength regions are accessible via OPCPA, provided that broadband phase matching can be achieved. Several approaches have been explored for broadband phase matching of OPCPA in the mid-IR. These include work based on periodically poled
lithium niobate (PPLN) [93, 30, 94, 52], BPM [95, 110, 111], and aperiodic PPLN (APPLN; also referred to as chirped QPM devices) [96, 58, 31]. This latter approach is attractive because it allows for broadband phase matching despite the material’s dispersion and damage threshold.

Assuming 1.064 µm pumping and a near-collinear beam geometry, phase matching from degeneracy up to 4.5 µm in MgOLN requires a range of QPM periods from 32.4 µm to 27.5 µm. Since the relative change in period is small, fabricating a grating whose period smoothly varies between these periods is possible with comparable lithographic poling procedures as used for PPLN devices. This property also means that APPLN devices are well suited for mid-IR seed generation via DFG [52, 112]. However, due to a number of design constraints that were explained recently [56], achieving broadband OPCPA supporting high-quality pulses is more challenging than obtaining a sufficient phase matching bandwidth. In particular, two of the constraints discussed in [56] motivated the present experiment. First, unwanted nonlinear processes involving sum- and difference-frequency mixing between the nominal pump, signal and idler pulses can be phase matched due to the broad range of QPM periods involved. Second, when large intensity-times-length products are involved, the device becomes more sensitive to random duty-cycle (RDC) errors in the QPM grating [57], which can lead to an unwanted enhancement of pump SHG, even if this process is not phase matched by the nominal grating design. Therefore a new amplification regime, one that is less sensitive to these effects, is called for.

In this chapter, we demonstrate OPCPA based on a noncollinear beam configuration in combination with QPM, implemented via PPLN. Similar phase matching schemes to the one discussed in this chapter have been deployed previously. Noncollinear QPM in PPLN was demonstrated in the context of 1.064-µm-pumped optical OPG of a broadband signal wave in the spectral range between 1.66 µm and 1.96 µm [113]. A related noncollinear phase matching scheme, based on an angularly-chirped input signal wave, was proposed for PPLN in [114]. Until now, however, such schemes have not been experimentally demonstrated for broadband QPM OPCPA. With a related approach, we generate mid-IR pulses at a center wavelength of 3.4 µm with a pulse duration of 44.2 fs (sub-four-
cycles), an energy of 21.8 µJ, and a repetition rate of 50 kHz (average power 1.09 W). The present system uses APPLN devices for simultaneous pre-amplification and parametric transfer of the 1.5-µm seed to the mid-IR, combined with a noncollinear PPLN power amplifier operated in an achromatic phase matching regime, and is the first realization of such a hybrid OPCPA configuration. The achieved pulse energy represents more than an 80% improvement compared to the result presented in chapter 3 using the same pump and seed lasers. Furthermore, the wide applicability of this scheme for broadband parametric amplification is shown and also how noncollinear APPLN devices can resolve the issues for aperiodic QPM devices mentioned above, enabling octave-spanning gain bandwidths. This chapter is based on a manuscript which has been published in [32].

4.1 Experimental setup

A schematic of the present OPCPA system is shown in Fig. 4.1. Here, a new final power amplifier stage is employed, while keeping the seeding and pre-amplification layout the same as the system reported in chapter 3 of this thesis and [31]. For the sake of completeness, an overview of the system layout is also given. The OPCPA system is pumped at 1064 nm by an industrial laser (Time-Bandwidth-Products, "Duetto") which produces 12-ps pulses at a repetition rate of 50 kHz and an average power of 10 W. Approximately 6 W from this laser is used for pumping the APPLN-based pre-amplifiers, while the remaining power is used to seed a home-built Innoslab amplifier [103], which currently provides up to 17.8 W when operated at 50 kHz. For seeding the OPCPA system, a femtosecond fiber laser (Toptica FFS pro), which produces 65-fs pulses at 80-MHz at a center wavelength of 1560 nm, is used. These seed pulses are spectrally broadened in a DS-fiber. After this fiber, seed-pulse chirping is performed with a 4f pulse shaper followed by a silicon prism pair thereby the seed pulses exhibit a similar pulse duration as described in section 3.1 of chapter 3 for the case of the all-collinear OPCPA system. After the pulse chirping arrangement, two collinear pre-amplifiers based on apodized APPLN gratings [54] are employed and denoted as OPA1 and OPA2, respectively. As mentioned above, the seeding, chirping, and pre-amplification components of the sys-
Figure 4.1: Experimental setup of the mid-IR OPCPA. The “dispersion management unit” includes a stage for spectral broadening, a 4f pulse shaper and silicon prism pairs for dispersion compensation. The angular-dispersion-free idler extracted from the pre-amplifier OPA2 is used as seed for the noncollinear PPLN power amplifier (OPA3) shown in the dashed box. Adapted from [32].

...tem are the same as described in the previous chapter and in [31]. For the system covered by this chapter, the power amplifier was modified, denoted OPA3. The new amplifier configuration is depicted in the dashed box of Fig. 4.1. The collinear pre-amplifier OPA2 is seeded by the 1.5-µm signal output of OPA1, and generates a broadband and angular-chirp-free 3.4-µm idler output suitable for seeding OPA3. OPA3 is based on a PPLN grating and uses a noncollinear pump beam geometry, described further in section 4.2. Compression of the amplified idler is performed in a 50-mm sapphire rod as it is the case for the all-collinear OPCPA system. The spectral phase of the signal seed is parametrically transferred to the generated idler: by adjusting the signal pulse chirp before OPA1 via the 4f pulse shaper and silicon prism pair, the remaining chirp on the amplified idler pulses after the sapphire rod is compensated.

4.2 Phase matching and parametric amplification

The phase matching scheme employed in OPA3 is depicted in Fig. 4.2. Rather than the conventional non-critical configuration enabled by QPM (where phase matching is insensitive to the incident angles), an achromatic configuration based on noncollinear beams is used, in which the phase mismatch $\Delta k$ is insensitive to wavelength. In contrast to APPLN devices used in a non-critical configuration, periodic QPM gratings in an achromatic configuration are less sensitive to parasitically phase matched pro-
cesses and RDC-errors, which can lead to unwanted pump SHG, although the process is not nominally phase matched. In fact, this is the reason for the choice of phase matching configuration in a periodic QPM device. In this configuration, QPM offers two unique advantages over conventional BPM: the ability to achieve phase matching for almost any beam geometry while still using the largest nonlinear coefficient $d_{eff}$ of a material; and the ability to use aperiodic QPM devices for even larger gain bandwidths and customized gain spectra. The corresponding phase matching diagram is shown in Fig. 4.2(a), assuming mid-IR idler seeding collinear to the constant grating k-vector $K_g$, and a narrow-band noncollinear pump incident at an angle to the crystal c-axis. The inset shows the frequency-dependent spread of the longitudinal phase-mismatch $\Delta k$ by zooming in on the circled region. For comparison, Fig. 4.2(b) shows the phase matching diagram for two spectral components (3 $\mu$m and 4 $\mu$m) in the collinear APPLN pre-amplifiers (the scale and changes in $K_g$ are exaggerated). The schematic of a chirped QPM grating shown next to these phase matching diagrams, illustrates how these spectral components are amplified around different positions in the grating (3 $\mu$m near the lower, input-side of the grating; 4 $\mu$m near the upper, output-side). In Fig. 4.2(c), the corresponding frequency-dependent phase mismatch in the absence of a QPM grating is shown, denoted $\Delta k_0(\lambda)$, for both collinear and noncollinear cases. The

![Figure 4.2](image-url)
4. Noncollinear achromatic QPM OPCPA power amplifier

The noncollinear case (green curve) shows a relatively small spread of $\Delta k_0(\lambda)$: in this case, a single QPM period in a short device combined with an intense pump can amplify the 3- to 4-$\mu$m spectral range, as in OPA3. In the collinear case (blue curve), the wider spread of $\Delta k_0(\lambda)$ can still be amplified by a longer, chirped grating having the corresponding range of QPM periods, as in the pre-amplifiers (see dot-dashed lines around the blue curve). The potential of combining a chirped QPM grating with the noncollinear arrangement is illustrated by the dot-dashed lines around the green curve: in this case, an octave-spanning gain spectrum is supported using a similar spread of grating periods as already used in the current pre-amplifiers OPA1 and OPA2. This possibility and the importance and constraints of combining chirped QPM gratings with noncollinear geometries, is discussed in more detail in section 4.4. To illustrate the role of the pump angle in more detail, in Fig. 4.3(a), the frequency-dependent quasi phase matching period as a function of wavelength for several internal angles of the pump beam is plotted. Generally, the broadest bandwidth from a PPLN device is obtained near the turning point in QPM period with respect to wavelength; this turning minimizes the spread of $\Delta k$ [Fig. 4.2(a) inset], and also corresponds to GV-matching of the signal and idler waves. By varying the pump angle (see legend of Fig. 4.3(a)), the turning point can be tuned to any wavelength beyond degeneracy. In the experiment, an internal pump angle of $\approx 6.25^\circ$ and a QPM period of 26 $\mu$m, is used as indicated by the dashed black line in Fig. 4.3(a). Figure 4.2 together with Fig. 4.3 show how the beam geometry can be optimized for bandwidth [minimizing the spread of $\Delta k(\lambda)$], and phase matching can be obtained at

![Figure 4.3](image-url)

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**Figure 4.3:** (a): Phase matching period for several different angles of the pump beam. The QPM poling-period of 26 $\mu$m used for OPA3 is indicated by the black dashed line. (b) Example small-signal gain spectrum enabled by such a device. Adapted from [32].
4.2. Phase matching and parametric amplification

whatever angle is optimum by adjusting the QPM period. The analogous procedure in BPM is to rotate the crystal axes, but in this case both the beam geometries and the nonlinear tensor elements that can be used are more strongly constrained by phase matching considerations, and there is no straightforward way of introducing a spatially varying phase mismatch.

Furthermore, beyond the experimental demonstration representing a proof-of-principle (described in section 4.3), the opportunities enabled by noncollinear pumping of chirped QPM devices is explained in section 4.4. It is important to note that small noncollinearities are typically employed in wavelength-degenerate OPA and OPCPA schemes for beam separation and to avoid the phase-sensitivity of true degenerate OPA; example systems using QPM include Refs. [93, 30, 94, 52]. The small noncollinearity in such systems only has a minor influence on the phase matching properties, unlike the present approach. This fact is also emphasized by the illustration in Fig. 4.3, when comparing the phase matching curves for 0° and 1.25°. There is no evidence for a significant change in the phase matching properties.

Next, to illustrate the characteristic features of OPCPA based on the phase matching curves shown in Fig. 4.2(c) and 4.3(a), an example gain spectrum for a mid-IR PPLN OPA device is shown in Fig. 4.3(b). For this example a moderate coupling rate between the signal and idler waves of $\gamma = 2 \text{ mm}^{-1}$ is assumed. The corresponding small-signal gain spectrum is given by

$$G_{ss} = \left| \cosh (gL) + i \frac{\Delta k}{2g} \sinh (gL) \right|^2,$$

where $g = [\gamma^2 - (\Delta k/2)^2]^{1/2}$ and $L$ is the crystal length. A finite $\Delta k$ at the center wavelength can broaden the gain spectrum, as for collinear and nearly-collinear degenerate OPA [52]. However, the phase mismatch should not be too large for a saturated amplifier, because the achievable pump depletion is also limited by the ratio $(\Delta k/\gamma)$. Assuming only two of the three waves are present at the input of the device (i.e. only the pump and the seed waves), and taking the high-gain OPA limit of such parametric interactions (i.e. by assuming a very weak input seed wave compared to the pump but a long enough crystal to fully deplete the
pump), the maximum plane-wave conversion efficiency is given by

$$\eta_{\text{max}} = 1 - \left( \frac{\Delta k}{2\gamma} \right)^2,$$

where this equation applies for $|\Delta k/2\gamma| < 1$, i.e. the requirement for parametric gain. This result can be obtained via the formalism described in [115], which provides an intuitive geometrical interpretation of the possible parametric conversion trajectories given arbitrary input conditions and $\Delta k$. A value of $\Delta k/\gamma = -0.6$ at the center wavelength thus provides a reasonable trade-off: the bandwidth is broadened according to Eq. (4.1) and Fig. 4.3(b), while the maximum efficiency (for an idealized plane-wave interaction) is still $> 90\%$ based on Eq. (4.2).

### 4.3 Experimental results

We implemented the proposed scheme as the final power amplification stage. The average powers for the seed and pump just before the AR-coated 2-mm long, 2-mm by 2-mm wide PPLN crystal are 31 mW and 17.8 W, respectively. The pump is focused to an intensity of $10.7 \, \text{GW/cm}^2$ ($1/e^2$-radius of 475 \(\mu\)m in the horizontal and 350 \(\mu\)m in the vertical axis).

In Fig. 4.4(a), the measured idler spectra before and after the power amplifier are presented. Next, to estimate the OPG background, the signal seed was blocked before the first APPLN pre-amplifier so that the noncollinear power amplifier was operated in an unseeded state. The spectrally resolved OPG background is shown in Fig. 4.4(b). The average power was also measured with a thermal power meter. The spectral characterization as well as the power measurements are within the noise floor of the respective detectors, indicating a negligible OPG background compared to the amplified pulses ($<300 \mu\text{W}$). Furthermore, no evidence for photorefractive effects was observed. The amplified idler pulses were compressed by the careful design of sign and amount of dispersion of the near-IR seed pulses prior to the APPLN pre-amplifiers, in combination with bulk-propagation of the idler after the final power amplifier through an AR-coated, 50-mm-long sapphire rod. This method yielded a pulse duration of 44.2 fs (less than four optical cycles). Figure 4.5 shows the measured and retrieved spectrograms, spectra and spectral phase from
an SHG-FROG characterization of the pulses. The residual fluctuation of the spectral phase results in a pedestal in the time domain. Note that, as discussed in [31] and also in chapter 3, the fluctuations in spectral phase most likely originate from the near-IR seeding scheme, particularly from the spectral broadening, rather than any of the OPCPA devices. The reconstructed temporal intensity profile is shown in Fig. 4.5(d). The energy of the compressed idler pulse was 21.8 µJ, corresponding to an average power of 1.09 W at the 50-kHz repetition-rate and a gain of 15.46 dB. The inset of Fig. 4.5(d) shows the beam profile of the compressed mid-IR idler. The fairly large noncollinear angle between the idler, signal and pump beams leads to some spatial walk-off within the crystal. This effect, combined with the slight ellipticity of our pump beam, explains the slight ellipticity of the idler beam, which potentially could reduce the beam quality.

Moreover, the spatial walk-off effect imposes a constraint on the peak power of the pump that is needed to obtain a high parametric gain (while using a round beam). For a characteristic noncollinear angle $\Theta_p$, the ratio of transverse spatial walk-off $w_{wo}$ to pump beam radius $w_p$ can be expressed as

$$\frac{w_{wo}}{w_p} = \frac{\gamma L \tan(\Theta_p)}{\gamma w_p} \approx \sqrt{\frac{P_0}{P_{p, pk}}} \text{arccosh} \left( \sqrt{G_{ss}} \right) \tan(\Theta_p). \quad (4.3)$$

Figure 4.4: (a): Measured spectra of the input idler (blue) and output idler (red) of the non-collinear OPCPA power amplifier. The inset shows the good input idler beam profile generated by the second APPLN pre-amplifier, OPA2. (b): Spectral characterization of the OPA background, measured by blocking the near-IR seed pulses before the first APPLN pre-amplifier, in comparison to the idler pulse spectrum. These measurements show that the energy of this OPG background is at least 34.6 dB lower than the idler pulse energy. Adapted from [32].
In the first part of this equation, the value of $\Theta_p$ is determined by the GV-matching condition; the $(\gamma L)$ factor determines the small-signal OPA gain [116]; and, for a given nonlinear crystal, $(\gamma w_p)^2$ is proportional to the peak power of the pump. The second part of the equation applies these relations assuming $\Delta k = 0$ for calculating $G_{ss}$; $P_0$ is a characteristic peak power associated with the material and the wavelengths involved, and is given by

$$P_0 = \frac{n_i n_s n_p c \epsilon_0 \lambda_s \lambda_i}{16 \pi d_{eff}^2}, \quad (4.4)$$

and $P_{p,pk}$ is the value for the pump peak power. For a 50% duty cycle QPM grating and $d_{33} = 19.5 \text{ pm/V}$ for MgOLN [117], $d_{eff} = 2/\pi d_{33}$ and $P_0=17.2 \text{ MW}$. Consequently, with $\Theta_p = 6.25^\circ$ (the geometry used experimentally), a peak power of 14 MW is required to achieve a plane-wave small-signal gain of 30-dB while keeping $w_{wo}/w_p < 0.5$ (to avoid large walk-off relative to the pump beam size). The peak pump power used in OPA3 is 24.1 MW, satisfying this constraint. To satisfy the walk-off con-

**Figure 4.5:** (a) Measured spectrogram from SHG-FROG characterization. (b) Retrieved spectrogram. (c) Measured and retrieved spectrum and spectral phase and (d) retrieved temporal intensity profile with temporal phase showing a pulse duration of 44.2 fs. The inset shows the idler beam profile. The FROG- error of the reconstruction for a grid size of 512 by 512 points is 0.0125. Adapted from [32].
4.4 Implications for other QPM-media

The presented noncollinear, achromatic phase matching scheme employed to conventional PPLN devices may have the potential for the extension to chirped QPM devices. A possible route to this is discussed in this section.

Aperiodic QPM devices

Looking beyond OPCPA in periodic devices, our results open up the interesting possibility of combining achromatic QPM arising from noncollinear beams with aperiodic (chirped) QPM gratings. In this section, we show the advantages of this approach, and discuss the peak power levels required.

In section 6 of [56], the possibility of phase matching additional sum and difference-frequency mixing processes in chirped QPM devices is considered. If the QPM period for some unwanted processes (such as idler SHG) overlaps with the QPM periods required for the nominal OPA process, then this unwanted process may be efficient, potentially distorting the OPCPA output. Experimentally, these effects are avoided by the choice of QPM grating chirp rate [31, 56], but as the range of QPM periods is increased (to amplify a broader bandwidth), it becomes more difficult to avoid such processes in a collinear geometry. However, if the desired OPA process depends on the beam angles in a different way to the other DFG and SFG processes, then the beam geometry offers a way to reduce the influence of these other effects.

To illustrate this approach, Fig. 4.6 shows the phase matching period for several processes assuming different pump angles and wavelengths: for 0° and \( \lambda_p = 1.064 \, \mu m \) in Fig. 4.6(a), and for 7.5° and \( \lambda_p = 1.030 \, \mu m \) in Fig. 4.6(b); the legend applies to both Figs. 4.6(a) and 4.6(b). As an example, consider the OPA and idler SHG processes. To suppress idler SHG, this process should either be phase matched before the OPA process, or it should not be phase matched in the grating at all. However, if the QPM
4. **Noncollinear achromatic QPM OPCPA power amplifier**

![Figure 4.6](image-url)

**Figure 4.6:** Phase mismatch for unwanted processes for two pump wavelengths and angles geometries. (a): \( \Theta_p = 0^\circ \) and \( \lambda_p = 1064 \text{ nm} \), i.e. collinear beams, as currently used in our APPLN devices and discussed in [56]. (b): Noncollinear configuration with \( \Theta_p = 7.5^\circ \) and \( \lambda_p = 1030 \text{ nm} \). We assume idler seeding with angle \( 0^\circ \) to the grating wave vector. The plane of incidence includes the crystal c-axis. Several processes are shown: SHG of the nominal pump, signal and idler waves; SFG of the idler and signal with the pump; DFG between the signal and idler waves; and amplification of this signal-idler difference-frequency by the pump. The signal is discarded, so processes involving this wave are less critical. The curves are cut off when one of the wavelengths involved lies beyond 5 \( \mu \text{m} \), based on the high absorption and inaccuracy of the Sellmeier relation beyond this wavelength [118]. In (a), the idler SHG process cannot be avoided across the whole wavelength range shown, as discussed in the text. In (b), with a positive QPM chirp rate, corresponding to a QPM period increasing with position through the grating from 21.6 \( \mu \text{m} \) to 24.4 \( \mu \text{m} \), idler wavelengths from 2.3 \( \mu \text{m} \) to 5 \( \mu \text{m} \) (and possibly further, limited by absorption and accuracy of the Sellmeier equation) can be amplified while suppressing the parasitic processes shown. The legend applies to both (a) and (b). Adapted from [32].

period passes through idler SHG later in the grating than OPA, the idler will be intense (especially in an OPA stage with high pump depletion) and so idler SHG will be efficient. In Fig. 4.6(a), the curves for OPA and idler SHG cross, so the latter process cannot be avoided for all the wavelengths shown. In Fig. 4.6(b), these processes do not cross and the required QPM periods do not overlap.

For other processes, for example idler-signal DFG, the QPM periods do overlap, but are shorter than those for OPA. Thus, by choosing a positive QPM chirp rate \( \kappa > 0 \), corresponding to an increasing QPM period with position), these processes will be phase matched earlier than the OPA process. As such, most of the parasitic processes can be avoided in Fig. 4.6(b) by using a positive QPM chirp rate. There is an exception at 2.6 \( \mu \text{m} \) where the curves for OPA and signal SHG cross. Based on Fig. 4.6(b) and typical values of \( \gamma \approx 3 \text{ mm}^{-1} \) for APPLN OPCPA, we estimate that spectral components within around +/- 35 nm of this crossing have amplification.
regions that overlap with signal-SHG phase matching; such an overlap would lead to a somewhat reduced OPA efficiency for these spectral components. However, for components outside this narrow wavelength range, only the signal will be strongly affected by the signal SHG process, and this signal wave is discarded after the amplifier. Figure 4.6 shows how the OPCPA process can be favored over other parasitic processes such as idler SHG. The amplification bandwidth supported is as much as 2.3 \( \mu m \) to 5 \( \mu m \), more than an octave. The corresponding range of grating wave vectors \( K_g \) is 258 mm\(^{-1}\) to 291 mm\(^{-1}\) (QPM periods from 24.4 \( \mu m \) to 21.6 \( \mu m \)). Since this grating spatial frequency bandwidth is only slightly larger than that used in the current OPCPA experiments, the device should not be much more sensitive to the effects of RDC errors [56, 57].

Nonetheless, a potential issue with this noncollinear APPLN OPCPA scheme is spatial walk-off: if there is an idler-pump beam walk-off comparable to their beam sizes, then idler spectral components phase matched near the start of the chirped QPM grating will be displaced laterally compared to those phase matched near the end, leading to a spatial chirp. This spatial walk-off must therefore be minimized. To quantify this constraint, the walk-off in terms of the other device design parameters is expressed in analogy to Eq. (4.3). The small-signal gain is given, for a linearly chirped QPM grating, by \( G_{ss} = \exp(2\pi\Lambda) \) [43], where \( \Lambda = \gamma^2 / |\kappa| \) and \( \kappa = d(\Delta k)/dz = -dK_g/dz \) is the QPM chirp rate. The quantity \( \kappa^2 \) is proportional to pump intensity, so achieving a high gain requires, for a given intensity, a sufficiently small chirp rate \(|\kappa|\). Given this chirp rate, the product \(|\kappa|L\) must be large enough that the grating is swept through the range of wave vectors required to amplify the idler spectrum. However, the spatial walk-off increases with the grating length \( L \). By expressing the spatial walk-off in terms of these quantities, we find:

\[
\frac{w_{wo}}{w_p} \approx \left( \frac{|\kappa|L}{\gamma} \right) \left( \frac{P_0}{P_{p, pk}} \right)^{1/2} \frac{\ln(G_{ss})\tan(\Theta_p)}{2\pi}, \quad (4.5)
\]

where \( P_0 \) was defined following Eq. (4.3). As a simple example, with \( \gamma = 3 \text{ mm}^{-1} \) (peak intensity of approximately 10 GW/cm\(^2\)), \( G_{ss} = 30 \text{ dB} \), \( \Theta_p = 7.5^\circ \), \( L \approx 4 \text{ mm} \), and \(|\kappa|L = 33 \text{ mm}^{-1}\) based on the wave vector bandwidth requirement stated above, the corresponding peak power re-
requirement to satisfy $w_{wo}/w_p < 0.2$ (imposing a stricter constraint than in the previous section because of the greater severity of a spatial chirp effect compared to an elliptical beam distortion) is $1 \text{GW}$. For a given phase matching bandwidth, this peak power can be reduced by increasing $\gamma$, but the maximum possible value of $\gamma$ is limited by the damage threshold. Our current pump laser has lower power ($25 \text{MW}$), but GW peak powers can be achieved with current 1-$\mu$m laser technologies. Demonstration of this regime may therefore require an upgraded pump, or alternatively elliptical input beams.

Note also that the GV-walk-off of the broadband idler relative to the noncollinear pump over 4 mm is, for pump angle $\Theta_p = 7.5^\circ$, between $\approx -200 \text{fs}$ (for 2.3 $\mu$m) and $\approx +1400 \text{fs}$ (for 4.9 $\mu$m); when designing an OPCPA using few-ps pump pulse durations, this temporal walk-off should be considered. Maintaining small beam walk-off for a relatively large noncollinear angle should also ensure suppression of gain guided noncollinear modes [56, 98], although some care should be taken in this respect, especially if using elliptical beams and lower peak powers than those suggested by Eq. (4.5). In particular, using a large beam size in the walk-off direction (major axis) but a small one in the other transverse direction (minor axis) increases the susceptibility to gain guided modes along the minor axis. A detailed study of this aspect of the problem is beyond the scope of this thesis.

**Orientation-patterned Gallium-Arsenide**

It is interesting to note that the demonstrated amplification technique is also applicable to other QPM media as well, including non-birefringent materials. For example, it can support broadband OPA in orientation-patterned gallium-arsenide (OP-GaAs) [119], with the potential for extending OPA and OPCPA into the far-IR spectral region [120].

To compare OP-GaAs to MgOLN, it is important to note that the 1.064-$\mu$m pump wavelength is somewhat longer than half the zero-dispersion wavelength (which is around 1.9 $\mu$m in MgOLN). An analogous configuration for OPA in OP-GaAs, whose zero-dispersion wavelength is around 6.6 $\mu$m [121], would be to use a pump wavelength of $\approx 3.4 \mu$m or longer. This pumping configuration could support GV-matched OPCPA for cen-
ter seed wavelengths from 7 to $\sim 18 \mu m$. Alternatively, using 2-$\mu m$ or 2.5-$\mu m$ pump lasers, GV-matching could be achieved for signal-seeded OP-GaAs devices. Therefore, this technique may prove to be important in developing widely tunable and few-cycle sources in the far-IR as well as the mid-IR spectral range.

### 4.5 Summary and conclusions

In conclusion we have demonstrated a broadband mid-IR hybrid OPCPA system based on APPLN pre-amplification and a new noncollinear GV-matched PPLN power amplifier. The idler seed pulses, centered around 3.4 $\mu m$, are generated from two weakly-saturated all-collinear APPLN pre-amplifiers. The pre-amplifiers take advantage of important features of chirped QPM media, including convenient and collinear parametric transfer of the near-IR (1.5 $\mu m$) seed to the mid-IR, and amplification of a broad bandwidth determined by the QPM grating structure. Such pre-amplifiers are also less sensitive to some of the parasitic processes shown in Fig. 4.6(a), since the signal and idler waves remain significantly weaker than the pump throughout the device. The idler pulses generated by OPA2 are used to seed OPA3, where they are amplified to 21.8 $\mu J$ (energy measured after pulse compression), representing an 80% improvement over our previous result. The pulses are compressed to 44.2 fs (corresponding to less than four optical cycles) by propagation through a bulk sapphire rod. Fine-tuning of the idler chirp is obtained by adjusting the pre-chirping of the 1.56-$\mu m$ signal seed before the pre-amplifiers. The GV-matched noncollinear QPM OPCPA technique demonstrated here is a highly versatile approach for broadband amplification in the mid-IR, and even the far-IR via OP-GaAs. The present results also demonstrate that this technique can be combined with APPLN amplifiers in a practical hybrid OPCPA system configuration. Furthermore, the discussion in section 4.4 has shown how the combination of noncollinear beam geometry and chirped QPM devices suggests octave-spanning amplification may be possible; the use of nonlinear chirp profiles would even enable customizable gain profiles across this amplification bandwidth [43, 99]. This approach, together with further power scaling achievable by the use of wide-aperture
QPM devices as the gain medium [55], thus provides a route to amplification of high-energy single-cycle pulses. This capability will enable a wide variety of strong-field experiments pumped across the mid-infrared wavelength region. Furthermore, the OPCPA system described in this chapter is used to examine strong-field ionization experiments. The setup and the results of these experiments are discussed in chapter 6 of the present thesis.
Chapter 5

**Broadband frequency-domain OPA**

The previous two chapters covered the generation of few-cycle mid-IR pulses by the help of QPM devices, i.e. longitudinally chirped QPM devices (APPLNs) in chapter 3 and conventional PPLN devices as power amplifier in chapter 4 respectively. It has been shown that these devices have the potential for very broad phase matching bandwidths and hence very short pulse durations, down to sub-four-cycle pulses, and that they are also suitable for the generation of energetic pulses in the tens of microjoule-regime [31, 32]. In addition, these devices allow for the reduction of parametric superfluorescence and therefore enable low-noise operation when the system design accounts for certain guidelines as proposed in [56] and experimentally verified in [31, 32].

In conventional OPCPA, amplification occurs at all instances of time and space of the pump pulse and could suffer from back-conversion of energy from the signal to the pump if the pump intensity is too high [17]. Thus, it is quite challenging to maintain the desired interaction across the spatial, temporal and spectral profiles of the interacting ultrashort waveforms. Recently, a complementary approach to OPCPA was theoretically introduced by Chen *et al.* [122] and experimentally verified in a related scheme by Schmidt *et al.* [123], termed frequency-domain optical parametric amplification (FOPA). In their experimental realization the seed pulse is spatially chirped by the use of a 4f pulse shaper arrangement, analogous to [124], and amplification takes place in the Fourier plane. By placing multiple birefringent phase matching crystals in the Fourier plane,
the phase matching condition for different spectral regions can be adjusted individually. Thus, one of the key constraints among conventional OPCPA can be relaxed. Moreover, the spatially dispersed seed pulse used for this technique offers great flexibility in terms of matching the effective seed pulse duration with the few-ps pump pulse and thus for efficient energy transfer as will be explained in section 5.2 of this chapter. However, this great flexibility also comes along with a major drawback: the optical path lengths through the individual crystals have to be precisely matched to wavelength precision. Furthermore, the system complexity increases with the number of crystals.

In this chapter, a new platform for nonlinear optics is introduced, in particular for the generation of few-cycle pulse durations in the mid-IR spectral range by means of FOPA based on two-dimensionally patterned (2D)-QPM media. Besides the ability for broadband amplifications, this technique can also simultaneously combine the separate functionalities of frequency transfer and pulse shaping purposes into one single monolithic and lithographically fabricated QPM device. This approach offers solutions to performance limiting issues among conventionally used techniques. These issues include complicated noncollinear beam geometries and high laser intensities close to the damage threshold that are normally needed to achieve phase-matched amplification over an ultra-broadband spectrum. Moreover, the 2D-QPM FOPA approach supports scaling in power and it overcomes drawbacks inherent in multiple-crystal approaches as used in [123]. Here, a proof-of-principle experiment is demonstrated by employing this new paradigm in nonlinear optics by the generation of few-cycle mid-IR pulses through OPA of spatially dispersed seed pulses in the Fourier-plane of a 4f pulse shaper. This chapter is based on a manuscript which has been published in [125].

5.1 Two-dimensional quasi phase matching devices

The sign of the nonlinear coefficient is periodically or aperiodically flipped in QPM devices as already described in section 2.1.3 of chapter 2. This patterning gives rise to an additional term $K_g$ augmenting the phase matching condition leading to the relation $|k_p - k_s - k_i - K_g| \approx 0$, where $k_j$
represent the wave vectors of the interacting waves. In contrast to birefringent phase matching (BPM), which relies on favorable material properties, phase matching in periodic or chirped QPM devices can be freely engineered via lithographic processes applied to LiNbO$_3$ or LiTaO$_3$. For the case of a chirped QPM device the grating wave vector is depending on the longitudinal spatial coordinate $z$, so that $K_g = K_g(z)$, representing a 1D-QPM device. This allows to extend the phase matching bandwidth well beyond the one of conventional periodic QPM devices [43, 112].

Here a much more general approach is presented: the deployment of fully two-dimensional quasi phase matching (2D-QPM) patterns to tailor the parametric interactions experienced by a spatially dispersed seed pulse. This technique relies on the ability to vary the QPM period in the transverse direction such that each spectral component is perfectly phase matched. Moreover, the QPM period can be varied continuously with no
Figure 5.2: (a): Selection of ferroelectric domain profiles for the QPM period mapping from Fig. 5.1(c) and (d). Every 20\textsuperscript{th} domain is shown. (b): Absolute phase $\phi_{QPM}(x, 0)$ at the input position corresponding to the domains shown in (a). (c): Stretched image of the 2D-QPM grating fabricated in a 1-mm thick MgOLN crystal. The image is constructed by a series of microscope images of the $+z$-facet of the crystal along the transverse direction. Adapted from [125].

inherent limitation in bandwidth. Figure 5.1 illustrates the ability of such a 2D-QPM device. A single monolithic plane-parallel crystal is used and consequently the linear optical properties of the QPM device remain homogeneous.

When considering the example Gaussian intensity profile shown in Fig. 5.1(b), then such changes in the pump intensity over the transverse direction would significantly change the gain for different spectral components. However, another great advantage of the 2D-QPM concept is that it also enables the variation of the QPM properties along the beam propagation direction, e.g. varying the effective length $L$ such that it is matched to the pump beam’s intensity profile in order to modify the nonlinear interaction and thereby flatten the small-signal gain spectrum. This general capability is shown in Fig. 5.1(b)-(d).

In order to design a practical 2D-QPM grating profile and to facilitate its capabilities, a smooth nonlinear variation in the QPM period is introduced. A corresponding map plotting the QPM period as a function of
the transverse and the longitudinal position is shown in Fig. 5.1(a).

To fabricate such a design, the corresponding absolute phase \( \phi_{\text{QPM}}(x, z) \) is introduced, given by

\[
\phi_{\text{QPM}}(x, z) = \phi_{\text{QPM}}(x, 0) + \int_{0}^{z} K_{g}(x, z') dz',
\]

(5.1)

where \( 2\pi/K_{g} \) is the local QPM period and \( \phi_{\text{QPM}}(x, 0) \) is an input phase profile. It can be freely chosen by the design. Once the QPM pattern is set by \( \phi_{\text{QPM}}(x, z) \), then the relation between the phase and the nonlinear coefficient satisfies \( d(x, z) = \text{sgn}(\cos(\phi_{\text{QPM}}(x, z))) \). This QPM phase is then transferred to any wave generated during the nonlinear interaction process and also implies that an arbitrary phase mask for pulse shaping purposes can be implemented. A more thorough discussion on this can be found in [125].

Fig. 5.2(a) shows several ferroelectric domains of the 2D-QPM device. For the design, an initial phase function \( \phi_{\text{QPM}}(x, 0) \) as shown in Fig. 5.2(b) was chosen. It is important to note that this domains do not have any discontinuities but exhibit a significant curvature. This feature is in strong contrast to conventional QPM devices utilizing multiple separate gratings for discrete tuning [126], or straight but fanned ferroelectric domains for continuous tuning [127].

Figure 5.2(c) shows an image taken from an inspection of the fabricated domain profile over the entire width. This confirms that these domain profiles can be successfully fabricated with high quality. There are no noticeable errors over the entire 1 mm-thick, 25 mm-wide and 12 mm-long MgOLN crystal. The figure is stretched along the longitudinal direction in order to make the individual curved ferroelectric domains visible.

### 5.2 Frequency domain parametric interactions in 2D-QPM-media

In this section the parametric process occurring in the 2D-QPM device is explained. Furthermore, it is shown how these processes can be affected and controlled by different system parameters. For this purpose a continuous-wave simulation involving the signal and the pump is shown in Fig. 5.3(a); however the simulation model includes the longitudinal vari-
5. Broadband frequency-domain OPA

ation of the interaction length of the grating. The simulation illustrates the exponential parametric amplification, followed by pump depletion and an intended rapid change of the QPM period in order to terminate the nonlinear interaction. It is shown for arbitrary parameters in a normalized fashion. The capability to control, to modify or to turn off the nonlinear interaction is unique to structured QPM devices and underlines the great potential and flexibility. In fact, such a simulation can also be performed in a frequency-dependent manner and is presented later in this section. Fig. 5.3(a) illustrates the fact that by turning off the interaction after the relevant effective length, the onset of back-conversion of energy from the signal to the pump is suppressed. Although frequency-dependent modifications can be achieved in conventional longitudinally-chirped QPM devices using single or multiple gratings [128, 43, 99], the performance of these devices is ultimately limited by coupling between different parts of the spectrum [56]. In the FOPA concept, the spatial chirp introduced by the 4f pulse shaper arrangement decouples the different spectral components more robustly and therefore enables a greater flexibility.

A better understanding of the extent of the decoupling can be gained when the complete spatiotemporal profile of real pulsed beams is considered. For this purpose it is necessary to get the relation between the spatial profile at the input of the diffraction grating of the 4f pulse shaper and the temporal profile in the Fourier plane of the FOPA. This relation can be expressed in terms of the beam and geometrical parameters as

$$\tau_{\text{eff}}(\lambda) \approx \frac{\lambda}{c} \frac{\Delta x}{\Delta \lambda} \frac{w_{\text{in}}(\lambda)}{f},$$

(5.2)

where $\Delta \lambda$ is the FWHM range of wavelengths contained by the pulse, $\Delta x$ the spatial extent of that wavelength range and $f$ the focal length of the 4f pulse shaper. The effective pulse duration of the spatially-chirped signal pulse $\tau_{\text{eff}}$ is a crucial system design parameter and has to be compared to the pump duration in order to estimate over the photon conversion efficiency. The conversion efficiency could suffer from a too small effective seed pulse duration $\tau_{\text{eff}}$ compared to the pump pulse duration.

For the purpose of visualizing the amplification process governed by the nonlinear dynamics, a numerical model for nonlinear mixing processes in 2D-QPM media was developed. The result, which shows the
input and the output of a full spatiotemporal simulation, is plotted in Fig. 5.3(c). It accounts for the propagation coordinate $z$, the transverse dimension $x$ and time $t$, representing a (2+1D) simulation. The pump at the input of the 2D-QPM FOPA (dashed red) has a pulse duration of 14 ps FWHM and the effective input signal duration $\approx 5.2$ ps FWHM (blue dashed). The simulation indicates that the region overlapping with the signal experiences strong depletion, but the significantly shorter signal duration in this example prevents the temporal wings from depletion. The impact on pump depletion can also be seen by integrating over the time coordinate as shown in Fig. 5.3(b). However, complete pump depletion does not occur. Moreover the impact of a fully 2D-QPM pattern is shown in Fig. 5.3(d) showing simulated output spectra for three different cases. The dashed blue line represents the input signal spectrum whereas the solid blue line corresponds to the fully amplified spectrum obtained from a 2D-QPM device. For the case of a fanout grating (solid red line in Fig. 5.3(d)), where the QPM period is varied transversely to the beam propagation direction but does not have a longitudinal variation: a substantial reduction of the bandwidth is observed. The bandwidth for a conventional periodic QPM grating is drastically reduced, since the constant grating wave vector $K_g$ can only perfectly phase match one single corresponding wavelength.

As mentioned above, another unique feature of these two dimensionally patterned QPM devices beyond the amplification capability, is to act as an arbitrary phase mask allowing for simultaneous gain and pulse shaping. Thereby pulse shaping is achieved by imparting the QPM phase $\varphi_{QPM}$ from Fig. 5.1(b) or any other arbitrary phase profile (within the fabrication capabilities) to the generated idler. Then the idler spectral phase can be approximated by

$$\varphi_i(\nu) \propto -\varphi_s(\nu_p - \nu) + \varphi_{QPM}(x_i(\nu), 0) - k_i(\nu)L,$$  \hspace{1cm} (5.3)

where $k_i(\nu)$ is the idler wave vector, $\nu$ the optical frequency and $\varphi_j(\nu)$ is the spectral phase of wave $j \in \{i, p, s\}$. The derivation and a more general expression of $\varphi_i(\nu)$ can be found in [125].
Figure 5.3: Modeling of frequency domain optical parametric amplification FOPA in a 2D-QPM medium. (a): Plane and continuous-wave interaction in a longitudinally-varying QPM grating, showing the procedure used to switch off the parametric amplification after a certain distance through the crystal. (b): A series of simulations like in (a), showing the evolution of the pump as a function of transverse and longitudinal position. At each transverse position, a separate plane and continuous wave simulation is performed, with the pump intensity and effective length according to Fig. 5.1(c). In comparison to (a), longitudinal position is shown in a non-normalized fashion. (c): Full spatiotemporal simulation of the FOPA process. The figure shows the output electric field envelopes of the pump and signal for the transverse position $x = 0$. (d): Output signal spectra for three cases: the 2D-QPM grating pattern introduced here; a simpler fanout pattern with no longitudinal variation; and the simplest case of a standard periodic grating. Adapted from [125].

5.3 Experimental setup and results

In this section, the experimental setup and the results achieved by the employment of this new technique are presented. For demonstration purposes, a FOPA based on a monolithic 2D-QPM device as the amplification medium in the Fourier plane was developed as a final stage of our few-cycle mid-IR OPCPA [31, 32]. A schematic of the setup is shown in Fig. 5.4. It consists of reflective ruled diffraction gratings with 75 lines/mm, designed for a blaze wavelength of 4 µm. The focal length used for the 4f arrangement is $f = 200$ mm. The pump beam is shaped by spherical and
5.3. Experimental setup and results

Figure 5.4: Schematic of experimental 2D-QPM-FOPA setup. The mid-IR idler generated from the preamplifiers (described in chapter 3 and 4) acts as the seed for the FOPA.

cylindrical telescopes in order to achieve an elliptical pump beam and is thereafter collinearly overlapped with the spatially dispersed mid-IR idler seed by a dichroic mirror. This dichroic is reflective for the pump and transmissive for the mid-IR idler seed. After the 2D-QPM crystal another dichroic mirror removes the pump and transmits the amplified mid-IR idler. The pump laser operates at a repetition rate of 50 kHz and has an average power of 16.5 W and a pulse duration of $\approx 14$ ps FWHM. A $1/e^2$ full-width beam size of 27.4 mm in the horizontal and 140 $\mu$m in the vertical direction is used. The mid-IR idler seed has an average power of 6.4 mW before the first diffraction grating at the input of the FOPA. At the Fourier plane, e.g. at distance $2f$ after the first diffraction grating, the spectral components of the mid-IR idler seed are spatially chirped according to Fig. 5.1(a). It is important to note that for efficient energy transfer from the pump to the idler, a cylindrical telescope prior the 4f arrangement is employed in order to improve the temporal overlap between the pump and mid-IR idler seed pulse, according to Eq. (5.2). With this telescope a relatively large $1/e^2$ full-width beam size of $\approx 12$ mm in the horizontal extension is obtained for the mid-IR idler seed. The measured spectra are shown in Fig. 5.6. A compressed output power of 1.03 W was measured after the second diffraction grating, which corresponds to 20.6 $\mu$J at 50 kHz repetition rate. When accounting for the losses from the diffraction grating ($\approx 31\%$), the dichroic mirror to remove the pump ($\approx 5\%$) and also the beam routing optics, the average power was estimated to be $\approx 1.65$ W directly after the AR-coated 2D-QPM device at the Fourier plane. This power value corresponds to 33 $\mu$J of pulse energy. When calculating the
Figure 5.5: (a): Measured mid-IR idler spectrum after the 4f arrangement without amplification. This represents the spectrum of the seed at the input of the 4f arrangement. (b): Amplified spectrum after the FOPA arrangement. The seed spectrum is scaled arbitrarily in order to make it visible in comparison to the amplified spectrum. Adapted from [125].

ratio of the photon fluxes of the generated mid-IR photons at 3.4 µm and the pump photons at 1.064 µm, e.g. the quantum conversion efficiency, then a photon conversion of 32% is obtained. This value in fact represents a substantial improvement over the previous OPCPA configuration which is based on noncollinear power amplification in a conventional PPLN covered in chapter 4 and [32].

Next, the OPG background was checked by operating the amplifier in an unseeded configuration, e.g. blocking the seed of the 2D-QPM FOPA. The result of this measurement is plotted in Fig. 5.5(b) and confirms that there is negligible OPG background in the output beam.

For the purpose of pulse compression, the dispersion of the 1.55 µm-preamplifier pulses is adjusted in the OPCPA front-end, such that the mid-IR idler pulses are compressed at the output of the 2D-QPM FOPA. The compressed pulses are characterized with SHG-FROG and the results are shown in Fig. 5.6. The measured and retrieved spectrograms (Fig. 5.6(a) and (b)) show good agreement which is also confirmed by the low FROG-error of 0.005 for a grid size of 512 × 512 points. The reconstructed spectrum together with an independently measured spectrum and spectral phase is plotted in Fig. 5.6(c). The spectra also show good agreement. Furthermore, the reconstructed temporal pulse profile is shown in Fig. 5.6(d), showing a compressed pulse duration of 53 fs FWHM. This duration is mainly limited by the available seed-bandwidth (43 fs transform-limit, corresponding to four optical cycles). The intensity fluctuations on the spec-
tra are likely a consequence of the spectral broadening of the 1.55 μm seed in the OPCPA front-end.

Regarding the photon conversion efficiency, it already exceeds the ones achieved from state-of-the-art mid-IR OPCPA systems [30, 52, 53, 32]. However, the estimated short FWHM seed pulse duration of \( \approx 6.35 \text{ ps} \) in the Fourier plane compared to the 14-ps pump pulses limits the achievable conversion efficiency [125]. In particular, this seed pulse duration can be estimated by inserting the FWHM beam size \( w_{\text{in}}(\lambda) \) into Eq. (5.2).

Figure 5.2(c) shows the domain poling quality for a certain region of the fabricated 2D-QPM device. There is no evidence for fabrication errors. Thus the conclusion is that QPM gratings with significantly larger widths up to 60 mm are feasible in terms of fabrication. Ultimately the main limitation is the available wafer width. Larger widths help to scale \( \Delta x \) accordingly and yield a corresponding increase in seed pulse duration as indicated by Eq. (5.2). Potentially this can lead to a further improvement in terms of conversion efficiency.

All in all, the obtained results demonstrate and show the promise of

![Figure 5.6](image-url)

*Figure 5.6:* (a) Measured spectrogram from SHG-FROG characterization. (b) Retrieved spectrogram. (c) Measured and retrieved spectra and spectral phase and (d) retrieved temporal intensity profile with temporal phase showing a pulse duration of 53 fs (43 fs transform-limit). The inset shows the beam profile of the compressed pulse after the second diffraction grating at the output of the 2D-QPM FOPA. Adapted from [125].
the 2D-QPM FOPA technique, and pave the way to the new paradigm of 2D-QPM frequency domain nonlinear optics.

5.4 Summary and conclusions

In summary, a new platform for the purpose of frequency-domain nonlinear optics has been demonstrated. It combines spatially-chirped seed pulses with a two-dimensionally patterned QPM crystal (2D-QPM). The unprecedented flexibility overcomes limitations inherent in conventional BPM and QPM devices. The capability for high fidelity fabrication of 2D-QPM devices with curved domains throughout the full width has been shown. These devices allow for scaling in bandwidth, even for extremely broad bandwidths, by matching the QPM period trajectory according to the spatially-chirped input wave. In fact, this approach is applicable to a wide range of nonlinear optical processes such as harmonic generation and OPA.

A 2D-QPM FOPA consists of a continuum of narrow-band OPAs across the transverse direction of the nonlinear crystal and therefore offers the freedom to adjust the interactions across the seed spectrum via the QPM pattern. This is in strong contrast to OPA performed in conventional devices. In addition to the transverse variation, we introduced a longitudinal variation in the QPM pattern to compensate for the transverse spatial Gaussian pump beam shape (non-flat top beam profile) and flatten the gain experienced by spectral components at different transverse positions. Moreover, a wide variety of other longitudinal variations are possible in these devices, for example introducing a longitudinal chirp in the QPM period, or adding a second QPM segment for additional functionalities.

A proof-of-principle experiment has been presented by employing a 2D-QPM and demonstrating its capabilities as the amplification medium in the Fourier plane of a broadband mid-IR FOPA for the first time. At the output of the 2D-QPM FOPA a compressed average power of 1.03 W corresponding to a pulse energy of 20.6 µJ at a repetition rate of 50 kHz and a pulse duration 53 fs (43 fs transform-limit) (Fig. 5.3) was achieved. When accounting for linear losses an average power of 1.65 W was estimated at the Fourier plane directly after the 2D-QPM crystal. This average
power corresponds to a quantum conversion efficiency of 32% characterized in terms of photon fluxes and represent a substantial improvement compared to our previous power amplifiers [31, 32]. Moreover, it has been shown that the FOPA arrangement also helps to avoid parametric superfluorescence (OPG) as shown in Fig. 5.2(b).

In conclusion, the results demonstrated by this proof-of-principle experiment together with the capability of bandwidth scaling are very promising for future high-repetition rate, few-cycle pulse light sources. This feature together with the slab-like geometry of the FOPA, supporting a one-dimensional heat flow and therefore making it power scalable, make it also very promising for high average-power light sources for the purpose of attosecond science.
Chapter 6

Applications in strong-field physics

The availability of intense laser fields operating over a wide range of wavelengths in combination with few-cycle pulse durations has paved the way for a new domain among studies of light-matter interactions. The field strengths achievable with these lasers are comparable to the Coulomb forces inherent in atomic systems. Exposing these systems to intense laser fields alters the conditions and therefore can lead to the occurrence of phenomena, such as strong-field ionization [75, 74]. Probing these phenomena could reveal some new insights and lead to a deeper understanding of extreme light-matter interactions. For this purpose a state-of-the-art, few-cycle mid-IR OPCPA operating at 3.4 μm has been developed within the framework of this thesis [31, 56, 32].

The present chapter discusses the results which have been obtained from strong-field ionization experiments. These experiments were performed using few-cycle mid-IR pulses delivered from our OPCPA-system. Due to the fairly long central laser wavelength of 3.4 μm and the high peak intensities, we were able to perform experiments close to the region where the breakdown of the dipole approximation in strong-field ionization is predicted. Under the assumption of the dipole approximation, magnetic field effects are neglected in all considerations of strong-field ionization, which has already been introduced in section 2.2.2 of chapter 2. The influence of the magnetic field component of the laser pulses acting onto the electrons during the process of strong-field ionization was probed by recording complete two-dimensional photoelectron momentum distribu-
Figure 6.1: Intensity-wavelength parameter space in strong-field ionization. The green-dotted area indicates the parameter space where the dipole-approximation is considered as valid and is referred to as the “dipole oasis”. The short wavelength limit of the dipole approximation is indicated by the violet line “upper dipole limit”. The long wavelength limit arises when the onset of magnetic field effects become evident. This is indicated by the brown-dashed line “magnetic displacement”. The red triangles indicate the parameter space used for the experiment discussed in the current chapter 6. These parameters are close to the theoretically predicted long wavelength limit of the dipole approximation. Adapted from [36].

6.1 Experimental setup

For the purpose of the investigation of strong-field ionization, a mid-IR few-cycle OPCPA has been developed as described in chapters 3 and 4. This system delivers few-cycle pulses with a pulse duration of 44 fs corresponding to sub-four optical cycles, a pulse energy of 21.8 µJ at a repetition rate of 50 kHz, corresponding to an average power of 1.09 W. The output of this laser is linearly polarized in the horizontal plane by the help of a broadband HWP and afterwards guided into a velocity-map imaging spectrometer VMIS [129, 130, 131]. The laser beam is focused into the interaction region by a dielectric mirror in a back-focusing geometry and with a focal length of 15 mm. The photoelectrons originating from strong-field ionization are mapped by electrostatic lenses onto a microchannel plate and are thereafter imaged by a phosphorous screen.
6.2 Results and discussion

Full 2D-PMDs were recorded from the noble gases xenon, argon, neon and helium for intensities ranging from $2 \times 10^{13}$ W/cm$^2$ to $8 \times 10^{13}$ W/cm$^2$. The recorded PMDs show an asymmetry along the beam propagation axis, i.e. the z-axis, with respect to the reference, which is the center spot as indicated in Fig. 6.3(a). This center spot corresponds to low-energy electrons being field ionized by the spectrometer’s electrostatic field. They come from highly excited states, which populated by the interaction with the laser pulse [83, 132, 133]. The static electric field applied in the spectrometer for imaging purposes, ranges from 0.5 to 1 kV/cm and therefore can field ionize excited states with a binding energy corresponding to a principal quantum number of $n = 21$ or higher [134]. In fact, these elec-
trons do not interact with the laser pulse and do not gain kinetic energy in the detector plane. Consequently these electrons can serve as reference for zero momentum of the photoelectrons [83].

Afterwards, the recorded PMDs are divided into three regions as shown in Fig. 6.2(a). The three distinct regions are individually projected onto the beam propagation direction, i.e. z-direction. Thereafter, the peak offset is extracted by comparing with the peak from the projection of the central part, representing the reference. The central region consists of a slice of width $\Delta p_x = 0.05$ a.u. to isolate the reference spot corresponding to zero momentum, e.g. $\Delta p_z = 0$ a.u.. Then the projections of the two outer regions exhibit an offset in the peak along the beam propagation direction as shown in Fig. 6.3(b). Moreover, this offset is recorded as a function of the applied laser intensity. For the method of quantifying the peak offset, the error was estimated to be in the order of a camera pixel size.

The applied laser intensities were first calibrated by the width of the PMDs obtained from the measurements for the case of circularly polarized light and second via the longitudinal width of the PMDs obtained from semi-classical calculations, which are going to be described in a subsequent section. This procedure was chosen in order to exclude any influence from interferences which can occur for the case of linearly polarized light.

For the purpose of excluding the possibility that the observations are introduced by an experimental artifact, PMDs in the same geometry were

Figure 6.3: (a): Typical projection of a 2D-PMD of xenon for an intensity of $6 \times 10^{13}$ W/cm$^2$ obtained by the use of a VMIS and linearly polarized few-cycle pulses at a center wavelength of 3.4 $\mu$m. Adapted from [36].

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recorded at a wavelength of 800 nm. The results are shown in Fig. 6.4(d). This measurement was performed in helium at a significantly higher intensity of $1.4 \times 10^{14} \text{W/cm}^2$ than used for the mid-IR-experiments. Nevertheless, the photoelectron image does not show any measurable asymmetry. Therefore, it can be excluded that any experimental artifact is causing the observations at mid-IR wavelengths.

In the following paragraph the procedure used for the classical trajectory Monte Carlo (CTMC) simulations of electrons is explained. These simulations serve for comparison with the experimentally obtained data using a semiclassical two-step model [23, 25, 135, 136]. The initial conditions for the simulations originate from the tunnel exit calculated in parabolic coordinates [137, 138], the ionization rate and initial momentum distribution from Ammosov-Delone-Krainov (ADK)-theory [139, 140]. Although theoretical models for the description of tunneling ionization beyond the dipole approximation exist [141, 142, 143], the validity of these models providing the initial conditions for the CTMC simulations was questioned [79, 81]. Thus, the simulations were tested on their robustness against variations of the spatial starting point for the propagation and also against variations in the ionization rate beyond the change expected for relativistic tunneling. For this purpose the exit point was varied in between 60% and 300% and the ionization rate by at least 5 orders of magnitude. In particular, the outcome of these simulations was very robust against these variations therefore any significant influence due to deviations from the initial conditions can be excluded. Furthermore, the robustness of the simulations was also verified by checking for differences in the simulations, potentially being induced by the geometry or the dynamics of the ionization step for different gas species. Although the first atomic ionization potential considerably varies, the same trend in offset was observed for all the different species. This strongly suggests that the observation of the asymmetry in the PMDs is mainly governed by the propagation of the liberated electrons under the influence of the combined Coulomb and laser field.

The applied model fully includes the magnetic field component of the laser pulse and the Coulomb field of the residual ion during the propagation. For each electron trajectory the simulation is performed until the
Figure 6.4: Photoelectron momentum distributions (2D-PMDs) recorded for xenon and helium and their projections onto the beam propagation direction. The upper figures represent measured and simulated data. (a): Experimentally obtained PMDs of xenon for the intensities $3 \times 10^{13}$ and $6 \times 10^{13}$ W/cm$^2$. (b) Corresponding simulated PMDs at the intensities as the ones used for the experiment in (a). The negative offset is reproduced through the combined influence of the Coulomb potential of the residual ion and the full electromagnetic laser field. (c): Simulated PMD excluding the magnetic field component of the laser field. The projection does not exhibit an offset. (d): Measured PMD of helium at a wavelength of 800 nm and an intensity of $1.4 \times 10^{14}$ W/cm$^2$. For the shorter wavelength the dipole approximation is valid and thus no offset is visible. Adapted from [36].

end of the laser pulse and the final asymptotic momenta are calculated by the use of Kepler’s analytical formula [144, 145]. However, numerical problems potentially occur in connection with the $1/r$-Coulomb potential. To avoid this, electrons which come closer than 0.5 a.u. to the parent ion are filtered out in the simulations. This affects only a small number of electrons, e.g. 0.25% of the trajectories. It has also been verified that the filtering process does not have any impact to the outcome of the simulations.

For each laser intensity ranging from $2 \times 10^{13}$ W/cm$^2$ to $1 \times 10^{14}$ W/cm$^2$, $10^6$ trajectories are calculated and thereafter binned in the momentum space with a bin size of $10^{-3}$ a.u.

In accordance to the experiment, the maximum of the simulated PMDs is projected onto the beam propagation direction and identified by Lorentzian fits of the central part of the distributions. In all the PMD-simulations the central spot is absent since field ionization of highly excited Rydberg
states by the spectrometer field is not included in the simulations. For the simulations, the reference for zero momentum is anyway intrinsically known. Furthermore, the error for peak identification is estimated from the bin size used for the simulations.

The extracted peak offset of the PMDs show a clear trend with respect to intensity for the experimental and theoretical data points, particularly an increasing offset for increasing intensities for all the different target gases can be observed. This feature is also directly visible in the PMD shown in Fig. 6.4(a). Surprisingly, the offset is shifted towards negative values, i.e. opposite to the beam propagation direction. This observation seems to be counterintuitive as it is in contradiction to the behavior one would expect for a free electron, without the influence of the Coulomb potential, because the behavior of a free electron is expected to be governed by the radiation pressure that is exerted onto it by the Lorentz force. This picture was also used by Smeenk et al. in [83], for the interpretation of an observed shift of the peak in the PMD into direction of beam propagation. In contrast, the present experiments show that the behavior for the case of linearly polarized light is altered by the influence of the combined action of the magnetic field and the Coulomb potential. In the present case the electron can be driven back to the ion core by the laser field and thereafter can interact with the residual ion’s Coulomb potential [23, 25].

The experimental observations are compared with the CTMC calculations taking into consideration both the magnetic field of the laser pulse and the Coulomb potential of the residual ion. There is good agreement between the calculations and the experimental data as shown in Fig. 6.4(b). The influence of the magnetic field is also confirmed by the calculations; the asymmetry along the beam propagation direction vanishes when the magnetic field is neglected in the calculations.

Figure 6.5 shows the extracted peak offsets from the experiments for different target gases together with the ones from the calculations. An increase in momentum offset for increasing laser intensity can be observed. Moreover, excellent agreement between experiment and calculations was observed. A simple intuitive explanation for these observations could be that one might think of electrons being pushed in a first step into the beam propagation direction by the magnetic field component of the Lorentz
Possible momentum transfer of the order $I_p/c$ onto either the ionized electron, the ion or the electron-ion system was recently discussed in [141, 142]. In accordance to these studies, a momentum kick of the electron into the $z$-direction was included in the simulations and the results were compared with the ones without initial momentum kick. It was found that the measurements for the case of linearly polarized light are not sensitive enough to resolve consequent signatures in the photoelectron momentum spectra.

### 6.3 Summary and conclusion

In summary, few-cycle pulses delivered from a state-of-the-art mid-IR OPCPA were used to perform strong-field ionization of the different noble gases xenon, argon, neon and helium in a VMIS. The laser pulses had a pulse energy of 21.8 µJ at a repetition rate of 50 kHz and a pulse duration of 44 fs at a center wavelength of 3.4 µm and were focused to intensities in the order of $2 \times 10^{13}$ W/cm$^2$ to $8 \times 10^{13}$ W/cm$^2$. The recorded PMDs showed an asymmetry along the beam propagation direction. They were
divided into three regions and individually projected on to the beam propagation direction. The peak offset from the two outer parts were extracted by comparing with the peak of the inner part, which served as reference. The peak offsets were recorded as a function of the laser peak intensity and an increasing offset for increasing intensity was observed. Semi-classical calculations were in good agreement with experimental data and also confirmed the same trend for all the different gas species. When the magnetic field of the laser pulse was omitted in the calculations, no offset was evident anymore. In fact, these observations represent the breakdown of the dipole approximation in strong-field ionization in the long wavelength limit at moderate intensities. The results show that the electron dynamics is significantly influenced by the magnetic field component of the laser field and by the Coulomb potential of the parent ion, respectively.

The fact that the Coulomb potential of the parent ion also plays an important role for the electron dynamics for the case of linearly polarized light, challenges the previously used radiation pressure picture by Smeenk et al. [83]. Thus, the influence of non-dipole effects has to be considered in theoretical concepts describing the physics beyond the dipole approximation [146, 147]. Moreover, the observations are rather induced during the quasi-classical dynamics in the continuum than during the ionization step and therefore a direct insight into the physics of the initial ionization step is obstructed. This fact was concluded since the simulations are robust against any variation to the starting conditions. Consequently the presented results pose new challenges for the theoretical description of strong-field processes driven by long wavelength light sources in the limit or beyond the dipole approximation.
Chapter 7

Conclusion and Outlook

Energetic few-cycle pulses are a necessary prerequisite for experiments in attosecond and strong-field physics. All these experiments rely on the process of strong-field ionization, which requires peak intensities in the order of $10^{13} \text{ W/cm}^2 \sim 10^{15} \text{ W/cm}^2$. Furthermore, recent developments in attosecond science and strong-field physics clearly point towards long wavelength driving lasers due to their capabilities for generating soft x-ray higher-order harmonic (HH) radiation. Optical parametric chirped-pulse amplification (OPCPA) is ideally suited for transferring energy from one spectral region to another via parametric frequency down conversion, broadband amplification and the generation of few-cycle pulses, leading to the desired peak intensities in the order of $10^{13} \text{ W/cm}^2 \sim 10^{15} \text{ W/cm}^2$. In this thesis the development of an OPCPA-based long wavelength driving light source for the purpose of strong-field experiments has been presented. In the following paragraphs, the main achievements are summarized and an outlook of potential future directions is given.

The choice of nonlinear crystal plays a central role when designing an OPCPA system. It is of considerable importance that its optical properties are favorable in terms of transparency and phase matching capabilities for the targeted wavelength range. For mid-infrared (mid-IR) wavelength generation, LiNbO$_3$ is a suitable candidate [118]. Periodic poling of its ferroelectric domains enables collinear quasi phase matching (QPM) among the involved waves. However, perfect phase matching can only be
obtained for one certain set of wavelengths, i.e. pump, signal and idler wavelengths. Such crystals are referred to as periodically poled lithium niobate (PPLN) devices. Moreover, when a chirp is applied to the poling period, phase matching for different spectral components can be achieved at different positions within the crystal. These crystals are then referred to as aperiodically poled lithium niobate (APPLN) devices. The capabilities of such PPLN and APPLN crystals, for the purpose of mid-IR few-cycle pulse generation by means of OPCPA, has been studied in this thesis.

The first part of this thesis demonstrates an all-collinear mid-IR OPCPA, exclusively employing APPLN devices as nonlinear crystals throughout the whole system. The system consists of three stages, two preamplifiers and one power amplifier. This includes in a first step to externally broaden the seed spectrum centered at 1.56 µm in order to support shorter pulse durations. The next step is about dispersion precompensation so that the mid-IR idler pulses at the output of the three-stage amplifier can be compressed via bulk propagation. For this purpose the seed pulses are sent through a 4f pulse shaper and a subsequently arranged pair of silicon prisms. At this stage, the seed pulses are sent to the first preamplifier. The amplified signal is guided to the second preamplifier stage which uses as pump the power transmitted from the first preamplifier. The idler is discarded and the amplified signal is used as seed for the final power amplifier. Parts from the primary 1.064 µm pump light are further amplified in a home-built Innoslab amplifier and used as the pump for the power amplifier. Finally, the generated idler is extracted and compressed via bulk-propagation through a 50 mm long anti-reflection (AR) coated sapphire rod. The care taken to redesign the OPCPA system in terms of the seeding and amplification stages resulted in a sub-four-cycle pulse duration of 41.6 fs (one optical cycle is ≈ 11.3 fs at a center wavelength of 3.4 µm). Furthermore a pulse energy of 12 µJ was achieved, corresponding to an average power of 600 mW at a repetition rate of 50 kHz. The 41.6 fs-pulses represent a record short pulse duration from a high repetition-rate OPCPA at 3.4 µm central wavelength. Additionally, good internal conversion efficiency of 24.5% was achieved and the optical parametric generation (OPG) background was minimized [31].
Despite the short pulse durations of 41.6 fs obtained from the APPLN-based all-collinear OPCPA, the yield of photoelectrons from the examination of strong-field ionization was too low for the targeted experiments. Therefore, we implemented modifications in our system to increase the pulse energy of our mid-IR few-cycle light source. A noncollinear, achromatic QPM power amplifier was investigated in this goal and demonstrated for the first time in a proof-of-principle experiment. In contrast to the previous power amplifier, here a conventional PPLN is employed with a seed and pump interacting in a noncollinear fashion. Furthermore, idler seeding instead of signal seeding is employed. At the output after bulk compression, pulse energies of 21.8 µJ together with pulse durations of 44.2 fs were achieved. This corresponds to an average power of 1.09 W at a repetition rate of 50 kHz. While improving the pulse energy by more than 80% over the previous result, also the OPG background was greatly reduced [32]. This technique offers great flexibility since it can also be combined in future experiments with chirped QPM devices, such as APPLNs, potentially allowing for octave-spanning phase matching bandwidths. With the obtained parameters, the yield of photoelectrons from strong-field ionization was greatly enhanced so that experiments with sufficient photoelectron yield and good statistics were performed.

Before summarizing the results obtained from the strong-field ionization experiments, the demonstration of a new concept for driving nonlinear optics is briefly recapitulated. The new concept takes advantage of the great flexibility of structured QPM devices. The great potential of two-dimensionally structured QPM is shown by means of the demonstration of broadband parametric amplification in the Fourier-plane of a 4f pulse shaper. This technique is referred to as 2D-QPM frequency-domain optical parametric amplification (FOPA). The 2D-QPM device placed in the Fourier plane of a 4f pulse shaper varies the grating wave vector $K_g$ in the transverse direction and also the interaction length in the longitudinal direction at different transverse positions. This enables to phase match individual spectral components of a spatially chirped seed, i.e. the idler centered at 3.4 µm in our case. A 2D-QPM FOPA is constituted by a se-
ries of individual narrowband optical parametric amplifiers (OPAs) along the transverse direction and helps to decouple the amplification process for individual spectral components more robustly, enabling greater flexibility. A proof-of-principle experiment was presented by employing a 2D-QPM and demonstrating its capabilities as the amplification medium in the Fourier plane of a broadband mid-IR FOPA for the first time. Thereby pulses with an energy of 20.6 µJ at a repetition rate of 50 kHz, corresponding to 1.03 W were achieved at the output of the 2D-QPM FOPA. A pulse duration of 53 fs was measured with a bandwidth supporting 42 fs transform-limit (TL). The pulse energy of 20.6 µJ at the output corresponds to an energy of 33 µJ at the Fourier plane and a photon conversion efficiency of 32%. Furthermore, the interaction in the Fourier plane was numerically modeled by a series of plane and continuous wave interactions in a longitudinally-varying QPM and a full spatiotemporal simulation of the FOPA process was performed. With these simulations we gained important insights into the relation between the spatial profile of the seed at the input of the 4f pulse shaper arrangement, the temporal profile in the Fourier plane and the photon conversion efficiency. Due to its great flexibility, 2D-QPM devices can also be used for a variety of other nonlinear processes such as subsequent harmonic generation while still using a monolithic crystal. Furthermore the slab-like geometry of the device supports scaling in power and bandwidth.

Some of the demonstrated light sources in this thesis enabled to drive strong-field physics experiments and shined light on important questions. The dipole approximation is a concept which is widely used for the theoretical description of strong-field phenomena. However, the breakdown of the dipole approximation in strong-field ionization can be expected for driving wavelengths which become comparable to the target size. This limit is also known as the short-wavelength dipole limit. Moreover, there is also a lesser-known limit towards the longer wavelengths which is referred to as the long-wavelength limit. In the goal of investigating this limit in an experiment, linearly polarized mid-IR few-cycle pulses, generated by the OPCPA described in chapter 4, were guided into a velocity-map imaging spectrometer (VMIS) and focused into an interaction re-
region by a dielectric mirror to intensities in the order of $3 \times 10^{13} \text{ W/cm}^2$ to $8 \times 10^{13} \text{ W/cm}^2$. By recording and studying 2D-photoelectron momentum distributions (PMDs) from the noble gases xenon, argon, neon and helium, an asymmetry in the PMDs along the beam propagation direction was observed. It turned out that the outer parts of the distributions were shifted into the opposite of the beam propagation direction with respect to the inner part. Classical trajectory Monte Carlo (CTMC) simulations confirmed these observation only when the magnetic field of the laser pulse and the Coulomb potential of the residual ion was included in the model. When the magnetic field was neglected, no asymmetry was observable. These experimental results together with the semiclassical calculations, in fact represent the observation of the breakdown of the dipole approximation in strong-field ionization in the long-wavelength limit. Consequently, these observations pose new challenges for the theoretical description of strong-field processes beyond the dipole approximation, for example the generation of HHs with long wavelength driving lasers [36].

Future directions in the development of short- and long wavelength driving light sources build on the great flexibility of OPA/OPCPA. However, the trend is pointing towards a master-oscillator approach accompanied by high repetition rate OPCPA. In a master-oscillator approach the seed and the pump, which are used for the operation of a subsequent amplifier, are derived from one and the same oscillator. Such an approach could be implemented by taking first one part of the output power from a high-power thin-disk laser [148, 149] or a high-power fiber laser [150] for supercontinuum generation (SCG). Second, the residual power is used as pump for the amplification of the targeted spectral range of the supercontinuum (SC) by an OPA/OPCPA [151]. An alternative approach is to use titanium-doped sapphire (Ti:sapphire) oscillators with an additional output at 1030 nm besides the standard 800 nm output. Due to the broad emission cross section of Ti:sapphire, no further SCG or external spectral broadening is necessary in order to support few-cycle pulse durations. The 1030 nm output is then further amplified by regenerative [152] or Innoslab-amplifiers [104] and thereafter acting as pump for OPCPA [29]. Furthermore, a master-oscillator approach comes along with the
great advantage of passive carrier envelope phase (CEP) stability when difference-frequency generation (DFG)/optical parametric amplification (OPA) schemes are used for mid-IR generation [153]. Complex yet promising is also the coherent combination of multiple OPA/OPCPA channels operated in different spectral regions resulting in octave-spanning synthesized light transients [154, 155].

Wide-aperture QPM devices have become available [55], allowing QPM technology to support very high energies. The above mentioned schemes together with the bandwidth and energy scaling capabilities of wide-aperture, conventional and chirped QPM devices (PPLNs, APPLNs and twodimensionally structured QPM devices), are a very promising combination for the generation of energetic mid-IR few-cycle or even single-cycle pulses. Such light sources will open up the way for table-top soft x-ray light sources with attoseconds of pulse duration. Soft x-ray attosecond pulses will allow a new class of experiments in and beyond the water window including multidimensional microscopy on the atomic level [156].
Literature

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