Generation of mid-infrared laser using solid state laser and nonlinear optical frequency conversion

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NANYANG
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**GENERATION OF MID-INFRARED LASER
USING SOLID STATE LASER AND NONLINEAR
OPTICAL FREQUENCY CONVERSION**

TAN LIHAO
SCHOOL OF PHYSICAL AND MATHEMATICAL SCIENCES
2016

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USING SOLID STATE LASER AND NONLINEAR
OPTICAL FREQUENCY CONVERSION**

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Abstract

Coherent MIR radiation has several applications such as remote sensing, laser surgery, nonlinear spectroscopy, countermeasures, etc. Generation of coherent MIR radiation is achieved by solid state laser and quantum cascade laser with direct MIR lasing, or nonlinear frequency conversion from laser with shorter wavelength. In this thesis, we developed several different laser configurations to access the MIR wavelengths.

Firstly, a high repetition rate high power Ho:YAG laser was developed to generate high average power MIR ns pulse in an intra-cavity ZGP OPO configuration. A high energy Ho:YAG MOPA laser pumped ZGP crystal in an extra-cavity OPO configuration was demonstrated. A novel uncoated wedge ZGP OPO configuration was studied, addressing the AR-coating limitation encountered in conventional ZGP OPO.

Secondly, Coherent Polarization Locking configuration was implemented in Ho:YAG oscillator. Studies show the Coherent Polarization Locking configuration improves the beam quality in CW operation and reduces the risk of intra-cavity optical damage in pulse operation. Improvement in beam quality is due to the reduction of thermal related effects in the oscillator. Splitting the intra-cavity intensity in the Coherent Polarization Locking cavity reduces the possibility of intra-cavity optical damage.

Lastly, a high energy Yb:CaF₂ regenerative amplifier was developed as the pump source for a multi-stage ZGP OPA setup to generate fs MIR laser pulses. To increase the efficiency of fs MIR generation, a high energy Ho:YAG regenerative amplifier was explored as the direct pump source for ZGP OPA system.

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Chapter 1 Introduction

1.1 Motivation

Since the first demonstration of laser action by Maiman [1] using ruby in 1960, there has been an intensive research and development in the field of solid state laser, in terms of different wavelengths (ultraviolet to infrared), different laser gain medium, different operation modes (CW, pulse, nonlinear optical conversion, etc.) and different pump geometries.

Mid-Infrared (MIR) region covers the part of the electromagnetic spectrum between the wavelength range of 2.5-10 μ m. In this wavelength range, there are interesting and attractive applications owing to the following unique features:

- a) Several atmospheric transparent windows fall within the MIR region
- b) Various molecules have strong rotational-vibrational absorption lines within the MIR region, which create the molecular fingerprint region of the electromagnetic spectrum

However, these applications, such as remote sensing, laser surgery, nonlinear spectroscopy, trace gas monitoring, countermeasures, etc., require to use the MIR laser due to its coherent light radiation properties. Different applications require the laser source to possess different properties such as wavelength tunability, emission bandwidth, beam quality, CW or pulse operation, pulse width, output power/energy etc. Thus there

is a strong motivation in the research and developmental work for MIR laser to cater for different applications.

1.2 Objective

In this thesis work, its main objective is in the development of coherent MIR radiation sources using the solid state laser and nonlinear optical frequency conversion technology. The work done on the coherent MIR radiation source is used as a contribution to the application in the defence industry. The laser properties required to achieve are high energy and high average power MIR pulse laser, emitting between 3-5 μm , with near diffraction-limited beam quality. Pulse width of interest covers the nanosecond (ns) regime and the femtosecond (fs) regime. Pulse energy spans from hundreds of micro-Joules (μJ) to tens of milli-Joules (mJ) range, with pulse repetition rate of several hundreds hertz to several kilo-hertz (kHz).

1.3 Organisation of Thesis

This thesis is organized as follows:

Chapter 1 provides a brief introduction to the research work of this thesis.

Chapter 2 introduces the fundamental principle of the solid state laser theory, particularly on the quasi-three-level laser system. The theoretical background of nonlinear optical frequency conversion and chirped pulse amplification are presented.

Chapter 3 describes the developmental work of the 2 μ m high repetition rate Q-switched Ho:YAG solid state laser pumped with Tm:Fiber laser. A customised high energy Ho:YAG master-oscillator power-amplifier laser is developed by a commercial collaborator. Evaluations of direct diode-pumped Ho:YAG slab oscillator and Tm:Fiber pumped ceramic Ho:YAG gain medium are performed.

Chapter 4 investigates the applications of the Ho:YAG laser to pump the Zinc Germanium Phosphide (ZGP) optical parametric oscillator (OPO) in both intra-cavity and extra-cavity configuration. High repetition rate MIR laser pulses in the intra-cavity configuration and high energy MIR laser pulses in the extra-cavity configuration are generated. A novel uncoated wedge ZGP OPO configuration is also explored.

Chapter 5 discusses the concept of Coherent Polarization Locking (CPL) technique for coherent beam combining application. The CPL technique is applied to perform coherent combining of two Ho:YAG laser. The advantages of implementing CPL in quasi-three-level Ho:YAG oscillator, operating in both CW and pulsed, is presented.

Chapter 6 explores the high energy compact diode-pumped Yb:CaF₂ regenerative amplifier seeded with commercial Yb fs oscillator. The 1.03 μ m fs laser pulses output from the Yb:CaF₂ regenerative amplifier is subsequently used as the pump source for a ZGP optical parametric amplifier (OPA) to generate fs MIR radiation. The experimental configuration and results of the OPA setups are presented. The last section of this chapter investigates the use of Ho:YAG gain medium in a regenerative amplifier configuration to generate high energy 2 μ m fs laser pulses.

Chapter 7 summaries the key works of this thesis and recommends the potential future work that may be performed.

Chapter 2 Technological Background

2.1 Fundamental Laser Theory of Quasi-Three-Level Laser System

Laser operation occurs by the stimulated emission of atoms. Population inversion, where the population of the atoms in the upper energy level is larger than the population of the atoms in the lower energy level, is required for the laser oscillation. To describe the laser dynamics, such as population inversion and the radiation density, happening in a laser gain medium, a pair of coupled differential equations, known as the rate equations, are used. Other important information, such as the laser efficiency, can be derived by solving the rate equations. In this section, the rate equation theory is introduced for both the ideal four level and three-level system. The derivation of the rate equation to describe the laser dynamics for quasi-three-level laser system is discussed.

2.1.1 Rate Equation for Four Level Laser System

Figure 2.1 shows an ideal four level laser system. Atoms are pumped from the ground level 0 to the upper level 3. The pumping process is usually through the absorption of photons from a flashlamp, laser diode or another solid-state laser. For example, assuming the atoms are excited from level 0 to level 3 at a pump rate of W_p . Upon reaching level 3, the atoms decay rapidly to level 2. They stay at level 2 for a sufficient long time until laser action occurs between level 2 and level 1. At level 1, the atoms are again quickly decay to level 0. The system is assumed to be isolated and in equilibrium

and there are no collision allowed among the atoms. Therefore, the decay mechanism of level 3, level 2 and level 1 are through spontaneous emission with lifetimes of τ_{30} , τ_{32} , τ_{21} and τ_{10} respectively.

Figure 2.1 Transition in a four level laser system

The rate equations describing the atomic transitions between the energy levels can be written as [2]

$$\frac{dn_3}{dt} = W_p n_0 - \frac{n_3}{\tau_{30}} - \frac{n_3}{\tau_{32}} \quad (2.1a)$$

$$\frac{dn_2}{dt} = \frac{n_3}{\tau_{32}} - \frac{n_2}{\tau_{21}} - n_2 \sigma c \phi + n_1 \sigma c \phi \quad (2.1b)$$

$$\frac{dn_1}{dt} = \frac{n_2}{\tau_{21}} - \frac{n_1}{\tau_{10}} - n_1 \sigma c \phi + n_2 \sigma c \phi \quad (2.1c)$$

$$\frac{dn_0}{dt} = -W_p n_0 + \frac{n_1}{\tau_{10}} + \frac{n_3}{\tau_{30}} \quad (2.1d)$$

$$n_{tot} = n_0 + n_1 + n_2 + n_3 \quad (2.1e)$$

$$\frac{d\phi}{dt} = n_2 \sigma c \phi - n_1 \sigma c \phi - \frac{\phi}{\tau_c} \quad (2.1f)$$

where n_{tot} is the total population density, n_i is the population density at level i , σ is the emission cross-section of the photons, ϕ is the photon density, c is the speed of light and τ_c is the cavity lifetime in the cavity. Assuming that the decay from level 3 to level 2 and from level 1 to level 0 is very rapid

$$\tau_{32} \approx 0 \quad (2.2a)$$

$$\tau_{10} \approx 0 \quad (2.2b)$$

$$\tau_{30} \approx \infty \quad (2.2c)$$

Thus the population density in level 3 and level 1 is approximately zero. The rate equations in (2.1) are thus simplified to

$$\frac{dn_2}{dt} = W_p n_0 - \frac{n_2}{\tau_{21}} - n_2 \sigma c \phi \quad (2.3a)$$

$$n_1 \approx n_3 \approx 0 \quad (2.3b)$$

$$n_{tot} = n_0 + n_2 \quad (2.3c)$$

$$\frac{d\phi}{dt} = n_2 \sigma c \phi - \frac{\phi}{\tau_c} \quad (2.3d)$$

The rate equations in (2.3) can be further simplified to

$$\frac{d\Delta n}{dt} = W_p (n_{tot} - \Delta n) - \frac{\Delta n}{\tau_{21}} - \Delta n \sigma c \phi \quad (2.4a)$$

$$\frac{d\phi}{dt} = \left(\Delta n \sigma c - \frac{1}{\tau_c} \right) \phi \quad (2.4b)$$

where $\Delta n = n_2 - n_1 \approx n_2$, the population inversion density.

At the beginning of the laser oscillation, the photon density is very small, so it can be considered to be negligible. And thus the threshold condition of the laser oscillation can be achieved by

$$\Delta n \sigma c - \frac{1}{\tau_c} \geq 0 \quad (2.5)$$

And at the oscillation threshold, the population inversion threshold, Δn_{th} , can be derived from (2.5) as

$$\Delta n_{th} = \frac{1}{\sigma c \tau_c} \quad (2.6)$$

At steady state laser oscillation, population inversion and photon density remain constant, thus the equations in (2.4) is equal to zero

$$\frac{d\Delta n}{dt} = W_{p(n_{tot}-\Delta n)} - \frac{\Delta n}{\tau_{21}} - \Delta n \sigma c \phi = 0 \quad (2.7a)$$

$$\frac{d\phi}{dt} = \left(\Delta n \sigma c - \frac{1}{\tau_c} \right) \phi = 0 \quad (2.7b)$$

The steady state photon density can be derived as

$$\phi = \frac{W_p \tau_{21} (n_{tot} - \Delta n_{th}) - \Delta n_{th}}{\Delta n_{th} \sigma c \tau_{21}} \quad (2.8)$$

Given that the final output power of the laser is determined by the lifetime of the photon caused by the output coupling τ_t

$$P_{out} = \frac{h\nu\phi V}{\tau_t} \quad (2.9)$$

where h is the Planck constant, V is the cavity volume and ν is the emission frequency.

Combining (2.8) and (2.9), the final output power is described as

$$P_{out} = \frac{h\nu V}{\tau_t} \cdot \frac{W_p \tau_{21} (n_{tot} - \Delta n_{th}) - \Delta n_{th}}{\Delta n_{th} \sigma c \tau_{21}} \quad (2.10)$$

2.1.2 Rate Equation for Three Level Laser System

Figure 2.2 Transition in a three level laser system

The atomic transitions in an ideal three level laser system is described in Figure 2.2. Similar to the four level system, we assume that the system is in equilibrium with no collision and is isolated. With rapid decay from level 3 to level 2, the population density in level 3 is approximately equals to zero. Thus the rate equations describing the three level laser system are [2]

$$\frac{dn_2}{dt} = W_p n_1 - \frac{n_2}{\tau_{21}} - n_2 \sigma c \phi + n_1 \sigma c \phi \quad (2.11a)$$

$$\frac{dn_1}{dt} = -W_p n_1 + \frac{n_2}{\tau_{21}} + n_2 \sigma c \phi - n_1 \sigma c \phi \quad (2.11b)$$

$$n_{tot} = n_1 + n_2 \quad (2.11c)$$

$$\frac{d\phi}{dt} = n_2 \sigma c \phi - n_1 \sigma c \phi - \frac{\phi}{\tau_c} \quad (2.11d)$$

These equations can be further simplified by substituting $\Delta n = n_2 - n_1$

$$\frac{d\Delta n}{dt} = W_p (n_{tot} - \Delta n) - \frac{n_{tot} + \Delta n}{\tau_{21}} - 2\Delta n \sigma c \phi \quad (2.12a)$$

$$\frac{d\phi}{dt} = \left(\Delta n \sigma c - \frac{1}{\tau_c} \right) \phi \quad (2.12b)$$

Using the same techniques as applied for the four level laser system, the threshold condition and the steady state output power of the three level laser system can be derived as

$$\Delta n_{th} = \frac{1}{\sigma c \tau_c} \quad (2.13a)$$

$$P_{out} = \frac{h\nu V}{\tau_t} \cdot \frac{W_p \tau_{21} (n_{tot} - \Delta n_{th}) - (n_{tot} + \Delta n_{th})}{\Delta n_{th} \sigma c \tau_{21}} \quad (2.13b)$$

2.1.3 Quasi-Three-Level Laser System

A quasi-three-level laser system cannot be classified as an ideal four or three level laser system. Trivalent Ytterbium and Holmium laser materials behave as a quasi-three-level laser system. At room temperature operation, the lower laser level has a certain amount of thermal population. In order to describe the quasi-three-level laser system, the rate equations must be modified. With reference to the four level laser system described in (2.1) and consider that the lower laser level population is a fraction (f_l) of the ground level population and the upper laser level population is a fraction (f_u) of the excited state population, the modified rate equations are

$$\frac{dn_2}{dt} = W_p n_0 - \frac{n_2}{\tau_{21}} - n_2 \sigma c \phi + n_1 \sigma c \phi \quad (2.14a)$$

$$n_1 = f_l n_0, n_3 = f_u n_2 \quad (2.14b)$$

$$n_{tot} = n_0 + n_1 + n_2 + n_3 \quad (2.14c)$$

$$\frac{d\phi}{dt} = n_2 \sigma c \phi - n_1 \sigma c \phi - \frac{\phi}{\tau_c} \quad (2.14d)$$

When f_l and f_u is zero, (2.14) reduces to the four level laser system described earlier. The rate equations in (2.14) is a simplified version of the quasi-three-level laser system. In order to fully describe the laser dynamics in quasi-three-level laser system, the upper

laser level and the lower laser level should be a thermal fractional portion in each of the manifolds that they belong to [3] - [6].

2.2 Nonlinear Optical Frequency Conversion

In this section, the basic foundation of nonlinear optical frequency conversion is presented. The theory of nonlinear optics is already well documented in literature [7]. This section begins with a brief introduction of the physics of nonlinear optics and the nonlinear interactions of plane waves. This is followed by introducing the important concepts of frequency conversion, such as phase-matching conditions and conversion efficiency of second harmonic generation. Finally, the theory is expanded to describe optical parametric amplification and oscillation.

2.2.1 Physics of Nonlinear Optics

In a linear dielectric medium, the linear relationship between the polarization and the electric field applied to the medium is [7]

$$P = \varepsilon_0 \cdot \chi \cdot E \quad (2.15)$$

where ε_0 is the permittivity of free space and χ is the dielectric susceptibility. Before the invention of laser and the related intense electric fields, all media are considered to have the linear response as described by (2.15). However, with the use of laser radiation, nonlinear effects such as frequency doubling [8] and intensity-dependent refractive indices start to appear. The optical nonlinearity of a dielectric medium transposes into a

nonlinear relationship between the polarizations induced in the medium and the applied electric field. Figure 2.3b describes this nonlinear relationship.

Figure 2.3 Relationship between polarization and applied electric field for (a) a linear dielectric medium and (b) a nonlinear dielectric medium.

When a small electric field is applied, the relationship between P and E is assumed linear. However, when E increases toward the interatomic electric field values, typically at 1kV/cm, highly nonlinear relationship occurs. One can perform power series expansion to (2.15) to represent the polarization in a nonlinear function of E

$$P = \epsilon_0 \cdot (\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \dots) \quad (2.16)$$

where the first term describes the usual linear polarization and the rest are the high order terms known as the nonlinear polarization

$$P_{NL} = \epsilon_0 \cdot (\chi^{(2)}E^2 + \chi^{(3)}E^3 + \dots) \quad (2.17)$$

Next the nonlinear interactions of plane waves are discussed. From the Maxwell's equations, the wave equation describing a plane wave propagating in a dielectric isotropic medium is derived as

$$\nabla^2 E - \frac{1}{c_0^2} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2} \quad (2.18)$$

where c_0 is the speed of light in vacuum. Substituting (2.16) and (2.17) into (2.18), splitting the polarization between linear and nonlinear terms and using c as the speed of light in medium, the nonlinear wave equation is obtained as

$$\nabla^2 E - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{NL}}{\partial t^2} \quad (2.19)$$

The coupled-wave approach is used to solve for the different fields propagating through the medium. Taking the example of three waves mixing, assume that the total electric field E is a superposition of three E fields of frequencies, ω_1 , ω_2 and ω_3 , propagating in the z direction

$$E(z, t) = \sum_{n=1,2,3} E_n(z) e^{i\omega_n t} + c. c. \quad (2.20)$$

where $E_n(z) = A_n e^{-ik_n z}$ is the complex amplitude of the field as it propagates through the material and $k_n = \omega_n/c$. The second-order nonlinear polarization in (2.17) can be written as

$$P_{NL} = 2\varepsilon_0 d_{\text{eff}} E^2 \quad (2.21)$$

where d_{eff} is the effective nonlinear coefficient which is commonly used in place of $\chi^{(2)}$ in equations for three wave mixing. Substituting (2.20) into (2.21), the resulting nonlinear polarization obtained is a sum of all the cross-product terms

$$P_{NL} = 2\varepsilon_0 d_{\text{eff}} \sum_{n,p=1,2,3} E_n E_p e^{i(\omega_n + \omega_p)t} \quad (2.22)$$

providing a series of different frequency components. By substituting (2.20), (2.21) and (2.22) into (2.19), a single differential equation with all the frequency components is obtained. If the frequencies, ω_1 , ω_2 and ω_3 , are distinctive, three separate equations can be used to equate the terms on both side of (2.22) at each of the frequency separately.

The result is presented in the form of three Helmholtz equations with its associated sources

$$(\nabla^2 + k_1^2)E_1 = -S_1 \quad (2.23a)$$

$$(\nabla^2 + k_2^2)E_2 = -S_2 \quad (2.23b)$$

$$(\nabla^2 + k_3^2)E_3 = -S_3 \quad (2.23c)$$

where $S_{1,2,3}$ is the amplitude of the source term associated with each frequency. For the three waves to be coupled by the medium, their frequencies must be related to one another, such as $\omega_1 + \omega_2 = \omega_3$. Hence, the following three waves mixing coupled equations are derived

$$(\nabla^2 + k_1^2)E_1 = -2\mu_0\omega_1^2\varepsilon_0d_{\text{eff}}E_3E_2^* \quad (2.24a)$$

$$(\nabla^2 + k_1^2)E_2 = -2\mu_0\omega_2^2\varepsilon_0d_{\text{eff}}E_3E_1^* \quad (2.24a)$$

$$(\nabla^2 + k_1^2)E_3 = -2\mu_0\omega_3^2\varepsilon_0d_{\text{eff}}E_1E_2 \quad (2.24a)$$

It is convenient to normalise the complex envelopes of the plane waves by introducing the variables $a_p = A_p / (2\hbar\omega_p / \varepsilon_0 c_0 n_p)^{1/2}$, with n_p the refractive index of the material at frequency ω_p . The electric field can be written as

$$E_p(z) = \left(\frac{2\hbar\omega_n}{\varepsilon_0 c_0 n_p} \right)^{1/2} a_p e^{-ik_p z} \quad (2.25)$$

The intensities of the waves can be written as

$$I_p = \hbar\omega_p |a_p|^2 \quad (2.26)$$

where $|a_p|^2$ equals to the flux of photons at frequency ω_p . Assuming the coupling between the waves is weak, the variation of the amplitudes with distance is slow and thus taken as approximately constant over the length of the wavelength. This is known

as slowly-varying envelop approximation and it shows that the second order derivative terms are negligible. Thus the three wave mixing coupled equations become

$$\frac{da_1}{dz} = -iga_3a_2^*e^{-i\Delta k \cdot z} \quad (2.27a)$$

$$\frac{da_2}{dz} = -iga_3a_1^*e^{-i\Delta k \cdot z} \quad (2.27b)$$

$$\frac{da_3}{dz} = -iga_1a_2e^{-i\Delta k \cdot z} \quad (2.27c)$$

where

$$g = \left(\frac{2\hbar\omega_1\omega_2\omega_3d_{\text{eff}}^2}{n_1n_2n_3\epsilon_0c_0^3} \right)^{1/2} \quad (2.28)$$

and

$$\Delta k = k_3 - k_2 - k_1 \quad (2.29)$$

Δk is the phase vector mismatch, representing the error in the phase-matching condition.

2.2.2 Second Harmonic Generation Conversion Efficiency and Phase-Matching Conditions

A degenerate case of three-wave mixing is second harmonic generation, where the frequencies are related by

$$\omega_1 = \omega_2 = \omega \text{ and } \omega_3 = \omega_1 + \omega_2 = 2\omega$$

Two photons of frequency ω add up to form a photon at frequency 2ω . Two coupled wave equations in (2.27) describe the interaction

$$\frac{da_1}{dz} = -iga_3a_1^*e^{-i\Delta k \cdot z} \quad (2.30a)$$

$$\frac{da_3}{dz} = -i\frac{g}{2}a_1^2e^{-i\Delta k \cdot z} \quad (2.30b)$$

$$g = \left(\frac{4\hbar\omega^3 d_{\text{eff}}^2}{n_1^2 n_3 \varepsilon_0 c_0^3} \right)^{1/2} \quad (2.30c)$$

$$\Delta k = k_3 - 2k_1 \quad (2.30d)$$

Integration on these equations and using the appropriate boundary conditions, such as $a_3(0) = 0$, yield the relationship of the amplitudes along z axis. By assuming perfect phase-matching where $\Delta k = 0$ and the photon number $|a_1(z)|^2 + 2|a_3(z)|^2$ is conserved, the following is derived

$$a_1(z) = a_1(0) \operatorname{sech} \frac{g a_1(0) z}{\sqrt{2}} \quad (2.31a)$$

$$a_3(z) = -\frac{i}{\sqrt{2}} a_1(0) \operatorname{sech} \frac{g a_1(0) z}{\sqrt{2}} \quad (2.31b)$$

The conversion efficiency of the second harmonic generation can be written as

$$\frac{I_3(L)}{I_1(0)} = \tanh^2 \frac{\gamma L}{2} \quad (2.32a)$$

$$\gamma = \frac{2g a_1(0)}{\sqrt{2}} \quad (2.32b)$$

For a long device, large input intensity or large nonlinear coupling, γL is large. This results in the conversion efficiency approaches one and implies that all the input power is converted into the second harmonic power. For small γL , $\tanh^2 x \sim x$, thus the conversion efficiency can be expressed as

$$\frac{I_3(L)}{I_1(0)} = \frac{8\pi^2 d_{\text{eff}}^2}{n_1^2 n_3 \varepsilon_0 c_0^3 \lambda^2} L^2 I_1(0) = C^2 \frac{L^2}{A_{\text{eff}}} P(0) \quad (2.33a)$$

$$C^2 = \frac{8\pi^2 d_{\text{eff}}^2}{n_1^2 n_3 \varepsilon_0 c_0^3 \lambda^2} \quad (2.33b)$$

The above equation is expressed using $P(0) = I(0) \cdot A_{\text{eff}}$, the input power and the effective cross-sectional area of the nonlinear interaction. C^2 is a normalized conversion efficiency that is proportional to d_{eff}^2/n^3 , which is the parameter of the nonlinear material. Thus d_{eff}^2/n^3 can be used as a figure of merit to compare different nonlinear materials.

Figure 2.4 Tuning curve results from the sinc^2 term as a function of dephasing from the optimal phase matching condition.

The above derivation assumes perfect phase-matching where $\Delta k = 0$. This condition is usually not fulfilled in materials. The effect of phase mismatch can be studied by considering the low conversion scenario, assuming the amplitude of the fundamental wave remains approximately constant across the nonlinear material. The conversion efficiency of the second harmonic generation is expressed as

$$\eta = \frac{I_3(L)}{I_1(0)} = C^2 \frac{L^2}{A_{\text{eff}}} P(0) \text{sinc}^2\left(\frac{\Delta k L}{2\pi}\right) \quad (2.34)$$

where $\text{sinc}(x) = \sin(x) / x$. The effect of phase mismatch is to reduce the efficiency of the interaction by the $\text{sinc}^2(\Delta k L / 2\pi)$ factor. When $\Delta k = 0$, the sinc^2 factor is one, thus the

second harmonic power grows quadratically with the fundamental wave power. When $\Delta k \neq 0$, the sinc^2 factor is smaller than one, as shown in Figure 2.4. The width of the curve is inversely proportional to the length of the material, implying that increase in material length will narrow the phase matching condition. The length, $L_c = 2\pi / \Delta k$, is the coherent length, where it is defined as the maximum length over which nonlinear interaction is efficient.

Figure 2.5 Birefringent phase matching in a negative uniaxial material, with fundamental propagating with ordinary index, and second harmonic with extraordinary index.

Naturally, all linear optical materials exhibit dispersion of the refractive index, thus it is impossible to obtain perfect phase matching in these media. So in general, $\Delta k \neq 0$, and the conversion efficiency is very small. To obtain perfect phase matching, the phase matching condition can be fulfilled by using the birefringence of nonlinear materials [9]. Figure 2.5 shows the dispersion curve of a negative uniaxial material in both the ordinary and extraordinary wave. As shown, perfect phase matching can be achieved by propagating the fundamental and second harmonic wave with orthogonal polarizations.

It is worthy to note that nonlinear coefficients only couple certain field directions depending on the symmetry of the materials. Birefringent phase matching only occurs under these constraints.

2.2.3 Optical Parametric Amplification and Oscillation

Figure 2.6 Schematic of optical parametric amplification process

Next, the results from previous section is further implemented to the conversion efficiency of OPA, similar to the process known as difference frequency generation (DFG), illustrates in Figure 2.6. In OPA, a high power pump beam at frequency ω_3 is combined with a lower power signal beam at frequency ω_1 , yielding a large pump to signal intensity ratio. In contrast, the intensity between the pump and signal in the DFG process is comparable. The pump frequency is higher than the signal frequency. Their nonlinear interaction generates photon at frequency $\omega_2 = \omega_3 - \omega_1$, known as the idler beam. Conventionally, the signal beam has a higher frequency than the idler beam. Using the coupled wave equations for three wave mixing in (2.27) and assuming the undepleted pump approximation is valid, the conversion efficiency is derived as

$$P_2 = C^2 \frac{L^2}{A_{\text{eff}}} P_3 P_1 \text{sinc}^2 \left(\frac{\Delta k L}{2\pi} \right) \quad (2.35a)$$

$$C^2 = \frac{8\pi^2 d_{\text{eff}}^2}{n_1 n_2 n_3 \varepsilon_0 c_0^3 \lambda_2^2} \quad (2.35b)$$

The conversion efficiency obtained is similar to efficiency for second harmonic generation. Similar to second harmonic generation, one need a larger pump and signal powers and longer crystal with phase matching conditions satisfied to achieve higher conversion efficiency. Besides generating the idler frequency, the decomposed pump photons also produce photon at the signal frequency. This provides amplification at both the signal and idler wavelengths, and thus called parametric amplification.

When the nonlinear medium performing OPA is placed in a resonator with the pump source, the parametric gain oscillates within the resonator. Thus an OPO is formed. The parametric gain of the nonlinear material in the OPO can be expressed as

$$\gamma = \left(\frac{8\omega_1 \omega_2 d_{\text{eff}}^2}{n_1 n_2 n_3 \varepsilon_0 c_0^3} I_3 \right) \quad (2.36)$$

The parametric gain depends on the pump intensity. Similar to laser threshold, there is also a threshold intensity in OPO in which the gain equals to the losses in the cavity. When above threshold, quantum noise fluctuations of the signal and idler photons begin to amplify in the resonator. The oscillator can either be singly resonant or doubly resonant. In singly resonant oscillator, either the signal or the idler is oscillating in the cavity. In contrast, both signal and idler are oscillating in a doubly resonant oscillator. OPOs are important in many applications as it is capable to generate a widely tuneable MIR coherent radiation sources and providing more power than a simple OPA setup.

2.3 Chirped Pulse Amplification

Figure 2.7 Typical layout for a chirped pulse amplification system, although there are many additional structures that can vary from system to system, shown here are the essential pieces.

[11]

Typical mode-locked fs oscillator delivers laser pulses with energy in the nJ range. In order to increase the pulse energy to μJ , mJ or even Joules range, it requires optical amplification. However, the amplification mechanism needs to deal with the optical damage and nonlinear effects associated with the high peak intensities during the amplification process. The typical method currently used for the very high intense laser pulse amplification is the chirped pulse amplification (CPA) of mode-locked solid state laser system. CPA first introduces additional chirp to the fs laser pulse, which disperses the frequency components of the pulse temporally. This results in a reduction of the pulse peak intensity as the temporal pulse duration increases. Amplification of the stretched pulse intensity by multiple orders of magnitude can be realised without the

risk of undesired non-linear effects and optical damage. After amplification of the nJ pulse to the desired pulse energy, the laser pulses are recompressed by temporally overlapping the frequency components, thereby obtaining a much higher peak intensities [10]. A typical schematic layout of the CPA system is shown in Figure 2.7.

2.3.1 Pulse Stretcher

In the schematic layout shown in Figure 2.7, a mode-locked oscillator first creates the ultrashort fs pulses. The ultrashort fs pulses emitted from the oscillator are temporally stretched using diffraction gratings and an imaging system. This is the same principle used for the compensation of dispersion in an ultrafast fs oscillator. The exception is that the diffraction gratings used in the stretcher have a much greater angular dispersion as compared to the prisms used in the oscillator. This allows a large amount of dispersion to be introduced to the fs pulse [12]. In addition, positive dispersion can be introduced to the pulse using an imaging system between the gratings.

The pulse stretcher can be implemented in several different ways. The most common stretcher arrangements are the Martinez and the Offner stretchers. The use of grating based stretchers is first suggested by Martinez [13]. Figure 2.8 shows the schematic setup of the Martinez grating stretcher. Different spectral components take a different optical path length through the stretcher. Martinez demonstrated that a grating pair with a pair of lenses forming a telescope between the gratings can be used as positive dispersion stretcher. To understand this visually, we can recognize that the telescope forms an image of the first grating behind the second grating, resulting in a negative path length and therefore opposite signed-dispersion. The amount of positive or negative

group delay dispersion added to the pulse can also be manipulated based on the geometrical properties of the stretcher. Thus, as long as the distance of the gratings to the telescope are each less than the focal length of the lenses, positive dispersion can be satisfied. The original Martinez configuration is, however, the least commonly used in the current stretcher configurations. This is because Martinez stretchers use lenses rather than curved mirrors as imaging components. The lenses lead to more serious aberrations to the pulse wavefront and can distort ultrafast pulses [14].

Figure 2.8 Schematic setup of a Martinez grating stretcher.

Typically, positive dispersion is introduced in stretcher setup while negative dispersion for compressor setup. This is partly due to the past implementations where there are good reasons to use pulses with positive chirp. Historically, CPA was first implemented with the use of a long optical fibre for pulse stretching. At the typical laser wavelengths of 800-1064 nm for most CPA systems, materials are normally dispersive. This means that the longer wavelengths propagate faster (i.e. experience lower index of refraction) than short ones. Thus, fibre propagation naturally yields a pulse with positive chirp. This positive chirp can be counteracted by the negative chirp of a grating pair. Grating compressors are simple, and use only reflective optics, thus avoiding nonlinear pulse distortions. There are also other reasons to employ positively chirped, rather than negatively-chirped, in the CPA. The amplifier generally contains optical components

with positive dispersion. As the negatively-chirped pulse propagates through the amplifier system, it tends to shorten in duration. This makes it more difficult to avoid the problems that CPA is intended to overcome, and thus makes it necessary for the pulse stretcher to introduce substantially more negative chirp on the pulse to compensate the positive chirp introduced by the amplifier. In contrast, positive dispersion further lengthens a positively-chirped pulse.

2.3.2 Regenerative Amplifier

The geometries for solid state laser amplifiers vary greatly depending on the material and energy level expected to achieve. Amplifier geometries can vary from single-pass amplifier to multi-pass amplifier, and even cavity based amplifier such as regenerative amplifier. The choice of amplifier configurations depends greatly on the geometrical shape of the gain media itself. Long rods and slabs are more typically used in a single-pass amplifier while thinner transmission or reflection based gain media are more likely to be used in a multi-pass amplifier. Combination of different amplifier arrangements are also often used [10].

The schematic setup of a regenerative amplifier is shown in Figure 2.9. The regenerative amplifier works like a cavity with mirror at both ends. A pulse is coupled into the amplifier cavity using a Pockels cell, a $\lambda/4$ -plate and a thin film polarizer as the optical switch. It is then amplified by making multiples passes in the cavity before it is extracted out by the optical switch, a Faraday rotator and another thin film polarizer. A regenerative amplifier operation comprises of three phases [2]:

- Pump phase, with Pockels cell voltage at zero
- Amplification phase, with Pockels cell switched to $\lambda/4$ retardation
- Cavity dump phase, Pockels cell voltage switched back to zero voltage

Figure 2.9 Schematic setup of a regenerative amplifier. A Pockels cell, combined with a $\lambda/4$ -plate and a thin-film polarizer, acts as an optical switch. The Faraday rotator is used for separating input and output pulses [16].

At the pump phase, the Pockels cell voltage is at zero. The laser crystal is either CW pumped or pulsed pumped. During the pump phase, the laser oscillation is suppressed by introducing a 90° polarization rotation to the p-polarised radiation from the Brewster-cut laser slab, using $\lambda/4$ -plate and rear-mirror combination. A p-polarized pulse from a fs mode-locked oscillator passes through the first thin film polarizer, rotated by 45° rotation at the $\lambda/2$ -plate and a further 45° rotation at the Faraday rotator, to s-polarization. It is reflected by the second thin film polarizer into the cavity. The pulse is later rotated to p-polarization after double-passed through the $\lambda/4$ -plate and reflected from the first rear mirror. Thus, the pulse passes through the second polarizer and completes its first round trip. At the second round trip, the pulse is rotated again to s-polarized and reflects

off the second polarizer. The s-polarized pulse, after a 45° rotation from the Faraday rotator and compensated by the $\lambda/2$ -plate, remains as s-polarized pulse and reflects off the first polarizer. The small-signal double pass gain is measured by taking the ratio of the average power at the output with and without the pump source turned on.

At the amplification phase, the Pockels cell is switched to $\lambda/4$ retardation. A pulse is trapped inside the cavity when the $\lambda/4$ voltage is applied to the Pockels cell. This is performed as soon as the pulse leaves the Pockels cell and travels towards the direction of the amplifier crystal. As the $\lambda/4$ voltage is applied to the Pockels cell, the returning pulse from the amplifier crystal does not experience the polarization rotation described during the pump phase. Thus the pulse stays within the cavity and amplifies to a maximum pulse energy. The duration of build-up for the amplification depends on the gain of the system and also the characteristic of the Q-switched envelope of the cavity dumped pulse. Higher gain requires shorter build-up time, with typical time period of less than 100ns in a very high gain system, to a few hundreds of ns for a lower gain system. When the Pockels cell introduces $\lambda/4$ retardation, new pulses that arrive at the cavity similar do not experience the polarization rotation, and thus reject from the amplifier after the reflection from the first cavity mirror.

At the cavity dump phase, Pockels cell voltage switches back to zero voltage. After multiple passes in the cavity, gain saturation results in a pulse energy that reaches its maximum. At this stage, the Pockels cell voltage is switched to zero. Like previous case, the switching starts when the pulse just leave the Pockels cell. Upon returning, the polarization of the pulse is rotated by 90° to the s-polarization and is reflected by the two polarizer to the output of the regenerative amplifier.

2.3.3 Pulse Compressor

Figure 2.10 Schematic setup of a Treacy grating compressor.

After amplification, the stretched pulses are typically recompressed back to ultrafast fs pulses using a grating compressor as shown in Figure 2.10. The dispersion acquired by passing a laser pulse through a pair of parallel grating is described by Treacy in [15]. He showed that for any angle of incidence and grating separation, the group velocity dispersion is always negative. The frequency components are angularly dispersed and travel through different optical path lengths in order to re-overlap them longitudinally, producing high energy ultrashort pulses [12]. It should be noted that either Treacy or Martinez design can be used interchangeably as a stretcher or compressor. However the Treacy design is normally used as a compressor since it has the least number of optical elements. In order to avoid large amount of energy loss in the compressor, gratings with high efficiency are selected. In contrast, low efficiency gratings in the stretcher are of little importance as the lost energy can be recovered in the amplifiers stages.

Chapter 3 Development of Ho:YAG Laser Sources

Solid state 2 μ m lasers, such as Ho:YAG laser, are widely used in various scientific and technical applications today. It is also an efficient pump source for MIR OPO with ZGP crystal. The trivalent rare earth ions Holmium (Ho³⁺) doped materials, which works as a quasi-three-level laser system, are commonly used to obtain the 2 μ m lasers. ZGP is a very promising nonlinear crystal to perform down-conversion of wavelength to the MIR range due to its excellent mechanical and thermal properties. It also has high non-linear coefficients. However, the transparency of ZGP crystal only begins from wavelength beyond 2 μ m. Thus, in order to achieve nonlinear optical conversion using ZGP crystal, pulsed laser with emission wavelength of >2 μ m is essential. Here, the development of 2 μ m Ho³⁺ laser as the primary pump source for the ZGP OPO is studied.

In this chapter, the trivalent Holmium ions are introduced, with in depth discussion of its properties in YAG host medium. Subsequently, four different classes of Ho:YAG laser sources are demonstrated, which comprise of the following:

- High repetition rate / high average power Tm:Fiber pumped Ho:YAG oscillator.
- High pulse energy Tm:Fiber pumped Ho:YAG Master Oscillator Power Amplifier (MOPA).
- Direct diode-pumped Ho:YAG oscillator in CW and pulse operation.
- Tm:Fiber pumped CW Ho:YAG ceramic oscillator.

The development of a Tm:Fiber pumped Ho:YAG laser oscillator operating at high repetition rate of 10kHz is discussed in detail. A variable output coupling configuration

was used to optimize and achieve the highest possible output power of the oscillator. This laser cavity was subsequently applied as the pump source for an intra-cavity ZGP OPO for high average power MIR generation. A customised Ho:YAG laser with high pulse energy of $>80\text{mJ}$ operating at 100Hz built by a commercial collaborator is described. This high pulse energy Ho:YAG laser source, operates in a MOPA configuration pumped by two Tm:Fiber lasers, was used for pumping the conventional extra-cavity ZGP OPO to achieve high energy MIR pulses. This laser source was also used to investigate MIR generation in an uncoated wedge ZGP OPO, which will be presented in the next chapter.

In addition to the Tm:Fiber laser pumped Ho:YAG single crystal, we also evaluate the potential of two unconventional classes of Ho:YAG laser which use diode laser as the pump source and holmium doped ceramic as the gain medium. Diode laser is relatively smaller in size as compared to the high power Tm:Fiber laser. By replacing the Tm:Fiber laser with a diode laser, a compact direct diode-pumped Ho:YAG laser system can be developed. As compared to high power diode laser emitting in the wavelengths between $700\text{-}1000\text{nm}$, the technology of diode laser with emission wavelength in the Ho:YAG absorption band at $1.9\mu\text{m}$ is less mature. By using the high power $1.9\mu\text{m}$ fiber-coupled diode laser, which became recently available in the commercial market, we performed preliminary evaluations on direct diode-pumped Ho:YAG single crystal oscillator in both CW and ns pulsed operations.

Research in ceramic laser gain medium has made significant impact due to the capabilities in synthesizing of highly transparent polycrystalline ceramics. As compared to single crystal gain medium, ceramic material can be produced in large quantities and high doping concentration. Production costs for ceramics are also relatively cheaper as

compared to single crystals. Ceramic gain medium can be made with inhomogeneous doping concentration profile. This means that engineering on the doping concentration profile of the laser ions can be achieved. Proper designing of the doping concentration profile can improve the absorption efficiency of the pump source and reduce thermal lensing issue of the gain medium. We evaluated a CW Tm:Fiber pumped ceramic Ho:YAG oscillator and compared its performance with typical single crystal Ho:YAG oscillator.

3.1 Properties of Trivalent Holmium Ions

The first demonstration of Holmium laser was a Holmium doped Er_2O_3 host crystal in 1966 [17]. This is a co-doped system, where the Erbium co-doping provides a broad absorption band for lamp pump configuration. To reduce the threshold pump power, where it is closely related to the thermal population in the Holmium lower laser Stark level, the laser operation was performed at cryogenic temperature of 77K. Since the 1960s [17]-[18], the Holmium laser has progress in terms of the output power, slope efficiency and laser configurations.

Yttrium Aluminium Garnet (YAG) is an optically isotropic crystal which is mechanically and chemically robust with good thermal and optical characteristics. These properties have led it to be one of the host laser material chosen for Holmium. The absorbance and emission of Ho:YAG gain medium is shown in Figure 3.1 and Figure 3.2, respectively [19]. The peak absorption wavelength of Ho:YAG is at 1908nm. The upper laser level lifetime of Ho:YAG is 8.5ms [20], which is $\sim 30\times$ longer than Nd:YAG [21]. Henceforth, this provides an excellent energy storage in the crystal and

therefore has the potential for high energy Q-switched pulses. An in-band pumped Ho:YAG, where the crystal is excited with a pump wavelength that couples the ground energy state with the metastable upper laser, can be possible with low quantum defect of ~9%. Due to the low thermal contribution from the low quantum defect, potential for good power-scalability and efficient operation can be realised. Other thermal problems, such as lensing, higher ground state population, depolarization and thermal fracture, will also be minimised.

Figure 3.1 Ho:YAG Absorbance for unity concentration per meter with respect to wavelength

[19].

Figure 3.2 Ho:YAG Normalized emission ($\times 10^{-24}$) with respect to wavelength [19].

In conjunction with good atmospheric transmission, the Holmium laser emitting at eye-safe wavelength in the $2\mu\text{m}$ regime is an excellent source for environmental monitoring [22]-[24] and remote sensing [25]. As compared to Thulium based laser, the longer lasing wavelength from Holmium laser is an important pump source for ZGP and GaAs based OPO, generating MIR wavelengths between $3\text{-}5\mu\text{m}$. This MIR window has various applications for gas sensing, laser ranging and infrared countermeasures in the military sector [26]-[29] and use as a surgical tool in the medical sector [30]-[32].

The trivalent Holmium active ions can achieve infrared laser oscillation around $2.1\mu\text{m}$ with the $^5\text{I}_7 \rightarrow ^5\text{I}_8$ transition and around $2.9\mu\text{m}$ with the $^5\text{I}_6 \rightarrow ^5\text{I}_7$ transition [33]. The energy-level diagram of Ho^{3+} ions shown in Figure 3.3 describes the transition of the pump and the laser that contribute to the $2.1\mu\text{m}$ and $2.9\mu\text{m}$ oscillation. Laser emission around $2.1\mu\text{m}$ in Ho^{3+} ions requires optical pump wavelength at $1.9\mu\text{m}$. This wavelength

is not available as standard commercially high power laser diodes. However, the co-doping of Tm-Ho in a laser host material allows the direct pumping using high power laser diodes. Thulium has strong absorption band near 780-800nm, which can be easily pumped by laser diodes. Thus, Thulium is used as a sensitizer ion that absorbs the strong pump photon and subsequently transfers these absorbed energy to the Ho^{3+} ions.

Figure 3.3 Energy-level diagram of Ho^{3+} ions. Pump wavelength of $\sim 1.9\mu\text{m}$ ($\sim 1.1\mu\text{m}$) excites atom from $^5\text{I}_8$ level to $^5\text{I}_7$ ($^5\text{I}_6$) level. The corresponding emission wavelength of $\sim 2.1\mu\text{m}$ ($\sim 2.9\mu\text{m}$) is emitted when the atom relaxes back from the high energy level to the lower energy level.

An alternative to Tm-Ho co-doped laser gain medium to achieve $2.1\mu\text{m}$ radiation is by direct laser pumping of the Ho^{3+} ions from $^5\text{I}_8$ to $^5\text{I}_7$ using Thulium solid state or fiber laser emitting at $1.9\mu\text{m}$. This direct laser pumping method is preferred to avoid the average power limitation of the sensitized Tm-Ho laser system. Several advantages such as low quantum defect, elimination of energy transfer, low upconversion losses, high

short-pulse extraction and reduced sensitivity of gain versus temperature are offered by the direct laser pumping approach.

3.2 High Repetition Rate Tm:Fiber pumped Ho:YAG Laser with Variable Output Coupling

3.2.1 Introduction

The Ho:YAG laser is typically pumped by diode-pumped Tm solid state laser or Tm:Fiber laser tuned to centre wavelength of 1908nm, corresponding to the peak absorption wavelength of Ho:YAG. The trivalent Holmium has relatively long upper-level lifetime and large stimulated emission cross-section of $1.0 \times 10^{-19} \text{cm}^2$. These properties are key parameters for generating high energy nanosecond laser pulse output by applying Q-switching technique. In this experiment, we used a commercial Tm:Fiber laser to pump a Ho:YAG rod. Nanosecond pulses were generated using electro-optics Q-switch modulator operating at 10kHz. Output coupler for the Ho:YAG laser was optimized using a set of $\lambda/4$ -plate and a thin film polarizer (TFP). The output coupling of the laser can vary by rotating the slow axis of the $\lambda/4$ -plate with respect to the polarization axis of the TFP. The effective reflectivity of the laser output coupler can be expressed using

$$R = \cos^2(2\theta)$$

where θ is the angle between the slow axis of the $\lambda/4$ -plate and the polarization axis of the TFP. With this configuration, the output coupling of the laser can be easily optimized to achieve the maximum output power from the laser cavity.

3.2.2 Experimental Setup

Figure 3.4 Schematic Setup of Ho:YAG Laser. M1: HR Mirror at 1.908 μm and 2.09 μm ; M2: Dichroic Mirror HT@1.908 μm , HR@2.09 μm ; M3: HR Mirror at 2.09 μm ; L1: Bi-convex lens, $f=850\text{mm}$.

The schematic diagram of the experimental setup is shown in Figure 3.4. The pump source is a single mode 50W Tm:Fiber laser (Model TLR-50-1908 commercially available from IPG Photonics) with its emission wavelength centred at 1908nm, close

to the maximum of the Holmium absorption line. The Tm:Fiber laser emits a collimated beam with approximately 1.2mm diameter and is imaged into the Ho:YAG rod by lens L1 ($f = 850\text{mm}$). M2 is a flat dichroic mirror with high transmission at $1.9\mu\text{m}$ and high reflection at $2.1\mu\text{m}$ at a 45° angle of incidence. M1 is a flat mirror with high reflection for both $1.9\mu\text{m}$ and $2.1\mu\text{m}$, providing a double-pass pumping configuration. A Ho:YAG laser rod with length 30mm, diameter 3mm and doping concentration of 0.7% is used in this experiment. The Ho:YAG laser cavity is formed by mirror M1 and M3, and lens L1 is used to create a stable cavity between M1 and M3. The resonator mode radius is approximately $600\mu\text{m}$ at M3. M3 is a flat mirror with high reflection for $2.1\mu\text{m}$. The Ho:YAG rod is thermal-electric cooled (TEC) and its temperature is maintained at 20°C .

The variable output coupling of the Ho:YAG laser is performed by the TFP and the $\lambda/4$ -plate within the cavity. The TFP forces the Ho:YAG laser to oscillate in the linearly p-polarized direction between the TFP and M3. The p-polarized beam, after passing through the $\lambda/4$ -plate towards the Ho:YAG rod gets reflected by M1 and passes through the $\lambda/4$ -plate for the second time. The polarization of the beam re-orientates after passing through the $\lambda/4$ -plate twice. Thus the beam consists of both p- and s-polarized components. When the beam reaches the TFP, the p-polarized component transmits through while the s-polarized component is reflected as the Ho:YAG laser output. By adjusting the angle of the slow axis with respect to the horizontal axis, the output coupling of the Ho:YAG laser can be adjusted. This allows the optimization of the output coupling of the laser without changing any output coupler or optical components in the cavity. The Ho:YAG laser is Q-switched using a thermally compensated double-crystals RbTiOPO_4 (RTP) Pockels cell. The RTP Pockels cell operates as a $\lambda/4$ -plate when high voltage is applied during hold-off. The Q-switch operates at 10kHz repetition rate.

3.2.3 Results and Discussion

Figure 3.5 Ho:YAG output power vs $\lambda/4$ -plate Axis Angle. Blue curve shows the output power of the Ho:YAG laser, pumped with 46W of Tm:Fiber laser, with respect to the slow axis angle of the $\lambda/4$ -plate. Red curve shows the theoretical effective output coupler reflectivity.

CW operation of the Ho:YAG laser was achieved without the electro-optic Q-switching element. The CW output power of the Ho:YAG laser with respect to the angle between the slow axis of the $\lambda/4$ -plate and the polarization of the TFP at maximum pump power of 46W is shown in Figure 3.5. The solid curve shows the calculated reflectivity of the output coupling. Optimization of the output coupling of the laser was achieved by rotating the $\lambda/4$ -plate over an angle ranging from 0° to 35° . The optimum effective output coupling for this laser setup was around $R=45\%$. As the effective output coupling

increases, the output power decreases significantly. This gives a strong representation of the relationship between the changed in the effective output coupling and the rotation of the $\lambda/4$ -plate.

Figure 3.6 Ho:YAG Laser Output Power vs Tm:Fiber Pump Input Power. Red curve shows the performance of the Ho:YAG laser operating in CW mode. Blue curve shows the performance of the Ho:YAG laser operating in 10kHz pulse repetition rate.

The output power of the Ho:YAG laser as a function of the Tm:Fiber pump power incident onto the Ho:YAG rod is shown in Figure 3.6. The result shows both the CW and pulsed operation at a repetition rate of 10kHz. The performance of the pulsed operation was slightly better than the CW operation. This was due to the higher gain of the laser when operate in pulsing mode. The highest laser output power was measured at 21W with the maximum available pump power. The pump threshold power for CW and pulsed operation was approximately 15.7W and 13.3W respectively. The high pump

threshold was the result of a large pump beam diameter of 1.2mm. The slope efficiency for both operations was close to 65%. Figure 3.6 also shows the corresponding optical-to-optical efficiency of the laser. With the laser operating at pump power approximately three times above threshold, the optical-to-optical efficiency achieved was 44%. FWHM pulse width of approximately 100ns was measured at 10kHz with maximum pump power. The laser output beam was focused with a 125mm lens and its beam profile was measured over a distance using a pyroelectric camera. The M^2 of the output beam was calculated to be <1.1 , as described in Figure 3.7.

Figure 3.7 Ho:YAG Laser Beam waist as function of position after a focus lens with $f=125\text{mm}$.

The calculated M^2 , based on curve fitting of the beam propagation equation, is near diffraction limit of <1.1 .

3.3 High Energy Ho:YAG MOPA Laser

3.3.1 Introduction

In this section, we describe a customized commercial high energy Ho:YAG laser used as the primary pump source for high energy ZGP OPO. The high energy Ho:YAG laser was built in a MOPA configuration as described in [34] by a commercial collaborator, IBL Innovative Berlin Laser GmbH. It can achieve a maximum of pulse energy of 125mJ with 20ns pulse width, at a repetition rate of 100Hz.

3.3.2 Laser Configuration

Figure 3.8 Schematic layout of the high energy Ho:YAG MOPA laser.

Figure 3.8 shows the schematic layout of the Ho:YAG MOPA double-pass system developed by IBL Innovative Berlin Laser GmbH. The system consisted of an end-pumped Ho:YAG oscillator rod pumped with a 1.9 μ m 50W CW Tm:Fiber laser from IPG Laser in a double-pass pump configuration. Q-Switching operation in the oscillator was realized using RTP Pockels cell operating as a $\lambda/4$ -plate when a high voltage was applied. The Q-switched Ho:YAG oscillator consisted of highly reflective mirror, an RTP electro-optic modulator, a TFP, a folding mirror, two dichroic mirrors and an output coupler.

The output laser pulse from the oscillator entered the amplifier stage through a $\lambda/2$ -plate and a TFP, and reflected by a dichroic mirrors into the Ho:YAG amplifier rod. The Ho:YAG amplifier rod was pumped by a 1.9 μ m 70W CW Tm:Fiber laser from IPG Photonics Corporation in a double-pass pump configuration. The amplified pulse was directed back into the amplifier rod, through a $\lambda/2$ -plate and series of TFPs, for a second pass amplification before exiting the amplifier stage.

The actual Ho:YAG MOPA laser used for our ZGP OPO experiment was in a single-pass amplifier configuration. The TFP after the amplifier rod experienced the risk of encountering two concurrently high energy pulses incident on it at the same time. This had resulted in frequent optical damage on that TFP. Thus, in order for long term reliable operation, the TFP after the amplifier rod was removed.

3.3.3 Laser Performance and Parameters

Figure 3.9 shows the laser pulse energy and the corresponding pulse width of the high energy Ho:YAG MOPA laser at different pulse repetition rate between 100Hz to 1kHz.

The laser was capable of generating 15.6mJ of 2.09 μ m pulse energy with 66ns pulse width, at 1kHz operation, translating to an average laser output power of 15.6W. As the pulse repetition rate decreased, the pulse energy increased while the pulse width reduced. At 100Hz operation, the MOPA laser generated 82mJ of 2.09 μ m pulse energy with 18ns pulse width. These data converted to an average laser output power of 8.2W and pulse peak power of 4.56MW.

Figure 3.9 Ho:YAG MOPA laser pulse energy / pulse width with respect to pulse repetition rate.

M^2 measurement was performed for the Ho:YAG MOPA laser operation at 100Hz. A beamsplitter was used to leak a small portion of the pulse laser output for the M^2 measurement. The laser output beam was focused with a 125mm lens and its beam profile was measured over a distance using a pyroelectric camera. The M^2 of the output

beam from the Ho:YAG MOPA laser operating at 100Hz was calculated to be ~3.7, described in Figure 3.10.

Figure 3.10 Ho:YAG MOPA laser M^2 measurement, 82mJ pulse energy operation at 100Hz pulse repetition rate.

3.4 Direct Diode pumped Ho:YAG Oscillator

The experimental setup for a direct diode-pumped Ho:YAG slab laser is shown in Figure 3.11. The diode laser used are from BrightLock Ultra-500 series, manufactured from QPC Laser by Laser Operations LLC. The diode laser was fibre-coupled into a 600 μ m core size multi-mode fibre. The emission power was approximately 27W when

operating at a drive current of 45A. The emission wavelength of the diode laser, measured using NIRQuest spectrometer from Ocean Optics, is shown in Figure 3.12. As shown, the diode emission is centred at 1913nm, with a spectral width of 2-3nm. The peak emission wavelength of the diode laser is slightly off from the peak absorption wavelength of Ho:YAG, which is at 1908nm. Thus, the expected absorption efficiency of the diode laser will be lower as compared to the IPG Tm:Fiber laser.

Figure 3.11 Schematic setup of the diode-pumped Ho:YAG slab laser.

Two aspherical lenses were used to shape the pump beam to a diameter of ~1mm. The Ho:YAG slab used was a 10mm length single crystal, with 1% Ho-doping concentration. The input pump surface of the slab was AR-coated at 1.9μm and HR-coated at 2.1μm. This surface acted as the cavity mirror of the laser. The other surface of the slab was AR-coated at 2.1μm. A dichroic mirror was used to reflect the unabsorbed 1.9μm pump power back towards the gain medium, forming a double-pass pump configuration. The other cavity mirror was a concave mirror with HR coating at 2.09μm. For CW operation, the output coupling was formed by a TFP and a λ/4-plate, similar to the variable output

coupling configuration described in Section 3.2.1. Optimization of the output coupling was performed by rotating the $\lambda/4$ -plate.

Figure 3.12 Emission spectrum of 1.9μm QPC diode laser, measured using Ocean Optics NIRQuest spectrometer.

For Q-switched pulse operation, a RTP Pockels cell was inserted between the TFP and the $\lambda/4$ -plate, in a cavity dumping configuration. The Pockels cell functioned as a $\lambda/4$ -plate when the high voltage was applied. The $\lambda/4$ -plate was rotated with its slow-axis 45° with respect to the p-polarized oscillation. This results in a high loss cavity when the Pockels cell is OFF. A high voltage was applied to the Pockels cell for several

hundreds of μs . With this time frame, the cavity was in a low loss cavity state and thus laser oscillation occurred. After this time frame, the accumulated high energy pulse was reflected by the TFP.

Figure 3.13 LIV curve of 1.9 μm QPC diode laser.

Figure 3.13 shows the diode laser power and the corresponding voltage with respect to the drive current. The laser output power of the diode-pumped Ho:YAG slab laser, operating in CW mode, is shown in Figure 3.14. The CW operation generated 3.4W of 2.09 μm output power at a pump diode current of 45A, corresponding to an optical pump power of 26W. This resulted in an optical-to-optical conversion efficiency of ~13.1%. In Q-switched pulse operation, 1mJ pulse energy with pulse width of 51ns, operating at 1kHz, was generated. This result was achieved with pump diode current of 35A, which provided 20.3W of diode pump power. Higher pulse energy operation was limited due to optical damage on the HR concave mirror.

Figure 3.14 CW output power of diode-pumped Ho:YAG slab laser.

3.5 Tm:Fiber pumped Ceramic Ho:YAG Oscillator

A ceramic Ho:YAG crystal was provided by Professor Tang Ding Yuan, Nanyang Technological University School of Electrical and Electronic Engineering (NTU EEE), to evaluate their ceramic Ho:YAG sample for CW laser operation. The ceramic Ho:YAG had an aperture size of 2x3mm and length 9.3mm. The ceramic gain medium had a homogeneous holmium doping concentration of 1.5% a.t. Both aperture surfaces were polished and AR-coated for both the pump and laser wavelengths. The experimental setup of the ceramic Ho:YAG cavity is shown in Figure 3.15. The cavity was pumped with Tm:Fiber laser radiation, focused to a beam diameter of 0.6mm shown

in Figure 3.16. It was a linear cavity formed by an input coupler and an R = 40% output coupler. A dichroic mirror was placed after the gain medium to perform a double-pump-pass operation. The cavity length was ~57mm. The ceramic Ho:YAG gain medium was water-cooled at a constant 15°C.

Figure 3.15 Cavity setup of a Tm:Fiber pumped Ceramic Ho:YAG laser.

Description	Value	Units
Centroid X ISO	▼ 7.756639e+00	mm
Centroid Y ISO	▼ 5.998834e+00	mm
D4σX ISO	▼ 6.181e-01	mm
D4σY ISO	▼ 6.049e-01	mm
D4σ ISO	▼ 6.115e-01	mm
D%pkX	▼ 6.000e-01	mm
D%pkY	▼ 7.000e-01	mm
D%pk	▼ 6.482e-01	mm

Figure 3.16 Left: Measured pump beam diameter with Spiricon Pyrocam III. Right: Pump beam profile of Tm:Fiber laser for the ceramic Ho:YAG gain medium.

Figure 3.17 Experimental result of CW ceramic Ho:YAG laser. Blue curve shows the output power of the ceramic Ho:YAG oscillator with respect to the pump power. Red curve shows the corresponding conversion efficiency. Brown curve shows the extrapolation of the oscillator performance without saturation.

Figure 3.17 shows the experimental CW laser result of the ceramic Ho:YAG from NTU EEE. As shown, the laser generated 7.3W of 2.09 μ m output power with 16W of pumped power. This corresponded to a maximum optical-to-optical efficiency of 45.6%. A slope efficiency of 60% was achieved. The pump threshold calculated was approximately 1436W/cm². These data have clearly shown that the ceramic Ho:YAG performance was comparable with the typical published single crystal Ho:YAG laser. Thus, this shows that there is a great potential in ceramic Ho:YAG laser research. Output power saturation was observed when the pump power was further increased beyond 16W, described in Figure 3.17. The saturation in the output power was due to the saturation of absorption coefficient and strong thermal lensing. Further investigations are required to optimize

the performance of the ceramic Ho:YAG oscillator by cavity design and customize doping profile of the ceramic gain medium.

3.6 Conclusion

In conclusion, we have introduced the properties of the trivalent Holmium ions and the important parameters of Ho:YAG. The peak absorption and emission wavelength of Ho:YAG is 1908nm and 2.09 μ m respectively. The upper laser level lifetime of Ho:YAG is 8.5ms, providing an excellent energy storage in the crystal. Therefore, it has great potential for high energy Q-switched pulses. Four different classes of Ho:YAG laser sources are also presented, covering different configurations and laser specifications.

A Tm:Fiber pumped Ho:YAG laser oscillator operation at high repetition rate of 10kHz was discussed. Using the variable output coupling configuration the highest laser output power was achieved at 21W with the maximum available pump power. The pump threshold power for CW and pulse operation was approximately 15.7W and 13.3W respectively. The high pump threshold was the result of a large pump beam diameter of 1.2mm. The slope efficiency for both operations was close to 65%, with the optical-to-optical efficiency of 44% achieved for pulse operation. FWHM pulse width of approximately 100ns was measured at 10kHz at maximum pump power. The measured M^2 of the output beam was <1.1 .

A high energy Ho:YAG MOPA laser built by a commercial collaborator was introduced. The Ho:YAG MOPA laser generated 2.09 μ m laser pulses with high energy of 82mJ operation at 100Hz. The pulse width measured at 100Hz was 18ns. These data translated

to an average laser output power of 8.2W and pulse peak power of 4.56MW. The measured M^2 of the output beam at 100Hz was 3.7.

A diode-pumped Ho:YAG oscillator and a Tm:Fiber pumped Ceramic Ho:YAG oscillator was discussed. The CW operation of the diode-pumped Ho:YAG Oscillator generated 3.4W of 2.09 μ m output power at an optical pump power of 26W. This results in an optical-to-optical conversion efficiency of ~13.1%. In Q-switched pulse operation, 1mJ pulse energy with pulse width of 51ns, operating at 1kHz, was generated with 20.3W of laser power.

The Ceramic Ho:YAG oscillator generated 7.3W of 2.09 μ m output power with 16W of pumped power, corresponding to a maximum optical-to-optical efficiency of 45.6%. A slope efficiency of 60% was achieved. The pump threshold was ~1436W/cm², which was comparable to typical published single crystal Ho:YAG laser.

The high repetition rate/high average power Ho:YAG oscillator and the high energy Ho:YAG MOPA laser were subsequently employed to perform OPO operation using ZGP crystals to achieve laser pulses emitting at MIR wavelengths. Further evaluations are required for the direct-diode pumped configuration and ceramic Ho:YAG gain medium in order to benchmark with the specification that the Tm:Fiber pumped single crystal Ho:YAG laser is achievable.

Chapter 4 MIR Generation using Ho:YAG Laser pumped ZGP OPO

As mentioned in the previous chapter, ZGP is a very promising nonlinear crystal to perform down-conversion of wavelength to the MIR range. ZGP crystal has excellent mechanical and thermal properties and high non-linear coefficients. However, the transparency of ZGP crystal begins from 2 μ m onwards. Henceforth, the 2 μ m Ho³⁺ laser, discussed in previous chapter, is developed as the primary pump source for the ZGP OPO.

In this chapter, the properties of the ZGP crystal will be introduced. The motivation of using ZGP nonlinear crystal to generate MIR radiation and using Ho:YAG laser medium as the pumped source for the ZGP OPO will be discussed in detail. The results of ZGP OPO pumped by the Ho:YAG laser sources mentioned in the previous chapter will be presented. An intra-cavity ZGP OPO, pumped by the high repetition rate Ho:YAG laser, is built, generating Watt-level MIR wavelength laser radiation. The customized commercial high energy Ho:YAG MOPA laser is used to pump an extra-cavity ZGP OPO, with tens of mJ pulse energy in the MIR wavelength. Lastly, a novel uncoated wedge ZGP OPO configuration is shown, with its potential to achieve high energy MIR laser pulses.

4.1 Properties of ZGP Crystal

The chalcopyrite ZnGeP_2 was first suggested in 1971 for nonlinear optics [35]. In the late 1909s, high quality ZGP became available and it almost totally displaced the other crystals. ZGP has very high nonlinearity of $d_{\text{eff}} = 75\text{pm/V}$ [36], good thermal conductivity and acceptable damage thresholds. However, the transmission wavelength of ZGP cut-off at near $2\text{ }\mu\text{m}$, as shown in Figure 4.1. The exact position and shape of ZGP transmission curve is sensitive to impurities and defects in the crystal and hence, it is critical to have optimal crystal growth procedure [37].

Figure 4.1 Transmission curve of ZGP with a crystal thickness of 1cm. Data extracted from SNLO.

Due to the high nonlinearity of ZGP, its nonlinear figure of merit (d_{eff}^2/n^3) is several times larger than that of the other common nonlinear crystals such as KTP (Potassium

Titanyl Phosphate), PPLN (Periodically Poled Lithium Niobate), BBO (Beta Barium Borate), etc. The birefringence of ZGP is suitable to achieve Type I phase-matching over a large range of wavelengths. Figure 4.2 shows the theoretical Type I phase-matching tuning curve of ZGP OPO with pump wavelength of $2.09\mu\text{m}$, operating at room temperature. The degeneracy is at an internal angle of $\sim 55.65^\circ$, and the wavelength is tuneable between $3.37\mu\text{m}$ to $5.5\mu\text{m}$ by angle tuning of the crystal from the degeneracy point to $\sim 54.3^\circ$. Type II phase matching is also possible for generation of longer wavelength.

Figure 4.2 . Type I Phase matching curves for a ZGP OPO pumped by $2.09\mu\text{m}$ at crystal temperature of 300K. The phase-matched Signal (Idler) wavelength can be tuned from $\sim 3.7\mu\text{m}$ to $4.18\mu\text{m}$ ($5.5\mu\text{m}$ to $4.18\mu\text{m}$) by tuning the phase matching angle from $\sim 54.30^\circ$ to $\sim 55.65^\circ$. Data extracted from SNLO.

ZGP has an acceptable damage threshold of $>1\text{J}/\text{cm}^2$, in addition to its good thermal and mechanical properties. The damage threshold, with improved polishing techniques, can be improved to $2\text{J}/\text{cm}^2$ [38]. ZGP is often reported to have wide transparency range from $0.74\mu\text{m}$ to $12.4\mu\text{m}$, however, low absorption is only available for wavelength between $2\text{-}8\mu\text{m}$. This constraint leads to the need of pump wavelength longer than $2\mu\text{m}$ in order to achieve highly efficient ZGP OPO. The transmission losses between $1\text{-}4\mu\text{m}$ are highly dependent on the defects and impurities in the crystal. Thus, the crystal growth process is very critical in order to achieve high quality ZGP crystal [39].

4.2 High Repetition Rate Intra-Cavity ZGP OPO

4.2.1 Introduction

Typical ZGP OPOs are pumped in an extra-cavity configuration, where the OPO cavity is separated from the pump laser cavity. In this setup, we explored the intra-cavity configuration of ZGP OPO, which is similarly demonstrated by Zakel and his co-workers [40]. Zakel demonstrated the ZGP intra-cavity OPO using a Cr:ZnSe laser, where the emission wavelength of the Cr:ZnSe laser was tuneable from $2.35\text{-}2.6\mu\text{m}$. Cr:ZnSe laser was pumped at 1800nm , therefore, the quantum efficiency was lower as compared to Ho:YAG. In addition, the short upperstate lifetime of $6\mu\text{s}$ does not favour Q-switch operation to generate ns pulses. Thus, pulsed pump sources are required to achieve efficient ns pulsed operation. On the other hand, Ho:YAG has large gain and the optical loss due to the intra-cavity ZGP OPO does not pose an issue for effective pulsing operation.

4.2.2 Experimental Setup

Figure 4.3 Schematic Setup of Intra-Cavity ZGP OPO pumped by Ho:YAG Laser.

The intra-cavity ZGP OPO setup, shown in Figure 4.3 is a slight modification on the Ho:YAG laser described in Section 3.2. In this setup, Mirror M1 was replaced by an R=90% output coupler for the Ho:YAG laser and the $\lambda/4$ -plate was removed. The intra-cavity ZGP OPO cavity was formed by mirror M3 and M4. Mirror M3 was flat with high transmission at 2.1μm and high reflection at 3-5μm while M4 was an R=70% output coupler for the MIR OPO. Several ZGP crystals from Inrad were tested in this

experiment. The ZGP crystals have cutting angle between 53° - 56° for type-I phase-matching using Ho:YAG laser as the pump source. Both end surfaces of the ZGP crystals were AR-coated for wavelengths of $2.1\mu\text{m}$ and $3\text{-}5\mu\text{m}$.

4.2.3 Results and Discussion

Figure 4.4 Intra-Cavity ZGP OPO MIR output power vs Tm:Fiber pump Power.

The Ho:YAG laser beam properties were measured with the use of the $R = 90\%$ output coupler. The laser was operating at 10kHz pulse repetition rate. The FWHM pulse width was approximately 120ns at maximum pump power. The estimated pump beam diameter for the OPO was 1.2mm. Based on these laser properties, the intra-cavity ZGP

OPO was performed. Figure 4.4 shows the MIR output power as a function of the Tm:Fiber pump power. Optimization of the MIR generation was done by tuning the phase-matching angle of the ZGP and adjusting the cavity length of the OPO. The OPO was operating at near degenerate point. A maximum output power of 1W MIR was generated with the highest available pump power from the Tm:Fiber laser.

4.3 High Energy Extra-Cavity ZGP OPO

4.3.1 Introduction

High energy MIR laser pulses are desired for application such as long distance Lidar. In order to generate the high energy MIR pulses, high energy pump pulses are essential. In this section, we discuss the high energy extra-cavity ZGP OPO pumped using the high energy Ho:YAG MOPA laser described in Section 3.3.

4.3.2 Experimental Setup

The extra-cavity ZGP OPO was built based on the conventional OPO configuration, as shown in Figure 4.5. The cavity length was 50mm, formed by an input coupler and an $R = 40\%$ MIR output coupler. The ZGP crystal used in the experiment was grown in DSO National Laboratories. Both surfaces of the ZGP used were AR-coated at the pump and MIR wavelength, The AR-coating was performed by Layertec GmbH. The $2.09\mu\text{m}$ high energy pump laser was collimated, with beam diameter of $\sim 4\text{mm}$ as shown in Figure 4.6.

Figure 4.5 Schematic setup of extra-cavity ZGP OPO Cavity.

Figure 4.6 2.09 μm Pump Spot Profile at focus, D4 σ X = 3.968mm, D4 σ Y = 4.175mm.

4.3.3 Results and Discussion

Calibration on a 3-5 μm bandpass filter was performed before the measuring the power of the MIR generated. As shown in Figure 4.5, the MIR generated transmitted through

a high pass filter and a bandpass filter before it incident onto the power meter. The high pass filter was used to filter out any residue $2.09\mu\text{m}$ pump energy. After passing through the high pass filter, only MIR wavelengths were present. MIR power was also measured before and after the bandpass filter. This was to experimentally determine the MIR transmission of the bandpass filter. The transmission data was shown in Figure 4.7, with a transmission value of 84.8% for MIR wavelength. Based on this transmission value, the output power of the MIR was measured and the actual MIR pulse energy generated was re-calculated.

Figure 4.7 Transmission measurement of the $3\text{-}5\mu\text{m}$ MIR bandpass filter, $T\sim 84.8\%$.

Figure 4.8 Experimental data of high energy extra-cavity ZGP OPO, 100Hz pulse repetition rate.

Figure 4.9 M^2 measurement and beam profile of the MIR pulse at 10mJ pulse energy.

The experimental results of the DSO grown ZGP in an extra-cavity configuration, operating at 100Hz, is shown in Figure 4.8. As shown, 20mJ of MIR pulse energy was generated with a pump energy of 55mJ. A slope efficiency of 41.5% was achieved, with the maximum optical-to-optical efficiency of 36.5%. The limiting factor to achieve higher energy was due to the optical damage of the AR-coating on the ZGP crystals. M^2 measurement was performed for the 10mJ MIR pulse, described in Figure 4.9. The measured M^2 is ~ 7.3 . Figure 4.9 also shows the beam profile of the MIR pulses at 10mJ.

Figure 4.10 Experimental result of high energy extra-cavity ZGP OPO, 200Hz pulse repetition rate.

Using the same cavity, MIR generation was performed for operation at 200Hz pulse repetition rate. The generated MIR result is shown in Figure 4.10. As shown, 17mJ of MIR pulse energy was generated when pumped with 55mJ of 2.09μm pump energy, operating at 200Hz pulse repetition rate. A slope efficiency of 36.6% was achieved, with

the maximum optical-to-optical efficiency of 30.9%. By operating at 200Hz, the MIR average power had increased from 2W to 3.4W as compared to operating at 100Hz.

4.4 Novel Uncoated Wedge ZGP OPO

4.4.1 Introduction

The major contribution which limits the achievement of high pulse energy in a ZGP OPO setup is the laser induced optical damage cause by the high energy pump pulses onto the AR-coating of the ZGP crystal. Conventional linear OPO configuration requires the optical surfaces of the nonlinear crystal to be applied with multi-layer AR-coating for the pump, signal and idler wavelengths. For the case of ZGP crystals, this multi-layer AR-coating covers the wavelength of 2.1 μ m Ho:YAG pump and the MIR wavelength range between 3-5 μ m. Due to the immaturity in the development of such dual-band AR-coating of the ZGP crystal surfaces, these coatings often suffer from laser induced damage by the high energy pump laser pulses. Damage threshold of the AR-coating can be increased by expanding the pump beam size to a larger area, thus reducing the energy fluence experience by the AR-coating for a given pulse energy level. However, the increase of pump area relates to the increase in the pump threshold and also the decrease of the OPO output beam quality.

To overcome this issue, a novel uncoated wedge ZGP OPO configuration was examined. Typical well-polished ZGP crystal surfaces are able to withstand a laser damage threshold of $>1\text{J}/\text{cm}^2$. As compared to the well-polished ZGP crystal surfaces, typical dual-band multi-layer AR-coating for the MIR wavelength lies below $0.5\text{J}/\text{cm}^2$. This provides a strong motivation to explore a potential configuration which makes use of

the higher intrinsic damage threshold of ZGP crystal in the OPO configuration. However, the uncoated ZGP crystal input surface has a Fresnel reflection of $\sim 27\%$ for an incoming laser beam at normal incidence. This introduces a significant interface reflection loss in contrast to the AR-coating surfaces, where the AR-coating is designed to reduce the reflection loss to a negligible level. Instead of treating the Fresnel reflection as a purely optical loss, we make use of it as a fixed output coupling with reflection of $\sim 27\%$ in an OPO cavity.

4.4.2 Uncoated Wedge ZGP Crystal

The schematic design of the uncoated wedge ZGP crystal is shown in Figure 4.11. The ZGP crystal was designed with in a wedge configuration where the two optically polished surfaces were not parallel to each other. To design the cut angle of the ZGP crystal, the refractive indices of the ZGP crystal at different wavelengths were used. The important calculated parameters for the uncoated ZGP OPO crystal are shown in Table 4.1. The values were calculated based on the ZGP crystal with a cutting angle of 55.3° . This cutting angle fulfils the phase matching condition for a Type I nonlinear optics interaction with pump wavelength of $2.09\mu\text{m}$ at the degenerate point.

The fundamental concept of the wedge design relies on the Brewster angle of the ZGP crystal for the polarization of the generated MIR wavelength. Similar to the previous section, the linearly polarized $2.09\mu\text{m}$ pump laser was the ordinary wave with respect to the optics axis of the ZGP crystal while the generated MIR signal and idler wavelengths were the extraordinary waves. As shown in Figure 4.11, the extraordinary MIR wavelengths are p-polarized with respect to the wedge interface. By designing the

cutting angle of the ZGP crystals, the p-polarized MIR wavelengths incident on the wedge interface at the Brewster angle. This led to the transmission of the MIR wavelengths through the crystal/air interface without suffering from reflection losses.

Figure 4.11 Schematic of uncoated wedge ZGP crystal OPO configuration.

n_e (@4.18μm)	3.144574
n_o (@2.09μm)	3.144809
Ext. Brewster angle (@4.18μm)	72.36°
Int. Brewster angle (@4.18μm)	17.64°
Critical angle (@4.18μm)	18.54°
Critical angle (@2.09μm)	18.54°
Fresnel reflectance (@4.18μm)	26.77%
Fresnel reflectance (@2.09μm)	26.78%

Table 4.1 Important calculated parameters for the uncoated ZGP OPO crystal.

4.4.3 Experimental Setup

The OPO cavity was formed by a HR mirror and the input surface of the ZGP crystals as shown in Figure 4.12. This uncoated ZGP crystal surface acted as an output coupler for the MIR radiation with $R \sim 27\%$. The $2.1\mu\text{m}$ pumped beam was s-polarized and the resulted MIR signal and idler were p-polarized. The second surface of the uncoated ZGP crystal was cut at an angle where the MIR radiation incident on that surface at the Brewster angle. This reduced the reflection loss of the MIR radiation on this second surface. Due to Fresnel reflection from the uncoated ZGP surface, $\sim 27\%$ of the $2.1\mu\text{m}$ pump will be reflected. In order to prevent this back-reflection into the laser, tilting the pump beam slightly in the vertical plane by an angle of $<3^\circ$ was necessary. With the large refractive index of ZGP, the internal angle mismatch introduced by this tilt is $<1^\circ$. We tailored this tilt in the non-critical plane so that it did not impose an issue to the phase-matching condition.

Figure 4.12 Wedge ZGP OPO setup.

The high energy Ho:YAG laser described in Section 3.3 was used as the pump source for the novel wedge ZGP OPO cavity. The pump diameter used in the experiment was

~5.5mm. A 45° dichroic mirror, HR for the s-polarized 2.1μm pump beam and HT for the p-polarized MIR signal and idler, was used to separate the pump and MIR beam. A HR mirror at MIR wavelength was used to form the OPO cavity with the flat input surface of the wedge ZGP crystal.

The angle parameters of the wedge ZGP crystal used in the experiment is shown in Figure 4.13. As compared to Table 4.1, the cut wedge angle was slightly smaller than the internal Brewster angle of 4.18μm in ZGP crystal. The optics axis of the ZGP crystal was at 54.74°, which also defers slightly from the desired angle at 55.3°. It was a great challenge to achieve the desired angle parameters while ensuring the input surfaces are well polished.

Figure 4.13 Wedge ZGP cutting angle used in the experiment.

4.4.4 Results and Discussion

The preliminary data of the uncoated wedge ZGP OPO MIR generation is shown in Figure 4.14. 3.8mJ of MIR pulses were generated, operating at 100Hz. As the feedback

from the output coupler was only $R=27\%$, the threshold was relatively high. The pump energy shown was calculated, taking into the consideration of the 27% Fresnel reflection loss by the uncoated ZGP surface. The maximum pump energy that was used for the nonlinear conversion was $\sim 29\text{mJ}$. The resultant optical-to-optical conversion efficiency was 13%, with the OPO pumped at two times above threshold. The slope efficiency was $\sim 26\%$. Although the efficiency of this wedge OPO configuration was not impressively high, the OPO cavity was surprisingly stable. Any slight misalignment from the optimized position of the rear HR mirror only corresponds to a very minor deviation of the MIR beam within the crystal. By simple calculation using Snell's law, a misalignment of 1° external to the ZGP crystal corresponds to an internal misalignment of only $\sim 0.25^\circ$. Thus this uncoated wedge ZGP OPO cavity is highly robust and compact.

Figure 4.14 Experimental data of the uncoated wedge ZGP OPO MIR generation.

4.5 Conclusion

In conclusion, we had introduced the properties of the ZGP crystals and its important parameters for nonlinear frequency conversion. ZGP has very high nonlinearity of $d_{\text{eff}} = 75\text{pm/V}$, good thermal conductivity and acceptable damage thresholds of $>1\text{J/cm}^2$, Low absorption of ZGP crystal is only available for wavelength between $2\text{-}8\mu\text{m}$. Therefore, pump wavelength longer than $2\mu\text{m}$ is necessary in order to achieve highly efficient ZGP OPO.

An intra-cavity ZGP OPO was performed with a slight modification of the high repetition rate Ho:YAG laser developed in the previous chapter. The laser operated at 10kHz pulse repetition rate, with a FWHM pulse width of 120ns . A maximum output power of 1W MIR was generated, optimised by tuning the phase-matching angle of the ZGP and adjusting the cavity length of the OPO.

A high energy extra-cavity ZGP OPO was demonstrated using DSO grown ZGP crystal. The high energy Ho:YAG MOPA laser introduced in the previous chapter was used as the primary pump source for the high energy ZGP OPO, in the conventional ZGP OPO configuration. 20mJ of MIR was generated with a $2.09\mu\text{m}$ pump energy of 55mJ . This corresponded to an optical-to-optical efficiency of 36.5% . A slope efficiency of 41.5% was achieved, with a measured M^2 of ~ 7.3 for the MIR beam at output pulse energy of 10mJ . 200Hz ZGP OPO operation was demonstrated with 17mJ of MIR pulse energy generated when pumped with 55mJ of $2.09\mu\text{m}$ pump energy. A slope efficiency of 36.6% was achieved, with the maximum optical-to-optical efficiency of 30.9% . The MIR average power had increased to 3.4W as compared to 2W operating at 100Hz .

A novel uncoated wedge ZGP OPO configuration was demonstrated. Immature development of dual-band AR-coating on the ZGP crystal surfaces suffer from laser induced damage by the high energy pump laser pulses. The novel uncoated wedge ZGP OPO configuration, with a laser damage threshold of $>1\text{J}/\text{cm}^2$, gives the promise to overcome the coating damage issue. 3.8mJ of MIR pulses were generated with $2.09\mu\text{m}$ pump energy of 29mJ, operating at 100Hz. Optical-to-optical conversion efficiency achieved was 13%, pumped at two times above threshold. The slope efficiency was $\sim 26\%$. Due to the nature of the wedge configuration, the OPO cavity was highly stable from any slight misalignment on the rear HR mirror.

Chapter 5 Coherent Polarization Locking of Ho:YAG Laser

Coherent Beam Combining [41] is a promising approach for power scaling of lasers while maintaining good beam quality and preserving the coherent properties in lasers. Studies on coherent beam combining of laser diode arrays have been done for decades, with its main objective to combine the emissions from multiple diode emitters while preserving a fixed relative phase among them. Both active [42]-[43] and passive [44] phase locking mechanisms were demonstrated to improve the brightness of diode. Similar coherent beam combining was also demonstrated for solid state lasers [45]-[46]. Active coherent beam combining often requires the implementation of complicated electronic control in order to precisely maintain the optical path difference between the combining beams. On the other hand, passive coherent beam combining is a relatively simpler technique and it requires less control. Coherent Polarization Locking (CPL) [45]-[46] is one of the techniques to achieve passive coherent beam combining with near-perfect combining efficiency. Several beams in a cavity are coherently locked in phase to achieve a polarization that experiences minimal loss in the cavity. In this chapter, the theory of CPL is first described in detail. Subsequently, the implementation of CPL for quasi-three-level Ho:YAG laser operating in both CW and pulsed mode is demonstrated. The advantages of implementing CPL in quasi-three-level Ho:YAG laser is discussed.

5.1 Theory in Coherent Polarization Locking

Figure 5.1 Illustration of coherent polarization locking of two orthogonal polarized lasers. The gain mediums with laser oscillation in orthogonal polarization combined using a PBS. The combined beam passed through a polarizer with transmission axis at 45° . Both gain medium cavity shared a common output coupler.

Coherent Polarization Locking is a passive coherent beam combining technique based on the polarization properties of the combining beams. Figure 5.1 illustrates the two orthogonal polarized lasers, with the same wavelength, spatially combined using a polarization beam splitter. The two lasers, oscillating in p-polarization and s-polarization, respectively, share a common output coupler. We assume that both lasers possess the equal magnitude of electric field. The wave equations of the two lasers are described as:

$$E_p = E_o \cos(kx - \omega t + \varphi_p(t)) \quad (1)$$

$$E_s = E_o \cos(kx - \omega t + \varphi_s(t)) \quad (2)$$

The superposition of the two wave equations results in the polarization ellipse:

$$\frac{E_p^2}{E_o^2} + \frac{E_s^2}{E_o^2} - 2 \frac{E_p E_s}{E_o^2} \cos(\Delta\varphi(t)) = \sin^2(\Delta\varphi(t)) \quad (3)$$

where

$$\Delta\varphi(t) = \varphi_s(t) - \varphi_p(t) \quad (4)$$

Due to the time dependence of the relative phase difference $\Delta\varphi(t)$, the resultant polarization of the superposition laser is random in nature. When a linear polarizer, with transmission axis at 45° relative to the horizontal axis, is introduced in the laser cavity between the polarization beam splitter and the output coupler, both orthogonal polarized lasers individually experience a 50% polarization loss through the linear polarizer. However, if the relative phase difference is at either $\Delta\varphi(t) = 0$ or $\Delta\varphi(t) = \pi$, the resultant polarization of the superposition laser is 45° linearly polarized. This resultant laser transmits through the linear polarizer without experiencing any polarization loss, and thus oscillates within the cavity. In other words, the linear polarizer acts as a passive mechanism which forces the relative phase difference to be locked at either $\Delta\varphi(t) = 0$ or $\Delta\varphi(t) = \pi$. This explains the basic working mechanism of CPL which performs passive coherent beam combining.

5.2 CW Coherent Polarization Locked Ho:YAG Laser

5.2.1 Introduction

In this section, the demonstration of CPL in a CW Ho:YAG lasers is described. Ho:YAG lasers are quasi-three-level systems, emitting at a wavelength of $2.09\mu\text{m}$. Holmium lasers have attractive medical and remote sensing applications, and they are also suitable pump sources for nonlinear frequency conversion to generate mid-infrared radiation. In contrast to a four-level laser system, a quasi-three-level laser system has inherent thermal effects and its laser characteristics are strongly temperature dependent. This makes its high-power operation difficult. In addition, Ho:YAG experiences reabsorption loss at the laser wavelength due to the thermally populated lower laser level. This internal power loss in the gain medium creates an additional heat load in the crystal, leading to other common thermal degradation effects, such as thermal lensing, bifocusing, and induced birefringence. It is also known that Ho:YAG laser operation with high doping concentration can cause deleterious processes such as Ho:Ho upconversion, which limits the laser efficiency. Consequently, a lower doping concentration is more favourable. However, ground state depletion in low-doping crystal saturates the pump absorption under high-power pumping. Therefore, there is a strong motivation to achieve high-power laser operation with reduced thermal issues in Ho:YAG. In this experiment, we use CPL technique to combine several low-doping Ho:YAG lasers that are pumped with lower powers to overcome the thermal-optics effects and upconversion loss, while achieving power scaling of the laser.

5.2.2 Experimental Setup

The schematic of the coherent polarization locked Ho:YAG laser is shown in Figure 5.2. The system was pumped by a single linearly polarized Tm:Fiber laser emitting at $1.908\mu\text{m}$, corresponding to the peak absorption wavelength of Ho:YAG. The fiber output was split into two separate parallel pump beams using a $\lambda/2$ -plate (HWP1), a thin film polarizer (TFP1), and a folding mirror (FM1). By rotating HWP1, the splitting ratio of the pump beams can be changed, thus manipulating the gain of each individual path. These two pump beams propagated with a spatial separation of $\sim 10\text{mm}$. A 250 mm focal length lens placed before HWP1 focused both beams to a pump beam diameter of $\sim 800\mu\text{m}$ inside a single Ho:YAG slab (doping concentration of 0.75 at. %). The Ho:YAG gain medium was 10mm in length and was cooled at 20°C . The input surface was AR-coated for the pump wavelength and HR-coated for the laser wavelength. The coherent polarization locking cavity was formed by this HR-coated surface of the gain medium and a flat output coupler with $R = 60\%$, with a cavity length of $\sim 275\text{mm}$. The other surface of the gain medium was AR-coated for both pump and laser wavelengths. A dichroic mirror, highly reflective at the pump wavelength and highly transmissive at the laser wavelength, was placed after the Ho:YAG slab, allowing a double-pass pump configuration. A lens, a half-wave plate (HWP2), and a thin film polarizer (TFP3) were placed in the laser path. The laser output was collected by a lens and an output coupler (OC).

Figure 5.2 Schematic setup of cavity for CW CPL Ho:YAG laser.

The beams from Arm A and Arm B were spatially combined using a thin film polarizer (TFP2) and a folding mirror (FM2). TFP2 also forced the polarization of the lasing beam in Arm A and Arm B to oscillate in s-polarization and p-polarization, respectively. A 300 mm focal length lens was placed inside the cavity, 100mm from the output coupler, to provide a stable cavity configuration and good mode matching of the laser beam with the incident pump beam. Coherent polarization locking was achieved by a $\lambda/2$ -plate (HWP2) and a thin film polarizer (TFP3) within the cavity. The polarization of the coherent combined beam after TFP2 was rotated by HWP2 to horizontal polarization. Optimum combining efficiency could be achieved for any arbitrary power ratio between Arm A and Arm B by simply rotating HWP2. Consequently, it is possible to optimize the setup for maximum laser output power and combining efficiency by rotating both HWP1 and HWP2.

This same experimental setup also allowed laser oscillation in only one of the arms. This was achieved by having the pump power incident onto the gain medium at a single spot (a single arm) instead of splitting the pump laser into two. This allowed us to compare the laser performance and output beam profile for two different laser configurations, coherent combined laser and single arm laser, with similar pump spot diameter and cavity mode diameter.

5.2.3 Results and Discussion

Figure 5.3 shows the single-pass absorbed power for a single spot (single arm laser) and double spots (coherent combined laser) pump configuration with respect to pump power. Saturation in the pump absorption was observed in the single spot pump configuration

when the pump power exceeded $\sim 20\text{W}$, whereas the double spots pump exhibited a linear behaviour. This showed that by splitting the pump source into separate beams and focusing them onto different locations in the same crystal, the total absorbed pump power increased, thus increasing the pumping efficiency and gain of the entire laser system. In summary, this dual-pumping spot configuration doubled the effective pump area, which aid in mitigating the thermal issues of increasing the power in a single pump spot area by twice the amount. Compared to a conventional end-pumping configuration, increasing the pump mode size required the laser mode size to increase accordingly for the same overlapping efficiency. This led to the need for a longer cavity length. In contrast, coherent polarization locking allowed the effective pump area to increase, while maintaining a compact and short laser cavity.

Figure 5.3 Single-pass absorbed power for a single laser spot (Arm A alone) and coherently combined laser (Arm A and B).

Figure 5.4 Laser output power of single arm laser [blue curve] and coherent combined laser [red curve].

Figure 5.4 shows the output laser power from the single arm laser and coherent combined laser configuration. The curve of the single arm laser shows a rollover in the output power at our highest available pump power. The roll-off in the output power was due to the saturation of absorption coefficient and strong thermal lensing. This limited the power scaling of this laser configuration. In the case of a coherent combined laser, the maximum output power produced was $>10\text{W}$, with no observable output power saturation even at our maximum available pump power. Slope efficiency of the coherent combined laser was close to that of the single arm laser. The pump threshold of the coherent combined laser was approximately twice that of the single arm laser due to the doubling of the pump spot area. In contrast with the single arm laser, we prevented the saturation of the laser output by the coherent polarization locking technique. Saturation

of absorption coefficient was also avoided and thermal lensing was weaker in the coherent combined laser.

Using the same experimental setup, we measured the output power with respect to the pump power with a pump power ratio of $\sim 1:1$. Output power for independent Arm A and B, measured with the slow axis of HWP2 rotated to 0° and 45° , respectively, is shown in Figure 5.5. When the slow axis of HWP2 rotated to 0° with respect to p-polarization, Arm A oscillated, while Arm B experienced high loss on TFP3. Similarly, by rotating the slow axis of HWP2 to 45° , Arm B oscillated.

Figure 5.5 Output power of the independent Arm A [red curve] and B [blue curve].

The total output power generated by the independent Arm A and B was taken as a reference for incoherent combined power. This was compared with the output power

from the coherent combined laser to calculate the coherent combined efficiency. Figure 5.6 shows the generated output power from the coherent combined laser and the calculated coherent combined efficiency with respect to the total pump power. We achieved combining efficiency of $>96\%$ at maximum output power.

Figure 5.6 Output power [red (lower) curve] and combining efficiency [blue (upper) curve] for coherent combined laser with respect to pump power.

The output beam profile of the coherently combined laser and single arm laser were taken using a Spiricon Pyrocam III beam profiler. Figure 5.7 shows the beam profile of the output beam from the two laser configurations. An M^2 value of ~ 1.1 was measured at maximum power for the coherent combined laser, whereas a beam quality of $M^2 > 2$ was measured for the single arm laser. Therefore, we showed that the beam quality of the output laser was preserved to a near-diffraction-limited beam through coherent polarization locking. The improvement of the beam quality is mainly due to the

reduction in the thermal effects experience in a single laser arm and the requirement to achieve coherent polarization locking. Any higher order mode in one arm needs the same higher order mode in the other arm to survive and oscillate in the coherent polarization locking configuration. This hinders the survival of higher order modes in the cavity, thus leading to the improvement of the beam quality.

Figure 5.7 Far field beam profile of (a) coherent combined laser and (b) single arm laser at maximum output power.

We optimized the coherent combining efficiency by rotating HWP1 and HWP2, respectively, where it changed the total pump power, pump ratio, and gain of Arm A and B. With a total pump power of 30.6W, we obtained 9.6W of coherently combined laser power, with individual arm laser power of 6W and 3.6W, respectively. This corresponded to a near-perfect coherent combining efficiency. The output power of the combined laser was observed to be stable with power fluctuation of $<0.5\%$ over a period of 5mins. It should be noted that the optimum combining output achieved was not with a 1:1 power ratio, due to the imbalance in pump ratio and gain, but with an arbitrary ratio determined empirically in our experiment. This is an additional advantage of coherent polarization locking where it allows for the combination of lasers with different

power outputs with near-perfect combining efficiency. In contrast, coherent combining in a Vernier–Michelson-type cavity [1] required the individual laser output power ratio to be fixed based on the beam splitter ratio, in order to achieve near-perfect coherent combining efficiency.

5.3 Pulsed Coherent Polarization Locked Ho:YAG Laser

5.3.1 Introduction

In this section, the CW CPL Ho:YAG laser is extended to perform pulsed operation by including Q-switching element in the laser cavity. As mentioned earlier, Ho:YAG lasers are quasi-three-level systems, emitting at a wavelength of $2.09\mu\text{m}$. Holmium lasers have attractive medical and remote sensing applications, and they are also suitable pump sources for nonlinear frequency conversion to generate mid-infrared radiation. Q-switching operation is required to generate ns pulses for pumping optical parametric oscillators with high conversion efficiency. During high pulse energy operation, one key challenge is to avoid optical damage induced by the extremely high intra-cavity intensity generated within the laser cavity. Typical optical damage is easily observed on the multilayer thin-film coated optics, such as the polarizers, the gain medium, and the Pockels cell. High-damage-threshold thin-film coating for $2\mu\text{m}$ operation is not as readily available as compared to coating for $1\mu\text{m}$ operation. Thus, there is a strong motivation to decrease the intra-cavity intensity of high-energy pulse lasers while maintaining the high output energy of the laser pulses. In this experiment, we use CPL to combine two orthogonal Ho:YAG lasers in pulsed mode and investigate the advantage of using CPL to mitigate intra-cavity optical damage. Pulse operation is

operated in two different scenarios. In the first scenario, Q-switching is performed on the common arm of the coherent combining cavity. And in the second scenario, Q-switching is performed only on one of the polarization arms, while the cavity of the other polarization arm does not have any Q-switching element. For the second scenario, it is interesting to examine the dynamics involved in Q-switching coupled with the CPL operation in the cavity.

5.3.2 Experimental Setup

Figure 5.8 Schematic setup of cavity for Q-switched CPL Ho:YAG laser.

The schematic of the Q-switched CPL Ho:YAG laser is shown in Figure 5.8. The system was pumped by a single linearly polarized Tm:Fiber laser emitting at $1.908\mu\text{m}$, corresponding to the peak absorption wavelength of Ho:YAG. The fiber output was split into two separate parallel pump beams using a $\lambda/2$ -plate (HWP1), a thin film polarizer (TFP1), and a folding mirror. By rotating HWP1, the splitting ratio could be changed, thus manipulating the gain of each individual path. These two pump beams propagated with a spatial separation of $\sim 25\text{mm}$. The total pump power used was 28W. A 300 mm focal length lens, placed before HWP1, focused both beams to a pump beam diameter of $\sim 1\text{mm}$ inside two Ho:YAG rods (doping concentration of 0.36 at. %). Both water-cooled (at 20°C) Ho:YAG rods had a diameter of 3mm and length of 30mm. The laser

cavity was formed by two rear mirrors, one for each rod, and a common ROC 750mm concave output coupler with $R=40\%$ at the laser wavelength. The cavity length was 225mm. Dichroic mirrors, highly reflective at the pump wavelength and highly transmittive at the laser wavelength, were placed after the Ho:YAG rods, allowing a double-pass pump configuration. The beams from arm A and arm B were spatially combined using a thin film polarizer (TFP2) and a folding mirror. TFP2 also forced the polarization of the lasing beam in arm A and arm B to oscillate in p-polarization and s-polarization, respectively. CPL was achieved by a $\lambda/2$ -plate (HWP2) and a thin film polarizer (TFP3) within the cavity. The polarization of the coherently combined beam after TFP2 was rotated by HWP2 to horizontal polarization, and transmitted through TFP3 with low reflection loss. Pulsed operation was performed using a RTP Pockels cell. The Pockels cell was placed at two different positions, P1 and P2, respectively, as shown in Figure 5.8, for the two different scenarios mentioned earlier. At position P1, the Pockels cell was placed after TFP3 at the common arm. Q-switching operation occurred after the CPL operation was performed. Thus, the CPL operation was transparent to the Pockels cell. In other words, the Pockels cell perceived the CPL setup before TFP3 as a typical laser cavity. At position P2, the Pockels cell was placed at arm A, before the coherent locking element. Q-switching operation experienced the effect introduced by the coherent beam combing performed by the coherent locking element. The cavity for arm B did not have any Q-switching element for pulse operation. In this scenario, we examined the pulsed operation behaviour of the CPL cavity when Q-switching was performed only at arm A.

5.3.3 Results and Discussion

Figure 5.9 shows the pulse energy and the pulse width of the laser output at different pulse repetition rate achieved for the first scenario, where the Pockels cell was placed at position P1. We achieved pulse energy of 9.13mJ with 14ns pulse width, operating at pulse repetition rate of 800Hz.

Figure 5.9 Pulse energy/pulse width versus pulse repetition rate for CPL Q-switched Ho:YAG laser with the Pockels cell placed at P1.

We performed M^2 measurement by focusing the output beam, measured the beam size at several positions across the waist and curve fitted the data to the Gaussian beam propagation equation. Figure 5.10 shows the beam radius measured with respect to distance and the far-field beam profile captured using Spiricon Pyrocam III. The output beam was near diffraction limited, with a M^2 value of ~ 1.07 achieved at the highest

output pulse energy. No intra-cavity optical damage was observed during pulse operation at 800Hz.

Figure 5.10 M^2 measurement of Q-switched CPL Ho:YAG laser at the highest pulse energy. The inset shows the laser-output far-field beam profile.

For comparison, a conventional Q-switched Ho:YAG laser cavity setup was built with the same gain medium, pump mode size, and cavity design. With the same amount of pump power, the conventional laser generated an average power of 10W at a pulse repetition rate of 2kHz, corresponded to pulse energy of 5mJ. When operating below 2kHz, intra-cavity optical damage was observed on the multilayer thin film coated on the surface of the Ho:YAG rod. Therefore, the laser operation for the conventional setup was limited for pulse energies up to 5mJ. This showed a strong advantage of using CPL

in pulsed operation to mitigate intra-cavity optical damage. In the CPL cavity, the total intra-cavity intensity in the common arm was shared between the two orthogonal arms. TFP2 split the intra-cavity intensity experienced by the common arm to two lower intensities for each polarization arm. This mitigated the risk of intra-cavity optical damage to the optics placed along the individual arms, thus achieving higher output pulse energy. In addition to the lower intra-cavity intensity, the good beam quality of the coherent locked beam suppressed the existence of hot spikes, which was another common effect that can damage the optics. The risk of the intra-cavity optical damage was thus reduced.

Figure 5.11 Stable optical pulse train in Q-switched CPL Ho:YAG laser.

Next, we examined the pulsed operation of the CPL cavity for the second scenario, where the Pockels cell was placed at position P2 as described in Figure 5.8. Q-switching

was performed only in the cavity of arm A, as there was no Q-switching element in the arm B cavity. Pulsed operation was successfully achieved without any observation of pulse instability, any loss of laser coherent properties, or any drastic drop in the average power compared to the output power during CW operation. A stable output pulse train generated by this setup was observed on the oscilloscope using a fast photodetector as shown in Figure 5.11. The stable pulse train could be explained by examining the restriction imposed by the CPL technique. In a CPL cavity, coherent beam combining was achieved only when the combined gain from both arm A and arm B exceeded the total losses within the cavity. No laser oscillation was possible in an individual polarization arm without the existence of the other arm. By inserting a Q-switching element in one of the polarization arms, we manipulated the gain of that particular arm. However, the combined gain of the entire CPL cavity also changed dynamically according to the Q-switching element. This led to stable pulsed operation without loss of the laser coherent properties and the output power. In addition, the result showed that during ns pulse operation, the CPL mechanism was fast enough to respond to the dynamics introduced by the Q-switching element.

The average power, pulse energies, and pulse width of the laser output at different pulse repetition rate achieved for the second scenario are shown in Figure 5.12. Pulse energy of 7mJ, with 18ns pulse width, operating at pulse repetition rate of 800Hz, was achieved, with near diffraction-limited output beam quality. The decreased in the output pulse energy as compared to the first scenario was a result of the decreased in the coherent beam combining efficiency. With the RTP Pockels cell placed at position P2, the output beam profile of arm A was slightly distorted. The cavity mode size at position P2 was smaller, which introduced undesired thermal effects on the RTP Pockels cell. This

resulted in a decrease in the spatial beam overlapping efficiency of the two orthogonal arms, thus achieving a lower coherent beam-combining efficiency.

Figure 5.12 Pulse energy/Pulse width versus pulse repetition rate for CPL Q-switched Ho:YAG laser with the Pockels cell placed at P2.

5.4 Conclusion

In summary, we first introduced the basic concept of CPL, which is a passive coherent beam combining technique applied to a laser cavity. Polarizing elements within the laser cavity forced two orthogonal linearly polarized beams to oscillate in phase in the cavity. This resulted in a combined linear polarized beam oscillating at 45° . CPL technique was applied in a CW and Q-switched Ho:YAG laser oscillator.

A CW Coherent Polarization Locked Ho:YAG Laser was investigated. Single-pass absorption by a single Ho:YAG slab was saturated at 7.5W with the available pump power of 35W. By splitting the same pump power equally to excite two spatially separated spot on the single Ho:YAG slab, the single-pass absorption had increased by 66% to 12.5W with no saturation of absorption. Coherently combining the gain from the two pump spot in a CPL cavity enabled achieving a maximum output power of 10W, with a coherent combining efficiency of >96%. There was no observable output power saturation in the CPL cavity. In contrast, a single arm laser, with also 10W output power, exhibited a rollover in the output power. This showed that CPL had prevented the saturation of the laser output. Saturation of absorption coefficient was also avoided and thermal lensing was weaker in the coherent combined laser. The measured M^2 of the CPL cavity beam was ~ 1.1 , improved from the measured M^2 of >2 from the single arm laser. This proved that CPL had the capability to reduce thermal-optics problem and achieved excellent beam quality in a high power CW operation.

The technique of CPL in Ho:YAG laser was extended further in Q-switched ns pulse operation. Pulse energy of 9.13mJ with 14ns pulse width was generated, operating at pulse repetition rate of 800Hz. On the contrary, a conventional Q-switched Ho:YAG laser with the same cavity parameter generated an average power of 10W at a pulse repetition rate of 2kHz, limiting pulse energy to only 5mJ. The cause of the limitation is the intra-cavity optical damage observed on the multilayer thin film coated on the surface of the Ho:YAG rod at pulse repetition rate $<2\text{kHz}$. This showed that the pulsed CPL Ho:YAG laser cavity had the potential to mitigate intra-cavity optical damage. Pulse operation was also observed when the Pockel cell was placed only in one of the laser arm. The experiment showed that the pulsed operation was not affected by the fast

dynamic change in the gain within the cavity. This was possible due to the inherent fast coherent locking mechanism imposed by CPL.

Chapter 6 Femtosecond Regenerative Amplifier

pumped MIR OPA

MIR with fs pulse width is in active research in the recent years. Brida and his co-workers had demonstrated the generation of broadband fs MIR pulses using optical parametric amplification (OPA) configuration [47]. The setups began with a Ti:Sapphire fs laser, and subsequently involved several stages of white-light generation (WLG) and OPA. Since this setup used Ti:Sapphire laser, which required a bulky 532nm pump source from the second harmonic generation (SHG) of Nd:YAG laser, the entire system was relatively large. As compared to the MIR fs OPA setup pumped using a Ti:Sapphire fs laser, the Yb-doped fs laser has drawn a strong attraction as an alternative pump source for MIR fs pulse generation. Yb laser has strong absorption peak in the 940-980nm range, where mature, compact and high power diode laser emitting wavelengths between 940-980nm is easily available in the current market. Therefore, there is a strong motivation to develop the high energy diode-pumped Yb-doped fs laser with the overall laser system size significantly smaller than the Ti:Sapphire fs laser. In this chapter, the development of high energy Yb:CaF₂ fs regenerative amplifier setup is first discussed in detail. Subsequently, a ZGP OPA setup, pumped by the Yb:CaF₂ regenerative amplifier and generating MIR fs pulses, is described. Lastly, the research to develop the 2 μ m Ho:YAG regenerative amplifier is presented, with its motivation as the direct 2 μ m pumped source for the ZGP OPA setup.

6.1 Yb:CaF₂ Regenerative Amplifier

6.1.1 Introduction

Figure 6.1 Energy Level of Yb ions [48].

Figure 6.1 shows the energy level diagram of the trivalent Yb³⁺ ions. The lasing dynamics of the Yb³⁺ ions occurs between the various manifolds of the two levels, ²F_{7/2}

and $^2F_{5/2}$. As shown, the energy-level has a simple structure, with no concentration quenching, no excited-state absorption, no upconversion, no energy migration and no cross-relaxation. It has high electron-phonon coupling, which results in a broad gain bandwidth. The laser operation, pumping at 900-980nm and emission at 1030nm, exhibits low quantum defect. Thus Yb^{3+} is an ideal candidate for diode-pumped high power fs laser.

Figure 6.2 Thermal conductivity and emission bandwidth of different Yb-doped materials [48].

Figure 6.2 shows a graphical comparison of the thermal conductivity and emission bandwidth of different common host material doped with Yb^{3+} ions. The favourable host material should exhibit both high thermal conductivity and wide emission bandwidth. High thermal conductivity is an important factor for high power and high energy/high repetition rate laser operation. The transform-limited pulse width is related to the

emission bandwidth of the laser pulse. Thus the wider the emission bandwidth, the shorter the transform-limited pulse. Wide emission bandwidth supports short pulse width down to fs range. Among the common host materials, CaF₂ has the wide emission bandwidth of up to 80nm and thermal conductivity similar to YAG.

Figure 6.3 Absorption cross-section spectra of Yb:CaF₂ crystal at different temperature [48].

Figure 6.3 and Figure 6.4 shows the absorption and emission cross-section spectra of Yb:CaF₂ crystal at various temperatures, ranging from 290K to cryogenic cooled temperature of 77K. As shown, cryogenic cooling increases the absorption and emission cross-sections coefficient of the gain medium. There is a 2 times and 1.6 times increase in the absorption cross-sections coefficient at 980nm and the emission cross-sections coefficient at 1030nm respectively, as compared to room temperature. The emission

band at 1030nm remains wide at cryogenic temperature. Cryogenic cooling of the Yb:CaF₂ crystal also switches its energy laser scheme from a quasi-three-level energy scheme to a four level mode, thus decreases the re-absorption loss at the lasing wavelength. This motivates the use of cryogenic cooled Yb:CaF₂ gain medium to develop a regenerative amplifier that potentially can achieve high energy ultrashort laser pulses.

Figure 6.4 Emission cross-section spectra of Yb:CaF₂ crystal at different temperature [48].

6.1.2 Experimental Setup

The fs laser was based on the chirped pulse amplification (CPA) concept. The system comprised of a Kerr lens mode-locked seed oscillator, a pulse stretcher, a regenerative amplifier and a pulse compressor. In CPA, the seed pulses were stretched in the positive dispersion stretcher, prior introducing into amplifier. These stretched pulses were amplified in a cryogenically cooled regenerative amplifier based on Yb:CaF₂ gain medium. The amplifier operated at 1kHz repetition rate and produced an output pulse energy of 7.5mJ. After amplification, these pulses were recompressed down to <200fs using reverse process of stretching in a negative dispersion compressor.

Figure 6.5 Output Spectrum of Yb:KGW Seed Oscillator.

The frontend of the system was an fs seed oscillator from Light Conversion Ltd. The oscillator was based on the Yb:KGW gain medium, end-pumped with high brightness

laser diode. The seed oscillator operated at pulse repetition rate of 75.63MHz, with an output power of 750mW. This corresponded to ~10nJ pulse energy. Figure 6.5 shows the output spectrum of the Yb:KGW seed oscillator. The spectral bandwidth exceeded 16nm at FWHM, corresponding to a transform-limited pulse duration of ~80fs.

The temporal stretching of fs seed pulses was performed using a positive dispersion 4f-stretcher. The stretcher was based on a single transmission grating (1700l/mm, Wasatch Photonics) and R = -600mm spherical mirror. Schematic layout of stretcher is shown in Figure 6.6. In the stretcher, the distance between the grating and curved mirror was ~150mm. A folding mirror was placed in the Fourier plane of stretcher. The separation between curved mirror and folding mirror was 300mm. This distance was kept with high precision in order to avoid residual uncompensated angular dispersion in the output beam.

Figure 6.6 Schematic setup of pulse stretcher.

The throughput of the stretcher was 17.5%, which corresponded to output pulse energy of 1.7nJ. This rather low transmission efficiency was due to the centre wavelength of seed spectrum was not optimal for the pulse amplification in the regenerative amplifier.

The regenerative amplifier was based on Yb:CaF₂, which has its highest gain in the vicinity of 1030nm. Therefore, the grating stretcher was optimized to transmit seed spectrum at the centre of 1030nm by introducing high transmission losses for the blue spectral components. The seed spectrum after stretcher is shown in Figure 6.7. The seed pulses were stretched down to 270ps, measured using SHG autocorrelation function and assuming Gaussian pulse envelope. An amplitude shaper, consisted of manually adjustable screws which partially blocked the seed spectrum spatially dispersed in the Fourier plane of the stretcher, was introduced to compensate for spectral gain narrowing in the amplifier. The amplitude shaper partially blocked the spectral components which experienced highest gain in the amplifier, thus suppress the central gain peak around 1030nm. Spectral pre-shaping allowed broadening of the emission bandwidth from ~5nm to ~12nm simultaneously introducing small losses to the seed power.

Figure 6.7 Optical spectrum of Yb seed pulse before (black curve) and after stretcher (red curve).

The regenerative amplifier was based on a cryogenically cooled 2%Yb:CaF₂ crystal, pumped in a slab configuration, which allowed the suppression of thermally induced distortions and lensing when pumped with high power. The Brewster angle cut slab had a length of 4mm and input aperture of 3mm width by 1.2mm height. The crystal was wrapped with a layer of thin indium foil and mounted in a copper heat sink. The entire crystal assembly was placed inside the cryogenic chamber and cooled down to 110K using the closed-loop refrigerator.

Figure 6.8 Beam profile at the output of LD.

The laser crystal was pumped by two high brightness laser diodes from DILAS GmbH. Each of the diode delivered 60W of optical power at 70A driving current. The emission wavelength was stabilized at 976.5nm using the volume Bragg grating. The output

radiation from the laser bars was propagating in free space without coupling it into the fiber. This allowed the preservation of the linear output polarization and avoided fiber coupling losses. The emission radiation from the laser bars were collimated using fast/slow axis collimator in both the x- and y-axes. After collimation, the output beam was symmetrized by dividing the emission from the laser bars into two stripes, as shown in Figure 6.8. The beam size at the output of the laser diode was 5x5mm, with 6mrad divergence angle in both axes. This corresponded to a beam quality of $M^2 \sim 25$. The beam profile shown in Figure 6.8 is taken by imaging the output of laser diode onto the CCD beam profiler using an imaging system with $M = 0.3$ magnification ratio.

Figure 6.9 Schematic layout of end-pump geometry for Yb:CaF₂ RA.

The Yb:CaF₂ crystal was end-pumped from a single side using both laser diodes simultaneously. The pumping geometry is shown in Figure 6.9. The pump beams were spatially combined in order to preserve its linear polarization and avoid parasitic reflection losses from the Brewster angle cut surfaces of Yb:CaF₂. The pump beams were combined geometrically by folding the emission stripes as close as possible to each other, shown in Figure 6.9. The pump beams were focused onto the crystal using a

60mm lens. This 60mm lens had an Ø2mm hole drilled in the centre, which allowed lasing while ensuring collinear pumping geometry. The far field distribution of pump beam is shown in Figure 6.10. The beam size in the focus was 450x470µm ($1/e^2$), optimized to achieve good spatial overlap with the cavity mode. The unabsorbed pump light after single pass through the crystal was re-imaged onto the Yb:CaF₂ crystal using a 2f-2f configuration with spherical R = -75mm mirror.

Figure 6.10 Far field beam profile of the focused pump beam at the crystal.

The cavity configuration of the cryogenically cooled Yb:CaF₂ regenerative amplifier is sketched in Figure 6.11. The cavity consisted of two curved mirrors M2 and M5 with R = -1500mm, two folding mirrors M3 and M4 and two flat end mirrors M1 and M6. The total cavity length was approximately 1.9m. Special care was taken to set the proper mode size on each individual optical component. Figure 6.12 shows the mode size of

the regenerative amplifier cavity. The beam diameter ($1/e^2$) on the Yb:CaF₂ crystal and the Pockels cell was $\sim 500\mu\text{m}$ and $\sim 2000\mu\text{m}$ respectively. The mode was maximized at highly nonlinear β -BBO crystal in the Pockels cell to keep the level of overall B-integral low. Contrary to this, mode size on the gain material was kept rather small since this ensured high single pass gain and had weak influence for nonlinear phase accumulation due to low n_2 of CaF₂ crystal.

Figure 6.11 Schematic diagram of cavity design. WP- $\lambda/4$ -plate, P-thin film polarizer, PC-Pockels cell, FR-Faraday rotator.

Figure 6.12 The size of the mode based on cavity design of the regenerative amplifier cavity.

The coupling of pulses in and out of the amplifier was performed by the electro-optics switches. The switch comprised of a BBO Pockels cell, a $\lambda/4$ -plate and a thin film polarizer. The Pockels cell operated as a $\lambda/4$ -plate when a high voltage of $\sim 7.8\text{kV}$ was

applied. The duration of amplification stage was controlled by setting the time interval during which high voltage was applied to Pockels cell.

The amplified pulses were recompressed using two reflection gratings with 1750l/mm, arranged in a double pass Littrow-configuration, as shown in Figure 6.13. The overall transmission of compressor exceeded 95% due to high diffraction efficiency of the diffraction gratings. In the compressor, manual adjustment of separation between the gratings allowed the controlling of the negative or positive chirp for an output pulse.

Figure 6.13 Schematic of pulse compressor setup – double-pass Littrow-configuration .

6.1.3 Results and Discussion

The performance of regenerative amplifier cavity was first examined in CW laser operation. By switching off the voltage on Pockels cell and adjusting the $\lambda/4$ -plate for the highest output power, laser output power of 18.5W was obtained with an incident pump power of 61.5W. The dependence of output power as a function of pump power

is shown in Figure 6.14. The output power preserved linear dependence in the whole operation range. The upper limit of 18.5W was restricted by the available pump power. Linear behaviour of the curve was attributed as the direct indication of excellent thermal management of gain medium and proper design of laser cavity, which was insensitive for thermal lensing.

Figure 6.14 Dependence of CW output power on pump power for Yb:CaF₂ RA cavity.

In picosecond operation, the regenerative amplifier was seeded by the stretched 270ps pulses from the fs Yb:KGW oscillator. The regenerative amplifier operated at a pulse repetition rate of 1kHz and produced 6mJ pulses after pulse compression. The dependence of output power on the pump power in the case of picosecond operation regime is shown in Figure 6.15. The curve experienced exponential growth at moderate pump intensity and subsequently switched to linear dependence when the pump

intensity increased. The initial portion of the curve corresponded to the amplifier operation in small signal gain regime. Further increase in pump power led to the saturation of the amplification.

Figure 6.15 Dependence of output power on the pump power in the case of picosecond operation at repetition rate of 1kHz.

The recompressed pulses were characterized using second harmonic generation frequency-resolved optical gating (SHG-FROG). The results are presented in Figure 6.16. Measurement revealed 191fs pulse duration in the case of 5mJ output energy and rather flat spectral phase under whole emission spectral bandwidth. The pulse characterization was additionally performed at different amplification levels and fixed number of cavity roundtrips. According to the measurements, the pulse compressibility

was not affected by high output energies. This implied that the nonlinear phase was well under control.

Figure 6.16 Recompressed pulse characterization using SHG FROG for output energy of 5mJ from Yb:CaF₂ regenerative amplifier.

6.2 MIR fs Optical Parametric Amplifier based on ZGP Crystal

6.2.1 Introduction

Ultrashort fs laser pulses emitting at MIR wavelengths have several scientific and defence applications. As there is currently no efficient gain medium for laser emission in the MIR spectra capable to produce fs pulses, the alternate approach for fs MIR pulses

is through nonlinear optical down conversion from shorter wavelengths at the NIR (800nm, 1030nm) to generate MIR radiation. Our approach to achieve fs MIR pulse is based on the WLG/OPA pumped by the high energy Yb:CaF₂ regenerative amplifier described in the previous section. ZGP crystal is used for the nonlinear wavelength conversion. The ZGP OPA setup achieving 6μJ of ultrabroad spectrum around 6μm is described. The spectrum is capable to support 32fs pulse duration, which corresponds to a 1.5 optical cycle pulse at the carrier wavelength.

6.2.2 Experiment Setup

To extend the down-converted wavelength from an OPA beyond 4μm, dedicated MIR nonlinear crystals and IR short pulse lasers for pumping the OPA are essential. Due to the good optical quality, broad phase-matching bandwidth and high nonlinearity, ZGP is one of the most attractive crystals for the generation of ultrashort pulses in the MIR spectral range beyond 4μm. ZGP crystal needs to be pumped at a wavelength above 2μm due to the detrimental linear and two-photon absorption [49]. This can be achieved either by Ho- or Cr-based lasers, or by parametric frequency down conversion of Nd- or Yb-based laser light using an OPA. We demonstrated ZGP OPA to achieve broadband MIR spectrum using the latter approach.

The schematic layout of the 6-μm OPA system is given in the Figure 6.17. The frontend of the system was the Yb:CaF₂ regenerative amplifier described in the early section. The 6mJ output energy from the laser system was split for the generation of 2.1μm pump and 3.2μm seed pulses for the ZGP OPA. Generation of 2.1μm pump involved a WLG stage with sapphire plate, a SHG stage with BBO crystal and several BBO OPA stages.

WLG in a 4mm thick sapphire plate, pumped by 1.03 μ m pulses generated the 680nm source, which was seeded to the first BBO OPA stage. The first BBO OPA stage was pumped by the 515nm SHG of the Yb:CaF₂ source and seeded by the 680nm source, generating the idler 2.1 μ m pulses. This generated 2.1 μ m seed was subsequently amplified in two consecutive OPA stages based on Type I BBO crystals, pumped directly with the 1.03 μ m pulses.

Figure 6.17 Schematic setup of the MIR fs ZGP OPA.

As for the 3.2 μ m seed pulses, it was generated by a KTA OPA pumped by 1.03 μ m pulses and seeded by a 1.5 μ m source generated through WLG in a 6mm thick YAG crystal. The WLG spectrum generated in the YAG crystal, also pumped by 1.03 μ m pulses, extended up to 1.7 μ m. The KTA OPA consisted of two independently tuneable cascaded 6mm thick KTA crystals, which produced a broad 3.2 μ m seed spectrum. The generated 3.2 μ m seed pulses were matched in space and time with the 2.1 μ m pump pulses in a 12mm long ZGP crystal, cut for Type I collinear interaction with $\theta=57.1^\circ$.

6.2.3 Results and Discussion

Total pump pulses energy of $160\mu\text{J}$ was used to pump the ZGP OPA. The total output of the ZGP OPA was estimated at $17\mu\text{J}$, which corresponded to an approximately $6\mu\text{J}$ of idler ($6\mu\text{m}$) and $11\mu\text{J}$ of signal ($3.2\mu\text{m}$) pulses. The spectrum of the amplified $3.2\mu\text{m}$ and generated $6\mu\text{m}$ radiation are shown in Figure 6.18. The measured bandwidth of the idler wave, centred at around $6\mu\text{m}$, spanned an octave. The spectrum was strongly modulated due to the absorption of air. The difference in the shapes of the signal and idler waves manifested the impact that the broadband pump pulse at $2.1\mu\text{m}$ had on the shape and width of the $6\mu\text{m}$ spectrum. The ultrabroad spectrum was capable to support 32fs pulse duration, which corresponded to a 1.5 optical cycle pulse at the carrier wavelength.

Figure 6.18 Emission spectrum from the ZGP OPA.

6.3 Ho:YAG Regenerative Amplifier

6.3.1 Introduction

In the previous section, we demonstrated the potential of using ZGP nonlinear crystal in an OPA configuration to generate ultrabroad MIR spectrum, capable to support 32fs pulse duration, which corresponded to a 1.5 optical cycle pulse at the carrier wavelength of around 6 μ m. However, the conversion efficiency from the frontend 1.03 μ m Yb:CaF₂ regenerative amplifier pump was relatively low. This was because additional wavelength conversion and amplification stages were needed to generate 2 μ m pump source for the ZGP OPA stage. These additional stages can be replaced by a Holmium-based gain medium with direct emission in the 2 μ m regime. Thus, there is a strong motivation to design and build a regenerative amplifier based on Holmium gain medium, with output energy of several mJ and pulse width of fs range, as the primarily pump source for the ZGP OPA to achieve high energy MIR fs pulses with high conversion efficiency.

6.3.2 Experimental Setup

The frontend of the Ho:YAG regenerative amplifier setup was an Yb-based diode-pumped solid state laser pumped BBO OPA, shown in Figure 6.19, generating the seed 2.1 μ m pulse. The 1030nm fs pulse from the Yb fiber MOPA was split to two paths. A 1 μ J fraction of the 1030nm pulse energy was used to perform WLG in a 6mm thick undoped YAG crystal, generating wavelength between 680nm. The remaining of the 1030nm pulse energy was directed to a 1mm thick Type I BBO crystal for SHG. After the BBO crystal, the remaining 1030nm pulse energy and the second harmonic 515nm

pulse were separated by a dichroic mirror. The 515nm pulse was directed as the pump source for the Type I BBO crystal in the first OPA (OPA I). OPA I was seeded by the 680nm signal from the BBO WLG, generating idler wavelength at 2.1 μ m. To preclude angular chirp on the idler beam, the 680nm seed and 515nm pump beams were strictly overlapped collinearly. The remaining 1030nm pulse energy was directed as the pump source for the dual Type II KTA crystal in the second OPA (OPA II) to amplify the 2.1 μ m idler from OPA I. The thin film polarizer in OPA II reflected the s-polarized 2.02 μ m signal from the first KTA crystal and transmitted the p-polarized 1030nm pump and 2.1 μ m idler to the second KTA crystal. The second KTA crystal reversed the walk-off accumulated in the first KTA crystal and thus compensated spatial chirp on the amplified beam. The amplified 2.1 μ m pulse was used as the seed source for the Ho:YAG CPA system.

The Ho:YAG CPA system consisted of a stretcher, a Ho:YAG regenerative amplifier and a compressor, presented in Figure 6.19. The Ho:YAG regenerative amplifier was based on an AR-coated 0.5% doped $\varnothing$ 6x50mm Ho:YAG rod end-pumped by a 70W Tm:Fiber laser operating at a central wavelength of 1908nm. The end faces of the YAG rod were concave, with a -500mm radius of curvature, to offset the positive thermal lens induced by the pump. The Ho:YAG rod was water cooled at 18°C. Due to the unavailability of Faraday rotators capable to withstand high peak intensities, a ring-shaped RA cavity was designed. The injection and output beam ports were separated and the amplification of the injected beam was unidirectional. Due to the ring-cavity design, the RTP Pockels cell operated in the half-wavelength regime. The length of the RA cavity was 4m, corresponding to a roundtrip time of 13.5ns.

Figure 6.19 Layout of the Yb-fiber master oscillator power amplifier (MOPA) pumped OPA seed source and Ho:YAG CPA system. TFP - thin film polarizer, $\lambda/2$ -plate, WLG - white light generation unit, SHG - second harmonic generation unit, PC - Pockels cell, DM - dichroic mirror, CM - curved mirror, HR - high reflector.

6.3.3 Results and Discussion

The $2.1\mu\text{m}$ pulses with energy of $\sim 0.7\mu\text{J}$ and bandwidth of 40nm FWHM, shown in the insert of Figure 6.20, were stretched in a positive-dispersion Öffner scheme stretcher. The stretcher was based on a single 600lines/mm ruled diffraction grating and $R = -762\text{mm}$ spherical mirror. The transmission of the stretcher was $\sim 22\%$, which was determined by the efficiency of the diffraction grating and clipping of the spectral wings on the folding mirrors in the stretcher. The seed after the stretcher extended from 2080 to 2110nm had an energy of 160nJ .

Figure 6.20 Output spectra of the Ho:YAG RA for shaped (black solid line, cyan filled area) and unshaped (red dotted line, yellow filled area) seed. In the inset the output spectrum of the OPA (dashed blue line) and unshaped (red dotted line, yellow filled area) and shaped (black solid line, cyan filled area) spectra transmitted through the stretcher are presented. The fine structure of the shaped spectrum could not be resolved by our spectrometer (NIRQuest512).

Spectral shaping of the seed pulses plays a crucial role in the presented CPA system. Without spectral shaping of the input pulses, the amplified spectrum after the regenerative amplifier collapsed to a mere 2.4nm FWHM centred at 2090 nm. At higher

pump levels, a weaker second band emerged at the 2097nm gain peak of Ho:YAG, shown in Figure 6.20. To precompensate the gain narrowing in the regenerative amplifier cavity, a mechanical amplitude shaper was used in the Fourier plane of the stretcher, which suppressed the intensity of the seed light in the spectral region where the gain was the highest. The shaping was primarily associated with the suppression of the red part of the seed spectrum, corresponding to the 2090nm and 2097nm gain peaks of Ho:YAG. Despite the additional loss of 60% of the seed pulse energy, the `

Figure 6.21 Temporal profile and phase of compressed 2.1μm pulses as retrieved from SHG FROG measurements. The yellow-shaded contour shows the transform limited pulse.

The amplified pulses were recompressed in a negative dispersion compressor, consisted of two 600lines/mm diffraction gratings separated by 120cm. The transmission of the compressor was 33% and was mainly determined by low diffraction efficiency of the gratings. The pulse duration obtained from the SHG FROG measurements in Figure

6.21 was 530fs, whereas the Fourier transform limited pulse duration was ~ 440 fs. Before compression, the amplifier generated 3mJ pulses at a 5kHz repetition rate with 55W of pump power. The number of round trips in the regenerative amplifier was set to 22. The change of the slope at high pump levels shown in Figure 6.22 was due to the decreased in spatial overlap of the pump and laser mode volumes. Despite the fact that the cooling of the 50mm long $\varnothing 6$ mm Ho:YAG rod was done simply by running water through the holder, the polarized output of the regenerative exhibited an excellent spatial profile with a beam quality factor M^2 of 1.3, shown in the insert of Figure 6.22.

Figure 6.22 Dependence of the output energy on pump power in the case of 22 round trips in the RA cavity and seed energy of 85nJ. In the inset, a far-field beam profile of the RA output is shown.

6.4 Conclusion

To conclude, a cryogenic cooled Yb:CaF₂ regenerative amplifier was discussed. The physical properties of Yb:CaF₂ were introduced, along with the advantage of increasing the absorption and emission cross-sections coefficient of the gain medium at cryogenic cooled temperature of 77K. The regenerative amplifier operated at a pulse repetition rate of 1kHz and produced 6mJ pulses after pulse compression. The recompressed pulses were characterized using SHG FROG, revealed a 191fs pulse duration in the case of 5mJ output energy and rather flat spectral phase under whole emission spectral bandwidth.

A MIR fs OPA based on ZGP crystal was presented. As there is currently no efficient gain medium for laser emission in the MIR spectra capable to produce fs pulses, the alternate approach for fs MIR pulses is through nonlinear optical down conversion from shorter wavelengths at the NIR (800nm, 1030nm) to generate MIR radiation. The MIR fs ZGP OPA setup achieved 6μJ of ultrabroad spectrum around 6μm. The spectrum was capable to support 32fs pulse duration, which corresponded to a 1.5 optical cycle pulse at the carrier wavelength.

Development of high energy MIR fs pulses was extended further by developing a high energy 2.1μm fs laser setup for the MIR fs ZGP OPA. A Ho:YAG regenerative amplifier was investigated in a ring-cavity design. Before pulse compression, the amplifier generated 3mJ pulses at a 5kHz repetition rate with 55W of pump power. The pulse duration obtained from the SHG FROG measurements was 530fs, whereas the Fourier transform limited pulse duration was ~440fs. An excellent spatial profile with a beam quality factor M^2 of 1.3 was achieved.

Chapter 7 Summary and Future Work

7.1 Thesis Summary

Coherent light radiation covering the MIR spectrum between 2.5-10 μ m has extremely important applications in nonlinear spectroscopy, chemical sensing, laser surgery in medical field and remote sensing and countermeasures in defence. Different applications have their own unique requirements on the specification of the MIR coherent source in terms of wavelength tunability, emission bandwidth, beam quality, CW or pulse operation, pulse width, output power/energy etc. In this thesis, we developed different MIR coherent sources operating in ns and fs pulses through nonlinear optical frequency conversion using ZGP crystal.

The major contribution for high nonlinear conversion efficiency is the pump laser source for the nonlinear interaction in the ZGP crystal. In the ns pulse operation, four classes of Ho:YAG laser sources were developed. Two of the Ho:YAG laser sources were employed to pump the ZGP OPO generating MIR radiation. An uncoated wedge ZGP OPO was demonstrated with high robustness towards optical alignment. Coherent Polarization Locking, a coherent beam combining technique, was implemented in a Ho:YAG laser oscillator. We investigated the impact contributed by CPL to the Ho:YAG laser oscillator operating in both CW and pulse. In the fs pulse operation, a high energy Yb:CaF₂-based regenerative amplifier was developed as the frontend pump source for ZGP OPA system. Frequency conversion of the 1 μ m Yb:CaF₂ emission to 2 μ m was essential for the ZGP OPA. Due to the multiple frequency conversion, the

conversion efficiency from the 1 μ m to 6 μ m was relatively low. Thereafter, a Ho:YAG regenerative amplifier was studied and developed as an alternate direct pump source for the ZGP OPA system to generate MIR radiation.

Tm:Fiber pumped Ho:YAG laser oscillator was developed, operating at high repetition rate of 10kHz and achieving an average output power of 21W using the variable output coupling configuration. A high pump threshold of 13.3W was the result of a large pump beam diameter of 1.2mm. The slope efficiency was closed to 65%, with the optical-to-optical efficiency of 44%. FWHM pulse width of approximately 100ns was measured at 10kHz, with a measured M^2 of <1.1. Customized commercial high energy Ho:YAG MOPA laser, generating 2.09 μ m laser pulses with high energy of 82mJ and pulse width of 18ns operating at 100Hz, was introduced. These data translated to an average laser output power of 8.2W and pulse peak power of 4.56MW. The measured M^2 of the output beam at 100Hz was 3.7. Direct diode-pumped Ho:YAG oscillator was investigated, generating 3.4W of 2.09 μ m output power at an optical pump power of 26W. In Q-switched pulse operation, 1mJ pulse energy with pulse width of 51ns, operating at 1kHz, was generated with 20.3W of laser power. Tm:Fiber pumped ceramic Ho:YAG oscillator was demonstrated, generating 7.3W of 2.09 μ m output power with 16W of pumped power, corresponding to a maximum optical-to-optical efficiency of 45.6%. A slope efficiency of 60% was achieved. The pump threshold was $\sim 1436\text{W}/\text{cm}^2$, which was comparable with typical published data of single crystal Ho:YAG laser.

Intra-cavity ZGP OPO was performed with the high repetition rate Ho:YAG laser operating at 10kHz pulse repetition rate. Maximum output power of 1W MIR was generated, optimised by tuning the phase-matching angle of the ZGP and adjusting the cavity length of the OPO. High energy extra-cavity ZGP OPO, using DSO grown ZGP

crystal, was pumped by the high energy Ho:YAG MOPA laser operating at 100Hz in the conventional ZGP OPO configuration. >20mJ of MIR was generated with a pump energy of 55mJ, achieving an optical-to-optical efficiency and a slope efficiency of 36.5% and 41.5% respectively. M^2 of ~ 7.3 for the MIR beam with 10mJ pulse energy was achieved. 200Hz ZGP OPO operation generated 17mJ of MIR pulse energy, pumped with 55mJ pump energy. The MIR average power had increased to 3.4W as compared to 2W operating at 100Hz. A novel uncoated wedge ZGP OPO configuration, with a laser damage threshold of $>1\text{J}/\text{cm}^2$, was examined to overcome the coating damage issue. 3.8mJ of MIR pulses were generated with pump energy of 29mJ, operating at 100Hz. Optical-to-optical conversion efficiency achieved was 13%, pumped at two times above threshold. The slope efficiency was $\sim 26\%$. Due to the nature of the wedge configuration, the OPO cavity was highly stable from any slight misalignment on the rear HR mirror.

CW Coherent Polarization Locked (CPL) Ho:YAG Laser was investigated. Single-pass absorption by a single Ho:YAG slab was saturated at 7.5W with the available pump power of 35W. In contrast, the single-pass absorption for dual pump spot increased to 12.5W with no saturation of absorption. Coherently combined the gain from the two pump spot in a CPL cavity achieved a maximum output power of 10W, with a coherent combining efficiency of $>96\%$. No observable output power saturation in the CPL cavity. A single arm laser, with also 10W output power, exhibited a rollover in the output power. The measured M^2 of the CPL cavity beam was ~ 1.1 , improved from the measured M^2 of >2 from the single arm laser. This proved that CPL prevented the saturation of the laser output and was capable to reduce thermal-optics problem and achieve excellent beam quality in a high power CW operation. The technique of CPL in Ho:YAG laser was extended further in Q-switched ns pulse operation. Pulse energy of 9.13mJ with 14ns pulse width was generated, operating at pulse repetition rate of 800Hz. A

conventional Q-switched Ho:YAG laser with the same cavity parameter generated an average power of 10W at a pulse repetition rate of 2kHz, limiting pulse energy to only 5mJ. The intra-cavity optical damage observed on the multilayer thin film coated on the surface of the Ho:YAG rod limited the pulse repetition rate to <2kHz. The pulsed CPL Ho:YAG laser cavity had the potential to mitigate intra-cavity optical damage. Pulse operation was also observed when the Pockel cell was placed only in one of the laser arm. The pulsed operation was not be affected by the fast dynamic change in the gain within the cavity due to the inherent fast coherent locking mechanism imposed by CPL.

Cryogenic cooled Yb:CaF₂ regenerative amplifier was developed, operating at a pulse repetition rate of 1kHz and produced 6mJ pulses after pulse compression to a 191fs pulse. MIR fs OPA based on ZGP crystal was presented. The MIR fs ZGP OPA setup achieved 6μJ of ultrabroad spectrum around 6μm. The spectrum was capable to support 32fs pulse duration, which corresponded to a 1.5 optical cycle pulse at the carrier wavelength. Development of high energy MIR fs pulses was extended further by developing the high energy 2.1μm fs laser setup for the MIR fs ZGP OPA. Ho:YAG regenerative amplifier was investigated in a ring-cavity design. The amplifier generated 3mJ pulses before pulse compression at a 5kHz repetition rate with 55W of pump power. The pulse duration obtained from the SHG FROG measurements was 530fs, whereas the Fourier transform limited pulse duration was ~440fs. An excellent spatial profile with a beam quality factor M^2 of 1.3 was achieved.

7.2 Recommendation for Future Work

The performance of the ceramic Ho:YAG gain medium is promising as an alternative to single crystal gain medium. Ceramic material can be produced in large quantity and high doping concentration, with relatively cheaper production cost as compared with single crystal. The doping concentration profile of the laser ions in ceramic gain medium can also be engineered to improve the absorption efficiency of the pump source and reduce thermal lensing issue on the gain medium. CPL cavity has shown to improve the laser performance as compared to conventional linear cavity. By implementing the Brewster cut ceramic Ho:YAG gain medium in a CPL cavity configuration, there is a potential to generate high energy high repetition ns pulses at $2.09\mu\text{m}$. Brewster cut gain medium eliminates the need for AR-coating on the gain medium surfaces, thus reducing the risk of intra-cavity optical damage.

The conversion efficiency from the $1.03\mu\text{m}$ fs pulse to the MIR fs pulse in the ZGP OPA experiment discussed is relatively low. To improve the conversion efficiency, high energy Ho:YAG regenerative amplifier is developed as the pump source for the ZGP OPA experiment. The next step will be using the high energy fs pulse from the Ho:YAG regenerative amplifier to perform the MIR fs generation in the ZGP OPA setup, with slight modification on the schematic described in Figure 6.17. The $3.2\mu\text{m}$ signal for the ZGP OPA can be generated by using SHG of $2.09\mu\text{m}$ as the pump source for the YAG WLG and KTA OPA. The ZGP OPA will be directly pumped by the fs pulse from the Ho:YAG regenerative amplifier. Multiple ZGP OPA stages can also be used to further amplify the $3.2\mu\text{m}$ signal and $6\mu\text{m}$ idler.

List of Publications

Peer-Reviewed Journals

- 1) **L. H. Tan**, C. F. Chua, and P. B. Phua, "Preserving a diffraction-limited beam in Ho:YAG laser using coherent polarization locking," Opt. Lett. 37, 4621-4623 (2012).
- 2) **L. H. Tan**, C. F. Chua, and P. B. Phua, "Coherent polarization locking: an approach to mitigating optical damage in a pulsed Ho:YAG laser," Opt. Lett. 38, 1025-1027 (2013).
- 3) P. Malevich, G. Andriukaitis, T. Flöry, A. J. Verhoef, A. Fernández, S. Ališauskas, A. Pugžlys, A. Baltuška, **L. H. Tan**, C. F. Chua, and P. B. Phua, "High energy and average power femtosecond laser for driving mid-infrared optical parametric amplifiers," Opt. Lett. 38, 2746-2749 (2013) .

Conference Papers

- 1) "Generation of watt level mid-infrared wavelengths using intra-cavity ZnGeP₂ OPO within a 2.1 μ m Ho:YAG laser," **L. H. Tan**, and P. B. Phua, Proc. SPIE 7917,

Nonlinear Frequency Generation and Conversion: Materials, Devices, and Applications X, 79170O (February 21, 2011); doi:10.1117/12.876248.

- 2) S. Alisauskas, G. Andriukaitis, A. Pugzlys, A. Baltuska, A. Michailovas, **L. H. Tan**, H. J. Lim, P. B. Phua, and K. Balskus, "Micro-Joule Octave Spanning Pulses at 6 μm from a Hybrid Femtosecond OPA," in Lasers, Sources, and Related Photonic Devices, OSA Technical Digest (CD) (Optical Society of America, 2012), paper AM1A.6.
- 3) G. Andriukaitis, S. Alisauskas, A. Pugzlys, A. Baltuska, **L. H. Tan**, H. J. Lim, P. B. Phua, K. Balskus, and A. Michailovas, "Broadband 6 μm OPA Driven by Yb:CaF₂ DPSSL System," in CLEO: Science and Innovations, OSA Technical Digest (online) (Optical Society of America, 2012), paper CF3B.6.
- 4) P. Malevich, G. Andriukaitis, S. Alisauskas, A. Pugzlys, A. Baltuska, **L. H. Tan**, C. F. Chua, and P. B. Phua, "Femtosecond 3-mJ 5-kHz cw-pumped Ho:YAG CPA," in Frontiers in Optics Conference, OSA Technical Digest (online) (Optical Society of America, 2012), paper FW6B.10.
- 5) "Coherent polarization locking of thermal-sensitive Ho:YAG laser," **L. H. Tan**, C. F. Chua, and P. B. Phua, Proc. SPIE 8547, High-Power Lasers 2012: Technology and Systems, 85470U (November 8, 2012); doi:10.1117/12.970542.
- 6) P. Malevich, G. Andriukaitis, T. Floery, A. Verhoef, S. Alisauskas, A. Pugzlys, A. Baltuska, **L. H. Tan**, C. F. Chua, P. B. Phua, A. Fernandez, "Millijoule Femtosecond Pulses at 5 kHz from cw-Pumped Ho:YAG Regenerative Amplifier,"

in CLEO Europe, OSA Technical Digest (online) (Optical Society of America, 2013), paper CA-8.2.

- 7) "Coherent polarization locking of Q-switched Ho:YAG laser," **L. H. Tan**, C. F. Chua, and P. B. Phua, Proc. SPIE 8599, Solid State Lasers XXII: Technology and Devices, 85990I (March 6, 2013); doi:10.1117/12.2004393.
- 8) P. Malevich, A. Pugzlys, G. Andriukaitis, T. Flöry, A. Verhoef, A. Fernández, S. Ališauskas, A. Baltuska, **L. H. Tan**, C. F. Chua, and P. B. Phua, "High Energy and Average Power Femtosecond Laser for Pumping mid-IR OPAs," in CLEO: 2013, OSA Technical Digest (online) (Optical Society of America, 2013), paper CTh3H.5.
- 9) **L. H. Tan**, H. W. J. Lim and P. B. Phua, "Mid-Infrared Generation in an Uncoated Wedge ZGP OPO," in High-Brightness Sources and Light-Driven Interactions, OSA technical Digest (online) (Optical Society of America, 2016), paper MS4C.7.

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