Nanosecond tandem optical parametric oscillators for mid-infrared generation

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Licentiate Thesis

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Abstract

This thesis discusses a new scheme for generating radiation in the mid infrared spectral region, especially the 3.5-5 µm range. The scheme uses established Nd³⁺-lasers at 1.06 µm and down conversion in nonlinear optical crystals. The down conversion is made by two optical parametric oscillators (OPO) in series. The second OPO is a classical OPO using a zink germanium phosphide (ZGP) crystal. ZGP is the best nonlinear material available for the 4-8 µm spectral range, but it is absorbing below 2 µm. The new development presented in this thesis is the OPO used to convert the 1.06 µm laser radiation to a suitable OPO pump near 2 µm.

The OPO uses a type I quasi phase-matched crystal, which accesses high nonlinearities and avoids walk-off. The problem with type I OPOs close to degeneracy is the broad bandwidth of the generated radiation, which reduces the efficiency of a second OPO. This has been solved with a spectrally selective cavity using a volume Bragg grating output coupler. Unlike other bandwidth limiting schemes this introduces no intracavity losses and thus efficient OPO operation is achievable.

Narrow linewidth (~0.5 nm) OPO operation has been achieved with periodically poled LiNbO₃ (PPLN) and periodically poled KTiOPO₄ (PPKTP) while locking the signal wavelength at 2008 nm and simultaneously generating an idler at 2264 nm. A high average power PPLN OPO with 36 % conversion efficiency and 47 % slope efficiency is reported. Operation very close to degeneracy at 2128 nm with the narrowband signal and idler peaks separated by 0.6 nm was demonstrated in a PPKTP OPO. Both the signal at 2008 nm and the combined signal and idler around 2128 nm from the PPKTP OPOs have been used to show efficient pumping of a ZGP OPO. The maximum conversion efficiency from 1 µm to the mid-IR demonstrated is 7 % with a slope efficiency of 10 %. This is not quite as high as what has been presented by other authors, but the experiments reported here have not shown the optimum efficiency of the new scheme. Relatively simple improvements are expected to give a significant increase in conversion efficiency.
Preface

The studies treated in this thesis have been carried out as a part of my employment at the Department of Laser Systems in the Division of Sensor Technology of the Swedish Defence Research Agency (FOI) located in Linköping, Sweden during 2004 to 2006. Supervision and collaboration has been provided by the Department of Applied Physics at the Royal Institute of Technology, Stockholm, Sweden.

The thesis work was part of research projects sponsored by the Swedish Armed Forces and by internal funding at the Swedish Defence Research Agency.

The thesis contains an introductory part reviewing the basic theory and summarizing the accomplishments and reprints of three scientific papers.
List of publications

Publications included in the thesis


Publications not included in the thesis


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First I would like to thank Professor Fredrik Laurell at KTH for accepting me as a Ph.D. student. I would also like to thank Fredrik and Valdas Pasiskevicius for providing interesting ideas for experiments, explanations for unexpected phenomena and supervision in general.

Further I am very grateful for the support from my local supervisor at FOI, Lars Sjöqvist and from the head of the Department of Laser Systems, Ove Steinvall. Especially Lars has had to bear my frustration and unanswerable questions when the experiments did not work as hoped. He has also had to take the time when I just needed to tell someone about the new results when something finally worked. Acknowledgements should also go to the previous and present heads of the Division of Sensor Technology at FOI, Svante Ödman and Lena Klasén for approving economical support of my studies.

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1 Introduction

1.1 Background

Many different limits have been put on the mid-infrared wavelength region, and the exact figures are not the same everywhere, but often the 2-20 µm range is included. In this thesis the emphasis is on optical parametric oscillators generating radiation in the 3.5-5 µm sub range. This spectral region holds many applications for laser radiation but few practical laser sources are available.

The sources that exist have limitations in performance, operating conditions (such as demanding cryogenic cooling) or are too complicated. A comprehensive review of existing solid state mid-IR laser sources is given in a review book edited by Sorokina and Vodopyanov. The main performance limitations are in output power and in wavelength coverage. The first pulsed mid-IR laser was demonstrated by Sorokin and Stevenson in 1960 based on uranium doped calcium fluoride and operated at 2.6 µm. Laser transitions in the 2-4 µm region were in the following years demonstrated with a number of ions, for example U³⁺, Dy³⁺, Co²⁺, Ni²⁺, Tm³⁺, Ho³⁺ and Er³⁺. Host materials have been various crystals and more recently also fibers. Further advances came with the color center lasers based on vibronic transitions that spanned the 0.8 to 5 µm region. Host materials for long wavelength laser transitions are, however, a problem. Common oxide based host materials have phonon energies that introduce absorption at the interesting wavelengths. Lower phonon energies, for example in fluoride materials, give lower thermal conductivity that limits the performance of the lasers.

Other laser technologies used in the mid-IR include gas lasers, such as He-Ne, HF and DF, that can be used to generate both high energy pulses and continuous wave power. Gas lasers however suffer from practical problems regarding size, corrosion safety and reliability. There are also semiconductor lasers in the mid-IR, both heterojunction laser diodes and quantum cascade lasers. The peak powers from these lasers are very modest, and they generally require cryogenic cooling for efficient operation.

The available lasers do not fulfill all requirements posed on mid-infrared laser sources. Thus the interest turned to nonlinear optics and parametric down-conversion. The first optical parametrical oscillator (OPO) was reported in 1965 by Giordmaine and Miller using a LiNbO₃ crystal, and since then the field has evolved when new and better quality nonlinear crystals and pump lasers have become available. The nanosecond pulse regime is very suitable for nonlinear optics and several groups have shown mid-IR generation by difference frequency generation (DFG) and by OPOs. This is an attractive approach since high quality near infrared lasers that are suitable pump sources for wavelength conversion have been developed for other applications. Most notable are Nd³⁺-lasers operating at 1.06 µm, which are in abundant use for remote sensing and for materials processing. From the visible out to wavelengths of about 3.5 µm pulsed OPOs are now an established technology that is commercially available. Due to properties of available nonlinear optical materials high average power OPOs producing wavelengths longer than 4 µm need to be pumped at wavelengths longer than 2 µm (see section 2.4). Common approaches for generating this 2 µm laser radiation include Ho³⁺-lasers and
OPOs pumped at 1.06 µm. An illustration of the different schemes is given in figure 1-1, while a review of different 2 µm pump sources is given in section 4.1.

![Diagram of different ways to generate mid-IR radiation.](image)

**Figure 1-1.** Illustration of different ways to generate mid-IR radiation.

The main topic of this work is a new OPO scheme for efficient generation of narrowband radiation near 2 µm and subsequent use of this radiation to pump a second OPO generating radiation in the mid-IR. Chapter 1.2 will give a brief overview of the applications and chapter 2 presents the basic theory of nonlinear optics. Chapter 3 contains some important properties of laser beam propagation. In chapter 4 the design and construction of the OPO converting the wavelength from 1 to 2 µm is presented while chapter 5 discusses the mid infrared OPO.
1.2 Applications

The interest in the mid-infrared spectral region is motivated by two contradicting phenomena, low and high absorption. The main gases in the atmosphere (N₂, O₂, H₂O and CO₂) have low absorption in several parts of the mid-infrared region, as shown in figure 1-2, making remote sensing applications possible. On the other hand many trace gas molecules show discrete absorption lines in this region making it suitable for spectroscopy, both remotely through lidars (light detection and ranging)⁶, and in enclosed chambers¹. Using a tunable or broadband laser source the gas species can be determined through the absorption line fingerprint and the concentration can be calculated from the difference in on and off resonance intensity from DIAL (differential absorption lidar) measurements.

![Figure 1-2. Transmission for 10 km propagation through subarctic winter atmosphere as calculated by MODTRAN⁷.](image)

One important application driving the development of high average power sources is directed infrared countermeasures for protection of aircraft⁸. Many small shoulder-fired heat seeking missiles use the atmospheric transmission windows in the 2-5 µm range for homing. In the 1.9-2.6 µm spectral window the radiation is mainly generated by hot metal parts such as the nozzle, whereas in the 3-5 µm range there is also significant radiation from the hot gasses in the exhaust plume emitted from the engine. By directing a properly modulated laser on the missile to dazzle the sensor or create deceptive target signals in the seeker the countermeasure system may cause the missile seeker to loose track of the target. The result may be that the missile starts to wobble and breaks up in the free air, or simply that it passes the target at some distance. Missile threats are significant for military aircraft and helicopters in peace keeping operations. There is also a possibility that civilian aircraft may be targeted by terrorist attacks during take-off and landing.

High absorption in human tissue for several wavelength bands makes laser surgery possible. Photothermal ablation with laser pulses delivered through an optical fiber provides an effective and selective tool for cutting with minimum invasiveness⁹,¹⁰.
Cutting effectiveness varies depending on wavelength, pulse structures and tissue type. So far mainly the water absorption peak at 2.94 µm\textsuperscript{11} has been targeted by the use of Er:YAG lasers, but access to suitable sources at arbitrary wavelengths should increase the efficiency and selectivity, reducing collateral tissue damage. Especially the wavelength 6.45 µm, where protein absorption is stronger than the still high water absorption\textsuperscript{12}, is drawing interest as a way to reduce collateral damage. Experiments have been performed with a free-electron laser and with a nanosecond OPO source\textsuperscript{13}. There may also be wavelengths that are absorbed selectively in specific kinds of tissue, for example fat, but to a lesser degree in the body in general.
2 Nonlinear optics for mid-IR generation

The field of physics that describes propagation of electromagnetic radiation under conditions where the amplitude of the field influences the propagation is called nonlinear optics. This chapter contains a short introduction to a small part of the field that is relevant for this thesis. For a more comprehensive review textbooks are recommended\cite{14, 15}.

2.1 Nonlinear optics

Electromagnetic radiation passing through a crystal will apply a force on the electrons and ions in the material. The force causes primarily the electrons to oscillate and induces a time-varying electronic polarization in the material. For small electric field strength the material response in the frequency plane can be linearized

\[ P_L = \varepsilon_0 \chi^{(1)} E, \quad (2.1) \]

where \( \varepsilon_0 \) is the permittivity of vacuum, \( E \) is the electric field and \( \chi^{(1)} \) is the linear susceptibility, which is described by a second-rank tensor. For larger field strengths, for example when the field is a high intensity laser beam, nonlinear contributions to the electronic polarization have to be taken into account. The total induced polarization can then be described by

\[ P = \varepsilon_0 \chi^{(1)} E + \varepsilon_0 \left( \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots \right) = P^L + P^{NL}, \quad (2.2) \]

where \( E \) is the electric field and \( \chi^{(m)} \) is the \( m \):th order susceptibility tensor with rank \( m+1 \). An illustration showing the difference between linear and nonlinear polarization responses is given in figure 2-1. Susceptibility tensors of even orders have non-zero components only in non-centrosymmetrical materials, so the second order nonlinearities that are the strongest are present only in certain crystals. Third order nonlinearities are, on the other hand, available in all materials, even in air.

![Figure 2-1](image)

**Figure 2-1.** In the case of linear response the polarization will be proportional to the driving field (left), whereas when the material response is nonlinear the polarization will be distorted and contain contributions at harmonic frequencies.

The second order susceptibility \( \chi^{(2)} \) is a third order tensor and thus has rank 3 and 27 components. If all involved frequencies are far away from resonances in the material and the loss is neglected permutation or Kleinman symmetry holds. The second order
susceptibility can then be contracted to a more easily handled $3 \times 6$ matrix, called $d$, according to

\[
\begin{bmatrix}
    \begin{bmatrix}
        p_{\alpha\beta}^{(2)}
    \end{bmatrix}_x \\
    \begin{bmatrix}
        p_{\alpha\beta}^{(2)}
    \end{bmatrix}_y \\
    \begin{bmatrix}
        p_{\alpha\beta}^{(2)}
    \end{bmatrix}_z
\end{bmatrix} = 2\varepsilon_0 K \begin{bmatrix}
    d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\
    d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\
    d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36}
\end{bmatrix}
\begin{bmatrix}
    \begin{bmatrix}
        E_{\alpha\beta}
    \end{bmatrix}_x \\
    \begin{bmatrix}
        E_{\alpha\beta}
    \end{bmatrix}_y \\
    \begin{bmatrix}
        E_{\alpha\beta}
    \end{bmatrix}_z
\end{bmatrix},
\] (2.3)

where each coefficient $d_{ij}$ is dependent on the frequencies of the involved electrical fields. Depending on material symmetries several coefficients $d_{ij}$ can be zero or dependent on each other, and in a material with total inversion symmetry all of them will disappear. The constant $K$ is the degeneracy factor that is $\frac{1}{2}$ if two field components are degenerate in frequency and polarization and 1 if all three involved field components are distinguishable.

The frequency components of the polarization that is generated via the second order nonlinear susceptibility by an incoming field having two different frequency components are given by squaring the incoming field and can be expressed as

\[
P^{(2)} = C_{\text{SHG}1} E_1^2 e^{2i\omega_1} + C_{\text{SHG}2} E_2^2 e^{2i\omega_2} + C_{\text{SFG}} E_1 E_2 e^{i(\omega_1 + \omega_2)} +
\]

\[
+ C_{\text{DFG}} E_1 E_2 e^{i(\omega_1 - \omega_2)} + C_{\text{RECT}1} E_1^2 + C_{\text{RECT}2} E_2^2 + \text{c.c.,}
\] (2.4)

where $E_1$ and $E_2$ are the amplitudes corresponding to the frequency components $\omega_1$ and $\omega_2$ in the incoming field, the $C$ factors are efficiencies depending on the material, the involved frequencies and on degeneracy and c.c. means complex conjugate. The processes involved are second harmonic generation (SHG) of both field components, sum-frequency generation (SFG), difference frequency generation (DFG) and rectification (RECT) of both fields, and some of them are illustrated in figure 2-2.
The difference frequency generation process where the shortest wavelength beam (called pump) is much stronger than the longer wavelength input beam is so important that it usually treated separately and called optical parametric amplification (OPA). In a DFG process one photon in the pump beam will split into one photon of the frequency of the longer wavelength input beam and one photon of the difference frequency as illustrated in figure 2-3. In a parametric process the shorter wavelength generated (or amplified) beam is by convention called signal and the longest wavelength beam is called idler. In an OPA this is used to at the same time amplify the signal (idler) beam and generate an idler (signal) beam. Later on during propagation in the crystal the generated photons may interact with another pump photon so that the signal and idler grow exponentially until the pump starts to be depleted. The process of applying only a pump beam and letting it amplify the vacuum field, generating frequencies that were not present initially, is usually called optical parametric generation (OPG) and is mostly used for ultrashort (picosecond or femtosecond) pulses as very high intensities are needed to reach efficient conversion. For nanosecond laser pulses and even continuous wave beams an optical cavity can be used to feed-back and amplify the generated beams in an optical parametric oscillator (OPO). OPOs are the main topic of this work and will be treated more thoroughly in section 2.3.
In dielectric media where there are no free charges and the magnetic permeability can be neglected Maxwell’s equations can be reduced to the wave equation with a nonlinear driving term

\[ \nabla^2 E - \epsilon_0 \mu_0 (1 + \chi^{(1)} \chi^{(1)}) \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}. \]  

This equation can in general not be solved analytically, but by using some assumptions much more suited for calculations can be found. Assume that the electric field and the polarization can be described by sums of a finite number of quasi-monochromatic plane waves propagating along the x-axis according to

\[ E(x,t) = \frac{1}{2} \left[ E(x,\omega) \exp\left(i(\omega t - kx)\right) + c.c. \right] \] 

and

\[ P(x,t) = \frac{1}{2} \left[ P(x,\omega) \exp\left(i(\omega t - kx)\right) + c.c. \right], \]

where \( k = n(\omega)c/\omega \) is the wave vector with \( n(\omega) \) being the refractive index and \( \omega \) the angular frequency of the optical wave. The frequency domain wave equation for propagation along the x-axis, obtained through Fourier transformation, is

\[ \left( \frac{\partial^2}{\partial x^2} + \frac{\varepsilon(\omega)(\omega^2)}{c_0^2} \right) \tilde{E}(x,\omega) = -\mu_0 \omega^2 \tilde{P}_{NL}(x,\omega), \]  

where \( c_0 \) is the speed of light in vacuum and \( \varepsilon(\omega) = 1 + \chi^{(1)}(\omega) = (n + i\kappa)^2 \) with \( \kappa \) representing the losses in the material. Further we use the slowly varying envelope approximation (SVEA) where it is assumed that the variation of the field amplitude (due to linear and nonlinear processes) is small on the scale of the wavelength. This makes it possible to simplify the relation from a second to a first order derivative in the propagation direction. The material is assumed to be lossless, i.e. \( \kappa=0 \). The end result for a second order process, using equation 2.3 to deduce the polarization, is a system of three coupled first order differential equations:

\[ \frac{\partial E_1}{\partial x} = \frac{i\omega}{n_1c} K d_{eff} E_3 E_2^* e^{i(k_1-k_2)x}, \]

\[ \frac{\partial E_2}{\partial x} = \frac{i\omega}{n_2c} K d_{eff} E_3 E_1^* e^{i(k_1-k_2)x}, \]

\[ \frac{\partial E_3}{\partial x} = \frac{i\omega}{n_3c} K d_{eff} E_1 E_2 e^{-i(k_1-k_2)x}, \]  

where \( K \) is the degeneracy factor and \( d_{eff} \) is the effective nonlinearity that, in addition to the material properties, depends on the propagation direction, the optical frequencies and the polarization planes of the involved waves. For all nonlinear processes energy conservation must apply which gives the condition \( h\omega_3 = h\omega_1 + h\omega_2 \).

For a SHG process with \( \omega_1=\omega_2=\omega \) and \( \omega_3=2\omega \) the system is further reduced to two nonlinearly coupled differential equations. At low efficiency where the intensity of the driving field can be assumed to be constant it collapses to one linear first order equation that can be solved analytically in a plane wave approximation. For this simplified case the second harmonic intensity is:
\[ I_{2\omega}(L) = \frac{\varepsilon_0 n_{2\omega} c^2}{2} |E_{2\omega}|^2 = \frac{2\omega^2 d_{2\omega}^2 L^2 I_{\omega}^2}{\varepsilon_0 n_{\omega}^2 n_{2\omega} c^3 \sin^2 \left( \frac{\Delta k L}{2} \right)} \],

where \( \Delta k = k_{2\omega} - 2k_{\omega} \) is the phase mismatch of the process, \( L \) is the length propagated in the nonlinear crystal, \( I_{\omega} \) is the pump intensity, and \( n_{\omega} \) and \( n_{2\omega} \) are the refractive indices for pump and second harmonic, respectively.

### 2.2 Phase matching

It follows from equation 2.9 that second harmonic generation is most efficient when \( \Delta k = 0 \). As the wave vector \( k \) is proportional to the refractive index this is a matching of the phase velocities of the pump and the generated wave. If the two beams propagate with the same phase velocity newly generated radiation will be in phase with the radiation that was generated in another part of the crystal and the amplitudes will add. If the velocities, on the other hand, are different the phases will alternately interfere constructively and destructively and the end result will be a very low conversion efficiency. In the general case the phase-matching condition is:

\[ \Delta k = 2\pi \left( \frac{n_1}{\lambda_1} - \frac{n_2}{\lambda_2} - \frac{n_1}{\lambda_1} \right) = 0 , \tag{2.10} \]

that also can be interpreted as a momentum conservation condition since \( p = \hbar k \). If nothing is done to ensure phase-matching the generated waves will start to interfere destructively with waves generated at an earlier position after the coherence length

\[ L_c = \frac{\pi}{|\Delta k|} , \tag{2.11} \]

which in general is a very short distance. At the same time energy conservation

\[ \Delta E = \hbar c \left( \frac{1}{\lambda_1} - \frac{1}{\lambda_2} - \frac{1}{\lambda_1} \right) = 0 , \tag{2.12} \]

has to apply. In a degenerate process such as SHG the only way of fulfilling both these conditions is to have equal refractive indexes for the fundamental and harmonic waves. In bulk materials, far from resonances where the material is absorbing, the refractive index is decreasing with increasing wavelength. The dispersion is often modeled by the Sellmeier equation:

\[ n^2(\lambda) - 1 = \sum_{i=1} A_i \frac{\lambda_i^2}{\lambda^2 - \lambda_i^2} , \tag{2.13} \]

where \( \lambda_i \) are the resonance wavelengths and \( A_i \) their oscillator strengths. Most materials may be satisfactory modeled by two UV resonances and one far IR resonance. The result of this monotone dispersion is that the phase-matching conditions can not be fulfilled in the case where all waves have the same polarization. By using birefringent materials where the refractive index depends on the polarization it is often possible to fulfill the phase-matching condition by using the proper propagation direction in the crystal.

Birefringent crystals can be divided into uni- and biaxial crystals depending on the number of optic axes. For propagation along the optic axes all transversally polarized electromagnetic fields will sense the same index of refraction. In a uniaxial crystal the electric field can always be decomposed into one component that is polarized
perpendicular to the optic axes and one orthogonal component in the plane spanned by the optic axes. The component that is polarized perpendicular to the optic axes will always sense the ordinary index of refraction \( n_o \) whatever the propagation direction is, and is called the ordinary beam. The other component, called the extraordinary beam, has a nonzero projection on the optic axes and senses an index \( n_e(\theta) \) depending on the propagation angle according to

\[
\frac{1}{[n_e(\theta)]^2} = \frac{\cos^2 \theta}{n_o^2} + \frac{\sin^2 \theta}{n_e^2},
\]  \hspace{1cm} (2.14)

where \( \theta \) is the angle between the optic axes and the propagation direction.

In a positive (negative) uniaxial crystal the extraordinary index is increasing (decreasing) with the angle \( \theta \) from \( n_o \) for propagation along the optic axes to \( n_e \) for propagation perpendicular to the optic axes. As shown in figure 2-4 this can be used to find a propagation direction \( \theta \) where the extraordinary index for the fundamental wave is equal to the ordinary index for the second harmonic wave, thus compensating for the chromatic dispersion.

In the case where propagation is not along one of the crystal axes the birefringence will lead to Poynting vector walk-off, where the extraordinary beam propagates at an angle \( \rho \) to the phase fronts, with \( \rho \) given by:

\[
\tan \rho = -\frac{1}{n_e(\theta)} \frac{dn_e(\theta)}{d\theta}.
\]  \hspace{1cm} (2.15)

In a uniaxial crystal the walk-off can also be written as

\[
\tan \rho = -\frac{1}{2} [n_e(\theta)]^2 \left[ \frac{1}{n_e^2} - \frac{1}{n_o^2} \right] \sin(2\theta).
\]  \hspace{1cm} (2.16)
Walk-off will reduce the spatial overlap between the ordinary and extraordinary beams and thus has a detrimental impact on conversion efficiency when using birefringent phase-matching (BPM). The effects of walk-off can be partly compensated by using two crystals oriented in a walk-off compensating scheme\textsuperscript{16}, as illustrated in figure 2-5. Using twin crystals in a walk-off compensated scheme also has the advantage of removing beam translation during angle tuning.

Figure 2-5. The extraordinary beam exhibits walk-off from the ordinary beam with the angle $\rho$ when propagating in a birefringent crystal. This can be compensated with a second crystal inverted in the right manner.

Another way of compensating for the phase mismatch caused by the chromatic dispersion is quasi phase-matching (QPM), first introduced by Armstrong \textit{et al.} in 1962\textsuperscript{17}. The principle of QPM is to change the phase of the generated radiation instead of adjusting the phases acquired through propagation. By changing the sign of the nonlinear coefficient $d$ a phase shift of $\pi$ is introduced. Quasi phase-matching introduces a sign reversal every coherence length $L_c$ so that $d$ varies with a period $\Lambda = 2L_c$ according to

$$
d(z) = d_0 \text{sign} \left( \cos \left( \frac{2\pi z}{\Lambda} \right) \right) = d_0 \sum_{m=\pm \infty} G_m e^{ik_m z} . \tag{2.17}
$$

This modulation of $d$ adds an extra vector in the phase space that if the period is chosen correctly, i.e. two coherence lengths, cancels the phase mismatch for order $m=1$ according to

$$
\Delta k - k_{QPM} = 0 . \tag{2.18}
$$

In certain cases the phase mismatch is so large that periods short enough are not possible to manufacture and higher orders of $m$ have to be used, even though the power efficiency is proportional to $1/m^2$. Only odd orders $m$ can be used for QPM if the duty cycle is 50 \%, as even orders would introduce the phase shift at zero generated power. This is also evident from the expression for the $G_m$ coefficient, which is zero for even orders.

By inserting the Fourier expansion of $d$ into the coupled wave equations using the same assumptions as in equation 2.9 the following efficiency for second harmonic generation with QPM is found:

$$
I_{2\omega}(L) = \frac{e_0 n_{2\omega} c}{2} E_{2\omega}^2 = \ldots = \frac{2\omega^2 L^2}{e_0 n_{2\omega}^2 n_{\omega} c^5} \left[ \left( \int_{-L/2}^{L/2} d_0 \sum_{m=\pm \infty} G_m e^{ik_m z} \right) e^{-i\Delta k z} dz \right]^2 = \frac{2\omega^2 L^2}{e_0 n_{\omega}^2 n_{2\omega} c^5} L^2 d_0^2 \left( \frac{2}{m\pi} \right)^2 + \text{osc. terms, for } \Delta k = k_m . \tag{2.19}
$$

The generated intensity is equal to the phase matched case with $d_{QPM} = 2d_0/(m\pi)$. The Fourier terms that are not matched to the phase mismatch will only cause an oscillation in...
power with crystal length. An illustration of the gains for the different phase matching cases is shown in figure 2-6. Despite the $2/\pi$ reduction in efficiency when using QPM it is often more efficient than birefringent phase matching as polarization combinations with higher $d_{ij}$, that are not accessible with birefringent phase matching, can be used. Normally the crystal directions are defined so that $d_{33}$ is the largest component and all electric fields should thus be aligned along the z-axes of the crystal. An additional advantage of QPM is that the propagation direction can always be selected to be along the crystal axes so that walk-off is avoided.

![Figure 2-6](image_url)

**Figure 2-6.** Second harmonic intensity with different types of phase-matching assuming equal $d_{\text{eff}}$.

Periodic poling of crystals for use in QPM nonlinear devices utilizes the ferroelectricity of the materials. Ferroelectric materials have a spontaneous electronic polarization. By applying a strong electric field this polarization can be inverted by movement of ions between two positions in the crystal lattice. At the same time the sign of the nonlinear optical coefficient $d_{\text{eff}}$ will change. By depositing a periodic pattern of electrodes on one face of the crystal and applying high voltage a permanent domain grating suitable for quasi phase-matching can be produced.

A distinction is made between two possibilities of phase-matching. If the two lower frequency beams, that are input beams in an SFG process or are generated in a parametric process, have the same polarization the scheme is denoted type I phase-matching. If, on the other hand, the two low frequency, and hence longer wavelength, beams have different polarizations the phase-matching is of type II. One point to be made is that the wavelengths for the two polarizations cross at the angle where a parametric process produces degenerate frequencies for type II interaction (figure 2-7), but for type I interaction phase-matching is possible only on one side of the degenerate angle (figure 2-8). To be noted is also that the effective nonlinearity varies as a function of propagation angle. There is a possibility that even if the interaction is phase-matched $d_{\text{eff}}$
may be zero because of unfortunate material symmetries. This is, for example, the case for type I interaction in KTP\textsuperscript{18}.

![Figure 2-7. Phase-matching curves for type II interaction in KTP with a 1064 nm pump beam (solid). The effective nonlinearity for the relevant propagation angle and polarization combination (dashed).](image1)

![Figure 2-8. Phase-matching curves for type I interaction in ZGP with a 2128 nm pump beam (solid). The effective nonlinearity for the relevant propagation angle and polarization combination (dashed).](image2)
2.3 Optical parametrical oscillators

When a nonlinear crystal is placed in a cavity and pumped by a strong laser beam the parametric fluorescence is fed back to the crystal and amplified. This device is the optical parametric oscillator (OPO). OPOs pumped by nanosecond laser pulses are a mature technology and several reviews of their physics and applications have been presented\textsuperscript{19,20}.

If the parametric process is phase matched the amplification during one round-trip may exceed the cavity losses. When using nanosecond laser pulses for pumping the peak intensity is in general so high that there is a large net gain. The signal and idler will thus experience exponential growth. A nanosecond OPO will in general never leave the transient state but the generated fields will grow until the pump intensity goes down and the gain is reduced. In accordance with a laser the pump level where the gain equals the cavity losses is called the threshold for the continuous wave (cw) OPO operation. In a pulsed OPO a peak power only slightly above this level will never have time to produce a measurable output before the pump pulse ends. The threshold of the pulsed OPO is therefore defined as the pulse energy that is needed to produce a measurable output. The growth rate of the generated output above the threshold is called the slope efficiency, whereas conversion efficiency signifies output power divided by pump power.

The single pass parametric power gain in the nonlinear crystal is

\[ G = \Gamma^2 L^2 \frac{\sinh \left( \sqrt{\frac{\Gamma^2 - \Delta k^2}{4} L} \right)}{\left( \frac{\Gamma^2 - \Delta k^2}{4} \right)^2 L^2}, \]  

(2.20)

where \( \Gamma \) is the parametric gain factor given by

\[ \Gamma^2 = \frac{2 \omega_s \omega_i d_{\text{eff}}^2 I_p}{\epsilon_0 n_s n_i n_e c^3}, \]  

(2.21)

when only signal and pump beams are incident on the crystal\textsuperscript{21}. In the gain factor defined by eq. 2.21 the quotient \( d_{\text{eff}}^2 / n^3 \) can be identified. This expression contains all material dependence and is thus often called the nonlinear figure of merit for the material.

The losses in the OPO cavity originate from absorption, scattering and parasitic reflections in the cavity, but primarily from the finite reflections of the cavity mirrors. In fact for the OPO to produce any output at least either the signal or idler must have some fraction transmitted out of the cavity to generate the desired beam. Depending on if the cavity mirrors are reflecting both or only one of the signal and idler waves the OPO is called doubly or singly resonant (DRO or SRO), respectively. A DRO has a lower threshold than a SRO, but as each resonant wave in the OPO also has to fulfill the cavity conditions for longitudinal modes the DRO is less stable. For continuous wave (CW) operation the expense of the active cavity length control, that is required for stable operation, may be motivated by the reduced threshold. Another reason to use DROs is that for near degenerate operation it can be difficult to distinguish between the signal and idler at the cavity mirrors, but here the spectral width is so wide that jumps between the longitudinal modes are not significant if no line narrowing is introduced and the cavity may be left free-running. Another distinction between different cavity types is if the
pump passes only once through the crystal, single pass pumping, or if the undepleted pump is reflected back to pass a second time through the crystal and provide amplification also in the other direction, double pass pumping. Double pass pumping will increase the efficiency but may cause deterioration of the beam quality.

The simplest cavity configuration is the linear SRO cavity consisting of only the nonlinear crystal and two mirrors. This and some more complicated configurations are illustrated in figure 2-9. The incoupling mirror transmits the pump and reflects the signal, and the outcoupling mirror is partially reflecting for the signal and transmits pump and idler. The mirrors can be flat or curved to match the resonant mode to the pump waist. The L-shaped cavity has the possibility of introducing additional components, for example etalons for line narrowing, in the cavity without exposing them to the high pump intensity. The ring OPO has the advantage of ensuring that no pump radiation is reflected back to the laser, thus removing the need for an optical isolator. The bowtie ring cavity is mainly used in singly resonant continuous wave OPOs where long crystals and tight pump focusing is used because of the superior mode control that is possible through curved mirrors in this setup. The long cavity also increases the build-up time making it unsuitable for pulsed OPOs. Pico- and femtosecond OPOs where the roundtrip time is longer than the pulse length can be synchronously pumped by a high repetition rate laser so that the resonated signal travels with a new pump pulse on each roundtrip. All of these basic configurations come in a large number of variants, including the use of several crystals and walk-off compensating schemes.

![Diagram of OPO cavity configurations](attachment:image.png)

**Figure 2-9.** Examples of different OPO cavity configurations.

The theoretical threshold of singly resonant optical parametrical oscillators, for single and double passing of the pump, was derived by Brosnan and Byer\textsuperscript{22}. The threshold energy can vary between a few µJ\textsuperscript{23} and several mJ depending on nonlinear crystal, mirror reflectivities, wavelengths, focusing conditions and other parameters of the pump pulse and of the OPO cavity. The maximum efficiency of an OPO with a Q-switched
wide focus Gaussian pump beam and plane mirrors was calculated by Bjorkholm already in 1971 and is 71% in the singly resonant case. Parameters preventing 100% efficiency include energy in the low intensity spatial wings of the Gaussian beam, and the OPO build-up time.

The wavelengths generated by an OPO are determined by energy conservation and phase matching, as discussed in section 2.2. One of the main advantages of an OPO is that the phase matching conditions can be changed and thus the OPO is a tunable source of laser radiation. Basically every parameter that influences the index of refraction of the nonlinear medium can be used for tuning. The most common parameters to use are the crystal angle, the QPM domain grating period or the crystal temperature. In addition it is possible to vary the signal and idler wavelengths by keeping the crystal parameters constant and changing the wavelength of the pump. An example depicting how the wavelengths change with temperature and domain grating period in periodically poled LiNbO₃ (PPLN) is shown in figure 2-10. Explanations for the mismatch between theoretical and experimental data could include temperature measurement errors, period offsets in fabrication and inaccuracy in the refractive index model.

![Figure 2-10. Signal and idler wavelengths for a PPLN OPO depending on temperature. The solid curves represent theoretical tuning and are labeled by the domain grating periods in µm. The stars show experimental data.](image)

Perfect phase-matching is achieved for only one specific wavelength. There are two different quantities that both are called acceptance bandwidth. One is the maximum bandwidth for the pump to be able to amplify the same signal/idler pair. The other is the maximum signal bandwidth that can be amplified in an OPA. The latter one is also coupled to the bandwidth of the signals generated in an OPO.

If the pump is multi longitudinal mode the pumping efficiency will decrease with increasing bandwidth. Often an acceptance bandwidth is defined to give an indication on the acceptable pump linewidth for the specific material and pumping wavelength. The full width half maximum (FWHM) acceptance spectral width $\Delta\lambda$ can be defined so that
\[
\text{sinc}^2 \left( \frac{\Delta kL}{2} \right) = 0.5 \quad \text{for} \quad \lambda = \lambda_0 \pm \Delta \lambda / 2.
\]
The phase mismatch is calculated by a Taylor expansion of the refractive index around the phase matching wavelength, neglecting changes in the signal and idler wavelengths due to pump tuning, so that

\[
\Delta k = \left. \frac{2\pi}{\lambda} \frac{\partial n}{\partial \lambda} \right|_{\lambda = \lambda_0} \Delta \lambda + \left. \frac{2\pi}{\lambda} \frac{\partial^2 n}{\partial \lambda^2} \right|_{\lambda = \lambda_0} \Delta \lambda^2,
\]
where the second term in general is small compared to the first. To define a parameter for the material and not for a specific crystal generally \(\Delta \lambda L\) is the value used. For example, considering a ZGP OPO pumped at 2128 nm the acceptance bandwidth is 6.5 nm × cm. Similar relations can be used to calculate the sensitivity to a certain parameter in a fixed wavelength nonlinear process, e.g. an OPO with intracavity wavelength selective elements, second harmonic generation or difference frequency generation. Important parameters include wavelength, temperature and crystal angle.

The acceptance bandwidth for the signal with a fixed pump wavelength is calculated in analogy to the pump acceptance bandwidth by using a Taylor expansion of the phase mismatch, but the expansion is commonly carried out in angular frequency instead of wavelength

\[
|\Delta k| = \left| \frac{\partial k_s}{\partial \omega_s} \Delta \omega_s + \frac{\partial k_i}{\partial \omega_i} \Delta \omega_i \right| = \left| \left( \frac{\partial k_s}{\partial \omega_s} - \frac{\partial k_i}{\partial \omega_i} \right) \Delta \omega \right| = \frac{1}{c_0} \left| (n_{g,s} - n_{g,i}) \Delta \omega \right|
\]
in a first order expansion where the relation \(\Delta \omega_s = -\Delta \omega_i = \Delta \omega\) is a result of energy conservation and \(n_g\) signifies group index. It is evident that when the group velocities are equal the first order term disappears and the acceptance bandwidth will be determined by higher order terms in the expansion. This explains the very high bandwidths generated in a type I OPO close to degeneracy.

### 2.4 Nonlinear materials for mid-IR generation

There are several parameters that have to be taken into consideration when selecting what nonlinear crystal to use. They include for example transparency range, damage threshold, thermal conductivity, phase matching possibility, mechanical hardness, nonlinear figure of merit, crystal availability and price.

For the shorter wavelength part of the mid-infrared spectral range from 2 to approximately 4 µm there are a number of oxide based crystals that are possible to use. The most common ones are congruently grown LiNbO\(_3\) (LN) and KTiOPO\(_4\) (KTP). LN is mainly used with periodical poling (PP) for quasi phase matching\(^{26}\), whereas KTP is used both in bulk form with BPM and as PPKTP\(^{27}\). PPLN is the dominating crystal for low peak power applications as it has wide transparency and high gain (\(d_{33}=25.2\)\(^{28}\)). Long crystals lengths (up to 50 mm) are available, but PPLN has low damage threshold and the thickness is limited to about 1 mm due to a high coercive field for poling. PPLN is therefore suited for low peak power operation, but is not suitable for scaling to high pulse energies. PPLN OPOs generally have to be operated at elevated temperatures to avoid damage caused by the photorefractive effect. Doping with a few percent of MgO reduces the sensitivity to photorefractive effects\(^{29}\).
Figure 2-11. Transmission curves for KTiOPO$_4$, RbTiOAsO$_4$ and LiNbO$_3$.$^{18}$

KTP has a higher damage threshold than LN. The coercive field for periodic poling of KTP is also lower than that of LN and thus poled crystals as thick as 3 mm suitable for high energy applications can be fabricated.$^{30}$ With bulk crystals the nonlinear susceptibility component used is $d_{24}=3.9$ for type II phase matching. Using QPM it is possible to access the stronger nonlinear coefficient $d_{33}=15.3$$^{31}$. Alternatives to KTP are its isomorphs KTiOAsO$_4$ (KTA), RbTiOPO$_4$ (RTP) and RbTiOAsO$_4$ (RTA), where the arsenates are especially interesting due to their longer transparency range.$^{32}$ Transmission curves for KTP, RTA and LN are given in figure 2-11 for the region 1 to 5 µm.$^{18}$

At wavelengths longer than 4 µm the situation is more complicated. There are no materials suitable for conversion from the established Nd$^{3+}$-lasers to wavelengths longer than 4 µm. The main problem is transparency, but for the few nonlinear materials that are transparent in the interesting wavelength region other considerations such as crystal quality, damage thresholds and thermal effects make efficient OPO operation impossible.$^{33}$ Pumping of OPOs for generation of mid-IR radiation thus has to be done at wavelengths longer than 1.5 µm and in most cases even longer than 2 µm. The materials AgGaSe$_2$ (AGSe) and AgGaS$_2$ (AGS) that were used in early experiments$^{34-37}$, suffer from low damage thresholds and poor thermal conductivity and were not suitable for high repetition rate OPOs$^1$. Due to improvements in growth technique during the last ten years they have been replaced by ZnGeP$_2$ (ZGP). ZGP has high nonlinearity ($d_{eff}=75$), good thermal conductivity and acceptable damage thresholds. The short wavelength transmission cut-off is near 2 µm, as seen in figure 2-12, but the exact position and shape is sensitive to impurities in the crystal and the growth is thus very critical. ZGP will be further discussed in section 5.
Other less used nonlinear materials for the mid-infrared include CdSe, GaSe and CdGeAs$_2$ (CGA). CdSe has been used for OPOs$^{38}$, whereas the crystal quality of the soft layered GaSe has not allowed OPO operation. CGA has an extremely high figure of merit, 57 times that of PPLN, but the transparency is not that good and OPO operation has only been shown with 4.8 µm pumping$^{39}$.

Several semiconductor materials as GaAs, GaP and ZnSe show high nonlinearities, large transparency ranges, good damage thresholds and high thermal conductivity. They, however, have no birefringence and are not ferroelectric to allow periodical poling. Experiments have been done with stacks of thin material sheets for QPM$^{40}$, but problems with slicing the materials thin enough and low loss bonding of the stacks limit the efficiency. In the last few years progress has been made on orientation patterned growth of GaAs showing promise for future availability of a QPM material for longer wavelengths$^{41}$, but it is not yet commercially available. GaAs has an absorption band edge at 0.9 µm but high two-photon absorption limits pumping with ns laser pulses to above 1.8 µm. GaP on the other hand has its transparency edge at 0.5 µm and pulsed pumping of GaP by Nd$^{3+}$-lasers at 1.06 µm could become possible in the future, but so far no one has shown QPM of GaP$^{33}$. 

**Figure 2-12.** Transmission graphs for ZGP, AGS and AGSe$^{18}$. 

![Transmission graphs for ZGP, AGS and AGSe](image_url)
3 Laser beam propagation

The transversal intensity cross-section of a laser beam is often approximated with a 2-D Gaussian profile according to

\[ I(x, y) = \frac{2P}{\pi w^2} \exp\left(-2\frac{x^2 + y^2}{w^2}\right), \]  

(3.1)

where \( P \) is the total power in the beam and \( w \) is the 1/e\(^2\) intensity radius. For a propagating beam the beam radius increases according to

\[ w(z)^2 = w_0^2 + \left(\frac{M^2 \lambda (z - z_0)}{\pi w_0}\right)^2, \]  

(3.2)

where \( w_0 \) is the 1/e\(^2\)-radius at the focus, \( z_0 \) is the position of the focus and \( M^2 \) is a beam quality parameter. The \( M^2 \)-parameter is equal to 1 for an ideal Gaussian beam and larger for beams containing higher order modes\(^{42}\). For non ideal beams \( w(z) \) and the beam radius at the focus \( w_0 \) have to be generalized to twice the second moment of the beam \( w_{x,y} = 2\sigma_{x,y} \). This definition has become the international standard of beam width\(^{43}\). Even beams that have high \( M^2 \) often have an approximately Gaussian shape, as illustrated in figure 3-1 that shows a broadband beam with \( M^2 \) of 6 and 8 in the horizontal and vertical directions, respectively [III].

The first moment is the center of gravity of the beam and is defined according to

\[ x_0 = \frac{\int \int xI(x, y)dxdy}{\int \int I(x, y)dxdy}, \]  

(3.3)

in \( x \) and analogously in \( y \). The second moment that provides a measure for the beam width is given by

\[ \sigma_x^2 = \frac{\int \int (x - x_0)^2 I(x, y)dxdy}{\int \int I(x, y)dxdy}, \]  

(3.4)

in \( x \) and analogously in \( y \). In theory the second moment is easy to calculate from any camera image of the beam. Because the intensity is scaled by the squared distance to the center of the beam the calculation is however very sensitive to noise in the image. It is therefore common to use a Gaussian fit of the beam to determine the beam width when measuring the beam quality parameter \( M^2 \). Sometimes it is not possible to put a camera in the beam, for example due to the high intensity in a focus or because of limited availability of cameras for longer wavelengths, and other methods for measuring the beam width are used. The knife edge measurement technique assumes a Gaussian beam profile and the power transmitted past a knife edge scanned across the beam is measured. The beam radius is then defined as \( w = 0.78 \times |x_{90} - x_{10}| \), where \( x \) means position in a direction perpendicular to the beam propagation and the index on the \( x \) is the percentage of the total power that is transmitted past the knife edge.
An important parameter for the pump of an OPO is the Rayleigh length, defined as the distance from the focus where the beam radius has increased by a factor $\sqrt{2}$, or the peak intensity has been reduced by a factor 2. The Rayleigh length is given by

$$z_r = \frac{\pi w_0^2}{M^2 \lambda}.$$  \hfill (3.5)

To be able to use flat mirrors in a cavity the pump mode focus should be large enough that the cavity length is substantially shorter than the Rayleigh length. For tighter focusing it is advisable to use concave cavity mirrors matching the curvature of the wavefront. Nonlinear crystal longer than twice the Rayleigh length are of limited advantage as the edges will give a very small contribution to the conversion efficiency because of the drop in peak intensity.

The far field divergence of a laser beam (half angle) is

$$\theta = \frac{M^2 \lambda}{\pi w_0}.$$  \hfill (3.4)

For long range applications it is thus very important to have low $M^2$ as the beam size at the target scales directly with the beam quality parameter. By using a beam expander with a collimated output a beam radius at the focus of several cm is possible and accordingly divergence angles below 100 µrad can be obtained even in the mid-IR. Smaller divergences are generally not useful as the pointing accuracy is of this order of magnitude in high performance systems. The laser spot size after propagation through the atmosphere will not only be dependent on the natural divergence caused by diffraction. In addition the beam is also perturbed by turbulence in the air causing beam broadening and speckle patterns\textsuperscript{44}. The position of the beam also changes with time depending on pointing jitter in the laser and the OPO, vibrations in the platform where the laser is mounted (e.g. an aircraft) and because of refraction in large scale turbulence cells. The perturbations close to the plume from a jet engine are especially severe [IX,X].
4 Pump sources at >2 µm

4.1 Competing technologies

There have been a number of studies on OPO pumping at or above 2 µm reported in the literature. Examples of direct pumping by lasers include, but are not limited to, Ho\(^{3+}\)-lasers pumped by Tm-YLF lasers\(^{45}\) and Tm-fiber lasers\(^{46}\). The reason for this double laser scheme is that Tm\(^{3+}\) has a low emission cross-section and is not suitable for Q-switching, and Ho\(^{3+}\) lacks absorption bands that coincide with emission spectra from efficient diodes. An alternative is pumping with a Cr:ZnSe laser, that has the interesting emission band from 2.1-2.8 µm, making pump tuning of the mid-IR OPO possible\(^{47}\). Cr:ZnSe lasers cannot be Q-switched but need to be pumped by a pulsed laser near 1.9 µm for gain switching which makes a complex system. The advantage of pump tuning over angle tuning of the OPO is that the position and angle of the generated radiation does not change with the wavelength, something that may be important for long range transmission.

The most well developed solid state lasers are Nd\(^{3+}\)-lasers that are used for lidars and material processing. By using an OPO the 1.06 µm wavelength of these lasers can be converted to the 2 µm range suitable for pumping ZGP. The standard scheme is to use a type II bulk KTP OPO. Different configurations such as linear cavities with a single crystal\(^{48}\) and with walk-off compensated crystal pairs\(^{49}\), ring cavities\(^{50}\), polarization rotating cavities\(^{50}\) and intracavity OPOs\(^{52}\) have been demonstrated. By changing the angles of the crystals in a walk-off compensated pair of KTP crystals pump tuning is possible without realigning the second OPO\(^{53}\).

Because of the higher nonlinearities and the absence of walk-off QPM OPOs are very interesting. Near degenerate OPOs applying QPM crystals, however, have a very wide gain bandwidth. For example, a bandwidth of 210 nm (FWHM) around degeneracy was measured in a PPKTP OPO pumped at 1064 nm in a reference measurement for article III. Solutions to this problem include pumping with a 1.3 µm Nd:YAG laser to generate a signal at 2.12 µm without being close to degeneracy\(^{54}\), and type II QPM with a polarization rotating cavity, that uses a polarization combination with a smaller nonlinear coefficient\(^{55}\).

The most straightforward scheme is to make the cavity spectrally selective to reach threshold only for a small bandwidth. Traditional methods are etalons, diffraction gratings in Littman and Littrow configurations and birefringent filters\(^{22,56}\). All of these methods suffer from the problem that they also introduce extra loss at the desired wavelength. Thus the threshold is raised and the slope efficiency decreases. By using a low efficiency, low power OPO to generate a beam with good spatial and spectral quality and then amplifying it in an OPA a good high energy pump source can be obtained, as shown by Saikawa \textit{et al.} with a pair of MgO:PPLN crystals\(^{57}\). It is also possible to use the low power beam to seed an OPO.

The interest in type I OPOs operating very close to degeneracy is also motivated by the possibility to use both signal and idler to pump the same ZGP OPO. This was first realized by Perrett \textit{et al.} with a surface diffraction grating in an L-shaped cavity limiting.
the bandwidth of the double resonant OPO to 2 nm at degeneracy\textsuperscript{56}. Phua \textit{et al.} demonstrated the use of both signal and idler from a type II bulk KTP OPO by a wavelength dependent polarization rotator that aligned the polarization planes of both waves\textsuperscript{52}. Since the signal and idler from the KTP OPO were separated by 60 nm the end result was, however, two separate OPOs in the same ZGP crystal.

After the realization of the first narrowband OPO with a volume Bragg grating output coupler by Jacobsson \textit{et al.} in 2005\textsuperscript{58} we realized that the technique has a potential of providing a very good pump source at 2 µm in the form of a narrowband QPM OPO.

\section*{4.2 Volume Bragg gratings}

Longitudinal diffraction gratings, called volume Bragg gratings, bulk Bragg gratings or volume holographic gratings, having a periodical modulation of the refractive index can be used to selectively reflect only one wavelength with the reflectance bandwidth orders of magnitude lower than what is possible with coated filters. Several different materials can be used to produce longitudinal gratings, but the most successful so far are photothermo-refractive (PTR) glasses. In the alumo-sodia-silicate glass materials that are doped with silver, cerium and fluorine, illumination by ultraviolet light starts a photochemical process that after heat treatment results in a change of the refractive index\textsuperscript{59}. By illuminating with a holographic technique or a phase mask a grating according to \( n = n_0 + n_1 \sin(2\pi x / \Lambda) \) can be created, where the modulation \( n_1 \) is up to \( 10^{-3} \) in magnitude.

\begin{figure}[ht]
\centering
\includegraphics[width=0.5\textwidth]{bragg_grating.png}
\caption{A volume Bragg grating has a sinusoidal index modulation in the propagation direction.}
\end{figure}

The glasses and the generated gratings are mechanically, chemically and thermally (to at least 400 °C) stable, show damage thresholds similar to normal mirror substrates and are non-absorbing in the visible and near IR (up to 2.7 µm). The reflected wavelength is determined by the Bragg condition

\[ \lambda_B = 2n\Lambda. \] (4.1)
The reflectance bandwidth is calculated according to
\[
\frac{\Delta \lambda}{\lambda} = \frac{\lambda}{2nd} = \frac{\Lambda}{d} = \frac{1}{N},
\]
where \(d\) is the grating thickness and \(N\) is the number of grating periods in the material. The bandwidth can range from 0.01 nm to a few nm. This is many times narrower than the bandwidths achievable with multilayer coatings. The reflectance properties of thick hologram gratings were derived by Kogelnik. The peak diffraction efficiency is
\[
R = \tanh^2 \left( \frac{\pi n_1 d}{2n_0 \Lambda} \right).
\]
Maximum practical diffraction efficiencies are around 99% for grating thicknesses larger than 10 mm. As \(n_1\) and \(d\) are both adjustable parameters the bandwidth and the reflectivity can be varied independently.

Volume Bragg gratings are used as external feedback to diode lasers for wavelength stabilization. They have also been used as output couplers for solid state lasers with \(\text{Nd}^{3+}\), \(\text{Er}^{3+}\) and \(\text{Tm}^{3+}\) gain media.

### 4.3 Volume Bragg grating coupled OPOs

By using a volume Bragg grating as output coupler in the OPO the feedback in the cavity is limited to the desired wavelength. The output coupling is thus 100% at all wavelengths that are not within the narrow reflection bandwidth of the volume Bragg grating, and the threshold is never reached for broadband operation. At the resonance wavelength of the Bragg grating the radiation is, on the other hand, reflected back with the diffraction efficiency \(R\) and the output coupling is thus 1-R. If the surfaces of the grating are properly antireflection (AR) coated the grating introduces no extra losses in the cavity compared to a standard mirror output coupler.

![Figure 4-2. Spectrum of a PPLN OPO using volume Bragg grating [black] and mirror [gray] output couplers [II].](image)
Jacobsson et al. demonstrated a factor 20 decrease in signal linewidth at 975 nm compared to operation with a mirror when pumping at 532 nm\textsuperscript{58}. In article II a PPLN OPO with a signal at 2008 nm pumped by a Nd:YVO\textsubscript{4}-laser was demonstrated with 0.44 nm linewidth, a reduction by a factor 80 compared to operation with a mirror. A comparison of the spectra with mirrors and with a volume Bragg grating is presented in figure 4-2. Similar linewidths were also achieved with a PPKTP OPO pumped by a Nd:YAG laser. A picture of this OPO is shown in figure 4-3.

![Figure 4-3](image)

**Figure 4-3.** Photograph of the PPKTP OPO in [I] with the volume Bragg grating in the metal adapter to the left, the PPKTP crystal on the temperature controlled copper heat-sink in the middle and the incoupling mirror to the right.

In paper III a volume Bragg grating resonant at 2128 nm, very close to degeneracy was used to lock a PPKTP OPO. The signal and idler peaks were not fully resolved in the measurement, but were estimated to be separated by 0.6 nm, figure 4-4. All energy was contained in a 2 nm region and the bandwidths were estimated to 0.46 and 0.39 nm for signal and idler, respectively. The reference OPO with a mirror output coupler showed a bandwidth (FWHM) of 210 nm.
Figure 4-4. The output spectrum of a PPKTP OPO locked near degeneracy with a volume Bragg grating.

Tuning of an OPO with a Bragg grating output coupler has been demonstrated by tilting the volume Bragg grating used as a folding mirror in an L-shaped cavity. This method has the disadvantage that the generated signal is emitted in two different directions on the two reflections in the Bragg grating.

Because of low sensitivity to temperature changes of the optical pathlength in the PTR-glass the OPO tunes very slowly with temperature, only around 7 pm/K in the near IR. The result is high wavelength stability when using volume Bragg gratings for locking the wavelength. Changing the temperature of the nonlinear crystal, the common tuning method for QPM OPOs, will not cause the wavelength of the OPO to change. The result is instead a variation in OPO efficiency because of decreased overlap between the gain peak and the feed-back wavelength. In the PPLN OPO with a volume Bragg grating resonant at 2008 nm an acceptance temperature range of 5.2 K was measured at low pump power [II]. The PPKTP OPO close to degeneracy was less sensitive to temperature changes because of the wider gain bandwidth.
5 Mid-IR ZGP OPO

The main source of high average power mid-infrared radiation today is OPOs using ZGP crystals. ZGP has high nonlinearity \( (d_{\text{eff}}=75 \text{ pm/V}) \), which leads to a nonlinear figure of merit \( (d_{\text{eff}}^2/n^3) \) about 9 times larger than for PPLN. The birefringence is suitable for type I phase-matching over a large range of wavelengths. For long wavelength generation type II phase matching is also possible. In addition ZGP has good thermal and mechanical properties, as well as an acceptable damage threshold (>1 \( \text{J/cm}^2 \)). The transparency range is often reported as 0.74-12.4 µm, but low absorption is available only in the 2-8 µm region. The loss in the 1-4 µm region is dependent on defects and impurities in the crystal and the growth is thus very critical. There have been a large number of ZGP OPOs reported in the literature, see for example references 48-54.

The acceptance bandwidth of a ZGP OPO is 6.5 nm×cm for pumping at 2.13 µm. The acceptance bandwidth is the limit where the pumping efficiency has decreased by a factor two for the power at the edges of the spectral pump profile. The pumping efficiency will thus increase further with narrower bandwidth. Perrett et al. reported a decrease of slope efficiency from 33 % to 9 % when the pump bandwidth increased from 2.3 to 8 nm.56

In figure 5-1 examples of theoretical tuning curves for pumping at three different wavelengths are shown. It is evident that in order to phase-match an OPO simultaneously for the signal and idler from a 1 µm pumped OPO the first OPO has to operate very close to degeneracy. For a type I OPO this is possible only with the help of linewidth narrowing elements in the cavity.

![Figure 5-1](image_url)

**Figure 5-1.** Phase matching curves for a ZGP OPO pumped by 2.1 (dash-dotted), 2.128 (solid) and 2.158 (dashed) µm. The 2.1 and 2.158 µm wavelengths could be a signal-idler pair from an OPO pumped by a 1.064 µm Nd³⁺-laser.
We have demonstrated pumping of a ZGP OPO using the 2008 nm signal from a PPKTP OPO with a volume Bragg grating to limit the bandwidth to 0.5 nm [I]. The ZGP OPO had a slope efficiency of 41 % and a threshold of 38 µJ. The idler at 2264 nm was discarded as the available ZGP crystal had a coating and a cut that was not suitable for pumping with this wavelength.

To increase the efficiency volume Bragg gratings at wavelengths closer to degeneracy were used and a narrowband OPO at 2128 nm was demonstrated (see section 4.3). The output from this OPO was used to pump a ZGP OPO in a setup depicted in figure 5-2. The slope efficiency from the total power at 2.13 µm to the mid-IR was 43 % when the pump in the PPKTP OPO was single passed and 41 % when the dichroic mirror was aligned to double pass the pump. The total conversion efficiency from 1.06 µm to the mid-IR was 7 %.

![Figure 5-2](image)

Figure 5-2. The experimental setup that was used in [III]. The setup in [I] was very similar with the main difference being the resonance wavelength of the volume Bragg grating and the filtering between the two OPOs.

The ZGP OPO was tunable from degeneracy at 4.26 µm to signal wavelengths as low as 2.9 µm. The idler correspondingly increased in wavelength and probably reached 8 µm as expected from energy conservation, but the acquired spectrums in figure 5-3 are limited to 5.2 µm due to detector sensitivity. The ZGP crystal used was cut at 60° to the c-axis, but the phase matching angles for pumping with 2128 nm radiation range from 54.6° at degeneracy to 50.8° for a signal wavelength of 2.9 µm. This 3.8° change in phase-matching angle corresponds to a 12° change in external angle as the index of refraction in ZGP is 3.14 for the pump. Due to the unfavorable cut of the ZGP crystal, which was available from an earlier experiment, the external angle of incidence for the pump ranged approximately from 17° to 29°.

Other authors have presented ZGP OPOs with conversion efficiencies well above 50 %, and slope efficiencies above 80 %. Even though the total bandwidth from the Bragg grating locked OPO is larger than what is used in these papers and may limit the efficiency, the ZGP OPO presented here has opportunities for big improvements. Obvious ideas are to use a ZGP crystal cut for plane incidence, to use two crystal in a walk-off compensating configuration and optimization of reflectivity and curvature of the OPO mirrors.
Figure 5-3. Spectra showing the tuning range of the ZGP OPO in [III]. The measurement was limited to 5 µm, but from the measured signal wavelengths idler wavelengths out to 8 µm would be expected.

The bandwidth of the generated signals is very broad close to degeneracy and decreases when tuning away from the degeneracy point. This is typical for all type I OPOs as the parametric gain bandwidth is inversely proportional to the difference in group velocity of the signal and idler, as seen from equation 2.22, and at degeneracy this difference goes to zero.
6 Description of the included papers

Article I

ZnGeP$_2$ Parametric Oscillator Pumped by a Linewidth Narrowed Parametric 2 µm Source
Markus Henriksson, Mikael Tiihonen, Valdas Pasiskevicius and Fredrik Laurell

This paper is the first description of pumping a ZGP OPO with the output of an OPO with a volume Bragg grating output coupler. A PPKTP OPO pumped by a 20 Hz PRF Nd:YAG laser produced a 0.5 nm bandwidth signal at 2008 nm through the use of a volume Bragg grating output coupler. This is a factor 80 reduction of the bandwidth compared to a reference OPO using a mirror output coupler. The signal was used to pump a ZGP OPO generating broadband radiation in the 3-5 µm region. The ZGP OPO had a slope efficiency of 41 %, and the conversion efficiency of the whole tandem OPO setup was 4 % generating 170 µJ of output energy.

Contribution by the author:
The candidate performed the experiments together with M. Tiihonen. The candidate wrote the paper with assistance from M. Tiihonen.

Article II

Narrow linewidth 2 µm optical parametric oscillation in periodically poled LiNbO$_3$ with volume Bragg grating outcoupler
M. Henriksson, L. Sjöqvist, V. Pasiskevicius, F. Laurell

This paper describes the first high repetition rate (10 kHz) OPO using a volume Bragg grating. The PPLN OPO with a signal at 2008 nm is also the so far most efficient volume Bragg grating OPO with 47 % slope efficiency. The signal at 2008 nm had a bandwidth of 0.44 nm.

Contribution by the author:
The candidate performed the experiments and wrote the paper.

Article III

Efficient pumping of ZGP OPO by narrowband near degenerate PPKTP OPO
M. Henriksson, M. Tiihonen, V. Pasiskevicius and F.Laurell
manuscript submitted to Optics Express November 2006.
In this work we show a PPKTP OPO operating very close to degeneracy. The signal and idler peaks are 0.6 nm apart, and the full energy is contained in a 2 nm region. We also show efficient pumping of a ZGP OPO by signal and idler from the PPKTP OPO simultaneously with a maximum slope efficiency of 43 %. The total slope efficiency from 1 µm to the 3.5-5 µm region was 10 % and the conversion efficiency was 7 %. Tuning from degeneracy to a signal wavelength of below 3 µm with a corresponding idler wavelength at 8 µm was demonstrated.

Contribution by the author:
The candidate performed the experiments and wrote the paper.
7 Conclusions

This thesis describes a new and efficient way of generating laser radiation in the mid-IR through wavelength conversion from 1 µm in an improved tandem OPO setup. By using a volume Bragg grating as a wavelength selective cavity mirror in an OPO relatively narrow bandwidth is easily achieved even with the very wide gain bandwidth of a quasi phase-matched OPO. The narrow bandwidth makes the radiation from the OPO suitable for pumping a second OPO stage.

The new design with a volume Bragg grating output coupler can be used in any OPO, but is especially beneficial at near degenerate operation with quasi phase-matched crystals. A decrease in bandwidth from 210 nm to around 1 nm for the combined signal and idler waves was measured when changing from a mirror to a Bragg grating output coupler in a PPKTP OPO.

The possibility of using QPM instead of birefringent phase-matching in the first OPO step exhibits several advantages. The general QPM advantages of higher nonlinearities and no walk-off are of course valid. In addition birefringently phase-matched OPOs for conversion from 1 to 2 µm are commonly using type II configuration where signal and idler have different polarizations. In a QPM OPO all interacting waves may, however, have the same polarization and thus both signal and idler can be used to pump the second OPO stage.

In this thesis a ZGP OPO pumped by the 2008 nm signal from a PPKTP OPO using a volume Bragg grating was presented. Moreover, in an improved configuration pumping of a ZGP OPO with the signal and idler from a very near degenerate PPKTP OPO at 2128 nm was demonstrated.

The highest efficiencies presented in this thesis are 10 % slope efficiency and 7 % conversion efficiency from 1.06 µm to the 3.5-5 µm region. Other authors have presented 14 % conversion efficiency from a tandem OPO system using a type II ring cavity OPO with bulk KTP crystals\(^5^0\). By using an AR-coated crystal with larger single pass gain in the first OPO and optimizing the ZGP OPO I see opportunities for large improvements of the efficiency in my setup. As both signal and idler are used in both OPO steps each pump laser photon can in theory generate four photons in the mid-IR, and no energy is discarded in earlier steps. I thus believe that my setup should generate higher conversion efficiency than setups using type II phase matching in the first OPO step.
References

18. SNLO nonlinear optics code available from A. V. Smith, Sandia national laboratories, Albuquerque, NM 87185-1423.


