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ULTRAFAST HIGH POWER SOLID STATE LASERS

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in fulfillment of the requirements for the degree of
Doctor of Philosophy

2011

Declaration

I, Wei De Tan, hereby certify that this thesis has been written by me, that it is the record of work carried out by me and that it has not been submitted in any previous application for a higher degree.

I was admitted as a research student in January 2008 and as a candidate for the degree of Doctor of Philosophy in August 2011; the higher study for which this is a record was carried out in Nanyang Technological University between 2008 and 2011.

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Abstract

Diode pumped ultrafast high power solid state lasers are capable of delivering high power picosecond or femtosecond pulses with excellent beam quality and as such have widespread scientific and technological applications. To that end, it necessary to develop and improve performances of existing systems though laser engineering or new materials.

In this thesis, the CW and ultrafast laser performances of novel Yb: Sc₂SiO₅ (Yb: SSO) and Yb: CaYAlO₄ (Yb: CYA) are investigated. Using a board stripe free space diode as the pump source, Yb: SSO and Yb: CYA generated maximum CW output powers of 2.7 W and 2.3 W respectively with slope efficiencies of 70 % and 92 %. When mode-locked in the negative dispersion regime, solitary mode-locking was achieved. In the case of Yb: SSO, the laser was operated near the zero vicinity of zero group velocity dispersion such that 93 fs pulses with an output power of 20 mW at 1043.4 nm. When mode-locked at 1077 nm in which the net negative dispersion was slightly larger than in the 1043.4 nm case, the laser generated 125 fs pulses with an output power of 100 mW. In the case of Yb: CYA, the laser was mode-locked with a large net negative dispersion such that the laser generated 156 fs pulses with an output power of 740 mW. When mode-locked in the positive dispersion regime, Yb: SSO and Yb: CYA both demonstrated mode-locked spectra with steep edges with spectral bandwidths capable of supporting femtosecond pulses. The pulses obtained were strongly chirped with time bandwidth products many times above the Transform limit. This provides strong evidence of dissipative solitons in these lasers. Particularly in the case of Yb: CYA, the laser emitted 1.5 W of output power that was compressible down to 380 fs. In addition, the mode-locking of Nd: GdVO₄ was also demonstrated. By exploiting the birefringence and thermal lensing of the gain media, the laser was able to generate dual wavelength mode-locked emission with orthogonal polarizations using a V-shaped cavity. Such a device might find applications in laser interferometers.

Finally carbon based nano-materials such as semiconducting (7, 5) single walled carbon nanotubes (SWCNTs) and graphene were used to fabricate saturable absorbers (SAs) and were found to be capable of mode-locking ceramic Nd: YAG. In addition Z-scan investigation revealed that both novel SAs exhibited saturable absorption properties.

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List of acronyms

AFM	Atomic force microscopy
B	Breadth
CQGLE	Cubic-qintic Ginzburg-Landau equation
CVD	Chemical vapor deposition
CW	Continuous wave
DNA	Deoxyribonucleic acid
DS	Dissipative soliton
FOD	Fourth order dispersion
FWHM	Full width at half maximum
GDD	Group delay dispersion
GO	Graphene oxide
GTI	Gires-Tournois interferometer
GVD	Group velocity dispersion
H	Height
HWHM	Half width at half maximum
HR	High reflection
HT	High transmission
IDRI	Intensity dependent refractive index
IR	Infrared
KLM	Kerr lens mode-locking
L	Length
M	Mirror
M^2	Beam quality
ML	Mode-lock
Nd: YAG	Nd: $Y_3Al_5O_{12}$
NLSE	Non-linear Schrödinger equation
OC	Output coupler
PWE	Paraxial wave equation
QML	Q-switched mode-locked
RI	Refractive index
ROC	Radius of curvature

SA	Saturable absorber
SESAM	Semiconductor saturable absorber mirror
SAINT	Saturable absorber incorporating nano-tube
SHG	Second harmonic generation
SPM	Self phase modulation
SSL	Solid state laser
SWCNT	Single walled carbon nanotubes
T	Transmission
TEM	Transverse electromagnetic mode
TOD	Third order dispersion
Yb: BOYS	Yb: Sr ₃ Y(BO ₃) ₃
Yb: CALGO	Yb: CaGdAlO ₄
Yb: CYA	Yb: CaYAlO ₄
Yb: GSO	Yb: Gd ₂ SiO ₅
Yb: KGW	Yb: KGd (WO ₄) ₂
Yb: KYW	Yb: KY(WO ₄) ₂
Yb: LSO	Yb: Lu ₂ SiO ₅
Yb: SSO	Yb: Sc ₂ SiO ₅
Yb: SYS	Yb: SrY ₄ (SiO ₄) ₃ O
Yb: YAG	Yb: Y ₃ Al ₅ O ₁₂
Yb: YSO	Yb: Y ₂ SiO ₅
UV	Ultraviolet
VIS	Visible

List of commonly used symbols

a, b, c	Crystallographic directions
c	Velocity of light in vacuum
δ	Cavity losses
$\Delta\lambda$	Spectral bandwidth
ΔR	Modulation depth
ΔR_{ns}	Non-saturable absorption
$\Delta\tau_p, \Delta\tau$	Pulse width
D	Magnitude of group velocity dispersion
E_p	Pulse energy
g	Gain cross-section
κ	Thermal conductivity
λ	Wavelength
L_{prism}	Tip to tip prism separation
n	Refractive index
n_2	Non-linear refractive index
η_B	Beam overlap efficiency
η_{CW}	Continuous wave slope efficiency
$\eta_{femtosecond}$	Femtosecond mode-locked slope efficiency
η_{opt}	Optical to optical conversion efficiency
$\eta_{picosecond}$	Picosecond mode-locked slope efficiency
η_q	Quantum efficiency
P_{in}	Input power
P_{abs}	Absorbed power
P_{out}	Output power
σ_{abs}	Absorption cross-section
σ_{em}	Emission cross-section
τ_f	Fluorescence lifetime
τ_r	Radiative lifetime

Chapter 1

Introduction

1.1.: Background and aims

It has been more than fifty years since Maiman invented the ruby laser in 1960 [1] and since then great progress have been achieved in the use of laser light for different applications. The laser has enabled broadband connections to be possible in optical communication which spurred the development of the Internet in the 1990's as laser light is used as the information carrier in transatlantic fiber optics systems [2]. It would not be exaggerating to say that without the laser, the Internet would not be what it is today. The development of high powered lasers [3-4] has also changed the way work pieces are handled. These lasers have enabled the industry to machine products with complicated shapes and designs which would otherwise be impossible or difficult with mechanical machining tools. The usage of laser in the manufacturing process have also resulted in higher quality finishes and lowered fabrications costs for a great many products. The laser has also revolutionized the way medical procedures are carried out and have been used in corneal refractive surgeries [5], the treatment of varicose veins [6], cosmetic procedures and so on. Hence one can say that the laser has in certain ways changed how we live and work today.

Based on Maiman's initial design, scientists and engineers were able to build lasers based on different sources such as crystalline media, fibers, gases, dyes and semiconductors. These different types of sources have their own advantages and as such applications. Amongst which, the solid state laser (SSL) employing neodymium (Nd) and ytterbium (Yb) dopants ions for optical gain has generated much interest in recent years due to its high efficiency, high output powers, high beam quality and its ability to operate in different modes of operations (continuous wave (CW), Q-switched, picosecond, femtosecond etc.). The development of high brightness diodes in recent years have also

resulted in these SSLs to become more compact and economical to construct. By adding a mode-locker into the cavity, SSLs can be used to generate ultrashort pulses in the picosecond and femtosecond regime. Operation of lasers in these regimes, particularly in the femtosecond regime, allows one to probe the dynamics of molecular interactions that occur on this time scale. In addition, operating a laser in the picosecond and femtosecond regime allows one to produce better quality cuts as they introduce less or no damage to the work pieces. This is because thermal shock events in solids occurs on the nanosecond time scale [7] and by operating the laser at an even shorter time scale this problem can be sidestepped. This is particularly important to the singulation of thin wafers ($< 40 \mu\text{m}$) [7-8] into dies and the drilling of vias in the micro-electronics industry as thermal shock would further weaken the structure of the dies causing them to break easily in subsequent manufacturing processes. For the same reasons, the solar panel industry prefers to use pulsed lasers for their manufacturing processes. The demand for economical and compact ultrafast SSLs for both scientific and industrial applications has driven the development of this type of laser. However in SSLs, the dopants, the host materials and even the choice of mode-locker have bearing on the performance the subsequent laser constructed. As such, there is a growing interest in developing laser materials that would enhance the performance of laser systems. In recent years, much attention has been placed on the development of Yb-doped fiber lasers. In fact for CW generation, its output powers are comparable to solid state lasers ($\sim 50 \text{ KW}$ for industrial lasers [3, 9]). However for short pulse generation, due to non-linearities and fiber damage, Yb-doped fiber oscillators are limited to pulse energies of about 1 nJ while this figure can be much greater than 10 nJ in end pump oscillators [10] and in the microjoule regime for thin disk lasers. In addition, the shortest pulse widths obtainable for both Yb-doped bulk and fiber lasers are usually comparable ($\sim 35 \text{ fs}$ [11-12]).

One aim of this thesis is to investigate the CW and ultrafast high power performances of SSLs. This is primarily achieved through constructing well designed cavities using Nd^{3+} and Yb^{3+} ions as the optical amplifier to achieve high slope efficiencies and ultrashort pulse generation. In particular for femtosecond pulse generation, novel Yb^{3+} doped hosts are used and their performance was investigated in both the negative and positive dispersion regime. Another aim of this thesis is to study the performances of

carbon based mode-lockers such as carbon nanotubes and graphene as potential alternatives for current mode-locking technologies. The advantages of using carbon based mode-lockers are that they simplify the fabrication process and lowers the cost of construction. In addition, the extension of the operating wavelengths of carbon based mode-lockers can be achieved more easily than current technology based on semiconductors.

1.2.: Major contributions of this thesis

- Demonstration of mode-locking of Nd: GdVO₄ at 1065 nm.
- Demonstration a dual wavelength mode-locked Nd: GdVO₄ laser with orthogonal polarization controlled by thermal lensing.
- Mode-locking of Yb: SSO with 125 fs and 100 mW of output power from an oscillator.
- Mode-locking of Yb: CYA in with 156 fs and 740 mW of output power from an oscillator.
- Observation of dissipative solitons in Yb: Sc₂SiO₅ (Yb: SSO) and Yb: CaYAlO₄ (Yb: CYA). In the case of Yb: CYA, 340 fs with 0.29 W and 380 fs with 1.5 W of output powers were achieved from an oscillator in the positive dispersion regime.
- Observation of third and fourth order dispersion effects in Yb: SSO.
- Demonstration of mode-locking of ceramic Nd: YAG with (7, 5) SWCNTs and graphene.

1.3.: Organization of thesis

The organization of the thesis is as follows:

- Chapter 1 provides a simple introduction and discusses the aims of this thesis.
- Chapter 2 discusses some of the theoretical aspects of mode-locking and how certain parameters can have an effect on the final pulse width.
- Chapter 3 provides a discussion on the properties related to gain media and how they might affect laser performances. This chapter also summarizes most of the results reported in the literature in the last decade using certain Nd³⁺ and Yb³⁺ doped crystalline gain media. Recent results obtained with Yb³⁺ doped garnets (Yb: YAG etc.), sesquioxides (Yb: YSO, Yb: LSO, Yb: GSO etc.), vanadates

(Yb: YVO₄, Yb: YLuVO₄, Yb: YVO₄, Yb: GdVO₄ etc.) , double tungstates (Yb: KYW, Yb: KGW etc.) and disordered hosts (Yb: glass, Yb: BOYS, Yb: SYS, Yb: CALGO etc.) will also be discussed. It also contains aspects of laser engineering and cavity designs.

- Chapter 4 presents results obtained with a Nd: GdVO₄. By exploiting the birefringence of the host medium, the author was able to demonstrate a mode-locked laser in which the operating wavelength can be selectively chosen by exploiting the properties of the gain medium. The laser is also capable of generating a dual wavelength mode-locked laser with orthogonal polarizations.
- Chapter 5 presents the mode-locking results of novel Yb: SSO and Yb: CYA gain media in both the negative and positive dispersion regimes to generate femtosecond outputs. The CW performances of both novel gain media are also investigated. The mode-locked results are the first demonstrations of femtosecond pulse generation using these materials. The demonstration of dissipative solitons in these two materials also shows that dissipative solitons can be generated in solid state lasers.
- Chapter 6 provides the mode-locking results of a ceramic Nd: YAG with carbon based nano-materials such as semiconducting (7, 5) single walled carbon nanotubes (SWCNTs) and graphene.
- Chapter 7 summarizes the results obtained in this thesis and provides suggestions for future work.

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Chapter 2

Mode-locking

This chapter provides a general introduction to the concept and theory of mode-locking. The first section introduces the general concept of mode-locking and how shorter pulses could be obtained using this technique. The second section discusses the pulse shaping mechanisms involved in mode-locked lasers and how self-phase modulation (SPM) and group velocity dispersion (GVD) can lead to soliton formation. Finally the last sections provide further discuss on the physical aspects of SPM, GVD and saturable absorption how they can be exploited to generate ultrashort pulses.

2.1.: General considerations

Consider the case of a laser oscillating with a large number of longitudinal modes within a laser cavity. When the phases between the modes are random, the output is seen to be a periodic sequence of light pulses such as that shown in Fig 2.1.1. This corresponds to the case in which the laser is operating under CW operation.

Fig. 2.1.1.: The output of a laser with random phases between modes. This corresponds to the situation in which the laser is operating under CW operation.

To initiate mode-locking, it is necessary to introduce some sort of stable or time-independent phase difference between the modes such that they oscillate in unison. In a sense, the modes are “locked” together as they oscillate and thus the term mode-locking. Mode-locking is usually done by introducing an intra-cavity optical loss element which modulates the electric field and at the same time introduces a constant phase difference between the longitudinal modes. Mode-locking is typically classified as “active” or “passive”. In active mode-locking, an external signal controlled by electronics is fed to an acousto-optic or an electro-optic modulator which modulates the electric field and initiate mode-locking. Since the modulation signal is electronically controlled, the pulse widths are limited to the nanosecond/picosecond regime and as such not a popular method to obtaining ultrashort pulses. In passive mode-locking, a saturable absorber (SA) is used to modulate the electric field and initiate mode-locking. A SA usually introduces high losses for low intensity pulses and low losses for high intensity pulses. Since the highest intensity of a pulse occurs at its peak, they saturate the absorber more strongly than the low intensity wings, resulting in the generation of ultrashort pulses. SAs tend to have fast recovery times and are capable of supporting femtosecond pulses and are routinely used in many mode-locked systems today.

Fig. 2.1.2. : Longitudinal modes oscillating in phase in a mode-locked laser each separated by a frequency of $\Delta\omega$.

The concept of mode-locking can be understood by considering the output of a mode-locked laser in the frequency domain which contains $2m + 1$ longitudinal modes, each with amplitude of E_0 , oscillating in phase with one another as shown in Fig 2.1.2 which is only true if one assumes that the cavity has no dispersion. The phases of the modes are

assumed to have a constant phase relationship (note that in this case it results in transform limited pulses but is not necessary always true) which is given as:

$$\varphi_l - \varphi_{l-1} = \varphi \quad (2.1.1)$$

Hence the electric field, $E(t)$, at any point in the output beam, can be expressed as:

$$E(t) = \sum_{-l}^l E_0 \text{Exp}(i(\omega_0 + l \Delta\omega)t + l \varphi) \quad (2.1.2)$$

where ω_0 is the central frequency of the laser and $\Delta\omega$ is the frequency between modes. Rearrangement of (2.1.2) such that the equation is now written as:

$$E(t) = A(t) \text{Exp}(i\omega_0 t) \quad (2.1.3)$$

where,

$$A(t) = \sum_{-l}^l E_0 \text{Exp}((\Delta\omega t + \varphi)il) \quad (2.1.4)$$

$A(t)$ can be further expanded as a geometric progression sum to infinity such that it now reads:

$$A(t) = \frac{E_0 \sin\left(\frac{\Delta\omega t}{2}(2m+1)\right)}{\sin\left(\frac{\Delta\omega t}{2}\right)} \quad (2.1.5)$$

Rearrangement and squaring of (2.1.5) yields the intensity, $I(t)$ as:

$$I(t) = (A(t)/E_0)^2 = \left(\frac{\sin\left(\frac{\Delta\omega t}{2}(2m+1)\right)}{\sin\left(\frac{\Delta\omega t}{2}\right)}\right)^2 \quad (2.1.6)$$

Equation (2.1.6) looks very similar to the interference equation in classical optics and mode-locking can be classified as interference phenomena where the oscillating modes interfere to produce a train of evenly spaced light pulses. By, L'hôpital rule, a peak is obtained whenever the denominator in equation (2.1.6) vanishes. Hence consecutive pulses are separated when the argument in the denominator approaches integer multiples of π . Hence, two consecutive pulses that are separated in time, T_R , is given by:

$$T_R = \frac{2\pi}{\Delta\omega} = \frac{1}{\Delta\nu} = \frac{2L}{c} \quad (2.1.7)$$

where $\Delta\nu$ is the separation of the pulses in frequency between two pulses or the cavity repetition rate, L is the length of the laser cavity and c is the speed of light. T_R is also known as the cavity round trip time for a mode-locked laser.

Fig. 2.1.3.: Mode-locked pulse trains plotted using equation (2.1.6). As the number of modes being “locked” together increases, the output pulses become more intense and the pulse width reduces.

Fig. 2.1.3 shows a plot of mode-locked pulse trains modeled using equation (2.1.6). It is worth mentioning that as the numbers of modes being locked together increases, the output intensity of the mode-locked laser increases and the pulse width reduces as shown in Fig 2.1.3. In a mode-locked laser, the number of modes being “locked” together is related to the gain and emission bandwidth of the gain medium. In general, the larger the gain/emission bandwidth the larger is the number of modes being locked together and as such the narrower are the output pulses and the higher is the average power of the laser. This is however a simplistic view of things to demonstrate that pulse duration is inversely proportional to the available bandwidth. Typically, Yb^{3+} and Nd^{3+} doped gain media have gain/emission bandwidth of tens of nanometers and less than a nanometer respectively. Hence Yb^{3+} [1] and Nd^{3+} [2] gain media are typically used to generate pulse widths in the femtosecond and picosecond regime respectively. Ti: sapphire on the other hand has a

gain/emission bandwidth that is well over 200 nm and as such is capable of generating pulse widths as short as a few femtoseconds [3-4]. It should be noted that the gain bandwidth is proportional to the emission bandwidth of the gain media. Hence, the emission bandwidth is a good approximation to the gain bandwidth.

Fig. 2.1.4.: Envelope modulated longitudinal modes in the frequency domain

The basis of the argument used in the derivation of equation (2.1.6) assumes that the oscillating modes in the frequency domain to be of equal amplitude and identical phases to simplify matters, but in reality, they are usually modulated by an envelope as show in Fig. 2.1.4. A typical type of modulation in lasers and can be expressed as:

$$E_l = \sqrt{E_0 \exp \left(- \left(\frac{2l\Delta\omega}{\Delta\omega} \right)^2 \ln 2 \right)} \quad (2.1.8)$$

Since the electric field is still expressed as (2.1.2), the electric field amplitude, $A(t)$ is expressed in a similar way as (2.1.3) and is given as:

$$A(t) = \int_{-\infty}^{\infty} E_l \exp(i(l \Delta\omega)t) \quad (2.1.9)$$

Inspection of (2.1.9) shows that it is just the Fourier transform of the amplitude spectral, E_l , in which the result can be written as:

$$A^2(t) \propto \exp \left(- \left(\frac{2t}{\Delta\tau_p} \right)^2 \ln 2 \right) \quad (2.1.10)$$

The Full Width at Half Maximum (FWHM) of the Gaussian pulse, $\Delta\tau_p$, which is also the laser pulse width is given as:

$$\Delta\tau_p = \frac{2 \ln 2}{\pi \Delta\nu_L} = \frac{0.441}{\Delta\nu_L} \quad (2.1.11)$$

where $\Delta\nu_L$ is the laser bandwidth. This is an example of the time bandwidth product (TBP) in which the pulse width is inversely proportional to the laser bandwidth. The factor of proportionality depends on the shape of the pulse. For transform limited Gaussian shaped pulses this takes the value of 0.441 while that for Sech^2 shaped pulses this value is taken to be 0.315. When the TBP are close to the values stated, the laser pulses are said to be transform limited. If the values deviate from transform limited values, it indicates the presences of chirp within the system.

Now that the basic idea of mode-locking has been described, the basic pulse shaping mechanisms in mode-locking and their relation to the Haus' Master equation of mode-locking shall be discussed in the next section.

2.2.: Passive Mode-locking

Passively mode-locking occurs due to the opening of a net gain window during a cavity round trip time and can be largely classified into three types as shown in Fig. 2.2.1. The first of which is a result of the interplay between a slow saturable absorber (SA) with dynamic gain saturation, the second mechanism relies purely on the properties of a fast recovering SA while the third method manifest itself due to effects with a slow SA. Fast in this sense implies that the recovery time of the SA is much shorter than the pulse width and slow implies the pulse width of the pulses are either longer or comparable to the recovery time the SA.

Fig 2.2.1.: Three different passive mode-locking pulse shaping mechanisms using (a) a slow saturable absorber with dynamic gain saturation (b) a fast saturable absorber and (c) a slow saturable absorber [5].

The explanation for pulse shaping using a slow SA with dynamic gain saturation was put forward by New *et. al* [6]. He showed that when a slow SA recovers faster than a saturated gain medium, a small gain window, capable of supporting ultrashort pulses opens up as depicted in Fig 2.2.1 (a). A net gain window is opened by saturating the SA which is then closed by the dynamic saturation of the gain. As such, the SA would have to have a smaller saturation energy than that of the gain such that the SA saturates first. Hence, Pulse shaping in dye and some semiconductor lasers is dictated by this mechanism due to their small gain saturation energy. In solid state lasers (SSLs), this mechanism is harder to realize due to their saturation energies.

Pulses that are shaped using a fast SA do not require any dynamic gain saturation and are assumed to have a gain that is constant over the duration of the pulse as shown in Fig. 2.2.1 (b). This leads to the formation of a net gain window that depends only on the dynamics of a fast recovering SA as shown in Fig. 2.2.4 (b). Pulse shaping in Kerr lens mode-locked (KLM) SSLs are explained using this model. When first demonstrated, this form of mode-locking was deemed as “magic mode-locking” as there was no apparent SA within the cavity [7]. It was only understood latter on that the gain medium actually served as an artificial Kerr lens and together with a hard aperture forms a type of artificial SA [8]. Hence, the Kerr effect is regarded to be instantaneous and as such considered to be a fast SA. The instantaneous SA effect of KLM has allowed some of the shortest pulses to be demonstrated and been the most successful mode-locking technique employed in academic and commercial lasers for the last twenty years. However, the realization of a KLM laser required that the cavity to be operated near the limits of its

stability and as such required critical cavity alignment and rigid designs. In addition, these types of lasers are non-self starting and require the system to be given a small disturbance typically in the form of a shake or a bang on the optical table for mode-locking to be initiated. Due to the success of KLM, it was thought that for a very long time that very short femtosecond pulses could only be achieved with fast SAs. In short both mode-locking using a slow SA with dynamic gain saturation and a fast SA results in the opening of a gain window that shuts just before and after the pulse during a cavity round trip time. The physical effect of this is that only the pulse experiences net gain and grows. Noises and perturbations that exist outside of this gain window are not allowed to grow and as such the mode-locking is stable.

Kärtner *et. al.* [9-11] however showed that very short femtosecond pulses were also possible with slow SAs but without the constraints set by KLM. Slow SAs take a longer time to recover when compared to the pulse width. This results in a gain window to exist after the pulse. One might assume the noise after the pulse maybe amplified and such results in the mode-locking to be unstable. This however is not the case as the noises are not intense enough to be amplified and are spreaded over a long period of time forming a so called continuum [9-11]. The low intensity continuum experiences negligible bandwidth broadening from self-phase modulation (SPM) and is further broaden in time by group velocity dispersion (GVD). In the end the continuum is either absorbed by the slowly recovering SA or it decays with time as it experiences less gain per round trip when compared to the pulse. Mode-locking in the presence of negative GVD with semiconductor SAs are best explained using this type of pulse shaping.

Haus' master equation of mode-locking

The three different mechanisms of mode-locking can be approximated by considering Haus' master equation [12] of mode-locking which takes into account the various pulse shaping effects that can modify the pulse envelope within the resonator of a passively mode-locked laser. The master equation of mode-locking is a time dependent analysis of the pulse in steady state. The Master equation of mode-locking takes the form of:

$$T_R \frac{\partial A(T,t)}{\partial T} = \sum_i \Delta A_i = 0 \quad (2.2.1)$$

where A is the pulse envelope, T_R is the cavity round trip time, t is the retarded time on the order of the pulse width, T is the time that develops depending on the time scale of the order of T_R and ΔA_i are the various pulse shaping effects within the cavity that make small changes to A . For passively mode-locked SSL, the various pulse shaping effects can be treated as small changes to the pulse envelope. Hence, after every round trip the pulse envelope can be considered unchanged as all the small changes due to the various pulse shaping effects sum to zero.

The pulse shaping effects are modeled as individual linearized operator in Haus' Master equation. Treatment of the operators in such a way allows one to study the effects of the various pulse shaping effects either separately or in tandem. For passively mode-locked lasers the commonly considered effects include gain, saturable absorption modulation, losses, GVD, SPM and gain filtering.

The linearized gain operator is derived by assuming that the gain medium under consideration. It is given as:

$$\begin{aligned}\Delta A &\approx [g + \frac{g}{\Omega_g^2} \frac{d^2}{dt^2}]A \\ &= [g + D_g \frac{d^2}{dt^2}]A\end{aligned}\tag{2.2.2}$$

where g is the gain, Ω_g is the Half Width at Half Maximum (HWHM) of the gain bandwidth and D_g is the gain dispersion or gain filtering.

The physical effect of GVD on an ultrashort pulse as it passes through a dispersive medium is to broaden it temporally resulting in longer pulses. This is usually detrimental to obtaining ultrashort pulses. In a passively mode-locked SSL system, the main dispersive medium is typically the gain medium. The linearized GVD operator is obtained by considering the wave number, $k_n(w)$, in a dispersive medium and Taylor expanding it. It is given as:

$$\Delta A \approx i \frac{1}{2} k_n'' z \frac{d^2}{dt^2} A$$

$$= iD \frac{d^2}{dt^2} A \quad (2.2.3)$$

where $k_n'' = \frac{\partial^2 k_n}{\partial \omega^2} |_{\omega=\omega_0}$ and is representative of the GVD of a dispersive medium and D is the dispersion parameter which is half of the total group delay dispersion per cavity round trip.

SPM is a non-linear physical phenomenon that arises due to the Kerr effect of the gain medium. As an ultrashort pulse propagates through the Kerr medium, it is broadened in the spectral domain such that additional frequencies are generated. The linearized SPM operator is given as:

$$\begin{aligned} \Delta A &\approx -i \frac{kn_z z}{L} |A|^2 A \\ &= -i\delta |A|^2 A \end{aligned} \quad (2.2.4)$$

where δ is the SPM coefficient.

The saturable absorption of a SA can be considered by assuming a simple two level atom. The SAM can be modeled as a linear operator by:

$$\Delta A = s(t) = \frac{s_0}{1 + \frac{I(t)}{I_{sat}}} \quad (2.2.5)$$

Where $s(t)$ is the saturable absorber loss, s_0 is the unsaturated loss, $I(t)$ is the intensity of the pulse and I_{sat} is the saturation intensity of the absorber. If the saturation of the absorber is weak, one can do a Taylor expansion such that:

$$\Delta A = s(t) = s_0 - s_0 \frac{I(t)}{I_{sat}} \quad (2.2.6)$$

If one normalizes $|A(t)|^2$ to be power, the equation (2.2.6) becomes:

$$\Delta A = s(t) = s_0 - \frac{s_0 |A|^2}{I_{sat} A_{eff}} = s_0 - \gamma |A|^2 \quad (2.2.7)$$

where γ is the self amplitude modulation (SAM) coefficient.

When all the relevant pulse shaping effects are considered together in equation (2.2.1), Haus' master equation of mode-locking takes the form:

$$T_R \frac{\partial A(T,t)}{\partial T} = \left[g \left(1 + \frac{1}{\Omega_g^2} \frac{d^2}{dt^2} \right) - l + iD \frac{d^2}{dt^2} + (\gamma - i\delta) |A(t)|^2 \right] A(t) = 0 \quad (2.2.8)$$

In Haus's master equation of mode-locking, the gain and its associated filtering effects due to a finite gain bandwidth are considered in the first term, the losses in the cavity are considered through l where s_0 in equation (2.2.6) has been absorbed into. Dispersion is taken into account by D , the saturable absorber action is represented by γ while SPM is considered through δ . Equation (2.2.8) is a type of complex Ginzburg-Landau equation which can be used to model various effects in science and engineering. Haus's master equation has a solution first purposed by Martinez *et.al.* [13]:

$$A(t) = A_0 \text{Sech}^{(1-i\beta)} \left(\frac{t}{\tau} \right) \quad (2.2.9)$$

where A_0 is the amplitude, τ is the pulsewidth and β is the chirp parameter. Substitution of equation (2.2.9) back into equation (2.2.8) results in normalized solutions for pulse width, τ_n , bandwidth, $\Delta\omega_n$, chirp parameter, β , and stability, $(l - g)$, respectively:

$$\tau_n = \frac{2-3D_n\beta+\beta^2}{\delta} \quad (2.2.10)$$

$$\Delta\omega_n = \frac{\sqrt{1+\beta^2}}{\tau_n} \quad (2.2.11)$$

$$\beta = -\frac{3}{2}\chi \pm \sqrt{\left(\frac{3}{2}\chi\right)^2 + 2} \quad (2.2.12)$$

$$l - g = (1 - \beta^2) - 2\beta D_n \quad (2.2.13)$$

Where D_n is the normalized dispersion and $\chi = \frac{\delta D_n - \gamma}{\delta + \gamma D_n}$. An analysis of equations (2.2.10) to (2.2.13) allows one to study the effects that the linear operators can have on a mode-locked laser. These equations are plotted as a function of D_n and shown in Fig 2.2.2.

Fig. 2.2.2.: (a) Plot of stability, $(l - g)$, (b) chirp, β , (c) normalized bandwidth, ω_n , and (d) normalized pulsewidth τ_n , as a function of normalized dispersion, D_n . The value of the SAM coefficient, γ , is kept at 1 while the value of the SPM coefficient is varied between 0 to 4.

Fig. 2.2.2 (a) shows too large an amount of SPM can result in the mode-locked system to become unstable. This is because for a stable system, the losses should be greater than the gain before and after the pulse such that noise is not allowed to grow to destabilize the laser system. Fig. 2.2.2 (b) shows that two distinct regimes of operations exist for positive and negative dispersions. In the negative dispersion regime, the SPM within the system can be very well compensated such that the amount of chirp is close to zero and as such the pulses obtained can be close to the Fourier transform limit. However as one moves into the positive dispersion regime, the chirp of the system increases significantly with increasing SPM as they become harder to balance and as such the pulses are far from the Fourier transform limit. Both cases will be examined latter in this thesis.

Fig. 2.2.3.: Schematic of the laser modeled by the Haus' master/ complex Ginzburg-Landau equation.

The implications of Fig. 2.2.2 (c) and (d) should be considered together as bandwidth is inversely proportional to pulsewidth due to their Fourier transform relationship. The bandwidth in Fig. 2.2.2 (c) increases because of the increasing SPM which results in the generation of extra frequencies and as the bandwidth increases, the pulsewidth reduces as shown in Fig. 2.2.2 (d) due to their Fourier transform relationship. In the absence of SPM, the bandwidths and pulsewidths are symmetrical about zero dispersion. The shortest pulse width occurs when dispersion in the system is zero because there is no SPM within the system causing any chirp and that zero dispersion does not stretch the pulse. As the amount of SPM in the system increases, extra bandwidth is generated and the symmetry as compared to the case where there is no SPM is broken. This is because the SPM generates a positive chirp in the system. When positive dispersion is added to the system it results in the pulsewidth to increase rapidly as shown in Fig. 2.2.2 (d). This is because the positive dispersion reinforces the positive chirp generated in the system. On the hand as the chirp generated by the system can be balanced with negative dispersion and under these conditions, the pulse is shaped by soliton effects and the pulse duration is minimized. Hence soliton shaping effects dominates in negative dispersion compensated mode-lock systems. For a more detailed analysis of Haus' master equation of mode-locking, the reader is referred to Haus *et. al.* [12, 14]. Fig. 2.2.3 summarizes the various linear operators considered in Haus' master equation of mode-locking.

The non-linear Schrödinger equation

By considering only the effects of SPM and GVD on an ultrashort pulse, Haus' master equation reduces to the non-linear Schrödinger equation (NLSE). Fig 2.2.3 shows a schematic of the laser modeled by the NLSE. The NLSE provides analytical insight into the effects that only SPM and GVD may have on ultrashort pulse propagation. The NLSE can be obtained by substituting the linearized operator for GVD and SPM into the

Master equation (i.e. equations (2.2.3) and (2.2.4) into equation (2.2.1)) such that one obtains:

$$T_R \frac{\partial A(T,t)}{\partial T} = iD \frac{d^2}{dt^2} A(t) - i\delta |A|^2 A(t) \quad (2.2.14)$$

Fig. 2.2.3.: Schematic of the laser modeled by the NLSE.

If an ultrashort pulse is modeled according to equation (2.2.14), a number of different outcomes are possible depending on the initial conditions such as intensity, pulse width, sign and value of the GVD and the size of the SPM term. Fig 2.2.4 shows the general trends in pulse propagation under different conditions. Fig. 2.2.4 (h) and (i) shows an interesting situation in which SPM is balanced by negative GVD resulting in pulses to remain unchanged as they propagate in both time and spectral domains. As mentioned before, such pulses are also known as solitons as the pulse shaping effects are dominated by negative dispersion and SPM. This is similar to the discussion in the previous section. ~~in which~~ The solutions to the NLSE and Haus' master equation are known as fundamental solitons or fundamental solitary pulses. The simulations shown in Fig 2.2.4. were plotted using a NLSE solver written by Agrawal [15].

The solution to the NLSE is a fundamental soliton and is given by a hyperbolic secant [16] which takes the form:

$$A(z, t) = A_0 \text{Sech} \left(\frac{t}{\tau} \right) \exp(-i\phi) \quad (2.2.15)$$

where A_0 is the peak value of the pulse envelope for a fundamental soliton, τ is the pulse duration parameter for a sech shaped pulse and ϕ is a phase shift. Solving the NLSE with (2.2.15) is non-trivial but yields the FWHM pulse duration of τ_p as:

$$\tau_p = 1.7627 \tau \quad (2.2.16)$$

and the corresponding TBP is given as:

$$\Delta\nu_p\tau_p = 0.315 \quad (2.2.17)$$

The TBP indicates the how close the measured pulse width is to the theoretical limit set by its spectral bandwidth. TBPs with values higher than 0.315 indicate the presence of chirp within the system. For a given negative GVD and pulse energy, E_p , the pulse duration obtainable is given as:

$$\tau_p = 1.7627 \frac{2|D|}{\delta E_p} \quad (2.2.18)$$

Negative GVD and SPM are strong pulse shaping effects and are sometimes termed as soliton formation effects [5] in the literature. Equation (2.2.18) shows simple relationship that when GVD and SPM are balanced the pulses propagate as solitons with the shortest possible pulsewidths. Equation (2.2.18) is also known as the soliton area theorem and is also valid for Haus' master equation of mode locking (equation 2.2.8). For the NLSE, the pulse shaping effects are only due to negative GVD and SPM while in the case of Haus' master equation, the pulse shaping effects are due to negative GVD and SPM but perturbed by the SAM of the SA, dissipative and gain filtering effects. In both cases, the dominating shaping effects are negative dispersion and SPM. When these two parameters dominate pulse shaping effects, the subsequent pulse is known as a soliton or solitary pulses. The mode-locking is as such termed as soliton mode-locking. The NLSE is a limiting case of Haus' master equation of mode-locking.

Fig. 2.2.3.: Stimulated pulse propagation using the NLSE in the time and frequency domains under various conditions. (a) and (b) corresponds to pulse propagation in the presence of only positive GVD. (c) and (d) corresponds to pulse propagation in the presence of only SPM. (e) and (f) corresponds to pulse propagation in the presence of positive GVD and SPM. (h) and (i) corresponds to pulse propagation in the presence of negative GVD and SPM.

Kärtner *et. al.* [9-11] used soliton perturbation theory by considering equation (2.2.10). He treated SAM, gain filtering effects and dissipative effects as small perturbations to the Haus' master equation of mode-locking and showed that soliton pulses and weak continuum pulses are formed due to soliton formation effects and small perturbation effects respectively. The continuum pulses can be thought of as competition effects that compete with the solitons for gain. Under favorable conditions, the continuum can reach lasing threshold and such destabilize soliton formation and by extension the mode-locking of the laser. The addition of a SA served to stabilize the mode-locking by introducing losses to the continuum such that it does not reach lasing threshold. In a sense, the continuum places a limit on the performance of ultrafast SSLs.

Fig 2.3.1.: Schematic of the electric field of a pulse before and after it passes through a positive dispersive medium.

2.3.: Pulse shaping effects

The discussion in section 2.2 indicates that GVD, SPM and saturable absorption are important effects in aiding the shaping of pulses and maintaining stable mode-locking operation. The origins and physical effects of these effects will be discussed in this section.

Dispersion and dispersion compensation

When an ultrashort pulse circulates within the laser cavity, it passes through a number of dispersive media such as mirrors and the gain medium. The net effect of these dispersive media is shown in Fig. 2.3.1 in which two physical effects can be observed. Firstly, the center of the pulse is delayed when compared to another one that has not transverse through any dispersive media. This is known as group delay and is not a pulse

broadening effect. Secondly, the pulse experiences a positive chirp in which the blue components are delayed with respect to the red components of the pulse. This changes the shape of the pulse by temporally broadening it. The second effect is detrimental towards realizing ultrashort pulses. Take the case of Yb: SSO lasers. Li *et. al.* [17] demonstrated 2.3 ps pulses in a dispersion compensation free setup while Tan *et. al.* [18] demonstrated 125 fs pulses in a dispersion compensated setup. Hence it is important to understand the origins of these dispersion effects and ways in which the effects of dispersion can be reversed. The origins of pulse broadening can be understood by taking note that as a pulse passes through a dispersive medium, it acquires a frequency dependent phase shift, $\phi(\omega)$, given by:

$$\phi(\omega) = kn(\omega)L \quad (2.3.1)$$

where k is the wave number, $n(\omega)$ is the refractive index and L is the length of the medium. The phase shift can be Taylor expanded about ω_0 , the central angular frequency such that:

$$\phi(\omega - \omega_0) = \phi_0 + \phi'(\omega - \omega_0) + \frac{\phi''(\omega - \omega_0)^2}{2} + \frac{\phi'''(\omega - \omega_0)^3}{6} + \dots \quad (2.3.2)$$

where ϕ_0 is a constant frequency term, $\phi'(\omega - \omega_0)$ is known as the group delay term and can also be written as $\frac{\partial \phi}{\partial \omega}$. This term does not modify the shape of the pulse but delays the pulse. $\phi''(\omega - \omega_0)^2$ which can also be written as $\frac{\partial^2 \phi}{\partial \omega^2}$ is known as the group delay dispersion (GDD) per unit length and is responsible for pulse broadening. It is related to the GVD when the total amount of dispersion contributed by the dispersive medium per unit length is considered. The terms positive dispersion and negative dispersion are related to the sign of the GVD term. For most optical materials in the 1 μm regime, GVD is usually positive and as such negative dispersion have to be applied. $\phi'''(\omega - \omega_0)^3$ which is also written as $\frac{\partial^3 \phi}{\partial \omega^3}$ is known as the third order dispersion (TOD). TOD is a higher order dispersion that usually does not manifest itself unless the pulse width become less than 30 fs [19] or when GVD is very well compensated.

To calculate the width of a Gaussian pulse after it has passed through a dispersive medium, consider its electric field, $E(t)$:

$$E(t) = \sqrt{A_t \exp\left(-\ln 2 \left(\frac{2t}{\tau_p}\right)^2\right)} \exp(-i(\omega_0 t + \phi(t))) \quad (2.3.3)$$

where A_t is the amplitude of the pulse, ω_0 is the central frequency of the pulse, τ_p is the pulse width and $\phi(t)$ is the phase. To determine the pulse width after passing through a dispersive medium would require one to solve a convolution integral which is not trivial. Hence it would be easier to work in the frequency domain by Fourier transforming (2.3.3). This yields:

$$E(\omega) = \sqrt{A_\omega \exp\left(-\ln 2 \left(\frac{2(\omega - \omega_0)}{\Delta\omega}\right)^2\right)} \exp(-i\phi(\omega - \omega_0)) \quad (2.3.4)$$

where A_ω is the spectral amplitude, $\Delta\omega$ is the spectral frequency and $\phi(\omega)$ is the spectral phase which describes the relationship between the frequency components of the pulse. Hence the output electric field, $E_{out}(\omega)$, is given by just the multiplication of the transfer function of the dispersive medium and equation (2.3.4), the input electric field, $E_{in}(\omega)$, which is:

$$E_{out}(\omega) = E_{in}(\omega)R(\omega) \quad (2.3.5)$$

where $R(\omega)$ is the transfer function of the dispersive medium is taken as 1 [20] if the material is a linear transparent medium. Only second order effects in equation (2.3.2) were considered and substituted along with equation (2.3.4) into equation (2.3.5) yielding:

$$E_{out}(\omega) = \sqrt{A_\omega \exp\left(-\ln 2 \left(\frac{2(\omega - \omega_0)}{\Delta\omega}\right)^2\right)} \exp\left(-i\left(\frac{\partial^2 \phi}{\partial \omega^2}\right) \frac{(\omega - \omega_0)^2}{2}\right) \quad (2.3.6)$$

To obtain the pulse width, it is necessary to work in the time domain and as such inverse Fourier transforming of (2.3.6) results in:

$$E_{out}(t) = \sqrt{A'_t} \exp\left(\frac{4 \ln(2) t^2}{2(\Delta t^2 + i4(\ln 2)^2 \frac{\partial^2 \phi}{\partial \omega^2})}\right) \quad (2.3.7)$$

Fig 2.3.2: Variation of $\frac{\Delta t_{out}}{\tau_p}$ with Δt as the GDD is increased from 200 fs^2 to 1000 fs^2 . Curve nearest to the axes represent the case in which GDD was 200 fs^2 and increases in steps of 200 fs^2 .

where $\frac{\partial^2 \phi}{\partial \omega^2}$ is the sum of the GDD of the material and τ_p is the original pulse width. To obtain the output pulse duration, t_{out} , it is necessary to normalize (2.3.7) and squaring it and then equating it to the general intensity of a Gaussian pulse such that:

$$\exp \left(-\ln 2 \left(\frac{2t}{\Delta t_{out}} \right)^2 \right) = \exp \left(\frac{4 \ln(2)t^2}{\tau_p^4 + 16(\ln 2)^2 \left(\frac{\partial^2 \phi}{\partial \omega^2} \right)^2} \right) \quad (2.3.8)$$

and solving for (2.3.8) for Δt_{out} yields:

$$\Delta t_{out} = \frac{\sqrt{\tau_p^4 + 16(\ln 2)^2 \left(\frac{\partial^2 \phi}{\partial \omega^2} \right)^2}}{\tau_p^4} \quad (2.3.9)$$

Fig 2.3.2 shows the variation of $\frac{\Delta t_{out}}{\Delta \tau_p}$ which is also the pulse broadening factor with $\Delta \tau_p$. For pulses about 100 fs, the pulse does not broaden significantly when passing through a dispersive material but when the pulse width falls below 50 fs, $\frac{\Delta t_{out}}{\Delta \tau_p}$, increases rapidly. Hence for measurement of pulses shorter than 50 fs, the thickness and material of the output coupler and the steering optics have to be chosen carefully to prevent the pulse

width from broadening significantly. For picosecond lasers dispersion compensation is unimportant as the pulse width can range from a few picoseconds to tens of picoseconds.

GVD within a cavity is usually mainly contributed by the gain medium. Contributions by mirrors are typically negligible if low GVD optics are used. The GVD introduced by the gain medium of length, L , can be determined in terms of measurables given by:

$$\frac{\partial^2 \phi}{\partial \omega^2} = \frac{\lambda^3 L}{2\pi c^2} \frac{\partial^2 n(\lambda)}{\partial \lambda^2} \quad (2.3.10)$$

where $n(\lambda)$ is the refractive index of the material and λ is the central wavelength. $n(\lambda)$ is measured using an ellipsometry and then fitted with a Sellmeier type equation to extract the required coefficients [21]. The equations are then differentiated to obtain the corresponding derivative required. Inspection of equation (2.3.10) indicates that it is positive. Hence GVD in the cavity contributed by the gain medium is positive.

Fig 2.3.3.: Brewster cut dispersion prism pair used to generate negative dispersion in laser cavities.

Fig 2.3.4.: Solid and broken lines are GDD calculations due to a piece of Yb : $YAl_3(BO_3)_4$ (Yb: YAB) 5 mm long and a pair of SF 10 prisms separated by 30 cm respectively. The dotted line represents the total the total GDD within the cavity.

The dispersion prism pair

The most popular method of introducing negative dispersion into a laser cavity is through the usage of a pair of Brewster angle cut intra-cavity dispersion prisms. The advantages of using a prism pair include its low costs, ability to introduce a variable of negative dispersion, low insertion losses and allows for a relatively compact setup. It introduces negative dispersion into the cavity through geometrical means as shown in Fig. 2.3.3. The amount of negative dispersion introduced is proportional to the distances between the two apexes of the prisms. The first prism angularly disperses the incident beam while the second prism collimates the beam. This provides for a longer path difference for the red than the blue wavelengths. This extra path difference allows the blue components to “catch up” with the red ones thus recompressing the pulse. The prism pair is aligned in such a way that both faces are parallel to each other and at the Brewster angle such that the minimum insertion loss is introduced into the cavity. During alignment of the prisms, care is taken such that both faces of the prisms remain parallel. This is much easier to achieve by allowing one of the prisms, usually the one closer to the

output coupler to be fixed and that the one further away to be translated into and out of the optical path of the laser. By doing so, it allows a variable amount of negative dispersion to be added to the cavity. For compact lasers, a pair of SF 10 prisms offers the best performances for obtaining pulses that ranged from tens of femtoseconds to hundreds of femtoseconds as they would require a smaller prism separation to obtain the same amount of negative dispersion as compared to a pair of prisms made of fused quartz. However, fused quartz tends to suffer less from higher order dispersion and is used if sub-10 femtosecond pulses are required. Lemoff *et. al.* [22] studied dispersion compensation using prism pairs made from different materials and found that by selecting the correct prism material, second and TOD can be fully compensated.

The total amount of negative dispersion, $\frac{\partial^2 \phi_p}{\partial \omega^2}$, introduced by a prism pair has been demonstrated by Salin *et. al.* [23] to be:

$$\frac{\partial^2 \phi_p}{\partial \omega^2} = \frac{4\lambda^3}{\pi c^2} \left[-L_p \frac{\partial n_p}{\partial \lambda}(\lambda) + e \left(\frac{n_p(\lambda) \frac{\partial^2 n_p(\lambda)}{\partial \lambda^2}}{1 + n_p^2(\lambda)} \right) + \frac{\partial n_p}{\partial \lambda} \left(1 - \frac{1}{n_p^2(\lambda)(1 + n_p^2(\lambda))} \right) \right] \quad (2.3.11)$$

where $n_p(\lambda)$ is the wavelength dependent refractive index of the prisms which can be obtained from the Sellmeier type equations [23], e is the perpendicular distance from the tip of the apex of a prism to the laser beam in the cavity and L_p is the prism apex to apex separation. An analysis of equation (2.3.11) shows that the first terms in the brackets is the dispersion contribution by due to the prism separation and that it is always negative due to the negative sign. The second term in equation (2.3.11) is additional positive dispersion added to the cavity by the prism material as the beam passes through it. The importance of equations (2.3.10) and (2.3.11) is that they allow one to calculate if the laser system was operating in the positive or negative dispersion regime. To initiate solitary mode-locking one needs to operate the laser in negative dispersion regime and as such set L_p to a distance that is long enough to compensate for the positive dispersion of the gain medium. However, it should be noted that Sellmeier type equations of certain gain medium may not always be available but fortunately, their GVD are typically in the range of a few hundreds of fs² and can be assumed to be in this region as such can compensated by L_p separation that yields that is at least a few hundreds of negative fs².

Fig. 2.3.4 shows calculations of the GDD due to a piece of 5 mm Yb: $\text{YAl}_3(\text{BO}_3)_4$ (Yb: YAB), the GVD due to a pair of SF 10 dispersion prisms and the total cavity dispersion using equations (2.3.10) and (2.3.11). The Sellmeier equation for Yb: YAB was extracted from ref. [24]. Fig. 2.3.4 shows that the laser is operating in the negative dispersion regime which is required for soliton mode-locking.

Fig 2.3.5.: Diffraction grating pair.

The dispersion grating pair

Dispersion compensation can also be provided by using a pair of diffraction gratings. It provides negative dispersion using the same principals as a pair of diffraction gratings by providing a longer optical path for red wavelengths as compared to the blue ones as shown in Fig. 2.3.5. Diffraction gratings are typically employed as extra-cavity compression elements due to the large losses they introduce and are used when the emitted pulse has non-transform limited output. The advantage of using a pair of diffraction gratings is that they provide dispersion compensation many times larger than that possible by dispersion prisms in very compact setups. The total GVD provided by a pair of dispersion grating is given by:

$$\frac{\partial^2 \phi_p}{\partial \omega^2} = \frac{-\lambda^3 L_g}{\pi c^2 \Lambda^2} (1 - (\frac{\lambda}{\Lambda} - \sin \theta_i)^2)^{-\frac{3}{2}} \quad (2.3.12)$$

where L_g is the grating spacing, Λ is the grating period and θ_i is the incident angle of the pulse to be compressed.

Fig 2.3.6.: SPM effects on a Gaussian pulse.

Self phase modulation

The Kerr effect of a gain medium is also known as the intensity dependent refractive index (IDRI). It is a third order non-linear optical effect that arises due to the third order

non-linear electric polarization of atoms of the gain medium. This effect gives rise to an IDRI, $n(I)$, which is written as:

$$n(I) = n_0 + n_2 I(r, t) \quad (2.3.13)$$

where n_0 is the linear refractive index, n_2 is the non-linear refractive index and $I(r, t)$ is the intensity of the pulse. The value of n_2 determines the strength of the Kerr effect within a gain medium and this value is typically in the order of $10^{-16} \text{cm}^2 \text{W}^{-1}$ for laser materials.

Closer inspection of equation (2.3.13) shows that the IDRI has both a radial and time dependence. The radial dependence gives rise to the phenomena of KLM [5] while the time dependence results in SPM [25] of a pulse. SPM is a change in the phase of the pulse due to IDRI of the gain medium and the most important effect in relation to mode-locking is that extra bandwidths are generated. The extra bandwidth generated results in the optical spectrum to be broadened and as such pulse compression techniques can be applied to generate ultrashort pulses. If the pulse is Gaussian, then the IDRI becomes:

$$n(I) = n_0 + n_2 (I_0 \exp(\frac{-t^2}{\tau_p^2})) \quad (2.3.14)$$

Since the phase of a pulse is defined to be:

$$\phi = \omega_0 t - \frac{n(t)\omega_0 z}{c} \quad (2.3.15)$$

where z is the distance propagated by the pulse. Therefore, the angular frequency, $\omega(t)$, which is defined as the rate of change of phase can then be written as:

$$\omega(t) = \frac{d\phi}{dt} = \omega_0 - \frac{dn(t)}{dt} \omega_0 z \quad (2.3.16)$$

Substitution of equation (2.3.14) into equation (2.3.15) after the pulse has travelled a distance of L yields:

$$\omega(t) - \omega_0 = \Delta\omega = \frac{4\pi L n_2 I_0 t}{\lambda_0 \tau_p^2} \exp(\frac{t^2}{\tau_p^2}) \quad (2.3.17)$$

This equation implies the generation of extra bandwidth. A plot of the frequency of a pulse under the effects of SPM can be obtained with equation (2.3.17) and is shown in Fig. 2.3.6. It shows the leading edge of the pulse shifting to lower frequencies while the trailing edge shifts to higher frequencies. It should be noted that SPM results in a linear increase in frequencies near the center of the pulse and when this increase is adequately compensated with the correct amount of negative GVD, fundamental solitons with transformed limited pulse widths can be generated as discussed in section 2.2.

Saturable absorbers

SAs are passive intra-cavity optical components added to a laser cavity to initiate mode-locking. A SA usually introduces high losses for low intensity pulses and vice-versa for high intensity pulses. Since the peaks of pulses are more intense, they saturate the absorber more strongly than the lower intensity wings. The resulting effect is that the high intensity pulses are amplified at the expense of the less intense ones such that ultrashort pulses are obtained. Lasers mode-locked through the usage of SAs are capable of generating picosecond and femtosecond pulses and as such the preferred technique used to achieve mode-locking. The semiconductor saturable absorber mirror (SESAM) is the most widely used SA for initiating mode-locking [8, 25]. Worth mentioning is that in recent years, carbon based nano-materials such as graphene and single walled carbon nanotubes (SWCNTs) have also been investigated as alternative SAs [25-26].

Fig 2.3.7.: Schematic of a SESAM.

Semiconductor saturable absorber mirrors

The SESAM was invented by Keller *et. al.* [28] in 1992 in which semiconductor quantum wells were embedded between two mirror structures such as that shown Fig 2.3.7. The SESAM is the most widely used SA as it offers superior and stable performances. Its properties and fabrication techniques are well understood to the extent of commercialization [29]. The operating wavelength of a SESAM can be varied by tuning the semiconductor composition and the type of semiconducting material used. Typically for wavelengths around 1000 nm, InGaAs is used. The saturation absorption mechanism in a SESAM is depicted in Fig 2.3.8 in which carriers in the valance band (Fig 2.3.8 (a)) absorb incident photon energy and are promoted into the conduction band. When all the carriers in the valance band are promoted into the conduction band the SESAM becomes saturated or bleached (Fig 2.3.8(b)). As there are no more carriers to be promoted into the conduction band, any excess incident photon energy just passes through the SESAM as it becomes highly transparent (Fig 2.3.8 (c)). The SESAM then partially recovers its absorption through processes such as thermalization, recombination and trapping [8]. These processes are related to the recovery times of the SESAM. After the SESAM recovers its absorption, the whole cycle of saturable absorption repeats.

Fig 2.3.8.: Saturable absorption mechanism in a SESAM. (a) Absorption, (b) saturation and (c) transparency.

Fig 2.3.8.: A plot of the nonlinear reflectivity of a SESAM.

The saturable absorption on the SESAM results in it to have a non-linear reflectivity in which as the intensity increases, the reflectivity increases and this is depicted in Fig. 2.3.9. Apart from showing the non-linear reflectivity of the SESAM, such a plot also shows other important parameters such as the modulation depth, ΔR , saturation intensity, I_{Sat} , and non-saturable losses, ΔR_{ns} . I_{Sat} measures the value in which the SESAM becomes saturated or bleached. Typically this value is extracted by fitting experimental data to a mathematical model for non-linear reflection. ΔR , measures the maximum change in reflectivity for low intensity and high intensity light and for mode-locked systems, the larger this value is, the shorter are resulting pulse widths [11]. Usually, as ΔR increases, so does ΔR_{ns} , which measures the losses introduced by a SESAM when it is fully saturated. To fully characterize, the SESAM, one would need knowledge of the recovery times. It consists of a fast component that occurs on the time scale of 60 – 300 fs [8] and is related to the thermalization processes and a slower component that occurs on a time scale of a few picoseconds to nanoseconds [8] and is related to interband trapping and recombination processes. These recovery times are usually measured using a pump probe system.

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Chapter 3

Laser Engineering

In this chapter, general aspects of laser engineering will be discussed. The first section provides a discussion on gain media and some of their properties related to laser performance. It also provides a survey of the results reported in the literature to date of some of the single crystal gain media. Ceramic gain media are not discussed in this thesis. The second section describes figure of merits that define laser performances. The third section discusses about laser cavity designs and Gaussian beam optics.

Fig 3.1.1: The electronic structure for (a) Nd^{3+} and (b) Yb^{3+} doped gain medium. Solid lines pointing upwards and downwards represent absorption and emission respectively. The broken arrow point upwards represents re-absorption. Energy levels are not drawn to scale.

3.1.: Neodymium and ytterbium gain media

Neodymium (Nd^{3+}) and ytterbium (Yb^{3+}) are both trivalent rare earth ions which when doped in a host matrix allows the generation of laser light around 1 μm . The development of SSL gain media started with Nd^{3+} doped hosts but it was not until the 1990's where a suitable diode pump source was developed did the Yb^{3+} doped hosts started to gain popularity. Today, both types of ions are still used to dope different host materials but their areas of application differ.

Subtle differences exist between the two ions. The Nd^{3+} doped gain medium is a four level system and has an electronic structure as shown in Fig. 3.1.1 (a). During pumping and lasing only the $^4\text{I}_{9/2}$, $^4\text{I}_{11/2}$, $^4\text{F}_{3/2}$ and $^4\text{F}_{5/2}$ manifolds are involved in transitions. A pump first pumps the ground state ions from the $^4\text{I}_{9/2}$ manifold to the metastable state at the $^4\text{F}_{5/2}$ manifold. Thermalization then relaxes the ions to the lowest levels of the $^4\text{F}_{3/2}$ manifolds and laser action occurs with the $^4\text{I}_{11/2}$ manifolds which are also known as the upper and lower lasing levels respectively.

Yb^{3+} doped gain media are three level systems and has a very simple electronic structure that consist of only two manifolds. When an ion is pump from the ground state $^2\text{F}_{7/2}$ manifold, it is excited to the $^2\text{F}_{5/2}$ manifold. Thermalization results in the ion to relax to the lowest level of the $^2\text{F}_{5/2}$ manifold where laser action then occurs between this manifold and the $^2\text{F}_{7/2}$ manifold. Hence the $^2\text{F}_{5/2}$ and $^2\text{F}_{7/2}$ manifolds serve as the upper and lower lasing levels respectively. A key difference to that of the Nd^{3+} ion is that the lower lasing level and the ground state are common. This results in re-absorbtion of the lasing wavelength as shown in Fig. 3.1.1 (b) and as leads to a reduction in gain. Moreover, Nd^{3+} doped gain media tend to have an emission cross-section, σ_{em} , that is at least an order of magnitude bigger than its Yb^{3+} counterparts and as such a larger gain. Take the example of $\text{Nd: Y}_3\text{Al}_5\text{O}_{12}$ (Nd: YAG) which has a σ_{em} of $28 \times 10^{-20} \text{ cm}^2$ [1] compared to that of $\text{Yb: Y}_3\text{Al}_5\text{O}_{12}$ (Yb: YAG) which has a σ_{em} of $2.1 \times 10^{-20} \text{ cm}^2$ [1]. In addition, the pumping threshold for such a three level system is much higher than that of a Nd^{3+} doped gain medium as it would require at least half of the ground state ions to be excited before lasing threshold can be reached [2]. Nd^{3+} gain media would have an advantage over its Yb^{3+} doped counterparts in terms of applications that require low CW

thresholds such as in laser pointers and in cases where extremely high power CW generation such as in laser fusion experiments [3] and certain industrial machining lasers.

The common ground and lower lasing level in Yb^{3+} gain media also results in its spectroscopic performances to be strongly host dependent [4]. As the host changes, the crystal field environment around the Yb^{3+} ion and its subsequent effects differ as well. In general, when a Yb^{3+} ion is introduced into a crystal field, the $^2\text{F}_{5/2}$ and $^2\text{F}_{7/2}$ manifolds would experience a host dependent Stark splitting. This will spread the energy levels across a distribution which has the effect of broadening the emission spectrum. In addition Yb^{3+} doped gain media can exhibit very strong electron-phonon coupling effects [4] which result in the emission spectra to be homogeneously broadened. These two broadening effects work in tandem resulting in Yb^{3+} doped gain media to have broadband emission spectra that can span several tens of nanometers allowing them to support the generation of femtosecond pulses. In Nd^{3+} doped gain media such effects do not affect their emission spectra as strongly as their Yb^{3+} counterparts and as such the emission spectra are generally limited to less than one nanometer which limits pulse generation to the picosecond regime. Yb^{3+} doped gain media are preferred over its Nd^{3+} counterparts for the generation of femtosecond pulses due to their superior spectroscopic properties. However Nd^{3+} doped gain media, due to its higher gain and longer lifetimes still finds applications as high power pico-second light sources [5-7] and Q-switched lasers [8-9].

Choice of gain medium

The choice of the gain medium used in SSLs depends on the application and has an important bearing on its performances. For high power operations, gain media with good thermal properties are often desired. This is because under steady state operation, the gain medium will emit heat which can have an adverse reaction on laser performance [10]. The dissipated heat can result in large thermal built-up and strong thermal lensing. This phenomenon can modify the laser spot size resulting in the cavity to becoming unstable [2] or degradation in the laser output beam quality [2]. The large thermal built-up can also result in the formation of a large thermal gradient between the internal and external parts of gain medium and uneven expansion can ultimately lead to stress fracture. Hence, it is desirable for a gain medium operating at high output powers to have

a large thermal conductivity, κ , such that any built-up heat can be dissipated rapidly. Hence the value of κ places a limit on the maximum achievable output power. Fig. 3.1.2 shows a plot of the emission bandwidth, $\Delta\lambda$, and their corresponding κ values for a number of Nd^{3+} and Yb^{3+} doped single crystal gain media. Laser materials with cubic structure such as Nd: YAG and Yb: YAG have extremely good thermal properties in which their κ are close to $11 \text{ W m}^{-1} \text{ K}^{-1}$ and have demonstrated mode-lock laser output with 14 W [11] and 8.1 W [12] respectively using a diode end pumping schemes. On the other hand, glass hosts, due to their highly disordered structure, have κ values of less than $1 \text{ W m}^{-1} \text{ K}^{-1}$ [4], could only generate output powers of 84 mW [13] and 64 mW [14] for Nd: glass and Yb: glass respectively. For high power generation, a high κ is required.

Fig 3.1.2.: A plot showing the $\Delta\lambda$ and the corresponding κ of Nd^{3+} and Yb^{3+} doped single crystal gain media.

The generation of femtosecond pulses however places an additional constraint to the choice of gain media as only materials with broad emission bandwidths can generate pulses of such narrow widths. Equally important is that near the peak of the emission spectrum, the curvature must be as gentle as possible [15]. This is because when a laser is mode-locked, it does so about the bandwidth near the peak due to the availability of

larger gain. If the curvature of the peak is smooth, more bandwidth can be involved in the mode-locking and as such generate shorter pulses. In addition to the broadening mechanisms discussed in the previous section, glass hosts introduce an additional broadening mechanism to their gain spectra and by extension their emission spectra through inhomogeneous broadening [16]. This results in emission bandwidths of 20 nm [4] and 35 nm [4] in Nd: glass and Yb: glass respectively. It is only through introducing a Nd^{3+} ion to a disordered environment can this type of ion have a broad enough emission spectrum capable of supporting femtosecond pulses. To that end, 60 fs [13] and 58 fs [14] pulses have been generated using Nd: glass and Yb: glass respectively as shown in Fig 3.1.3. Although ultrashort pulse generation has been demonstrated, high power output has yet to be realized using glass hosts. Low κ values of the glass hosts make high brightness and high power pumping impossible and as such limits their output powers. Clearly for the generation of high power ultrashort pulses, a gain medium should ideally have a broad and gentle emission bandwidth with a high value of κ . Most crystal media generally succeeds in one of these criteria but fails in the other one. This is because, broadband and smooth emission spectra are usually associated with a highly disordered environment around the laser ion which interferes with lattice vibrations and as such results in poor thermal properties.

Ordered hosts

In ordered hosts, the environment surrounding the active ions is similar and as such the crystal field effects are uniform around the ion. Due to its ordered structure, this class of host is also characterized by having very high κ values and as such is highly suitable for usage in high power applications. Prominent ordered hosts would include the aluminum garnets (e.g. Nd: YAG [11], Yb: YAG [12] etc.), the sesquioxides (Yb: Sc_2O_3 [17] and Yb: Lu_2O_3 [18]) the fluorides (Yb: CaF_2 [19]), the oxyorthosilicates (Yb: Y_2SiO_5 (Yb: YSO) [20], Yb: Lu_2SiO_5 (Yb: LSO) [20], Yb: Sc_2SiO_5 (Yb: SSO) [21] and Yb: Gd_2SiO_5 (Yb: GSO) [22] the vanadates (Nd: GdVO_4 [23], Nd: YVO_4 [24], Nd: YLuVO_4 [25], Yb: YVO_4 [26], Yb: LuVO_4 [27] and Yb: GdVO_4 [28]) and the double tungstates (Yb: $\text{KY}(\text{WO}_4)_2$ (Yb: KYW) [29] and Yb: $\text{KGd}(\text{WO}_4)_2$ (Yb: KGW) [30]).

Fig 3.1.3.: A plot showing the femtosecond pulse widths and their corresponding output powers of end pumped Nd^{3+} and Yb^{3+} doped single crystal lasers. Gain media in red are some of the results that will be demonstrated in this thesis.

Intrinsic properties of Nd^{3+} doped in ordered hosts result in very narrow emission bandwidths, limiting their pulse widths to the picosecond regime. However due to their high σ_{em} values, they are still used in high power applications for both CW and picosecond lasers. Particularly in the case of Nd^{3+} doped vanadates, where their σ_{em} are higher compared to that of Nd: YAG, are finding increasing applications in industrial lasers. Compared to Nd: YAG, whose output is unpolarized, the output of Nd doped vanadate is polarized and as such can be frequency doubled to act as a pump for Ti: sapphire lasers. Moreover, vanadates being birefringent material can be manipulated to generate high CW output powers at two wavelengths simultaneously [31]. In chapter 4, the author shall demonstrate through laser engineering and exploitation of its birefringence property and thermal lensing, a passively mode-locked dual wavelength Nd: GdVO₄ laser with orthogonal polarizations. Such a laser could be used in laser interferometry [32] where dual wavelengths with orthogonal polarizations are required. The wavelength separation in this laser can reach THz regime and as such could increase the scanning speeds for laser interferometers drastically.

Ordered Yb^{3+} doped gain media have been routinely used to generate femtosecond pulsewidths as shown in Fig 3.1.3 and as such finding increasing applications directly diode pumped femtosecond lasers. The most famous of the ordered Yb^{3+} doped gain media is most likely to be Yb: YAG. This material has one of the highest κ values among all Yb^{3+} doped gain media as shown in Fig 3.1.2 however, its emission bandwidth about the peak is extremely steep and sharp and limited to about 9 nm. Hence the usage of this material to generate sub 300 fs [4] proved to be difficult. This resulted in a search for alternate host materials for Yb^{3+} ions with superior spectroscopic properties while retaining high κ values had led to the development of Yb^{3+} doped sesquioxides, oxyorthosilicates, fluorides, vanadates and double tungstates. The Yb^{3+} sesquioxides were found to have excellent κ values reaching as high as $17 \text{ W K}^{-1} \text{ m}^{-1}$ and are the highest among all Yb^{3+} host material reported to date. However, the peak of their emission spectra remained sharp and steep while the emission bandwidths were only moderately broader (Yb: $\text{Sc}_2\text{O}_3 \sim 12 \text{ nm}$) than that of Yb: YAG ($\sim 9 \text{ nm}$). Despite this, sub 250 fs has been demonstrated with this type of gain media. This type of gain media due to its high κ values and large σ_{em} (compared to Yb: YAG) would be better suited for use in thin disk lasers setup in which Yb: Lu_2O_3 [33] have been demonstrated to emit 370 fs pulses with an output power 20.5 W. Due to its high average power and ultrashort pulse output such a laser would be tool for micromaching as it does not result in thermal shock to the workpiece and as such provide a sub-surface damage free finish [34].

Yb^{3+} doped oxyorthosilicates are hosts with monoclinic structures and have emission bandwidths that are broader than that of Yb: YAG and the sesquioxides. Moreover, Yb^{3+} doped oxyorthosilicates have reasonable κ values of at least $4 \text{ W K}^{-1} \text{ m}^{-1}$ with the exception of Yb: GSO which has a value lower than $3 \text{ W K}^{-1} \text{ m}^{-1}$. The most impressive results among the Yb^{3+} doped oxyorthosilicates have been demonstrated using Yb: LSO and Yb: YSO [20] which generated output powers of 2.6 W and 2.61 W respectively with pulsewidths of 260 fs and 198 fs. Although Yb: GSO has the broadest emission bandwidths among the Yb^{3+} doped oxyorthosilicates, the reported pulse width of 340 fs [22] is the broadest among this group of gain media. This is most likely to inadequate dispersion compensation and an unsuitable cavity design. Should these two factors be improved, much shorter pulse widths should be achievable. In this thesis, the

performance of Yb: SSO is presented. It shall be demonstrated in chapter 5 that this gain medium is able to generate pulses as short as 93 fs which is the shortest reported to date for Yb³⁺ doped oxyorthosilicates.

Yb³⁺ doped vanadates are a unique class of Yb³⁺ gain media in that they have emission spectra that are much broader than that of Yb³⁺ doped garnets, sesquioxides, and oxyorthosilicates. In fact, they have an emission bandwidth almost as broad as that of Yb: glass (> 30 nm) and that their emission spectra are glass like in the sense that there are no peaks and vary gently across a large bandwidths [26-27]. In addition, this class of gain media avoids the pitfalls of Yb: glass in that they have κ values of greater than 5 W K⁻¹ m⁻¹ making them suitable for high power operations without requiring to resort to complicated pumping and cavity designs [35]. High power femtosecond pulses have been demonstrated using Yb: YVO₄ [26] and Yb: LuVO₄ [27]. For Yb: GdVO₄, 3.1 ps pulses with 1 W average output power have been demonstrated. No femtosecond output has been demonstrated with this gain medium. However, given its wide emission spectrum and high κ value, the author speculates that this gain medium can generate high power sub-100 fs pulses.

Disordered hosts

In disordered hosts, the structure of the host media, as the name suggest, is disordered. This results in the active ion to be randomly surrounded by ions from the host material and this leads to considerable inhomogeneous broadening of the emission spectra. This broadening mechanism is very strong to the extent that even Nd³⁺ doped gain media which typically have very narrow emission bandwidths can be broadened to the extent of supporting femtosecond pulses [36]. Apart from being broadband, the emission spectra in disordered Yb³⁺ doped hosts also vary gently across a large bandwidth and are devoid of sharp and narrow peaks. In fact the emission spectra of disordered Yb³⁺ doped gain media is the broadest and smoothest amongst all types of Yb³⁺ doped gain media. This large broadening of the emission spectra however comes at a price as the disordered structure disrupts thermal flow and as such drastically reduces the κ values of this class of gain media.

Examples of this class of hosts would include Yb: glass [13], Yb: $\text{Sr}_3\text{Y}(\text{BO}_3)_3$ (Yb: BOYS) [7] and Yb: $\text{SrY}_4(\text{SiO}_4)_3\text{O}$ (Yb: SYS) [8] which have emission bandwidths that ranged from 30 nm to about 70 nm. Although this emission bandwidth would in theory allow the generation of sub-100 fs pulses their κ values of below $3 \text{ W K}^{-1} \text{ m}^{-1}$ would imply that their applications for high power applications maybe limited without the usage of complicated pumping systems or by artificially increasing the thermal conductivity of these host by bonding an additional crystal to serve as a heat sink [37]. It may seem that the low κ of disordered crystal would limit their uses in low power applications. However expectations to the trend in which a high amount of disorder in the hosts structure results in low κ exists and examples include Yb: CaGdAlO_4 (Yb: CAIGO) [9] and Yb: CaYAlO_4 (Yb: CYA) [10]. High disorder exists within these crystals, to the extent that both crystals have an emission bandwidth of about 80 nm which is the broadest amongst all Yb^{3+} doped gain media to be ever reported. However, these gain media still yielded values of κ that are higher than $3.7 \text{ W K}^{-1} \text{ m}^{-1}$. To that end, Yb: CAIGO and Yb: CYA demonstrated 68 fs and 156 fs pulses respectively with output powers of 520 mW and 760 mW. For CW output, both crystals generated lasers with average powers of greater than 2 W [9, 38]. These results are amongst the best ever reported for Yb^{3+} doped solid state lasers using an end pumped configuration. Results obtained with Yb: CYA crystal will be demonstrated in chapter 5. In addition, the author shall also demonstrate the high power femtosecond mode-locking of Yb: CYA in the positive dispersion regime in chapter 5.

Mixed crystal hosts

Mixed crystals are another class of host material commonly used in SSLs. The idea is to mix two types of ordered host together to obtain a new type of solid state mixture with partial disorder such that the emission bandwidth can be broadened but at the same time retain some of the κ value of the parent hosts. Mixed crystal hosts usually introduce a disorder that is less than what disordered crystal host would. A good example of a mixed crystal host would be Nd: YLuVO_4 [25] which is obtained by mixing solid state solutions of Nd: YVO_4 [24] and Nd: LuVO_4 [26]. Another example would be Yb: LuScO_3 [39] which is obtained by mixing Yb: Sc_2O_3 [17] and Yb: Lu_2O_3 [18]. In the case of Yb:

LuScO₃, the mixed crystal was capable of generating mode locked pulses as short as 74 fs, whereas Yb: Lu₂O₃ and Yb: Sc₂O₃ only emitted pulses of 220 fs and 230 fs duration, respectively. However, the κ of Yb: LuScO₃ drops to 3.5 W K⁻¹ m⁻¹.

3.2.: Performance of a laser system

In this section, figure of merits that govern the performances of a laser system will be discussed. Namely the laser slope efficiency which is applicable to CW and mode-locked systems and the pulse width which is only applicable to mode-locked systems.

The laser slope efficiency

The laser slope efficiency is a figure of merit on the performance of a laser. It tells how efficient the laser is in converting absorbed pump laser light into laser light. It is obtained from the slope efficiency curve of a laser like that shown in Fig 3.2.1 and modeled using:

$$P_{out} = \eta_s(P_{in} - P_{th}) \quad (3.2.1)$$

Fig. 3.2.1.: The slope efficiency curve of a laser.

where P_{out} is the output power by the laser, P_{in} is the input power, P_{th} is the threshold power and η_S is the laser slope efficiency and is given by the slope of the slope efficiency curve. Usually, P_{out} is measured at the end of the output coupler (OC), while P_{in} is usually measured after the input mirror and before the gain medium. However a number of factors can also affect the slope efficiency of the laser such that it can be expressed as:

$$\eta_S = \eta_C \eta_q \eta_B \quad (3.2.2)$$

where , η_C is the output coupling efficiency, η_q is the quantum efficiency of the laser and η_B is the beam overlap efficiency. η_C is a parameter that is largely controlled by the transmission (T) of the OC and typically increases with the T of the output coupler. However should the T of the OC be too large a value, this would affect the population inversion within the gain medium resulting in the gain to be reduce which in turn reduces the η_S .

The η_q is a very important parameter as it limits the maximum achievable slope efficiency by a laser. This value is material dependent and is fixed once the gain medium of choice is chosen and is written as:

$$\eta_q = \frac{\lambda_p}{\lambda_L} \quad (3.2.3)$$

where λ_p is the pump wavelength and λ_L is the lasing wavelength. A typical Nd^{3+} doped solid state gain medium absorbs pump light at 808 nm and lasers at 1064 nm. According to equation (3.2.3), this would imply a maximum η_q of 76 %. On the other hand, a Yb^{3+} solid state gain medium is pumped at 976 nm and lases at 1040 nm, implying a η_q value of 94 %. This indicates that as much as 24 % of the pump power pumping Nd^{3+} doped gain media is converted into heat while for Yb^{3+} doped gain media this value can be as low as 6 %. This implies that the heating effect is lower in Yb^{3+} doped gain media when compared to its Nd^{3+} doped counterparts and can have serious design considerations when scaling to very high output powers as excess heat can result in the deterioration of the laser performances [3].

The η_B is the only free parameter that can be varied when aligning a laser and is given by:

$$\eta_B = \frac{V_L}{V_p} \quad (3.2.4)$$

where V_L and V_p are the laser and pump volumes within the gain medium. During alignment, it is possible for lasing to occur due to a partial overlap between V_L and V_p and when this occurs the η_S will be reduced. By adjusting the positions of the mirrors within the cavity, the V_L can be adjusted such that there is maximum overlap between V_L and V_p and this would lead a maximum value of η_S . Equation (3.2.4) is also a statement that suggests that in order for the laser to achieve a maximum value of η_S , there should be an ideal mode-matching between the laser and pump modes. Hence it is important to use a set of focusing optics that reshapes the pumping beam in such a way that its mode size matches to that of the laser spot size. The selection of the optics for mode matching is done using the equations presented in section 3.3.

For a particular laser cavity design, the transmission of the output coupler could be varied such that a number of η_S curves can be obtained. The P_{th} values can then be extracted from the η_S curves and plotted against the corresponding natural log of the reflectivity of the output coupler used. This method is a variation of the famous Findlay and Clay method [3, 40] and can be used to estimate cavity losses and modeled as:

$$-\ln R = AP_{th} - \delta \quad (3.2.5)$$

where R is the reflectivity of the output coupler, A is a geometrical factor and δ are the total cavity losses. The δ values are extracted from the y-intercept of the curve plotted. In general, if the losses from the mirrors used are very small, the bulk of the losses can then be assumed to have originated from the gain medium itself. Hence, this method can be used to estimate losses from the gain medium. A low value would indicate that the gain medium used is of high quality.

Intensity autocorrelation

Another figure of merit that governs the performance of a laser system is the pulse width that it generates. It is often desirable to be able to generate shorter pulses as it would imply higher peak intensities and this would be beneficial towards micromachining and metrology applications. The question however is how does one

measure an event that in the femtosecond regime since such a feat would require the usage of an event that occurs on an even shorter scale. The fastest electronics available is capable of only measuring events that occur on the picosecond time scales and is unable to resolve events in the femtosecond regime.

Fig. 3.2.2.: Experimental setup of an intensity autocorrelator [41].

A standard way to sidestep this problem and measure the pulse widths of ultrashort pulses is to use optical techniques instead where the pulse is used to measure itself. This is typically carried out using an intensity autocorrelator such as that shown in Fig. 3.2.2. This method involves splitting up an ultrashort pulse into two pulses and allowing one of them to be delayed with respect to the other. The pulses are then allowed to overlap and interfere within a second harmonic generation (SHG) crystal which then generates a frequency double signal given by:

$$E_{sig}^{SHG}(t, \tau) \propto E(t)E(t - \tau) \quad (3.2.6)$$

where $E_{sig}^{SHG}(t, \tau)$ is the electric field envelope of the SHG signal, $E(t)$ is the electric field envelope of the undelayed ultrashort pulse and $E(t - \tau)$ is the delayed electric field envelope of the ultrashort pulse. Hence the corresponding intensity is given as:

$$I_{sig}^{SHG}(t, \tau) \propto I(t)I(t - \tau) \quad (3.2.7)$$

where $I_{sig}^{SHG}(t, \tau)$ is the intensity of the SHG signal, $I(t)$ is the intensity of the undelayed ultrashort pulse and $I(t - \tau)$ is the intensity of the delayed of the ultrashort pulse. The generated $I_{sig}^{SHG}(t, \tau)$ is then detected by a photo-detector which integrates the signal as their response time is much slower than the pulse width such that they measure an intensity autocorrelation, I_{ac} given by:

$$I_{ac} = \int_{-\infty}^{\infty} I(t)I(t - \tau)dt \quad (3.2.8)$$

Once the autocorrelation trace has been obtained, it must be fitted with a pulse shape function in order to extract the pulse width, $\Delta\tau_p$. Two intensity pulse shapes for fitting the autocorrelation traces are the Gaussian and the Sech^2 functions. The Gaussian function is given as:

$$\exp(-2.77 \left(\frac{t}{\Delta\tau_p}\right)^2) \quad (3.2.9)$$

For Gaussian shaped pulses, the autocorrelation trace is about 1.41 times the width of the pulse duration. The Sech^2 fitting function is given as:

$$\text{Sech}^2(1.763 \frac{t}{\Delta\tau_p}) \quad (3.2.10)$$

For Sech^2 shaped pulses, the autocorrelation trace is about 1.54 times the width of the pulse duration. Other techniques such as frequency resolved optical gratings (FROG) and SPectral Interferometry for Direct E-field Reconstruction (SPIDER) exist for measuring ultrashort pulse widths. These techniques have the advantage in which an intensity pulse shape would not have to be assumed for pulse width measurements which would be useful for the measurement of very short pulses which can have complicated intensity profiles that deviates away from the typical Gaussian or Sech^2 profiles.

Fig. 3.3.1: The propagation of laser around the cavity in one round trip. $S1$, $S2$ and $S3$ are the ABCD matrices of the input mirror, the stretch of air space between the mirrors and the output coupler respectively.

3.3.: Design of laser cavities

The laser cavity used requires careful design in order to achieve CW output and mode-locking. Proper cavity designs result in the laser to remain within the cavity after a large number of bounces and also give the designer knowledge of the laser spot sizes at different positions within the cavity. By having knowledge of the spot size within the laser crystal, the designer can choose proper beam reshaping optics such that optimum mode matching is achieved and that the laser output is efficient. In addition, to achieve mode-locking, the spot sizes within the gain media and on the SESAM must be small enough to saturate them but at the same time big enough to avoid optical damages. With the knowledge of the spot sizes within and on these critical elements, the designer can design a cavity that is suitable for CW generation, mode-locking or KLM.

Fig. 3.3.2.: A typical X-fold cavity used to mode-lock or initiate KLM of a solid state gain medium. $M1$, $M2$, $M3$ and $M4$ are high reflection (HR) mirrors while $M5$ is the output coupler.

The laser spot sizes within the cavity are given by:

$$\omega = \sqrt{\frac{\lambda}{\pi} \frac{2B}{\sqrt{4-(A+D)^2}}} \quad (3.3.1)$$

where ω is radius of the laser spot. A , B and D are elements of the ABCD matrix [42] multiplication of the cavity. This equation is obtained by solving the paraxial wave equation, consideration of Gaussian beam optics, ABCD matrices and the ABCD propagation laws. For the interested reader, the derivations can be found in ref. [42]. Equation (3.3.1) allows the designer to know the spot sizes at critical various positions within the laser cavity can be applied to test the stability of the cavity designed. If the ABCD matrix of the cavity results in negative values to appear under the square roots in equation (3.3.1), it indicates that the cavity design is unstable and cannot be used to generate a laser beam.

Fig. 3.3.3.: The radii of the spot sizes at the focus along $L1$ for the cavity shown in Fig.3.3.2. Calculations were conducted using ROCs of -100 mm and -300 mm for $M4$.

<i>Optical Element</i>	<i>ABCD Matrix</i>
<i>Air space of length, d.</i>	$\begin{bmatrix} 1 & d \\ 0 & 1 \end{bmatrix}$
<i>Gain medium of length l and refractive index n.</i>	$\begin{bmatrix} 1 & nl \\ 0 & 1 \end{bmatrix}$
<i>Lens of focal length, f</i>	$\begin{bmatrix} 1 & 0 \\ -\frac{1}{f} & 1 \end{bmatrix}$
<i>Mirror of radius of curvature (ROC), R</i>	$\begin{bmatrix} 1 & 0 \\ -\frac{2}{R} & 1 \end{bmatrix}$

Table 3.1.: ABCD matrices of various optical elements commonly encountered within a laser cavity.

The ABCD matrix of a cavity can be obtained by making references to Fig. 3.3.1. The cavity used consists of an input mirror, a stretch of open air and an output mirror which has individual ABCD matrices of $S1$, $S2$ and $S3$ respectively. By consideration the propagation of the laser in a single round trip such that the laser light starts to propagate from the input mirror and towards the OC and back to the input mirror, the ABCD matrix of this cavity is then given by:

$$\begin{bmatrix} A & B \\ C & D \end{bmatrix} = S1.S2.S3.S2 \quad (3.3.2)$$

Equation (3.3.2) can be expanded should the cavity now contain more optical elements. The additional ABCD matrices of the optical components should be considered in sequence as the laser traces a round trip within the cavity. The ABCD matrices of various optical components commonly encountered is listed in Table 3.1. The ABCD matrix for a lens is most commonly used for modeling the thermal lens effect under high power operations. But this effect can be difficult to consider as its value changes with different pump powers. Fig. 3.3.2 shows a typical cavity used to mode-lock solid state gain media. Application of equation (3.3.2) to find the ABCD matrix of the cavity and substitution into equation (3.3.1) results in a plot (Fig. 3.3.3) that shows beam waist values for stable cavity lengths. Fig. 3.3.4 shows that with a smaller ROC, the beam waist at the focus of L1 reduces which is the general trend with smaller ROCs. Regions not bounded by the

curves indicate that the cavity lengths are unstable. Hence such a plot allows the designer to know the radius of the beam waist at the focus of the cavity and if the cavity he/she had designed was stable. The same calculations can be repeated to obtain similar plots for the radii of the spot sizes at the output coupler and on the SESAM.

Typically in femtosecond mode-locked lasers, the radii of the spot sizes are kept small to suppress Q-switch mode-locking (QML) [43] in SSLs. In QML, bunches of mode-locked pulses are emitted instead and this effect can result in higher peak powers resulting in damages to the SESAM. This detrimental effect can be avoided if:

$$E_p^2 > E_{sat,L} E_{sat,A} \Delta R \quad (3.3.3)$$

where E_p is the intra-cavity pulse energy, $E_{sat,L}$ is the saturation energy of the gain medium, $E_{sat,A}$ is the saturation energy of the absorber and ΔR is the modulation depth of the SESAM. $E_{sat,L}$ is proportional to the laser spot size in the gain and $E_{sat,A}$ is proportional to the laser spot size on the SESAM. In solitary femtosecond lasers, the radius of the laser spots is usually no more than 50 μm while in picosecond mode-locked lasers this value can be as large as a few hundred μm . In the case of KLM cavities, the radius of the laser spot sizes in the gain medium is kept to between 10-40 μm [26, 44] to ensure that a strong Kerr effect is induced.

Gaussian Optics

Generally, a laser beam is expected to have a Gaussian intensity distribution that corresponds to a TEM_{00} mode. A TEM_{00} Gaussian beam is the lowest order or the fundamental solution to the Paraxial Wave Equation (PWE). By solving the PWE, many properties of the Gaussian beam can be understood. It is important to understand these properties as the cavity designer usually have to reshape the pump light and the output beam and an understanding of these properties would allow the selection of the best optics such that the performance of the cavity built can be optimized. To obtain the PWE, one must first derive Helmholtz's equation from Maxwell's equations in free space and are given as:

$$\nabla \cdot \mathbf{E} = 0 \quad (3.3.4)$$

$$\nabla \cdot \mathbf{B} = 0 \quad (3.3.5)$$

$$\nabla \wedge \mathbf{E} = - \frac{\partial \mathbf{B}}{\partial t} \quad (3.3.6)$$

$$\nabla \wedge \mathbf{B} = \varepsilon_0 \mu_0 \frac{\partial \mathbf{E}}{\partial t} \quad (3.3.7)$$

where $\mathbf{E}$ is the electric field vector, $\mathbf{B}$ is the magnetic field vector, ε_0 is the permittivity of free space and μ_0 is the permeability of free space. Equations (3.3.4), (3.3.5), (3.3.6) and (3.3.7) are Gauss's law, Gauss's law for magnetism, Faraday's law of electromagnetic induction and Ampere's law respectively. It is possible to derive the PWE by taking the curl on both sides of equations (3.3.6) and using the identity $\nabla \wedge (\nabla \wedge \mathbf{E}) = \nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E}$ such that it becomes:

$$\nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E} = - \frac{\partial}{\partial t} (\nabla \wedge \mathbf{B}) \quad (3.3.8)$$

Substitution of equations (3.3.4) and (3.3.5) into (3.3.8) results in:

$$\nabla^2 \mathbf{E} = \varepsilon_0 \mu_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} \quad (3.3.9)$$

Since $c = \frac{1}{\sqrt{\varepsilon_0 \mu_0}}$, equation (3.3.9) in spherical coordinates becomes:

$$\nabla^2 \mathbf{E}(\mathbf{r}, t) - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}(\mathbf{r}, t)}{\partial t^2} = 0 \quad (3.3.10)$$

By assuming a plane wave solution to equation (3.3.10) such that $\mathbf{E}(\mathbf{r}, t) = \boldsymbol{\epsilon}(\mathbf{r})e^{ikz}e^{-i\omega t}$, then the electric field can be separated into spatial and time parts and one is able to show that:

$$\nabla^2 \boldsymbol{\epsilon}_0(\mathbf{r}) + k^2 \boldsymbol{\epsilon}_0(\mathbf{r}) = 0 \quad (3.3.11)$$

where k is the wavenumber and equation (3.3.11) is Helmholtz's equation. To describe lasers using Helmholtz's equation, one requires a solution with the following criteria:

- The solution propagates along the z-axis with a plane wavefront.
- The solution is able to handle a small amount of wavefront curvature that changes as the wave propagates.

- The solution has a finite cross-section that changes as the beam propagates.

A solution to Helmholtz's equation which satisfies the criteria stated above takes the form:

$$\boldsymbol{\varepsilon}(\mathbf{r}) = \boldsymbol{\varepsilon}_0(\mathbf{r})e^{ikz} \quad (3.3.12)$$

The bulk of the z dependence will take the form of a plane wave which is handled by the e^{ikz} term in equation (3.3.12). The $\boldsymbol{\varepsilon}_0(\mathbf{r})$ plays two roles:

1. Expresses amplitude dependencies on the x, y coordinates. This term is expected to drop off smoothly as one moves away from the z -axis.
2. Handles any residual dependence on the z direction that may exist due to diffraction. It also handles the Guoy phase.

Substitution of (3.3.12) into Helmholtz's equation yields:

$$\nabla_T^2 \boldsymbol{\varepsilon}_0 + 2ik \frac{\partial \boldsymbol{\varepsilon}_0(\mathbf{r})}{\partial z} + \frac{\partial^2 \boldsymbol{\varepsilon}_0(\mathbf{r})}{\partial z^2} = 0 \quad (3.3.13)$$

where ∇_T is the transverse Laplacian which can also be written as $\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$ and k is the wavenumber. Equation (3.3.13) however can be further simplify by making the assumption that the variations of $\boldsymbol{\varepsilon}_0(\mathbf{r})$ and $\frac{\partial \boldsymbol{\varepsilon}_0(\mathbf{r})}{\partial z}$ are within the order of a wavelength in the z -direction are negligible, one can drop the $\frac{\partial^2 \boldsymbol{\varepsilon}_0(\mathbf{r})}{\partial z^2}$ from equation (3.3.13) such that one obtains:

$$\nabla_T^2 \boldsymbol{\varepsilon}_0 + 2ik \frac{\partial \boldsymbol{\varepsilon}_0(\mathbf{r})}{\partial z} = 0 \quad (3.3.14)$$

which is the paraxial wave equation. The equation assumes that one is considering on the region close to the axis of the laser. The fundamental solution to the PWE is given by the trial solution:

$$\boldsymbol{\varepsilon}(\mathbf{r}) = Af(z)e^{-ikz} e^{\frac{ik(x^2+y^2)}{2R(z)}} e^{\frac{-(x^2+y^2)}{\omega^2(z)}} e^{i\phi(z)} \quad (3.3.15)$$

where A accounts for the amplitude of the electric field, $f(z)$ is a factor that accounts for weaker fields where the beam becomes bigger (i.e. continuity), e^{-ikz} accounts for the plane wave properties of the solution, $e^{\frac{ik(x^2+y^2)}{2R(z)}}$ accounts for the change in the curvature of the wavefront as the beam changes in size, $e^{\frac{-(x^2+y^2)}{\omega^2(z)}}$ accounts for Gaussian intensity distribution and allows for beam spreading/focusing and $e^{i\phi(z)}$ is an arbitrary position dependent phase to account for the Guoy phase. R is the radius of curvature of the wavefront and ω is the radius of the beam waist.

Equation (3.3.15) can be simplified by recognizing that $e^{\frac{ik(x^2+y^2)}{2R(z)}} e^{\frac{-(x^2+y^2)}{\omega^2(z)}}$ can be grouped together by defining a q transformation parameter as:

$$\frac{1}{q(z)} = \frac{1}{R(z)} + \frac{i\lambda}{\pi\omega^2(z)} \quad (3.3.16)$$

Hence, the spot size information is now contained within the q transformation parameter. Equation (3.3.15) can then be simplified by substituting equation (3.3.16) and one obtains:

$$\mathbf{E}(\mathbf{r}) = Af(z)e^{\frac{ik(x^2+y^2)}{2q}} e^{i\phi(z)} \quad (3.3.17)$$

Fig. 3.3.3.: Schematic of the cross-section of a TEM_{00} Gaussian beam.

Solving of the PWE with equation (3.3.17) yields the fundamental Gaussian beam solutions, two of which are:

$$z_R = \frac{\pi \omega_0^2}{\lambda} \quad (3.3.18)$$

$$\omega = \omega_0 \sqrt{1 + \left(\frac{z}{z_R}\right)^2} \quad (3.3.19)$$

where ω_0 is known as the beam waist and is the radius of the beam at the focus and z_R is known as the Rayleigh range and is defined as the distance over which the laser had expanded by $\sqrt{2}\omega_0$. Equations (3.3.18) and (3.3.19) can be better understood by making references to Fig. 3.3.3.

Fig. 3.3.4.: Focusing of a TEM_{00} Gaussian beam.

Fig. 3.3.4 shows a beam of radius ω_i is focused down to ω_f after passing through a lens of focal length, f . Hence substitution of the parameters shown in Fig 3.3.4 into equations (3.3.18) and (3.3.19) results in:

$$\omega_f = \omega_i \sqrt{1 + \left(\frac{f}{\frac{\pi \omega_i^2}{\lambda}}\right)^2} \quad (3.3.20)$$

If one consider far field effects, then the second term inside the square root of equation (3.3.20) is very much larger than 1 and as such the equation can be simplified to:

$$\omega_f = \frac{f\lambda}{\pi\omega_i} \quad (3.3.21)$$

Hence, equation (3.3.21) allows one to calculate the spot size after it has been focused by a lens.

The equations shown or derived in this section thus far assumes that the laser beam is a TEM₀₀ Gaussian beam with a beam quality or M² of 1. Unfortunately, real laser beams are usually not Gaussian and have M² values larger than 1. For a properly aligned mode-locked laser, the output can be assumed to be nearly Gaussian and usually have M² values that range from 1.1 to 1.5. For high brightness fiber coupled diode lasers the M² values can have values that range from 20 – 100 depending on the diameter of the fiber core. To account for these non-Gaussian beams, their M² values would have to be considered and as such equations (3.3.18), (3.3.19) and (3.3.21) would have to be modified. Equation (3.3.19) now takes the form:

$$\omega = \omega_0 \sqrt{1 + \left(\frac{\lambda z M^2}{\pi \omega^2}\right)^2} \quad (3.3.22)$$

Equation (3.3.18) then becomes:

$$z_R = \frac{\pi \omega_0^2}{\lambda M^2} \quad (3.3.23)$$

and equation (3.3.21) becomes:

$$\omega_f = \frac{f\lambda M^2}{\pi\omega_i} \quad (3.3.24)$$

Closer inspection of equations (3.3.22) to (3.3.24) shows that non-Gaussian beams are more difficult to focus than Gaussian beams. It sets a limit as to how small such beams can be focused. Equations (3.3.22) to (3.3.24) can also be used to predict how the spot sizes would vary and are used to select the appropriate optics to optimize laser performances.

For most of the work presented in this thesis, the mode quality obtained for CW and mode-locked operations are generally TEM_{00} . Under CW operations the M2 of the beam can be greater than 2 while that of mode-locked lasers are usually less than 1.5. The output laser obtained can be astigmatic especially when pumped by single emitter diodes as this is not corrected for in the resonator designs.

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Chapter 4

Dual-wavelength passively mode-locked Nd: GdVO₄

Nd: GdVO₄ is a very well known birefringent gain medium with excellent laser properties and used in many commercial laser systems. It has emission peaks at both 1063 nm and 1065 nm but lases preferably at 1065 nm as the gain at this wavelength is much larger. The author demonstrates that it is possible to select the wavelengths which the laser operates and mode-locks. The author also demonstrates that it is possible to simultaneously mode-lock the laser at both wavelengths. Dual-wavelength lasers have many applications which shall be discussed in section 4.1.

4.1.: Dual-wavelength lasers

Applications such as laser interferometry [1] and differential absorption lidar (DIAL) [2] require two lasers with different wavelengths operating simultaneously. For such applications, it is desirable to have a single laser that generates two wavelengths simultaneously as it simplifies the setup and reduces the cost [3]. Different techniques were developed to achieve dual wavelengths emission of lasers. One approach was to use specially coated mirrors [4, 5] or via inserting wavelength filtering or selection components into the cavity [3, 6], while another was to use the special gain line property of disordered gain medium [7, 8]. Using the latter technique, Xie *et. al.* have recently demonstrated a diode pumped dual-wavelength synchronously mode locked solid-state laser [9] and Yu *et. al.* have shown the passive Q-switching of a dual wavelength laser [8]. A common characteristic of the dual-wavelength lasers so far reported is that they emit at a single polarization. For some applications this is an advantage, e.g. the beating of the beams could be exploited for generating THz beams [9]. However, for other applications such as laser interferometry, a laser with dual wavelengths and orthogonal polarizations

is required [10]. In addition, such a kind of laser is also used in precision metrology and has been demonstrated to be able to function as a nanometer ruler [11] and as a highly sensitive weight measuring machine [10]. Previously, CW dual-wavelength lasers with orthogonal polarizations have already been demonstrated in He-Ne [10] and microchip Nd: YAG [12] lasers. These lasers required the insertion of a piece of birefringent material such as quartz into the optical cavities. The purpose of this was to introduce two optical axes into the optical cavity. As the quartz was birefringent, the refractive index (RI) for both axes differs and this effectively introduces two optical paths allowing a laser with a single cavity to support dual wavelengths with orthogonal polarizations. The author shall demonstrate that this idea can be extended to SSLs with birefringent gain mediums (e.g. Nd: GdVO₄). The advantage of using a birefringent gain medium is that it does not require the insertion of addition optical components into the cavity leading to much larger output powers. In addition, the frequency separation for the wavelengths generated in SSLs can reach the THz regime which would make it an idea light source for laser interferometers as the scanning speeds can be improved significantly [10].

	<i>Nd: YAG</i>	<i>Nd: GdVO₄</i>
<i>Emission cross-section [cm⁻²]</i>	2.8×10^{-19}	7.6×10^{-19}
<i>Absorption cross-section [cm⁻²]</i>	0.77×10^{-19}	4.9×10^{-19}
<i>Fluorescent lifetime [μs]</i>	230	100
<i>Refractive index @ 1064 nm</i>	1.82	n_{σ} : 1.971 n_{π} : 2.191
<i>Thermal Conductivity [W k⁻¹ m⁻¹]</i>	10.7	11.4
<i>Emission Bandwidth [nm]</i>	0.45	0.6
<i>Polarization</i>	Unpolarized	π (E//c).

Table 4.1: Comparison of the properties of Nd: YAG and Nd: GdVO₄.

4.2.: Properties of Nd: GdVO₄

Laser action in Nd: GdVO₄ was first demonstrated by Zagumennyĩ *et. al.* [13] in 1992. The availability of high power diodes coupled with improvements in the optical quality of GdVO₄ crystals allowed this particular type of gain medium to attract attention. Due to its high absorption and emission cross-sections, these crystals are ideal for usage in miniature lasers. They can be frequency converted to generate highly efficient green

lasers. These miniature green lasers can be used in laser pointers and in laser television sets. In addition, the output of such a Nd: GdVO₄ laser being highly polarized can be used as a green laser pump for Ti: sapphire lasers.

Fig. 4.2.1: Structure of GdVO₄ crystal [14]

Nd: GdVO₄ is a type of vanadate crystal that crystallizes in a zirconium silicate (ZrSiO₄) structure. For pure GdVO₄, the Zr⁴⁺ and Si⁴⁺ are replaced by the Gd³⁺ and V⁵⁺ ions. Each V⁵⁺ is coordinated by four O²⁻ to form the vanadate structure. The Gd³⁺ ion is surrounded by eight O²⁻ ions. When doped with Nd³⁺ ions, some of the Gd³⁺ or V⁵⁺ are replaced. The structure of Nd: GdVO₄ is shown in Fig 4.2.1. Table 4.1 shows that Nd: GdVO₄ is strongly birefringent and as such has polarization dependent gain profiles [15]. Naturally birefringent crystals also have the advantage of eliminating depolarization losses [16] under high power laser operations. Zhang *et. al.* [15] carried out polarization dependent studies and found that the strongest emission line of Nd: GdVO₄ occurred at 1063.1 nm in π polarization ($E//c$). In the σ polarization ($E \perp c$), there are two emission lines, located at 1063.1 nm and 1065.5 nm, respectively. Because the 1063.1 nm emission line in the π polarization is about five times larger than that of the 1063.1 nm and 1065.5 nm lines in the σ polarization, gain competition suppresses lasing in the σ polarization. Recently Wu *et. al.* have shown experimentally that using a dual cavity configuration and polarization control, dual-wavelength (at 1063.1 nm and 1065.5 nm) dual-polarization

CW emission could be achieved in a diode pumped Nd: GdVO₄ laser [7]. The author shall demonstrate that a Nd: GdVO₄ laser that could not only mode lock at either 1063 nm or 1065 nm but can also do so at both wavelengths. In the case of dual-wavelength mode-locking, the two different wavelength laser beams have orthogonal polarizations. The transition from one mode locking state to the other is simply controlled by varying the pump power absorbed by the crystal.

4.3.: Experimental setup

The experiment was done using a three mirror V-shaped cavity as shown in Fig. 4.3.1. The input mirror M1 is a flat mirror and has high transmission (HT) coatings at 808 nm and high reflection (HR) coatings at 1064 nm. M2 serves as a folding mirror and an output coupler (OC) with a radius of curvature (ROC) of -500 mm. It has a transmission of 2 % at 1064 nm. M3 serves as the SESAM and an end mirror with HR at 1064 nm.

Fig 4.3.1: Experimental setup of the dual-wavelength mode-locked laser. M1: Flat input mirror, M3: Plano-concave mirror with ROC = -500 mm, M3: SESAM

The laser crystal used was a piece of a-cut Nd: GdVO₄ with 0.5% doping concentration and has dimensions of 6 (L) $\times$ 3 (B) $\times$ 3 (W) mm³ grown using the Czochralski method and was placed at the focus of the pump which was very close to M1 to optimize mode matching. The Nd: GdVO₄ crystal was grown using the Czochralski by collaborators from The Institute of Crystal Materials, Shandong University, China. The crystal absorbed about 80 % of the incident pump light. The crystal has high transmission (HT) coatings on both its facets at 808 nm and 1064 nm. During the course of the experiment,

the crystal was given a small tilt ($< 5^\circ$) with respect to optical axis of the system. This suppressed etalon effects and improved mode-locking stability. The crystal was wrapped with a piece of indium foil and housed in a water cooled copper housing where the temperature was maintained at 18°C . The crystal was pumped by a $400\ \mu\text{m}$ fiber core coupled diode laser (Jenoptik, JOLD-50-CPXF-2P) capable of delivering up to 50 W of pump power and is focused using a set of 1:1 focusing optics. The focusing optics focuses the pump light down to a diameter of $400\ \mu\text{m}$ within the gain medium. The M^2 of the fiber coupled pump was approximated using:

$$M^2 = \frac{\pi a}{\lambda} NA \quad (4.3.1)$$

where a is the radius of the fiber core, λ is the wavelength of the pump and NA is the numerical aperture of the fiber used which was 0.22. The M^2 of the pump used was approximated to be about 170. Hence the depth of focus of the pump which is defined by two Z_R is about 2 mm. The pump laser had a central wavelength at 808 nm which can be tuned by adjusting the operating temperature which was maintained at 30°C with cooling water for these experiments. The absorbed pump power was measured as it is typically done in the literature [8-10] in which a power meter was placed before the crystal and then after the crystal. The difference in the power values measured was taken as the absorbed pump power.

The mode-locking was initiated by a of commercial semiconductor saturable absorbing mirror (SESAM) purchased from BATOP with a central operating wavelength at 1064 nm, a ΔR of 1.2%, a relaxation time of 500 fs, and a saturation fluence of $60\ \mu\text{J cm}^{-2}$. ABCD matrix analysis of the cavity using equation (3.3.1) showed that the radius of the laser spot size on the SESAM was about $180\ \mu\text{m}$ when the cavity length between M1 and M2 was 55 cm. Light was coupled into two fast detectors simultaneously in which linear polarizers with orthogonal axes (π and σ directions) were placed in front of each detector (New Focus, 1 GHz 1611) to monitor the mode-locking of each wavelength simultaneously. As the autocorrelator (Femtochrome, FR103-XL) was only sensitive to the σ polarization, a half-wave plate was placed in front of an autocorrelator when the emitted laser was mode-locked at 1063 nm to measure the pulse width. The purpose of the half waveplate is to flip the polarization of the input beam by 90° such that that the

autocorrelator is able to detect the signal. The half-wave plate was replaced with a linear polarizer with its axis at 45° to the π and σ directions when simultaneous dual-wavelength mode locking was observed. The purpose of the linear polarizer was such that the components parallel to the σ directions of both wavelengths could contribute to the autocorrelation signal. This was done to ensure such that both wavelengths can contribute to the autocorrelation measurements.

Fig. 4.4.1: Autocorrelation traces obtained when the laser was mode-locked at (a) 1063 nm and (c) 1065 nm. The pulses were 10 ps and 5.9 ps respectively. The solid circles are experimental points while the solid lines are a Gaussian fits. (b) and (d) are the corresponding mode-locked optical spectrums at 1063 nm and 1065 nm respectively. Both spectrums have a FWHM of 0.47 nm.

4.4.: Mode-locking performances

The laser was aligned until mode-locking was observed. It was noticed that the mode-locking of the system was sensitive to the alignment of the OC and in particular the position and tilt of the SESAM. The author believes this was mainly due to the SESAM being damaged from pervious usage and as such the system only mode-locks when laser mode on the SESAM was focused on an undamaged area as the losses would be the smallest.

Fig. 4.4.1 shows the autocorrelation traces and the corresponding optical spectra when the laser was mode-locked at the wavelength of 1063 nm or 1065 nm, respectively. When the laser was mode-locked at 1063 nm (π polarized) the pulse was 10 ps and its corresponding optical spectrum was 0.47 nm. When the laser was mode-locked at 1065 nm (σ polarized), the pulse width was 5.9 ps and the corresponding optical spectrum was also 0.47 nm. In both cases, a Gaussian intensity profile given by equation (3.2.9) was used to carry out the fitting. For the laser mode-locked at 1063 nm, the TBP was about 1.53 which was about 6.4 times above the Fourier transform limit. For the laser mode-locked at 1065 nm, the TBP was 0.74 which was about 1.68 times above the Fourier transform limit. The TBPs would indicate that the pulses could be further compressed by using a pair of extra-cavity prisms.

Fig. 4.4.2: Varying output power with increasing absorbed pump power.

Fig. 4.4.2 shows the variation of the output power of the laser as the optical power absorbed by the Nd: GdVO₄ crystal was increased. The mode-locking at 1065 nm is in itself an interesting phenomenon since the gain at this wavelength is much smaller than the one at 1063 nm and is normally not expected to lase. To initiate laser action, losses must be introduced to the 1065 nm component. Wu *et. al.* [7] achieved this via the

introduction of a polarization beam splitter into the laser cavity such that both wavelengths with different polarizations are directed to two different optical paths and as such emitted CW laser at two different OCs simultaneously. When the pumping power was low, the system initially mode-locked at 1065 nm. Above the mode-locking threshold, the laser could be switched to mode-lock at 1063 nm or simultaneously at both 1063 nm and 1065 nm by increasing the absorbed pump power. Mode-locking action was continuously monitored through observation of the pulse trains on an oscilloscope and the optical spectrum analyzer. Mode locking at 1063 nm resulted in a picosecond mode-locked slope efficiency, $\eta_{\text{picosecond}}$ of 13%. When mode-locked at 1065 nm, $\eta_{\text{picosecond}}$ was 11%. The optical to optical efficiency, η_{opt} was about 5 % when mode-locked at only one wavelength was mode-locked. The region bounded by the broken lines in Fig. 4.4.2 indicates the range of absorbed pump powers in which simultaneous mode-locking at 1063 nm and 1065 nm was observed. When the absorbed pumped power was increased, the 1063 nm mode-locking component intensified at the expense of the 1065 nm mode-locking component. The changes in the $\eta_{\text{picosecond}}$ when mode-locked at different wavelength are an indication of mode size variations.

Fig. 4.4.3: Mode-locked pulse trains at 1063 nm and 1065 nm obtained simultaneously with two detectors.

Fig. 4.4.3 shows the pulse trains obtained simultaneously via two fast photodetectors. Though both pulse trains were similar, they are not identical. Slight variations exist between the two signals and can be approximated using the oscilloscope. This difference between the two pulse trains in time maybe used to measure the group refractive index of Nd: GdVO₄ as demonstrated by Liang *et. al.* [17] whom used a similar method to determine the group refractive indices of Nd: YVO₄.

Fig. 4.4.4: Simultaneously mode-locked autocorrelation trace and its corresponding spectrum (a) autocorrelation trace. Solid circles are experimental points while the solid line is a Gaussian fit (b) corresponding optical spectrum.

Fig. 4.4.4 shows the autocorrelation trace and its corresponding optical spectrum when both 1063 nm and 1065 nm wavelengths were simultaneously mode-locked. The pulse width was 7 ps while the FWHM of the optical spectrums at both bands were about 0.58 nm. No multiple spikes were observed on the autocorrelation trace [9], indicating that the mode-locking was simultaneous but not synchronous. If the mode-locking is synchronous, the two different wavelengths would result in a beating effect resulting in multiple interference spikes in the autocorrelation trace such as that demonstrated by Cong *et. al.* [19]. This type of interference can be modeled by a simple two beam interference equation given by:

$$I = I_1 + I_2 + 2\sqrt{I_1 I_2} \cos(2\pi\nu t) \quad (4.4.1)$$

where I is the intensity of the interfered beam, I_1 is the intensity of the first beam undergoing interference, I_2 is the intensity of the second beam undergoing interference and ν is the frequency separation of the two beams undergoing interference. Application of equation (4.4.1) results in Fig. 4.4.5 which clearly shows additional interference peaks

within the main autocorrelation trace. The separation in time between these interference peaks will correspond to the separation in frequency of the two interfering wavelengths. Since such an observation is missing as evident from the autocorrelation trace in Fig. 4.4.4, it can be concluded that the mode-locking observed in Nd: GdVO₄ is simultaneous but not synchronous. This conclusion is also not difficult to imagine because of the refractive index differences between the π and σ directions and as such the 1063 nm and 1065 nm mode-locking travel at different velocities within the cavity and as such unable to overlap spatially resulting in simultaneous but non-synchronous mode-locking. In the case of Xie *et. al* [9] synchronous and simultaneous mode-locking was observed because a proper amount of dispersion added into the cavity such that both beams can travel at the same velocity in the cavity and as such overlap spatially on the SESAM. In the case of Cong *et. al.* [18] where no dispersion was added into the cavity, synchronous and simultaneous mode-locking was attributed to the effect of the SESAM in which its effect was to trap both the wavelengths together resulting in stable pulse evolution in the cavity.

Fig. 4.4.5.: Simulated autocorrelation trace of a synchronous mode-locked pulsed formed by two interfering beams.

Fig. 4.5.1.: Calculated mode sizes for the 1063 nm (broken line) and 1065 nm (solid line) laser modes for the cavity used in the experiment. (a) the case for low absorbed pump powers where only 1065 nm was lasing (b) the case for moderate absorbed pump powers where both wavelengths were lasing and (c) the case for high absorbed pump powers where only 1063 nm was lasing.

4.5.: ABCD Matrix Analysis

Typically the thermal lens has a detrimental effect in high power lasers due to induced diffraction losses and beam quality degradation however in this laser, thermal lensing is exploited to allow the demonstration of dual wavelength mode-locking. This phenomenon can be explained by considering the overlap efficiency between the pump beams and the lasing modes [16]. The lasing mode diameter, Ψ , was calculated using the ABCD matrices. Analysis using the ABCD matrices showed that the pump induced RI changes yielded very little influence over the size of the lasing modes and can be neglected. It was found that thermal lensing effect however had a significant effect on the mode sizes. The thermal lens is usually a detrimental effect in high power solid lasers as it can result in beam degradation. The thermal lensing effect of Nd: GdVO₄ can be approximated using the lens matrix as shown in Table 3.1 at the position of the crystal when carrying out the ABCD matrix analysis of the cavity. The ABCD matrix analysis calculations for the 1063 nm and 1065 nm modes sizes within the laser crystal are shown in Fig 4.5.1. The focal lengths of the thermal lens used in these calculations were 0.23 m,

0.21 m and 0.19 m for the cases of low (7.4 W), moderate (8 W) and high (8.8 W) absorbed pump powers respectively. For low, moderate and high absorbed pump powers, the laser mode-locks at 1065 nm, 1063 nm and simultaneously at both wavelengths.

Mode-locking Wavelength	1063 nm Laser Mode		1065 nm Laser Mode	
	Ψ [μm]	$(\Psi/\emptyset)^2$	Ψ [μm]	$(\Psi/\emptyset)^2$
1063 nm (Low pump powers)	1155	8.3	1026	6.6
1063 nm and 1065 nm (Moderate pump powers)	786	3.9	765	3.6
1065 nm (High pump powers)	306	2.3	609	2.3

Table 4.2.: Summary of the results for the case where $L=0.48$ m. $\emptyset$ is the size of the pump beam within the crystal.

Table 4.2 summarizes the calculated results for the cavity length, $L=0.48$ m, used in the experiment. A $(\Psi/\emptyset)^2$ value closer to 1 would indicate better mode matching. $\emptyset$ is the pump mode size and takes the value of 400 μm which is the same as the diameter of the fiber coupled pump since the system is imaged using a 1 : 1 optical system. At low absorbed pump powers where the thermal lens effect is smaller, $(\Psi/\emptyset)^2$ for the 1063 nm mode was much larger than the 1065 nm mode and very much greater than 1. This implied that the mode matching for the 1063 nm mode is poor. This results in gain to be distributed to the 1065 nm mode despite it having an emission cross-section that is five times smaller than that of the 1063 nm mode and as such, the laser mode locks at 1065 nm. At moderate absorbed pump powers where the thermal lens effect is stronger, the $(\Psi/\emptyset)^2$ for the 1065 nm mode was still smaller than the 1063 nm mode, which is an indication that laser still prefers to mode-lock at 1065 nm. However compared to the case of low absorbed pump powers, the $(\Psi/\emptyset)^2$ values are closer to each other. Since the emission cross section of the 1063 nm mode is 5 times greater than that of the 1065 nm mode, this compensates for the larger mode mismatch and as such gain is distributed between the two modes and the laser is able to mode lock at 1063 nm and 1065 nm. Note that since the gain medium is birefringent, it allows two lasing wavelengths with orthogonal polarizations to be supported by the cavity due to different optical lengths along the two optical axes. Hence by exploiting the birefringence of the gain medium, dual wavelength generating can be realized in a single laser cavity. At high absorbed pump powers where the thermal lens effect is the strongest, one finds the $(\Psi/\emptyset)^2$ values

for both modes to be the same. Since there is no advantage for the laser to oscillate at 1065 nm and at the same time the emission cross section at 1063 nm is five times higher than that of 1065 nm mode, the 1065 nm oscillations are suppressed and the laser mode locks at 1063 nm. When the SESAM in Fig. 4.3.1 was replaced with a HR mirror at 1064 nm, CW light at 1065 nm was initially emitted. Upon increasing the absorbed pump power, CW emission at 1063 nm and 1065 nm were observed simultaneously. Further increases in pump powers results in only CW emission at 1063 nm. This observation is similar to that reported in ref. [7] but the authors in that work introduced polarization control in the form of a polarizing beam splitter into the cavity such that laser was generated using two different cavities simultaneously. They later modified their setup by introducing a half waveplate into the cavity that acted as a polarization selector that allows the laser to selectively emit at 1063 nm or 1065 nm but not simultaneous laser generation at both wavelengths. By exploiting the birefringence of the gain medium itself, dual wavelength laser emission can be realized in a much more compact setup. In the work of Ren *et. al.* [12] dual wavelength was achieved in isotropic Nd: YAG was also achieved through polarization control by artificially introducing birefringence into the laser cavity via two half wave plates. By doing so however results in a drastic reduction in the laser's output power.

The η_{opt} and $\eta_{\text{picosecond}}$ obtained in this work were much lower compared to that obtained by Chen *et. al.* [19]. The reason being for this was because the mode-locking at 1065 nm and that when both 1063 nm and 1065 nm were simultaneously mode locked makes use of the thermal lensing effect which results in the mode-matching within the cavity to become poor as seen from the $(\Psi/\emptyset)^2$ in Table 1. Even when mode-locked at 1063 nm, the mode-matching is far from idea as seen from the corresponding $(\Psi/\emptyset)^2$ values from Table 1. Thus poor mode-matching results in the lower efficiencies.

4.6.: Concluding remarks

In conclusion, the author has demonstrated a dual-wavelength mode locking in a diode pumped Nd: GdVO₄ laser. By exploiting the birefringence and thermal lensing properties of Nd: GdVO₄, the author have demonstrated the laser could be mode locked either at the 1063 nm (π polarized) or 1065 nm (σ polarized), or simultaneously at the both

wavelengths with orthogonally polarizations by simply changing the pump power. A simple explanation was provided in terms of mode-matching between the pump and lasing modes of 1063 nm and 1065 nm. This was also the first demonstration of mode-locking of Nd: GdVO₄ at 1063 nm and simultaneously at 1063 nm and 1065 nm. The author believes that such a phenomenon could also be observed in other lasers with a birefringent gain medium.

4.7.: References

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Chapter 5

Laser performances of Yb: Sc₂SiO₅ and Yb: CaYAlO₄

Yb³⁺ doped solid state gain media are excellent materials for ultrashort pulse generation. However their individual host medium dictates a lot of their properties and as such laser performance differs between types of Yb³⁺ doped gain media differ greatly. In this chapter, the laser performances of two promising novel Yb³⁺ doped gain media in ultrafast lasers are presented.

5.1.: Properties of Yb: Sc₂SiO₅ (Yb: SSO)

High power femtosecond lasers have many scientific and industrial applications such as nonlinear wavelength conversion, ultrafast spectroscopy, optical coherent tomography and micromachining [1]. The applications have continuously driven the search for more compact femtosecond lasers with higher efficiencies and lower costs. Yb³⁺-doped crystals are highly suitable for use as gain media for femtosecond lasers. Yb³⁺-doped gain media have high quantum efficiencies (> 90 %), large emission bandwidths and the absence of deleterious effects such as excited state absorption, up-conversion and concentration quenching, which allow for the construction of efficient femtosecond lasers. Moreover, the absorption bands of the Yb³⁺-doped gain media match the emission wavelengths of the commercially available high brightness InGaAs laser diodes. Direct diode pumping is possible, which enables compact designs and higher efficiencies of the femtosecond lasers. The main drawback of Yb³⁺-doped gain media is that they are a quasi-three level system. In most crystals the Stark splitting of the ²F_{7/2} ground manifold of Yb³⁺ is a few hundreds of cm⁻¹. Recently, it was shown that this drawback could be partly circumvented in the Yb³⁺ doped oxyorthosilicate crystals, where due to the large crystal

field strength, large $\text{Yb}^{3+} {}^2\text{F}_{7/2}$ ground manifold splitting could be obtained [2]. In addition, it was also found that not only broadband emissions could be obtained in Yb^{3+} -doped oxyorthosilicate crystals, but also the crystals could have large thermal conductivity [3-4], an essential property required for high power lasers. Of the Yb^{3+} -doped oxyorthosilicate gain media, Yb: YSO [5], Yb: GSO [6] and Yb: LSO [5] femtosecond lasers that generated stable pulses of 122 fs, 343 fs and 233 fs, respectively, with output powers of 0.41 W, 0.4 W and 2.6 W have already been demonstrated. Hence, Yb^{3+} -doped oxyorthosilicate gain media have proven themselves as viable alternatives as femtosecond source generators. Yb: SSO being also a type of Yb^{3+} -doped oxyorthosilicate crystal is also expected to be an excellent ultrafast gain medium. Its properties and laser performances will be described.

Absorption and emission spectra

Some of the properties related to laser performances of a laser gain medium can be understood by studying the corresponding absorption and emission spectra. The absorption spectrum of Yb: SSO was measured using a UV-VIS-NIR spectrometer (JASCO, V-570) with a Xe lamp as the pump source. To calculate the absorption cross-section, σ_{abs} , of Yb: SSO from the UV-VIS-NIR measurements require the application of Beer-Lambert's Law which states:

$$\frac{I}{I_0} = \exp(-\alpha L) \quad (5.1.1)$$

where I is the intensity of the beam after passing through the sample, I_0 is the intensity of the incident beam, α is the absorbance and L is the length of the sample. α is then given by:

$$\alpha = -\frac{2.303}{L} \log_{10}\left(\frac{I}{I_0}\right) \quad (5.1.2)$$

Finally the absorbance is related to the σ_{abs} by:

$$\sigma_{\text{abs}} = \frac{\alpha}{N} \quad (5.1.3)$$

where N is the number of Yb^{3+} per unit volume.

The emission spectrum of Yb: SSO was obtained by exciting the sample with a 940 nm laser diode and detecting the subsequent fluorescence intensities with a spectrofluorimeter (Jobin-Yvon TRIAX 550) at room temperature. The emission cross-section, σ_{em} , values of Yb: SSO can then be obtained using the Fchtbauer-Ladenburg formula which states:

$$\sigma_{em}(\lambda) = \frac{1}{8\pi n^2} \frac{1}{c} \frac{\lambda^5 I(\lambda)}{\tau_r \int \lambda I(\lambda) d\lambda} \quad (5.1.4)$$

where n is the refractive index of the gain medium, c is the speed of light and $I(\lambda)$ is the fluorescence intensity at a wavelength of λ . τ_r is the radiative lifetime and for Yb: SSO was determined to be about 1.64 ms [3]. Fig. 5.5.1 shows the σ_{abs} and σ_{em} spectra of Yb: SSO.

Fig. 5.1.1: Absorption and emission cross-sections of Yb: SSO. The broken curve corresponds to σ_{abs} while the solid curve corresponds to σ_{em} .

Strong emission peaks are observed at 1006 nm, 1036 nm, 1062 nm and 1087 nm with values of $2.6 \times 10^{-21} \text{ cm}^2$, $4.4 \times 10^{-21} \text{ cm}^2$, $3.8 \times 10^{-21} \text{ cm}^2$ and $1.0 \times 10^{-21} \text{ cm}^2$ respectively. When the emission peaks are Gaussian decomposed, the emission bandwidths were approximately about 33 nm, 20 nm, 15 nm and 23 nm at 1006 nm, 1036

nm 1062 nm and 1087 nm respectively. Although the broadest emission bandwidth occurs at the 1006 nm peak, laser action and mode-locking at this wavelength would be difficult due to the strong reabsorption occurring near this peak. Reabsorption of the laser wavelength drops off sharply for the 1036 nm, 1062 nm and 1087 nm peaks. Negligible reabsorption exists for the 1062 nm and 1087 nm peaks and as such it would be better to operate the laser at these wavelengths. In fact, operating the laser at the 1087 nm peak would prove to be advantageous for ultrashort pulse generation as the emission bandwidth near that peak is relatively board and that the curvature of the emission peak is gentler [7] compared to the other peaks. In addition, negligible reabsorption of the laser wavelength occurs in the vicinity of this 1087 nm emission peak. It shall be demonstrated in the subsequent sections that mode-locking can be demonstrated for wavelengths near the 1036 nm, 1062 nm and 1087 nm peaks. In fact the best mode-locking performance was observed near the 1087 nm peak.

Fig. 5.1.2: The gain cross-section spectrum. β increases from 0 to 1 in steps of 0.25.

From the absorption and emission cross-section spectra, the gain cross-section spectra can be determined using:

$$g = \beta \sigma_{em}(\lambda) - (1 - \beta) \sigma_{abs}(\lambda) \quad (5.1.5)$$

where g is the gain cross-section and β is the population inversion ratio. This equation assumes that all the Yb^{3+} ions are either at the ground state or at the upper lasing state to simplify calculation. When β approaches 1, the gain cross-section spectrum reduces to that of the emission cross-section spectrum. Likewise when β approaches 0, the gain cross-section spectrum reduces to that of the absorption cross-section spectrum. The gain cross-section spectrum plotted using (5.1.5) is shown in Fig. 5.1.2 (negative values implies absorption). When β approaches 0.5, laser action is possible between 950 nm to 1150 nm which is useful for laser tuning and ultrashort pulse generation.

Fig. 5.1.3: Structure of Yb: SSO [8].

Structure and physical properties of Yb: SSO

SSO crystallizes in the monoclinic system where Yb^{3+} enters the host matrix by displacing a small proportion of the Sc^{3+} . This results in the Yb^{3+} lining in two different distorted octahedral sites (Sc (I) and Sc (II)) surrounded by SiO_4 such as that shown in Fig 5.1.3. Sc^{3+} based hosts possess desirable laser properties because of the similarities in the ion radii of Sc^{3+} (0.75 Å) [9] and Yb^{3+} (0.868 Å) [9] and as such high crystal field strength leads to large $^2\text{F}_{7/2}$ or ground state splittings. In fact the $^2\text{F}_{7/2}$ splitting of Yb: SSO reaches value of 1027 cm^{-1} [2], which is amongst the largest in Yb^{3+} doped crystals. This is comparable to Yb: GSO which has a value of 1061 cm^{-1} [6] and larger than that of Yb: YSO and Yb: LSO which have reported values 700 cm^{-1} [5] and 960 cm^{-1} [5]

respectively. To that end Yb: SSO is expected to have lower thresholds values as its operation would become more four level like when compared to other Yb³⁺ doped gain media. In addition, the small ionic radius of Sc³⁺ provides minimal disruption to the phonon vibrations and as such this class of host is expected to have higher thermal conductivities. In fact, Yb: SSO was found to have a κ value of 7.5 W m⁻¹K⁻¹ [3] which is amongst the highest in Yb³⁺ doped solid state gain media. Other desirable properties of Yb: SSO include having large σ_{em} with broadband emission bandwidths. The large ground state splitting and relative long τ_r value imply a low lasing threshold. Hence the properties of Yb: SSO make it a very promising ultrafast high power gain medium. Table 5.1 summarizes and compares the properties of Yb: SSO.

	<i>Yb: SSO</i>	<i>Yb: YSO</i>	<i>Yb: LSO</i>	<i>Yb: GSO</i>	<i>Yb: YAG</i>
$^2F_{7/2}$ splitting [cm ⁻¹]	1027	700	960	1061	785
κ [W m ⁻¹ K ⁻¹]	7.5	3.6	5.3	3.0	10.7
τ_r [ms]	1.64	0.67	0.95	1.1	1.2
σ_{em} [$\times 10^{-20}$ cm ²]	0.44	0.4	0.6	0.46	2.2
Emission bandwidth [nm]	20	16	21	32	9
$\Delta\tau$ [fs]	93 [This work]	122	260	384	340
P_{out} [mW]	20 [This work]	410	2600	400	110

Table 5.1.: Properties and laser performances of Yb³⁺ doped oxyorthosilicate and YAG crystals.

Fig 5.2.1: A 5 at. % Yb: SSO boule.

5.2.: Crystal growth of Yb: SSO

The crystal used for experiments in this thesis was grown by collaborators from the Shanghai Institute of Ceramics using the Czochralski method in an inductively heated iridium crucible under nitrogen ambient environment. A piece of Yb: LSO was used as the seed crystal because it is isostucture with Yb: SSO. The seed crystal was grown along the b-direction as the optical properties of the as grown crystal are optimum in this direction. The SSO matrix was fabricated using high purity ($> 99.995\%$ purity) SiO_2 , Sc_2O_3 and Yb_2O_3 powder. The pulling speed was between $1\text{-}3\text{ mm h}^{-1}$ while the rotation speed of the seed crystal was about 15-30 revolutions per minute. A crystal boule of dimensions of $\phi 25 \times 40\text{ mm}^3$ was grown and allowed to cooled to room temperature in The crystal boule was then allowed to slowly cool to room temperature in 40 hours. Figure 5.2.1 shows a picture of the as-grown boule. A $10 \times 10 \times 10\text{ mm}^3$ crystal was cut from the boule and optically polished before being tested for optical quality with a 20 mW He-Ne laser. No scattering sites were observed within the crystal indicating that the crystal grown was of high optical quality which is devoided of defects and bubbles. The crystal boule was grown with a Yb^{3+} doping concentration of 5 %.

Fig 5.3.1: Schematic of the experimental setup used to generate cw laser with Yb: SSO as the gain medium. F1 and F2 are a pair of plano-convex lens with 75 mm focal length. M1, M2 and M3 have ROCs of -100 mm, -300 mm and -100 mm respectively. M4 and M5 have ROCs of ∞ . L1, L2, L3, and L4 are 23.2 cm, 50 cm, 88 cm and 5.5 cm respectively.

5.3.: CW experiments with Yb: SSO

A schematic of the experimental setup used to generate CW laser with Yb: SSO as the gain medium is shown in Fig 5.3.1. The crystal used for all Yb: SSO related laser experiments in this thesis was cut from the Yb: SSO boule grown. The Yb: SSO obtained is b-cut and has dimensions of 2.2 (L) $\times$ 3 (B) $\times$ 3 (H) mm³ and has a Yb³⁺ doping concentration of 5 at. %. The crystal was optically polished with HT coatings coated on both its facets at the pump and laser wavelengths to minimize the Fresnel reflection losses. The Yb: SSO crystal was wrapped in indium foil and placed in a water-cooled copper housing maintained at 18 °C throughout the experiment. The crystal absorbed about 90% of the pump light incident to it. The crystal was placed about 5.5 cm away from M1. As segregation coefficient of Yb³⁺ in SSO is 0.96 [5] (relatively high), and concentration quenching is absent in Yb³⁺-doped gain media, the doping level of the crystal can be further increased, resulting in larger absorption. The optical spectrum was obtained using a high resolution optical spectrum analyzer (Ando, AQ-6315B).

Yb: SSO was pumped using high brightness board stripe diodes as it offers superior performances compared to fiber coupled diode lasers. These diodes however have astigmatic output in which the divergence of the beam and beam quality in both axes differs. But nevertheless, they still offer better beam quality than their fiber coupled diode counterparts and can be focused to very small pump sizes. Due to their high brightness, these types of diodes are better suited for mode-locked pumping lasers.

Fig 5.3.2. High brightness pumping using a board stripe diode with a fast axis collimator micro-lens. F1 and F2 are plano-concave lens with focal lengths of 75 mm.

A high brightness board stripe diode (Axcel Photonics, CM-975-6000-250) with central wavelength at (975 ± 3) nm was used as the pump source. It is capable of delivering up to 6 W of pump power. The diode has an emitter size of $200 \mu\text{m}$ (slow axis) $\times$ $1 \mu\text{m}$ (fast axis) with M^2 values of more than 30 and 1 in the slow and fast axis directions respectively. Due to its' highly of astigmatic nature, a fast axis collimation micro-lens was placed right in front of the emitter in the diode. This limits the divergence of the pump light in the fast axis such that it can be collimated by a plano-convex lens without being clipped by placing the diode at the focal length of F1 as shown in Fig. 5.3.2. The pump light was then focused into the gain medium via a second plano-convex lens. The diameter of the pump light was focused down to a size of about $100 \mu\text{m}$ (fast axis) $\times$ $250 \mu\text{m}$ (slow axis) within the crystal. ABCD analysis of the cavity using equation (3.3.1) showed that the laser spot has a diameter of $100 \mu\text{m}$ in the crystal and is shown in Fig. 5.3.3. The mode matching is not ideal but acceptable. By making L4 shorter it would be possible to make the radius of the laser spot size tighter but the cavity becomes much harder to align as the range of stable L1 lengths reduces to a few millimeters.

Fig. 5.3.3.: Radius of the laser spot size within the crystal against stable lengths of L1.

CW performances of Yb: SSO

The CW laser performance of Yb: SSO was investigated using the setup shown in 5.3.1 using a variety of mirrors with various output coupling ranging from 0.8 % to 5 % and the slope efficiencies curves are shown in Fig. 5.3.4. The CW slope efficiencies, η_{CW} , were 26 %, 46 %, 52 %, 60 % and 70 % when the output couplers were 0.8 %, 1.6 %, 2.4 %, 3.5 % and 5 % respectively. The corresponding maximum output powers were 1 W, 1.9 W, 2.1 W, 2.5 W and 2.7 W. Higher output powers should be possible if more pump power was available as the slope efficiency curves in Fig 5.3.4 did not saturate. At the maximum output power, the output was a good TEM₀₀ beam. Previously, the maximum η_{CW} reported for Yb: SSO was 45 % [3]. The enhanced performance observed in this work was most likely due to better mode matching in the cavity and a better quality pump source used. The maximum η_{CW} obtained in this work was also comparable to that of Yb: YSO (67%) [10], Yb: LSO (62%) [10] and Yb: GSO (75%) [11]. The lasing threshold powers of Yb: SSO were relatively low and were less than 0.6 W for all the couplers used. This can be attributed to the large ground state splitting due to the high crystal field in Yb: SSO. The low threshold pump powers observed in Yb: SSO are also in agreement with that reported for other Yb³⁺ doped oxyorthosilicate crystals. Yb: LSO [10] and Yb: YSO [10] have a lasing threshold of about 1 W while that of Yb: GSO is about 1.6 W [11]. Hence the CW lasing results obtained suggest that Yb: SSO is an efficient CW gain medium.

An interesting point to be noted is that even though the σ_{em} at about 1040 nm was the largest, CW emission around this wavelength was not observed unless a prism was added into the cavity. The reason for this was that there was reabsorption by the Yb³⁺ ions around this wavelength. These additional losses suppressed CW laser oscillation near the 1036 nm emission peak. The reabsorption of Yb: SSO at around 1036 nm is relatively high as observed in Fig 5.1.1 and diminishes towards longer wavelengths. On the other hand, a strong emission peak occurs at around 1062 nm as shown in Fig 5.1.1 and as such the CW laser emissions observed in Fig. 5.3.4 occurred near 1063 nm. Fig. 5.3.5 shows the CW emission spectrum of the Yb: SSO laser when the output coupler was 5 %. The spectrum was measured under the condition of maximum output power. Compared to Yb:

YAG, the reabsorption cross-section is negligible [12] around 1030-1040 nm and as such these lasers tend to operate around this region.

Fig 5.3.4.: CW laser performances of Yb:SSO using output couplers of various transmissions.

Fig 5.3.5.: CW laser spectrum of Yb:SSO with a 5% output coupler.

Consideration of the lasing thresholds obtained using different output couplers given by Fig. 5.3.4 and the modified Clay and Findlay method [13] given by equation (3.2.5), the total cavity losses, δ , was found to be about 6 %. This is an indication that some losses still exist within the cavity. The CW performance can be further improved by reducing the cavity losses either through further improving the crystal quality or by improving cavity designs. Nevertheless, the demonstrated CW performance of Yb: SSO still shows that it is a highly efficient gain medium.

Fig 5.3.6.: Determination of round trip cavity losses with the laser thresholds under output couplers of different transmissions.

The tuning properties of Yb: SSO was also investigated by inserting a SF 10 prism by inserting a prism near M5. It was found that the laser was continuously tunable from 1010 nm to 1040 nm and from 1060 to 1065 nm as shown in Fig. 5.3.7. The laser was not tunable between 1040 nm to 1065 nm. This observation was similar to that observed in Yb: GSO [14] in which the tuning curve was also not continuously tunable using a silica prism. Li *et. al.* [14] then demonstrated that this problem can be sidestep by using a SF 14 dispersion prism and a continuously tunable curve could be obtained from 1010 to 1100 nm. The author believes that Yb: SSO can also be continuously tunable by using a dispersion prism with higher dispersion should one be available.

Fig 5.3.7.: Tuning curve of Yb:SSO performed with a SF10 prism.

Fig 5.4.1: Schematic of the experimental setup used to generate femtosecond laser with Yb:SSO. F1 and F2 are a pair of plano-convex lens with 75 mm focal length. M1, M2 and M3 have ROCs of -100 mm, -300 mm and -100 mm respectively. M4 and M5 have ROCs of ∞ . L1, L2, L3, and L4 are 23.2 cm, 50 cm, 22 cm, 5.5 cm and 26.5 cm respectively. L_{prism} was 41.5 cm.

Fig 5.4.2: Dispersion data for the Yb: SSO laser. Blue curve – GDD estimations for a piece of 2.2 mm Yb: SSO. Gold curve – Total cavity GDD. Purple curve – Negative GDD due to prism pair with a tip to tip separation of 41.2 cm.

5.4.: Mode-locking of Yb: SSO

Fig 5.4.1 shows the schematic of the cavity used to initiate mode-locking of Yb: SSO in the negative dispersion regime. The cavity used is essentially the same cavity used to conduct the CW experiments except that a pair of intra-cavity prisms and a SESAM has been used to replace M4. Similarly, the crystal was wrapped with indium foil and placed in a water cooled housing maintained at 18 °C throughout the experiment. The crystal was tilted to about 6° with respect to the principal axis of the cavity to suppress etalon effects. This also improves the stability of the mode-locking operation. A broad strip diode (Axcel Photonics, CM-975-6000-250) with central wavelength at (975 ± 3) nm was used as the pump source. The pump source was placed in close contact with a water cooled heat sink maintained at 14 °C throughout the experiment. The diode was maintained at this temperature mainly to protect the diode from burning up at too high operating temperatures. The positive group velocity dispersion (GVD) introduced by the Yb: SSO crystal was compensated by the prism pair with a tip to tip, L_{prism} , separation of 41.5 cm. The prism pair was aligned to Brewster's angle to minimize the cavity losses.

The total amount of negative GVD provided by the prism pair was estimated to be about -280 fs^2 to -300 fs^2 as shown in Fig. 5.4.2. Since there is no available experimental data on the total amount of positive dispersion due to Yb: SSO, the crystal dispersion was estimated based on the data of 5 at. % Yb: GSO [14] and was found to be about 180 fs^2 to 210 fs^2 for a crystal of similar length as that used in these experiments. In general Yb³⁺ doped oxyorthosilicate crystals have very similar dispersion properties [14]. The mirrors used to initiate mode-locking were low GVD mirrors purchased from Layertec GmbH. These mirrors have close to zero dispersion from 1040 nm to 1060 nm. In addition, the SESAMs used to initiate mode-locking at 1040 nm and 1064 nm have close to zero dispersion at these respective wavelengths. To that end, the crystal was assumed to be the main dispersive element in the cavity.

Fig. 5.4.3.: Radius of the laser spot size on the SESAM against stable lengths of LI.

Since the cavity used for CW operation of Yb: SSO is essentially the same as that used for passive mode-locking, the diameter of the laser mode size 5.5 cm away from M1 can be extrapolated and is about $100 \text{ } \mu\text{m}$ as shown in Fig. 5.3.3. On the other hand, ABCD matrices analysis shows that the diameter of the spot size on the SESAM is about $70 \text{ } \mu\text{m}$ as shown in Fig 5.4.3. SESAMs from BATOP GmbH were used to initiate the

mode-locking. SESAMs with central wavelengths at 1040 nm ($\Delta R = 0.5\%$, $\Delta R_{ns} = 0.5\%$, relaxation time, $\tau = 500$ fs and saturation fluence, $\Phi_{sat} = 90 \mu\text{J cm}^{-2}$), and 1064 nm ($\Delta R = 1.2\%$, $\Delta R_{ns} = 0.8\%$, $\tau = 500$ fs and $\Phi_{sat} = 90 \mu\text{J cm}^{-2}$) were used. While the SESAM with center wavelength at 1040 nm initiated mode-locking at 1041 nm, the SESAM with center wavelength at 1064 nm triggered mode-locking at 1060 nm and 1077 nm. Light leaking out of M3 was coupled into a low noise photodetector (New Focus 1611-FC-AC) and connected to a 1 GHz digital oscilloscope (Tektronix DP0714) to detect the mode locked signals. Unfortunately the pulse to pulse stability cannot be measured as oscilloscope lacked the required statistical package to do such an analysis. The optical spectrum was obtained using a high resolution optical spectrum analyzer purchased from Ando (AQ-6315B), while the pulse width was measured using a commercial autocorrelator (APE, PulseCheck). The autocorrelation traces measured in this work were all obtained with the absorbed pump powers maintained at their respective maximum output powers.

Fig. 5.4.4.: Mode-locked pulsetrain traces at (a) short and (b) long time scales with Yb: SSO mode-locked at 1077 nm.

Solitary mode-locking of Yb: SSO

Solitary mode-locking was conducted in the negative dispersion regime. To initiate mode-locking, M4 was replaced by a SESAM and a pair of SF10 dispersion prisms were

inserted into the cavity. L_{prism} was fixed at 41.5 cm throughout the experiment. M5 was changed to an OC with 0.4 % transmission for wavelengths of (1040 ± 80) nm. This was done to ensure that the intra-cavity power was high so that the SESAM can be easily saturated and that the shortest pulses could be obtained. Mode-locking was observed at three different wavelength bands of 1041 nm, 1060 nm and 1077 nm. The pulse trains for all the three wavelength bands had a repetition rate of about 90 MHz. This corresponded to the cavity repetition rate, indicating that the fundamental mode-locking had occurred at the three different wavelengths. Fig 5.4.4 shows the pulsetrains obtained when the laser was mode-locked at 1077 nm.

Fig 5.4.5. The autocorrelation traces when Yb: SSO was mode-locked at (a) 1041 nm, (b) 1060 nm and (c) 1077 nm with (d), (e) and (f) showing their corresponding spectra respectively. The blue solid line in the autocorrelation traces are Sech² fits.

By using a SESAM whose central wavelength was designed to be around 1040 nm, the author was able to demonstrate the mode-locking of Yb: SSO at 1041 nm. When the SESAM was replaced with one with a central wavelength to be 1064 nm, the laser mode-locks at 1060 nm and 1077 nm. When the laser is mode-locked at 1041 nm, 1060 nm and 1077 nm, the maximum output powers to be about 40 mW, 50 mW and 100 mW respectively. The corresponding autocorrelation traces and spectra are shown in Fig. 5.4.5. Fig. 5.4.5 (a) shows the autocorrelation trace when the laser was mode-locked at 1041 nm in which a pulse width of 145 fs was measured, assuming a Sech^2 shaped fitting using equation (3.2.10). Its corresponding optical spectrum has a FWHM of 8 nm (Fig. 5.4.5 (d)). The resulting TBP was 0.32, which indicates that the pulses are nearly transform-limited. As the SESAM used has the lowest losses at around 1040 nm, mode-locking at 1041 nm was possible despite the presence of reabsorption at the wavelength. Replacement of the SESAM with the one with central wavelength at 1064 nm results in the laser to mode-lock at 1060 nm. Fig 5.4.5 (b) shows the resulting autocorrelation trace and the pulse width was found to be 144 fs, assuming a Sech^2 shaped pulse. Its corresponding optical spectrum had a FWHM of 8.8 nm (Fig 5.4.5 (e)). The resulting TBP was 0.34, which indicates that the pulses are nearly transform-limited. By adjusting the position of the OC and the position of the SESAM, the laser was able to mode-lock at 1077 nm with a maximum output power of about 100 mW. Fig 5.4.5 (c) shows the autocorrelation trace obtained at this wavelength. If a Sech^2 shaped pulse is assumed, the pulses have a pulse width of 125 fs. The optical spectrum has a FWHM of 10.4 nm (Fig 5.4.5 (f)). Therefore, the TBP was 0.34, indicating that the pulses at this wavelength are also near transform-limited. The fact the pulses are close to being transform limited indicates that the SPM is also balanced with negative dispersion in the cavity and some of the observations maybe understood by making references to Haus' master equation of mode-locking (Equation (2.2.8)). This is because Haus' master equation of mode-locking contains all the pulse shaping and perturbation effects that are present in a negative dispersion mode-locked cavity. In the presence of negative dispersion, the SPM modulation within the system is almost balanced and as such the chirp in the system is close to zero such as that shown in Fig. 2.2.2 (b) resulting in pulses that are close to the transform limit. Since the pulse width obtained are yet to be limited by the fluorescence

bandwidth of the emission peaks in Yb: SSO, shorter pulses should be obtainable in the presence of stronger SPM effects. However such a situation would require a larger amount of negative dispersion for balancing the SPM.

Since solitary pulses are shaped by Haus' master equation of mode-locking or the NLSE, the output powers obtained at each wavelength can be understood by making references to equation (2.2.9) which can be rearranged such that it yields:

$$E_p = 1.7627 \frac{2|D|}{\delta_L \tau_p} \quad (5.4.1)$$

(5.4.1) is also a statement of energy scaling and that the pulse energy of solitary mode-locked systems follows a linear relationship with the net dispersion of the laser system. As shown in Fig. 5.4.2, the total net negative dispersion in the system increases as the wavelength increases. At around 1041 nm, 1060 nm and 1077 nm the laser has a net negative dispersion of about -60 fs^2 , -80 fs^2 and -100 fs^2 respectively and by (5.4.1), the pulse energy increases towards longer wavelength as observed in the results presented above and provides further evidence of solitary mode-locking in Yb: SSO. It should be noted that the output powers and pulse energy are also partly influenced by the σ_{em} and σ_{abs} values around each wavelength but such effects would have smaller bearing than the role played by dispersion in soliton mode-locked systems. Equation (5.4.1) also shows that as the dispersion of the system decreases, the pulse-width should reduce which would imply that the shortest pulsewidths should be observed near the 1041 nm emission peak. This would be the case if the emission bandwidths and curvatures near each emission peak are the same. For Yb: SSO however, the broadest emission peak occurs around 1087 nm (relative to the 1036 nm and 1062 nm emission peaks) and as observed in Fig. 5.1.1, the profile near the top of this emission peak is also much gentler than the others peaks as such more bandwidth would be available to participate in mode-locking. Hence, a shorter pulse was observed as the Yb: SSO laser mode-locks towards the 1087 nm emission peak. From the point of view of obtaining high power ultrashort pulses, it would be advantageous to mode-lock Yb: SSO towards the 1087 nm peak. Indeed, the shortest and highest power pulses were obtained at 1077 nm as the laser mode-locks towards the 1087 nm emission peak.

Fig 5.4.6.: The autocorrelation traces when Yb: SSO was mode-locked at 1043 nm with a total GVD of about (a) -40 fs^2 and (b) -20 fs^2 respectively with (c) and (d) showing their corresponding spectra respectively. The blue solid line in the autocorrelation traces are Sech^2 fits. The sidebands observed in (c) and (d) are a result of third order dispersion [TOD] and fourth order dispersion [FOD] effects [19].

The spectrum of the Yb: SSO laser (Fig. 5.4.5 (d)) when mode-locked at 1041 nm has a small satellite peak occurring to the right of its principal spectrum. Such satellite peaks have previously been observed Yb: $\text{KLu}(\text{WO}_4)_2$ [15], Yb: CaGdAlO_4 [16], Cr: YAG [17] and Ti: sapphire [18]. Herrmann *et. al.* [19] carried simulations and showed that such a phenomena occurs when Ti: sapphire is KLM and operated in the vicinity of zero GVD and as such higher order dispersions such as TOD becomes more influential. He showed that TOD causes one satellite peak to appear and the location of which side of the principal spectrum it appears is determined by the sign of the TOD. The appearance of the satellite peak in Fig. 5.4.5 (d) provides evidence that the GVD in Yb: SSO when mode-locked at 1041 nm is very well-compensated such that it was operating close to zero GVD which supports the accuracy of the GVD calculations shown in Fig. 5.4.2. Herrmann *et. al.* [19] also carried out simulations to show that on operating the laser even closer to zero GVD, fourth order dispersion (FOD) effects can become significant and the resulting laser spectrum is characterized by two satellite peaks on either side of the principal spectrum. To study the effects that FOD could have on the laser spectrum of Yb: SSO, the amount of negative GVD dispersion was reduced to force the system to

operate even closer to zero GVD. This was done by inserting more prism material into the cavity when the laser was mode-locked towards the 1040 nm emission peak. The prism in question was mounted on a high precision three dimensional stage and was carefully translated such that about 0.1 mm of prism material was further inserted into the cavity. By doing so, the system was estimated to be operating at about -40 fs^2 of negative GVD at 1043 nm. This resulted in 115 fs pulses with an average power of 30 mW to be generated as shown in Fig. 5.4.6 (a) while its corresponding laser spectrum is shown in Fig. 5.4.6 (c). The corresponding TBP is about 0.32. Clearly two satellite peaks on both side of the principal spectrum are clearly observed in line with the simulations done by Herrmann *et. al.* [19] and consistent with the observations of Stingl *et. al.* [20] in KLM Ti: Sapphire lasers. This implies that further reduction in GVD results in FOD to modify the laser spectrum. A further translation of the prism of 0.1 mm resulted in the laser to be operating at a GVD of about -20 fs^2 . This resulted in 93 fs pulses with an average output power of 20 mW to be generated as shown in Fig. 5.4.6(b) while its corresponding laser spectrum is shown in Fig. 5.4.6 (d). Similarly, two satellite peaks were observed about the principal spectrum indicating that FOD is the dominant dispersion effect in the cavity. The TBP is 0.316. The low output powers in both cases are in agreement with equation (5.4.1) in which the output power reduces when operating closer to zero GVD. Further reduction in the GVD of the system resulted in the mode-locking to be unstable. These results show that higher order dispersion effects can manifest themselves even when in pulses of about 100 fs and as such can place a limitation on the pulsewidth obtainable in Yb^{3+} doped SSLs. To the author's knowledge, the results presented are the first observations of FOD effects in a Yb^{3+} doped SSLs.

The laser outputs for all the results presented in this section were TEM_{00} beams. Higher output powers should be possible by improving the mode matching of the system and by replacing the dispersion prisms with Gires-Tournois interferometer (GTI) mirrors instead. Higher output powers were possible by switching to output couplers with larger transmissions, however this may result in longer pulse widths.

Fig. 5.4.7.: Schematic of the experimental setup. F1: Aspherical lens, $f = 13.86$ mm. F2: Plano-concave cylindrical lens, $f = -25.4$ mm. F3: Plano-convex cylindrical lens, $f = 100$ mm. F4: Plano-convex lens, $f = 75$ mm. M1, M2 and M3: Plano-concave mirrors, ROC = -100 mm, -300 mm and -100 mm, respectively. M4 and M5: Flat Mirrors. L1 = 23.2 cm, L2 = 50 cm, L3 = 5.5 cm and L4 = 88 cm. The SESAM and the output coupler take the position of M4 and M5 respectively.

Dissipative solitons in Yb: SSO

The mode-locking characteristics of Yb: SSO was also studied in the positive dispersion regime. This was done by removing the dispersion prisms. In addition, a new set of focusing optics were used to ensure better mode-matching and a new pump source was also installed to deliver higher pump powers. A schematic of the setup is shown in Fig. 5.4.7. The laser diode used is a board stripe diode with central emission at (975 ± 4) nm (Lumics GmBH). The emitter size is $94 \mu\text{m}$ wide and the diode is capable of emitting up to 12 W of pump power. Like the diode used in the solitary mode-locking of Yb: SSO, the beam output is astigmatic in the fast and slow axes directions. However as the stripe on the diode is smaller, the M^2 values are about 20 and 1 in the slow and fast axes direction respectively. The astigmatic output of the diode is corrected using the focusing optics shown in Fig. 5.4.7. This type of pumping scheme is a variation of the high brightness pumping used in the solitary mode-locking of Yb: SSO and is much better since it allows more control over the final spot size and also allows for a smaller pump size. The pump light was first beam shaped by an aspherical lens together with a telescope made up of a pair of cylindrical plano-concave and a cylindrical plano-convex lens pair and focused into the crystal via a best form plano-convex spherical lens. The purpose of the aspherical lens was to collimate the output from the beam and provide a reduction to spherical aberrations. The telescope serves the purpose of expanding the

beam by about four times and collimating the beam in both the fast and slow axis directions. The purpose of the beam expansion is such that a tighter focus can be obtained after being focused by the best form lens. In addition, the best form lens is further reduce spherical aberrations and of the pump beam. The best form lens provides the best performance possible by a singlet lens and as such focus the pump beam to a size as small as possible.

The pump diode was placed in close contact with a water cooled heat sink and maintained at 14 °C to protect the diode from burning out at maximum pump power. The pump beam size in the crystal was measured with the knife-edge method. It has diameters of about $(60 \pm 10) \mu\text{m}$ (fast axis, horizontal direction) $\times$ $(120 \pm 10) \mu\text{m}$ (slow axis, vertical direction). The slow axis of the diode was along the vertical direction. ABCD matrix calculations of the cavity showed that the diameters of the laser spot sizes are 100 μm and 70 μm in the crystal and on the SESAM respectively. M4 was replaced by a commercial SESAM (Batop GmbH) with central wavelength at 1064 nm ($\Delta R = 1.2\%$, $\Delta R_{\text{ns}} = 0.8\%$, $\tau = 500$ fs and $\Phi_{\text{sat}} = 90 \mu\text{J cm}^{-2}$) to initiate mode-locking. M5 was replaced by an output coupler with a transmission of 0.4 % at the lasing wavelength. The rest of the experimental parameters, optics and cavity mirrors used remained the same as in the case of solitary mode-locking of Yb: SSO. The positive dispersion of the mode-lock system comes from the crystal dispersion of Yb: SSO which from Fig. 5.4.2 is estimated to be about 180 fs² at around 1060 nm.

Fig. 5.4.8.: The slope efficiency curve of Yb: SSO mode-locked in the positive dispersion regime. The broken lines indicate the different modes of operations of the laser.

Fig. 5.4.9.: Pulsetrain of fundamentally mode-locked Yb:SSO in the positive regime.

Fig. 5.4.10.: (a) Autocorrelation trace and (b) laser emission spectrum of Yb:SSO mode-locked in the positive dispersion regime.

Fig. 5.4.8 shows the slope efficiency curve of Yb:SSO mode-locked in the positive dispersion regime. The laser initially emitted CW laser which upon increment of pump power, generated QML pulses. At a pump power of about 7.5 W, fundamental mode-locking occurred. The different modes of operation of the laser were continuously monitored by observing their pulsetrains through light leaking out of M3. When mode-locked, the slope efficiency was about 4 %. The efficiency is largely due to the low transmission of the output coupler. The purpose of using a low transmission output coupler was to ensure a high intra-cavity power and as such introduce a high SPM. Fig. 5.4.9 shows the corresponding pulse train when the laser fundamentally mode-locked. The pulse to pulse separation was about 11 ns which corresponded to the cavity repetition rate which showed that when mode-locked, a single pulse existed within the laser cavity. At the maximum output power, the laser generated 2.7 ps pulses if a Sech² shape pulse was assumed at a central wavelength of about 1063 nm as shown in Fig 5.4.10. It is

interesting to note however that the laser spectrum deviates from the typical Gaussian or Sech^2 profile as expected for lasers mode-locked in the positive dispersion regime and has more of a rectangular shaped with steep sides. In addition, without dispersion compensation added to the cavity, the laser bandwidth has a FWHM of about 7.5 nm which allows the generation of pulses of 160 fs. In addition, the pulse is highly chirped and has a TBP of 5.4 and is about 17 times about the transform limit. The observation of strongly chirped pulses in the positive dispersion regime is slightly similar to that of Li *et. al.* [21] who also reported the mode-locking of Yb: SSO in the positive dispersion regime. He generated 2.3 ps pulses but with a spectral bandwidth of only 3 nm. The pulses he obtained were less strongly chirped and the emission spectrum was still more or less Sech^2 shaped. This is largely due to a smaller SPM effect within their cavity. Highly chirped pulses with a laser spectrum with steep sides are actually hallmarks of dissipative solitons (DSs). The observation of DSs however previously observed in fiber lasers [22-25] and KLM Ti: sapphire lasers [26-27]. There have been no previous reports of dissipative solitons in Yb³⁺ SSL. The observation of DSs pulses can also be understood by making references to Haus' Master equation of mode-locking as described by equation (2.2.8). Haus' master equation of mode-locking contains all the pulse shaping and perturbation effects (SAM, dissipative effects and gain filtering) that helped to shape the pulses in both the positive dispersion regimes. In the Yb: SSO laser, strong SPM is induced via the usage of an output coupler with a small transmission and when enhanced by positive dispersion results in strongly chirped pulses as predicated by Haus' master equation of mode-locking and such a situation is depicted in Fig. 2.2.2 (b). Compared to work reported by Li *et. al.* [21], the chirp in their system is much weaker and this is most likely due to weaker SPM effects in their cavity. Further discussions on DSs will be delayed till latter sections as this phenomenon is also observed in Yb: CaYAlO₄ (Yb: CYA) where such the properties of DSs are stronger.

5.5.: Properties and crystal growth of Yb: CaYAlO₄ (Yb: CYA)

The doping of Yb³⁺ ions into disorder hosts results in extremely broadband emission spectra. Fig. 3.1.2 shows that amongst the Yb³⁺ doped crystal gain media, disordered crystal hosts exhibit the broadest emission bandwidths. The disordered structure however

disrupts the phonon vibrations and as such results in low κ values which would limit output powers of such disordered gain media. As such Yb^{3+} doped disordered gain media [16, 28-30] are known to have broadband emission bandwidths but with low κ values. The development of Yb^{3+} doped tetragonal disordered gain media however showed otherwise as it was possible to grown disordered crystal hosts with gentle and broadband emission bandwidths whilst having high κ values. Yb: CaGdAlO_4 (Yb: CALGO) [16, 28] have been demonstrated to have emission bandwidths as broad as 77 nm but at the same time having a κ value of $6.9 \text{ W m}^{-1} \text{ K}^{-1}$. At close to zero dispersion [16], Yb: CALGO have been demonstrated to be able to generate 47 fs pulses with an output power of 40 mW. At larger negative GVD, Yb: CALGO generated 68 fs pulses with an output power of 520 mW thus showing that this class of crystal can be used for ultrafast high power applications. Very recently Yb: CaYAlO_4 (Yb: CYA), a gain medium with properties similar to that of Yb: CALGO has been developed [29-30]. It is expected that ultrashort pulses with high peak power could be generated with this crystal. The laser performances and potential as an ultrafast high power and high efficiency gain medium of this crystal are investigated for this thesis.

Structure and disorder of Yb: CYA

CYA crystallizes with the K_2NiF_4 structure as shown in Fig. 5.5.1 in which the Al^{3+} and O^{2-} ions in the CYA matrix would take the positions of Ni and F cations respectively. The Ca^{2+} and Y^{3+} cations would take the positions of the K cations. Finally, Yb^{3+} can be substituted into any of the positions held by Ca^{2+} and Y^{3+} . As Ca^{2+} and Y^{3+} have similar ionic sizes, they can be randomly distributed around Yb^{3+} cations [31]. The charge differences between the Ca^{2+} and Y^{3+} cations result in large crystal fields which in turn inhomogeneously broaden the emission spectrum of Yb: CYA .

Fig. 5.5.1.: Structure of K_2NiF_4 .

Fig. 5.5.2.: Pictures of as grown Yb: CYA boules: (a) before annealing (b) after annealing.

Crystal growth of Yb: CYA

The crystals used for investigating the laser performances of Yb: CYA were grown by collaborators from Shanghai Institute of Optics and Fine Mechanics. The crystals were grown using the Czochralski method in an inductively heated iridium crucible under nitrogen and oxygen (4 %) ambient environment. The starting compounds used in the fabrication were high purity (99.999 %) Yb_2O_3 , CaCO_3 , Y_2O_3 and Al_2O_3 and were weighted according to the formula $\text{CaYb}_x\text{Y}_{1-x}\text{AlO}_4$. The compounds were then crushed, mixed and pressed into tablets. A rectangular $\langle 1\ 1\ 0 \rangle$ oriented crystal was used as the seed. The pulling rate was 1-2 mm/h while the rotation rate was maintained at 5-15 rpm. The crystal boule was then allowed to slowly cool in 60 hours to room temperature in order to obtain high quality boules. Fig 5.5.2(a) shows the picture of a boule obtained right after cooling. The boule grown was of high quality and devoid of cracks but brown in color. This color defect can be eliminated by annealing the boule in a hydrogen environment such that it becomes colorless and highly transparent as shown in Fig. 5.5.2(b) and under green laser illumination, no scattering sites could be observed which indicates the boule grown was of high optical quality. Boules of various Yb^{3+} doping concentrations were grown and cut into required dimensions for subsequent experiments. The segregation coefficient for Yb^{3+} ions in CYA is 0.74 [32].

Fig 5.6.1.: Fluorescence lifetimes of a 5 at. % Yb: CYA measured by powder concentration dilution method at room temperature. Blue closed circles – 0.5 % weight concentration, purple closed squares – 1% weight concentration, gold closed diamonds – 5% weight concentration and green closed triangles – 40 % weight concentration.

5.6.: Spectroscopic properties of Yb: CYA

Lifetime Measurements

The fluorescence decay lifetime, τ_f , of Yb: CYA was measured using a spectrometer (FSP920, Edinburgh instruments) with a 10 Hz OPO laser (Vibrant II, OPOTEK) tuned to 910 nm as the pump source. The pulse width for the pump source was 5 ns. As reabsorption processes occur in Yb³⁺ doped gain media, the measured values of τ_f are much longer than the τ_r values especially in crystals such as Yb: CYA which has strong overlap between the absorption and emission spectra. Hence a measurement of τ_f does not necessary reflect the true value of τ_r . To sidestep this problem, Yb: CYA with low doping concentrations are typically used to reduce the reabsorption effects. To investigate the levels of doping concentrations that could yield reabsorption effects large enough to affect the accuracy of spectroscopic measurements, the power concentration dilution method adopting refractive-index matched fluid [30] was employed. Sample preparation was carried out by using a H₂ annealed 5 at. % Yb: CYA crystal rather a 1 % sample such that one is able to determine the varying trend of the results from a higher doping concentration downwards. The doped sample was then grounded into fine powder together with a piece of pure CYA with weight concentrations of 40 %, 5 %, 1 % and 0.5 % where the weight concentration is defined to be the weight percentage of the doped powder. The grounded samples were then immersed into monochlorobenzene fluid which has a refractive index of 1.52 and were then subjected to τ_f measurements.

Fig. 5.6.1 shows the measured τ_f values for samples of different weight concentrations. The blue closed circles, purple closed squares, gold closed diamonds and green closed triangles represent samples with weight concentrations of 0.5 %, 1 %, 5 % and 40 % respectively. All the curves follow a nearly a nearly exponential decay behavior and the fitted life times for the 0.5 %, 1 %, 5 % and 40 % weight concentration samples are 0.428 ms, 0.426 ms, 0.429 ms and 0.484 ms respectively. Hence it can be observed that as the weight concentration reduces from 40 % to 5 %, the τ_f values decreases significantly. When the weighted concentration was reduced from 5 % to 0.5 %, the change in values of τ_f was modest. Hence it can be concluded that the 1 % sample shows negligible reabsorption effects and the corresponding value of 0.426 ms can be taken as τ_f

value of Yb^{3+} ions in CYA. For this reason, a 1 at. % Yb: CYA sample was used for subsequent spectroscopic measurements.

Absorption and emission spectra of Yb: CYA

All spectroscopic properties of Yb: CYA were measured using low doping concentrations (1% at. Yb: CYA) to limit reabsorption during spectral measurements. A point to note is that since Yb: CYA belongs to the tetragonal system the emission spectra are polarized. Hence, it is important to distinguish the polarization directions as π and σ . The π and σ directions defined according to how the exciting electric field is oriented to the crystallographic c axis of the crystal. For the π and σ directions, the polarizations of exciting electric fields are parallel and perpendicular to the crystallographic c axis of the crystal respectively.

The polarized absorption spectra were carried out using a Lambda 900 spectrophotometer (Perkin- Elmer Inc.) and excited using a Ti: sapphire laser tuned to emit at 910 nm. The subsequent polarized absorption cross-section spectra determined using (5.1.1) to (5.1.3) and shown in Fig. 5.6.2. For both polarizations, the absorption peaks at 979 nm with values of about $5.7 \times 10^{-20} \text{ cm}^2$ and $2 \times 10^{-20} \text{ cm}^2$ for π and σ polarized directions respectively. The absorption FWHM bandwidths for π and σ polarizations are 12 nm and 13 nm respectively. Compared to Yb: YAG [33], whose σ_{abs} at the pump wavelength is about $0.6 \times 10^{-20} \text{ cm}^2$ – considerably smaller than that of Yb: CYA. Hence Yb: CYA has excellent absorption properties and is highly suitable for diode pumping.

Fig. 5.6.2.: Room temperature polarized absorption cross-section spectra of a 1 at. % Yb: CYA crystal. Solid line: π polarization. Dashed line: σ polarization.

Fig. 5.6.3.: Room temperature polarized emission cross-section spectra of a 1 at. % Yb: CYA crystal. Solid line: π polarization. Dashed line: σ polarization.

The polarized fluorescence spectra were measured by exciting Yb: CYA with a Ti: sapphire laser tuned to emit at 910 nm. The subsequent polarized outputs were allowed to pass through a monochromator (1000 M, SPEX) and measured at an InGaAs detector. The polarized emission cross section area can then be calculated from the fluorescence intensities measured through application of the Fuchtbauer-Ladenburg formula as shown in equation (5.1.4). The subsequent polarized emission cross section spectra are plotted in Fig. 5.6.3. Fig. 5.6.3 shows that for the π polarized direction, the peak emission occurred at about 981 nm with a peak emission cross section value of about $2.1 \times 10^{-20} \text{ cm}^2$ which is comparable to that of Yb: YAG at 1031 nm which takes the value of about $2.1 \times 10^{-20} \text{ cm}^2$ [33]. However lasing at this wavelength will be difficult due to the large reabsorption as shown in Fig. 5.6.2. A plateau centered around 1000 nm and measuring 20 nm in bandwidths exists for the π polarized direction, which would be good for generation of ultrashort pulses but large reabsorption of the laser wavelength exists making it difficult to lase at this wavelength band. Towards the longer wavelengths, a strong emission peak centered at about 1050 nm with a bandwidth of about 30 nm exists. Reabsorption about this peak is negligible and the large emission bandwidth would make it good for ultrashort pulse generation. A distinct feature of the σ polarized direction emission cross section spectrum is that it contains a broadband plateau that extends from 990 nm to 1060 nm. As this region of the emission spectrum is relatively flat it would be possible to make use of a part of the bandwidth to generate very short pulses. Yb: CALGO also has such a plateau existing in its emission spectrum as such was able to generate pulses as short as 47 fs [16]. The author believes that if mode-locking were to be conducted for the σ polarized direction of Yb: CYA, very short pulses could potentially be obtained. A sharp and high emission peak exists at 981 nm for the σ polarized emission spectrum. Similar to the situation for the σ polarized emission spectrum, large reabsorption exist around this wavelength and as such it would be difficult to generate any laser around 981 nm.

Fig. 5.7.1.: Thermal conductivity values of a 1 at. % Yb: CYA crystal. Solid line: π polarization. Dashed line: σ polarization.

5.7.: Thermal conductivity measurements of Yb: CYA

The κ measurements for a 1 at. % Yb: CYA crystal measured along the a and c axes are shown in Fig. 5.7.1. The κ values along the a and c axes were determined by using:

$$\kappa = \rho c_p \alpha \quad (5.7.1)$$

where ρ is the density, c_p is the specific heat and α is the thermal diffusion coefficient along the a or c axis. c_p was measured using a commercial differential scanning calorimeter (NETZSCH DSC 204 F1) on a 80 mg sample while ρ was measured using the Archimedeian buoyancy method [30]. α values were measured using a commercial thermal diffusion measurement machine (NETZSCH Nanoflash LFA447) on 1 at. % Yb: CYA a- cut and c-cut crystals.

The κ values of 1 at. % Yb: CYA are shown in Fig. 5.7.1 where at room temperature, they take values of $3.6 \text{ W m}^{-1} \text{ K}^{-1}$ and $3.2 \text{ W m}^{-1} \text{ K}^{-1}$ along the a and c axes respectively. Compared to pure CYA, the κ values for the a and c axes are $3.7 \text{ W m}^{-1} \text{ K}^{-1}$ and $3.3 \text{ W m}^{-1} \text{ K}^{-1}$ respectively. Although these values are lower than that reported for Yb: CaGdAlO₄

[16, 28] which were reported to have κ values of $6.9 \text{ W m}^{-1} \text{ K}^{-1}$ and $6.3 \text{ W m}^{-1} \text{ K}^{-1}$ for the a and c axes respectively, these values are still much higher than those of other disordered crystals as observed from Fig. 3.1.2 and are adequate for high power and high brightness pumping without experiencing fracture. In addition, the author shall also demonstrate that the κ values Yb: CYA are adequate to withstand sub- 200 fs pulses in excess of 700 mW. Table 5.2 summarizes the key properties of Yb: CYA and compare against those of Yb: CALGO, Yb: SSO, and Yb: YAG .

	<i>Yb: CYA</i>	<i>Yb: CALGO</i>	<i>Yb: SSO</i>	<i>Yb: YAG</i>
$\kappa [\text{W m}^{-1} \text{ K}^{-1}]$	3.7 (a), 3.2 (c)	6.9 (a), 6.3 (c)	7.5	10.7
$\tau [\text{ms}]$	0.426	0.420	1.64	1.2
$\sigma_{em} [\times 10^{-20} \text{ cm}^2]$	0.8(σ), 0.6(π)	0.75(σ), 0.4(π)	0.44	2.2
<i>Emission bandwidth [nm]</i>	$\sim 80(\sigma)$, $\sim 30(\pi)$	$\sim 80(\sigma)$	20	9
$\Delta\tau_p [\text{fs}]$	156 fs [This work]	47, 68	93 [This work]	340
$P_{out} [\text{mW}]$	740 [This work]	38, 520	20 [This work]	110

Table 5.2.: Comparison of key properties and results of Yb: CYA against that of Yb: CALGO, Yb: SSO and Yb: YAG.

5.8.: CW operations of Yb: CYA

Fig. 5.8.1 shows a V-shaped cavity designed for a CW laser using Yb: CYA. The diode pump and focusing optics used were the same as those used in the positive dispersion mode-locking of Yb: SSO (see Section 5.4 and Fig. 5.4.7). The pump diode was temperature tuned to emit at a wavelength of $(979 \pm 4) \text{ nm}$ by keeping it in close contact with a water cooled heat sink. The crystal was wrapped with indium foil and housed in a water cooled housing that was maintained at 14°C throughout the experiment. It is capable of emitting a maximum pump power of 12 W. The pump beam size in the crystal was measured with the knife-edge method. It has diameters of about $(60 \pm 10) \mu\text{m}$ (fast axis, horizontal direction) $\times$ $(120 \pm 10) \mu\text{m}$ (slow axis, vertical direction). The slow axis of the diode was along the vertical direction. ABCD analysis of the cavity in Fig. 5.8.1 showed that the focus of the cavity occurs at the surface of M1 and that the laser diameter was about $70 \mu\text{m}$. The Yb: CYA used has an Yb^{3+} doping concentration of 3 at. % and it was cut along the axis with dimensions of 2 (L) $\times$ 3 (B) $\times$ 3 (H) mm^3 . The crystal absorbed about 50 % and 20 % of the pump light when orientated in the π and σ directions respectively. The crystal was HT coated at both the pump and

laser wavelengths. The optical spectra were monitored and measured with a high resolution optical spectrum analyzer (Ando, AQ-6315B), while the output powers were measured using a Coherent PowerMax (PM 30) detector connected to a Coherent FieldMax II powermeter. The output couplers (OCs) were purchased from LayerTec GmBH.

Fig. 5.8.1.: V-shaped mirror cavity schematic used to test the CW performance of Yb: CYA. LD: Laser diode, F1: Aspherical lens, $f = -13.86$ mm, F2: Plano-concave cylindrical lens, $f = -25.4$ mm, F3: Plano-convex cylindrical lens, $f = 100$ mm, F4: Best form plano-convex lens, 75 mm, M1: Input mirror, $ROC = \infty$, M2: HR mirror, $ROC = -100$ mm, M3: Output mirror, $ROC = \infty$.

Fig. 5.8.2. shows the slope efficiency curves obtained using the V-shaped cavity in Fig. 5.8.1. Both σ and π orientations were investigated using this setup. Figure 5.8.3 shows the corresponding laser spectra obtained for both σ and π orientations. With the crystal orientated in the π direction, Yb: CYA generated a laser with a η_{CW} of 92 % at a wavelength of about 1050 nm with a maximum output power of 2.3 W. With the crystal orientated in the σ direction, Yb: CYA generated a laser with a η_{CW} of 93 % at a wavelength of about 1051.5 nm with a maximum output power of 1.2 W. These efficiencies obtained are close to the η_q of 93.2 % of the laser considering that the peak absorption wavelength occurs at 979 nm and the lasing wavelength occurs at around 1050 nm. The slope efficiencies obtained are comparable to that of the cryogenically cooled Yb: YAG laser [34] and the room-temperature operation Yb: glass [35] laser which were

reported to have η_{CW} values of 90 % and 88 %, respectively. Under an absorbed pump power of 3.25 W the laser and with the crystal orientated in the π direction, the laser emitted a maximum output power of 2.3 W at 1050 nm. The corresponding η_{opt} was 70 %. Under an absorbed pump power of 2.8 W and with the crystal orientated in the σ direction, the laser emitted a maximum output power of 1.2 W at 1051.5 nm. The corresponding η_{opt} was about 43 %. The η_{opt} obtained when the crystal was orientated in the π direction was much larger than that of Yb: glass which reported an η_{opt} of slightly greater than 50 % [35]. Worthed noting is that even though the emission cross-section area in the σ direction is much larger than that of the π direction as shown in Fig. 5.6.3, the output powers obtained do not reflect this. This is because the absorption in the π (50 % of pump light) is very much larger than that of the σ direction (20 % of pump light) and as such is able to convert more of the pump light into useful laser light. For the generation of high power using Yb: CYA, it is advantageous to orientate the crystal to the π direction. The authors believe that even larger output power should be possible should larger pump powers be available as no sign of pump saturation was observed as seen from Fig 5.8.2. In addition, the amount of absorbed pump power can be further increased by optimizing the Yb³⁺-doping concentration since the segregation coefficient for Yb³⁺ ions in CYA is 0.74 [32] and such the doping concentrations can be further increased. The CW output powers are stable during the course of the measurements (~5 hours). However its output stability is most critically affected by the pump and thermal lensing which can be controlled with adequate cooling.

By analyzing the laser threshold values under OCs of different reflectivities, the total cavity losses were estimated using (3.2.5) using CW data from the V-shaped cavity orientated in the π direction as the output powers are much larger than when the crystal was orientated in the σ direction and as such the analysis would be more meaningful. The pump threshold, P_{th} , of Yb: CYA lasers under OCs with reflectivity of 99.2 %, 98.4 %, 97.6%, 96.5 % and 95% are depicted in Fig. 5.8.4 and fitted with (3.2.5) and resulted in a value of δ of 0.006 which corresponded to a total cavity round trip loss of 0.6 %. Assuming that the loss was purely caused by the Yb: CYA crystal, the corresponding loss coefficient, α , of the crystal is about 0.015 cm⁻¹. The values obtained are comparable to that of Yb: glass which reported a δ of 0.006 [35] and an α value of 0.012 cm⁻¹ [35]. This

shows that the Yb: CYA crystal used has excellent optical quality and as such the high η_{CW} . The η_{CW} values obtained demonstrates that Yb: CYA is an excellent high power and highly efficient CW gain medium.

Fig. 5.8.2.: CW slope efficiency curves of Yb: CYA in a V-shaped cavity when $T = 5 \%$. Solid line - π orientation, broken line - σ orientation.

Fig. 5.8.3.: CW spectra of Yb: CYA in a V-shaped cavity. Solid line - π orientation, broken line - σ orientation.

Fig. 5.8.4.: Determination of the round trip cavity losses of Yb: CYA in a V-shaped cavity with laser thresholds under output couplers of different transmissions.

Tunability of Yb: CYA orientated in the π orientation was experimentally investigated by placing a SF 10 prism near the OC in Fig 5.8.1. Fig. 5.8.5 shows the corresponding tuning curve measured under an absorbed pump power of 3.25 W with a 5 % transmission OC. The laser can be continuously tuned from 1008 nm to 1063 nm, giving a tuning range of 55 nm. This tuning range is much larger than that of the disordered Yb: NaGd (WO₄)₂ [36] and comparable to that of the Yb: CaF₂ [37]. More than 500 mW of output emission was available for wavelengths between 1015 nm and 1055 nm, indicating that Yb: CYA could be an excellent gain medium for tunable laser in the near infrared region. Much higher output powers should be achievable should higher pump power is available. The limited tunability towards the shorter wavelength is due to the strong reabsorption at these wavelengths while that towards the longer wavelengths could be due to losses introduced by the cavity mirrors.

Fig. 5.8.5.: The tuning curve of Yb: CYA orientated in the π direction. The output coupler has a transmission of 5 % and the absorbed pump power was fixed at 3.25 W.

The CW performances of Yb: CYA were also investigated using an X-fold cavity. The setup used was similar to that shown in Fig. 5.4.7 but with M4 replaced by a HR mirror,

L1 being 23.5 cm and L4 being 60 cm. The crystal used, focusing optics and experimental conditions such as the temperature of the diode and crystal were kept the same as that when the CW measurements were obtained for the V-shaped cavity. The crystal was oriented in the π direction in an effort to obtain higher output powers. ABCD analysis of this cavity showed that the laser had a spot size with a diameter of 180 μm within the crystal.

Fig 5.8.6.: The CW slope efficiency curves of Yb: CYA in an X-folded cavity under OCs of various transmissions.

Fig 5.8.7.: The CW laser spectrum of Yb: CYA in the X-fold cavity with an OC of transmission of 3.5 %.

The corresponding slope efficiency curves obtained using OCs of various transmissions is shown in Fig. 5.8.6. The highest output power obtained was 1.92 W under a pump power of 3.81 W. This resulted in a η_{CW} of 71 % and an η_{opt} of about 50 %. The slope efficiencies obtained using this cavity was lower than that obtained using the V-shaped cavity due to poorer mode-matching between the pump and laser beams. Fig. 5.8.7 shows the corresponding CW laser spectrum when the OC had a transmission of 3.5 %. It should be noted that in Fig. 5.8.6, pump power of about 8 W was focused into Yb: CYA into a spot size as small as $60 \times 120 \mu\text{m}^2$ and that no crystal fracture was observed. This shows that Yb: CYA has a κ value high enough for high brightness pumping.

Fig 5.9.1.: A schematic of the experimental setup used to mode-lock Yb: CYA in the negative dispersion regime. F1: aspherical lens, $f = 13.86$ mm. F2: plano-concave cylindrical lens, $f = -25.4$ mm. F3: plano-convex cylindrical lens, $f = 100$ mm. F4: plano-convex lens, $f = 75$ mm. M1, M2 and M3: Plano-concave mirrors, ROC = -100 mm, -300 mm and -100 mm, respectively. M4 and M5: Flat Mirrors. L1 = 23.5cm, L2 = 50cm, L3 = 5.5 cm, L4 = 20cm, L5 = 25cm and L6 = 40cm. $L_{\text{prism}} = 42$ cm.

5.9.: Mode-locking of Yb: CYA

The mode-locking of Yb: CYA will be investigated in this section. Fig 5.9.1 shows the schematic of the experimental setup used to mode-lock Yb: CYA in the negative dispersion regime to generate femtosecond pulses. For mode-locking of Yb: CYA in the positive dispersion regime, the dispersion prism pair from Fig. 5.9.1 were removed. However the parameters of the focusing optics used were different.

Solitary mode-locking of Yb: CYA

The crystal used to study the mode-locking of Yb: CYA in the negative dispersion regime was the same as that used to study the CW performances of Yb: CYA in the V-shaped and X-folded cavities. The crystal has a Yb^{3+} doping concentration of 3 at. % and was cut along the a-direction into dimensions of $2(L) \times 3(B) \times 3(H)$ mm³ orientated in the π direction for higher output powers. The orthogonal direction was not tested as the absorption (~ 20 %) too low and as such not expected to be able to generate high output

powers. The crystal was coated with antireflection coatings at both the pump and laser wavelengths. It was wrapped with indium foil and placed in a copper housing which was water-cooled at 14 °C throughout the experiments. To suppress etalon effects and to improve the mode-locking stability, the crystal was tilted at about 5°. The laser diode used is a board stripe diode with central emission at (975 ± 4) nm (Lumics GmbH). The emitter size is 94 µm wide and the diode is capable of emitting up to 12 W. The pump light was first beam shaped by an aspherical lens together with a telescope made up of a cylindrical plano-concave and a cylindrical plano-convex lens pair and finally focused into the crystal via a plano-convex lens. The pump beam size in the crystal was measured with the knife-edge method. It has diameters of about (60 ± 10) µm (fast axis, horizontal direction) $\times$ (120 ± 10) µm (slow axis, vertical direction). The slow axis of the diode was along the vertical direction. Based on the ABCD matrix calculation the laser beam has a radius of about 50 µm in the crystal and 30 µm on the semiconductor saturable absorption mirror (SESAM), respectively. The crystal was placed at the beam waist position. The laser spot on the SESAM has to be tightly focused to effectively bleach it. The crystal absorbed about 50 % of the pump light. As the segregation coefficient of Yb^{3+} in CYA is about 0.74 [13], it is believed that the amount of pump absorption can be further increased via increasing the Yb^{3+} doping concentration. Fig. 5.9.1 showed that the cavity had a repetition rate of about 89 MHz. The SESAM (BATOP GmbH) was designed to operate at 1064 nm with a modulation depth of 0.2 %, a non-saturable absorption of 0.3 %, a relaxation time of 500 fs, and a saturation fluence of $90 \mu\text{J cm}^{-2}$. To ensure high output power, the non-saturable losses should be kept to a minimum and as such a SESAM with a non-saturable loss of 0.3 % was chosen. A pair of SF10 prisms rotated to the Brewster angle was inserted into the cavity to provide dispersion compensation. It was however difficult to determine the amount of negative dispersion required to compensate for the positive dispersion of Yb: CYA as the dispersion data was not available. Unlike in the case of Yb: SSO where the dispersion data of similar crystals such as Yb: GSO [14] was available and as such could be used the estimate dispersion of Yb: SSO, this was not the case for Yb: CYA. The next crystal with properties most similar to Yb: CYA was Yb: CALGO [16, 28]. However, even for Yb: CALGO, no dispersion data on its dispersion was published. The mode-locking of Yb: CYA was

conducted with a large net negative dispersion regime by ensuring that the negative dispersion provided by the prisms was very much larger than the positive dispersion of the crystal. The negative dispersion provided by the SF10 prism pair with a L_{prism} separation of 42 cm was estimated to be about -1500 fs^2 . It should be noted that for most solid state gain media of about 2 mm long, the positive dispersion is typically less than 400 fs^2 . The cavity mirrors used to initiate mode-locking were low GVD mirrors purchased from Layertec GmbH. These mirrors have close to zero dispersion from 1040 nm to 1060 nm. In addition, the SESAMs used to initiate mode-locking has close to zero dispersion at the wavelengths concerned. The crystal was assumed to be the main dispersive element in the cavity. The mode locked pulse train was monitored with a low noise photodetector (New Focus 1611-FC-AC) and displayed on a 1 GHz digital oscilloscope (Tektronix DP0714). Unfortunately the pulse to pulse stability cannot be measured as oscilloscope lacked the required statistical package to do such an analysis. The optical spectrum was measured with a high-resolution optical spectrum analyzer (Ando, AQ-6315B), while the pulse width was measured using a commercial autocorrelator (APE, PulseCheck).

Fig 5.9.2.: Mode-locked pulsetrain of Yb: CYA in the negative dispersion regime with an OC of transmission of 2.4 %.

Fig 5.9.3.: The mode-locked slope efficiency curves of Yb: CYA using OCs of various transmissions. The closed circles are experimental data. The solid and broken lines are the linear fits.

Femtosecond mode-locking of Yb: CYA was investigated using OCs of various transmissions. It was noted that the mode-locking was sensitive to alignment of the OC and the SESAM. Fig 5.9.2 shows a representative mode-locked pulsetrain when Yb: CYA was mode-locked in the negative dispersion regime. The pulsetrain shows that the pulse to pulse separation was about 11 ns which corresponded to the cavity repetition rate of 89 MHz. This implies that fundamental mode-locking has been achieved and that a single pulse circulates within the cavity. It was noted that in the femtosecond mode-locking of Yb: CYA, the laser initially emits CW laser and upon reaching the mode-locking threshold, the mode of operation switches abruptly to that of mode-locking. No Q-switched mode locking was observed. Fig. 5.9.3 shows mode-locked slope efficiency curves using OCs of various transmissions. The maximum mode-locked output power was obtained when the OC had a transmission of 2.4 %. Under CW operations, η_{CW} was 16 %, when mode-locked, the femtosecond mode-locked slope efficiency, $\eta_{femtosecond}$, increased to 37 %. Under a maximum available absorbed pump power of 3.7 W, the mode-locked laser output power was 740 mW, which corresponded to η_{opt} of 20 %.

When mode-locked the laser has always emitted TEM₀₀ beam. The laser produced maximum mode-locked output powers of 560 mW and 180 mW when the OCs were 1.6 % and 0.8 % respectively.

Fig. 5.9.4 shows the corresponding autocorrelation traces and the optical spectra of the mode-locked pulses obtained using OCs of various transmissions. All the autocorrelation traces were obtained under maximum output powers. Fig. 5.9.4 (a) and (d) show that the autocorrelation trace and mode-locked spectrum obtained when the OC used has a transmission of 0.8 %. Assuming Sech² shaped pulses, the autocorrelation trace gives a pulse width of 153 fs. The spectrum is centered at 1045.3 nm with a FWHM bandwidth of 8.1 nm. The time-bandwidth-product of the pulses is 0.32, indicating that the pulses are near transform-limited. Under this configuration, the pulses had energies of 2 nJ and a peak power of about 13 KW. Fig. 5.8.4 9(b) and (e) show that the autocorrelation trace and mode-locked spectrum obtained when the OC used has a transmission of 1.6 %. Assuming Sech² shaped pulses, the autocorrelation trace gives a pulse width of 147 fs. The spectrum is centered at 1047.3 nm with a FWHM bandwidth of 8 nm. The time-bandwidth-product of the pulses is 0.32, indicating that the pulses are near transform-limited. Under this configuration, the pulses had energies of 6.2 nJ with a peak power of 42 KW. Fig.5.9.4 (c) and (f) show that the autocorrelation trace and mode-locked spectrum obtained when the OC used had a transmission of 2.4 %. Assuming Sech² shaped pulses, the autocorrelation trace gives a pulse width of 156 fs. The spectrum is centered at 1045.3 nm with a FWHM bandwidth of 8 nm. The TBP of the pulses is 0.34, indicating that the pulses are near transform-limited. Under this configuration, the pulses had energies of 8.2 nJ with a peak power of 53 KW. The efficiencies and peak powers obtained for Yb: CYA are relatively high amongst Yb³⁺ doped femtosecond end pumped solid state oscillators. For example, Yb: YVO₄ [38] reported mode-locked output powers of 300 mW with a peak power of 14.5 KW and Yb: Sc₂O₃ [39] reported output powers of 540 mW with a peak power of 25.6 KW. In addition, Druon *et. al.* [40] also reported 300 mW output powers from Yb: BOYS with a peak power 33 KW. The author would like to point out that although other Yb³⁺ doped femtosecond end pumped solid state oscillators such as Yb: CaF₂ [41], Yb: CALGO [28], Yb: LSO [5] and Yb: YSO [5] had reported higher peak powers (but not necessarily shorter pulses), it was mainly because these

lasers were pumped by fiber coupled diode lasers which are capable of delivering higher pump powers. In the case of Yb: CYA, the maximum output powers obtained were limited by the pump power available and it is the author's belief that should more pump power be available, higher output powers should be possible as the mode-locked slope efficiency curves in Fig. 5.9.3 did not saturate. But nevertheless, the results obtained demonstrate that that Yb: CYA is a promising gain medium for the generation of efficient high power femtosecond pulses.

Fig 5.9.4.: The autocorrelation traces when Yb: CYA was mode-locked using OCs with transmissions of (a) 0.8 %, (b) 1.6 % and (c) 2.4% with (d), (e) and (f) showing their corresponding spectra respectively. The blue solid line in the autocorrelation traces are sech^2 fits.

Reference to equation (5.4.1) (soliton area theorem) shows that a solitary laser would generate pulses of higher energy when the net dispersion in the cavity is made large. In the mode-locking of Yb: CYA, a large GVD of -1500 fs^2 was introduced into the cavity such that the laser mode-locked with a large net negative dispersion. The large dispersion resulted in large output powers to be obtained when Yb: CYA was mode-locked as shown from the results presented in this section. This was however done at the expense of obtaining longer pulses as the net dispersion in a solitary laser also scales with the pulse width as shown by equation (5.4.1). Hence a balance would have to be struck between obtaining an output power that is high enough and a pulse width of reasonable width. It is worth mentioning again that the crystal is orientated in the π direction. This was because the absorption of the pump power in this orientation is much higher than the orthogonal direction and as such the output power generated would be much higher and this was reflected in the CW results with the V-shaped cavity in Fig. 5.8.2. When orientated in the π direction, Yb: CYA would mode-lock towards the 1050 nm peak. The limited bandwidth close to the peak would otherwise limit the shortest achievable pulsewidth. Hence for the generation of even shorter pulses it would be better to use the σ direction instead where the broadband plateau would allow the generation of pulses that are much shorter than 150 fs. It is likely that due to the limited absorption in this direction the output powers generated would be lower than what was observed in Fig. 5.9.3 despite the higher σ_{em} in this direction. Hence for the generation of high output powers it would be more advantageous to use the π direction, however for shorter pulse generation, it would be better to use σ direction to exploit the plateau in the emission spectrum as shown in Fig. 5.6.3. However the absorption of σ direction would have to be increased. This should ideally be done by increasing the doping concentration of Yb^{3+} .

As in the case of Yb: SSO, some of the results obtained may also be understood by making references to Haus' master equation of mode-locking (equation (2.2.8)) and Fig. 2.2.2. Although the laser was operated with a large net negative dispersion to generate higher output power, nevertheless the output pulses obtained are still near transform limited. This is because even when the net negative dispersion is large, it can still compensate the SPM in the system and as such remove most of the chirp in the system as shown in Fig. 2.2.2 (b). Fig. 2.2.2 (b) also shows that even in the presence of strong

negative dispersion the chirp in the laser still stays close to zero and as such allow one to generate near transform limited pulses.

Dissipative solitons in Yb: CYA

Traditionally, the operation of femtosecond SSLs is usually designed to operate in the negative dispersion regime. The interplay between SPM and net negative dispersion results in the formation of solitary pulses shaped according to the NLSE/ Haus' master equation of mode-locking. Solitary soliton type shaping not only stabilizes the mode locking operation, but also significantly reduces the mode-locked pulse width and as the net cavity dispersion reduces the narrower the formed soliton pulses at the expense of output power as shown in the case of solitary mode-locking of Yb: SSO and Yb: CYA. However when operating in the negative dispersion regime, excessive non-linear phase shift due to SPM combined with negative GVD can distort a solitary pulse causing it to breakup and as such the laser operates in multi-pulsing mode causing a drop in the output power and increase in pulse width. Excessive SPM can also result in the mode-locking to be unstable due to the presence of noise as shown in Fig. 2.2.2 (a). Excess non-linear phase shift is given by the B integral which states:

$$B(t, L) = \frac{\omega}{c} \int_0^L n_2 I(t, z) dz \quad (5.9.1)$$

where $B(t, L)$ is the B intergral, ω is the angular frequency, c is the speed of light, n_2 is the non-linear refractive index of the gain medium, $I(t, z)$ is the intensity of the solitary pulses and L is the propagation distance. (5.9.1) shows that excess non-linear phase would build up when the peak intensity increases. As such as the power increases or when the pulse shortens, excess non-linear phase builds up. When the value of $B(t, L)$ increases beyond a threshold value, non-linear effects such as self-focusing can distort the beam and breakup a solitary pulse. One way to side-step this problem is to operate the laser in the positive dispersion regime in which DSs can form highly chirped pulses in the picosecond regime. As DSs are highly chirped everywhere within a laser cavity, their corresponding intensities would be significantly lower than femtosecond solitary pulses. To that end, their power and energy can be scaled to values higher than that are normally not permitted by solitary lasers. As the output of a DS is highly chirped, the pulses can be

compressed using an extra-cavity compressor to obtain high power femtosecond pulses. The rest of the section is devoted to the positive dispersion mode-locking of Yb: CYA in order to realize DSs. The author shall demonstrate that when mode-locked in the positive dispersion regime, DSs can also be demonstrated in Yb: CYA and that high power femtosecond pulses that would otherwise be difficult to demonstrate in solitary mode-locked lasers can be realized.

Fig 5.9.5.: Experimental setup used to investigate positive dispersion mode-locking in Yb: CYA. F1: Aspherical lens, $f = 8$ mm. F2: Plano-concave cylindrical lens, $f = -25.4$ mm. F3: Plano-convex cylindrical lens, $f = 125$ mm. F4: Plano-convex lens, $f = 75$ mm. M1, M2 and M3: Plano-concave mirrors, $ROC = -100$ mm, -300 mm and -100 mm, respectively. SESAM, OC: $ROCs = \infty$. $L1 = 23.5$ cm, $L2 = 50$ cm, $L3 = 5.4$ cm and $L4 = 90$ cm. OC: Output couplers: $T = 0.8\%$ or $T = 5\%$.

Fig. 5.9.6.: Slope efficiency curves of Yb: CYA mode-locked in the positive dispersion regime. Closed circles: $T = 0.8$ %. Closed squares: $T = 5$ %. The red, green and yellow lines represent the CW, q-switched mode-locking, and cw mode-locking, respectively.

Positive dispersion mode-locking in Yb: CYA was investigated using the setup shown in Fig. 5.9.5. The setup is similar to the one used to investigate the positive dispersion mode-locking of Yb: SSO but the focusing optics used are different. The Yb: CYA crystal used has a Yb^{3+} doping concentration of 8 at. % and dimensions of $2.5 \text{ (L)} \times 3 \text{ (B)} \times 3 \text{ (H)} \text{ mm}^3$ and was cut along the a-direction. The crystal was oriented in the σ direction to ensure that more bandwidth can be generated during mode-locking at the expense of obtaining lower output power due to the lower pump absorption in this direction. The problem of lower absorption however was offset by using a crystal of higher doping concentration. Both facets of the crystal were HT coated at the pump and laser wavelengths to minimize the Fresnel reflection loss. The crystal was wrapped with indium foil and placed in a water-cooled copper house whose temperature was maintained at 14°C . The crystal was set with a slight tilt of about $5\text{--}8^\circ$ to the cavity axis to suppress etalon effects. A board stripe diode laser (Lumics, GmbH) which emits at a central wavelength of $(976 \pm 4) \text{ nm}$ was used as the pump source. The diode laser has an emitter size of $94 \text{ }\mu\text{m}$ and its temperature was maintained at 15°C . The pump light is first collimated by an aspherical lens and magnified through a $5\times$ plano-concave and plano-

convex cylindrical lens pair before being focused into the crystal by a plano-convex lens. In this case F1 was changed from a focal length of 13.86 mm as in the case when Yb:SSO was mode-locked to one with a focal length of 8 mm but with bigger numerical aperture to allow more pump light to reach the crystal. This was done to off-set for the lower pump absorption when the crystal was orientated in the σ direction for these experiments. The tradeoff however of changing F1 to one with a shorter focal length was that the laser spot size would become larger and this would lead to poorer mode-matching. In anticipation of this, F3 was changed from 100 mm to 125 mm such that the beam expander now expands the pump beam by a factor of $5\times$ instead of $4\times$ such that a smaller pump beam was obtained when focused into the crystal by F4. The diameter of the pump size in crystal is about $(80 \pm 10) \mu\text{m}$ (fast axis, horizontal direction) $\times$ $(200 \pm 10) \mu\text{m}$ (slow axis, vertical direction). Based on the ABCD analysis the laser has a spot size of $100 \mu\text{m}$ in diameter in the crystal and $60 \mu\text{m}$ in diameter on the semiconductor saturable absorber mirror (SESAM), respectively. The crystal absorbed about 70 % of the incident light. A commercial SESAM (BATOP GmbH) was used to initiate the mode-locking. The SESAM was designed to operate at 1064 nm with a modulation depth of 0.2 %, a non-saturable absorption of 0.3 %, a relaxation time of 500 fs and a saturation fluence of $90 \mu\text{J cm}^{-2}$. A SESAM with low non-saturable loss was preferred to reduce losses in the cavity.

Fig. 5.9.7.: Mode-locked pulse trains of Yb: CYA in the positive dispersion regime (a) 50 ns/ div (b) 2 $\mu\text{s}/\text{div}$

Mode-locking of the laser under $T = 0.8 \%$ and $T = 5 \%$ were investigated. In both cases the laser initially emitted CW. As the pump power was increased, the laser emission then switched to the QML and CW mode locking as depicted in Fig 5.9.6. Once CW mode-locking has been achieved, the laser can maintain this state of operation for hours unless it is disturbed. With $T = 0.8 \%$ the maximum output power achieved was 0.29 W and $\eta_{\text{picosecond}}$ was 10 %. With $T = 5 \%$, the maximum output powers achieved was 1.5 W while $\eta_{\text{picosecond}} = 33 \%$. The cavity repetition rate was 89 MHz. The pulse energies were 3.3 nJ and 17 nJ, respectively. Even larger output power should be possible if the mode matching between the pump and laser beam can be improved through the usage of a long focal length large aperture aspherical lens. Fig. 5.9.7 (a) and (b) shows the mode-locked pulse trains obtained at maximum output power. Fig. 5.9.7 (a) show that the pulse to pulse separation was about 11 ns which corresponded to the cavity repetition rate. This implies that the laser was fundamentally mode-locked at the maximum output power. No double pulsing was observed during the course of the experiment.

Fig. 5.9.8.: Optical spectra of the mode-locked pulses under different pump powers with (a) $T = 0.8 \%$ and (b) $T = 5 \%$.

Fig. 5.9.9.: Autocorrelation traces when $T = 0.8\%$. a) Uncompressed trace b) Compressed trace. Closed circles are experimental data while the solid blue line is a sech^2 fit.

Fig. 5.9.10.: Autocorrelation traces when $T = 5\%$. a) Uncompressed trace b) Compressed trace. Closed circles are experimental data while the solid blue line is a sech^2 fit

Fig. 5.9.8 shows the optical spectra of the mode-locked pulses under different pump powers. The spectra exhibit characteristic sharp steep spectral edges which are characteristics of DSs. The pump powers that corresponded to the spectra shown in Fig. 5.9.8 were between 5 W to 5.5 W of the absorbed pump power. The broadest FWHM spectra measured at the maximum output powers were 13 nm centered at 1057 nm and 8 nm at 1055 nm when $T = 0.8\%$ and $T = 5\%$, respectively. The spectra had bandwidths capable of supporting femtosecond pulses as short as 90 fs and 140 fs with spectra bandwidths of 13 nm and 8 nm respectively. Fig. 5.9.9 (a) and Fig. 5.9.10 (a) further show the autocorrelation traces measured at the maximum output powers. Assuming that the pulses have a Sech^2 profile, the emitted pulse widths were 7.4 ps and 8.5 ps, respectively. The TBPs were 26 and 19, which were 80 and 60 times above the Fourier transform-limited value. It means that the pulses are strongly chirped. Using a pair of reflection gratings (Spectrogon, 1200 lines per mm) separated by 7 cm, the pulses were extra cavity compressed. The narrowest pulses obtained were 340 fs and 380 fs,

respectively, as shown in Fig 5.9.9 (b) and Fig 5.9.10 (b). Weak pedestals are clearly observable on the bases of the compressed autocorrelation traces, which indicate the presence of non-linear chirp which implies that dechirping of the pulses to the transform limit was not straight forward. It is likely that compression to the Fourier limit would result in autocorrelation traces with very strong pedestals. A similar observation and conclusion was made by Cho *et.al* [41] when Ti: Sapphire was mode-locked in the positive dispersion regime. The author believes that to obtain the transform-limited pulses from the Yb: CYA laser a non-linear pulse compression method would be needed, e. g. via a piece of optical fiber [42]. The highly chirped pulses obtain indicates the presence of strong SPM effects within the cavity and that when operated in the positive dispersion regime results in strongly chirped pulses as predicated by Haus' master equation of mode-locking (equation (2.2.8)) and Fig 2.2.2 (b). Compared to the case of Yb: SSO, the chirp in Yb: CYA is very much stronger although the cavity designs are similar. This may be attributed to stronger SPM effects in Yb: CYA and perhaps different amounts of gain filtering. However this can only be confirmed through rigorous computer simulations. In the case $T = 0.8 \%$, the SPM within the resonator is larger than that in the case where $T = 5 \%$ and in the presence of positive dispersion resulting in the pulses to be more strongly chirped (Fig. 2.2.2 (b)). In addition, the stronger SPM results within the resonator for the $T = 0.8 \%$ case generates a larger bandwidth as observed in Fig. 5.9.8 which allows the DS pulses obtained to be compressed to shorter pulses.

As in the case of Yb: SSO, the spectra closely resemble that of a rectangle and the pulses obtained were strongly chirped. The author would like to stress on the fact that the cavity used to initiate mode-locking of Yb: CYA did not contain any negative dispersion. In fact the only dispersion within the cavity came from the material dispersion of Yb: CYA which provided a net positive GVD. Yet the spectra obtained had bandwidths capable of supporting femtosecond pulses and were compressible to the femtosecond regime. This is in conflict with conventional wisdom in which femtosecond pulse could only be generated in the negative dispersion regime [25]. Clearly the results obtained demonstrate another type of mode-locking that is accessible to Yb^{3+} doped solid state lasers to generate femtosecond pulses. In fact the results obtained clearly exhibits all the hallmarks of DSs. DSs were previously been observed in fiber [22-25] and Ti: Sapphire

[26-27, 41-43] mode-locked lasers. Kalashnikov *et. al.* [26] showed theoretically, that previous observations of rectangular like spectra in positive dispersion mode-locked Ti: Sapphire lasers were chirped solitons and such systems are known as positive dispersion mode-locked lasers. However the author would like to point out that mode-locked laser systems are actually dissipative systems and as such, positive dispersion mode-locking in Ti: sapphire lasers would be more accurately described as DS lasers instead to better reflect the nature of mode-locked SSLs. In essence, the chirped pulses observed in Ti: Sapphire lasers should be called DSs as it would better reflect the nature of such systems.

Chong *et. al.* [23-24] and Renninger *et. al.* [44] carried out numerical simulations and found a wide range of stable spectra using a modified version of Haus's master equation of mode-locking. They also experimentally verified that the rectangular pulses observed in fiber lasers were DSs. From their works however, it was unclear if this mode of operation could be extended to Yb³⁺ doped SSLs as well. In particular, the theoretical work of Kalashnikov *et. al.* [26] showed that DSs were unlikely to exist in Yb³⁺ doped SSL as their emission bandwidths were considered to be too narrow. The experimental results demonstrated in this thesis for Yb: SSO and Yb: CYA however showed otherwise and that DSs can exist in Yb³⁺ SSLs. The author would also like to point out while there have been previous reports of mode-locking of Yb: KYW [45-46] in the positive dispersion in which rectangular like spectra with highly chirped pulses had been observed, the authors failed to point out what they observed were in fact DSs or even chirped solitons. To that end, the positive dispersion mode-locked results presented in this section are the first identification of DSs in Yb³⁺ SSLs.

It has been demonstrated that the cubic-quintic Ginzburg – Landau equation (CQGLE) [44] which is a variation of Haus' master equation for mode-locking as described by equations (2.2.8) in chapter 2 can accurately model DSs in fiber lasers. This was done by consideration of the cubic and quintic saturable absorption terms in addition to the effects already considered in the Ginzburg – Landau equation as shown in equation (2.2.10). Using the CQGLE, Chong *et.al.* [24] and Renninger *et. al.* [44] showed both numerically and experimentally cavity non-linearities due to SPM, spectral filtering and net positive dispersion GVD are major pulse shaping effects in DSs. Hence, the manifestation of DSs results due to the interplay between three factors which is also the reason as to why DSs

were not previously observed in Yb^{3+} SSLs and are seldom observed in optical systems [25]. In the case of Ti: sapphire [47], the spectral filtering arises due to gain bandwidth filtering by the laser system. Likewise in Yb: SSO and Yb: CYA, the spectral filtering arises due to gain bandwidth filtering in the mode-locked systems which has the same effect as spectral filtering. Recognizing the DS operation of a laser can help to design a better laser, for example, by exploiting the DS resonance [48], the energy of achievable mode-locked pulses could be significantly increased. Wu *et. al.* [49] have demonstrated DS resonance in an Er^{3+} doped fiber laser in which single pulse energy as large as 280 nJ was obtained. Compared to operation in the negative dispersion regime, the pulses are chirped every within the cavity [25] and as such the peak intensity within the cavity will not exceed the threshold intensity require for pulse breakup and distortion which would otherwise limit output powers and pulsewidths. In addition, the pulses are already strongly chirped and as such do not require a stretcher when amplifying the pulses. This can reduce the cost and complexity of amplifier systems. Hence the operation of a Yb^{3+} doped SSL as DSs to generate femtosecond pulses offers many advantages.

5.10.: Concluding remarks

The laser performances of novel Yb: SSO and Yb: has been demonstrated. Both gain media demonstrated highly efficient CW laser generation with output powers of 2.7 W and 2.3 W generated using Yb: SSO and Yb: CYA respectively which corresponds to η_{CW} of 70 % and 92 %.

The mode-locking performances of these novel gain media were also investigated under negative and positive dispersion mode-locking. Under negative dispersion mode-locking, solitary mode-locking was demonstrated in both Yb : SSO and Yb CYA. Under solitary mode-locking with the dispersion set close to zero net dispersion, the shortest pulse achieved was 93 fs with an output power of 20 mW at 1043 nm. The mode-locked spectrum exhibits satellite peaks on either sides of the principal spectrum which occurred due to FOD effects dominating. To the author's knowledge, this is the first demonstration of FOD effects being observed in Yb^{3+} doped SSLs. By operating at a slightly larger net negative dispersion, the laser mode-locked at 1077 nm with a pulse width of 125 fs with an output power of about 100 mW. Similarly, Yb: CYA was also mode-locked under negative dispersion mode-locking. The mode-locking was conducted under a large net

cavity dispersion and it resulted in the laser to mode-lock at 1045.3 nm with a pulse width of 156 fs and an output power of 740 mW.

When mode-locked under positive dispersion, both Yb: SSO and Yb: CYA demonstrated DSs. They exhibited mode-locked spectral that were non-Gaussian in shaped but had steep edges that resembles that of a rectangle which had bandwidths capable of supporting femtosecond pulses. The pulses obtained were strongly chirped such that the TBPs were tens of times above the Fourier transform limit. These are hallmarks of DSs. In particular, Yb: CYA demonstrated an output power of 1.5 W that was compressible to 380 fs. To the author's knowledge, the work presented demonstrates the first observations of DSs in Yb³⁺ doped SSLs. The results obtained also shows that DSs are an accessible regime for for Yb³⁺ doped SSLs. By operating the laser as DSs, femtosecond pulses of potentially higher energies and output powers could be obtained as the pulses are chirped everywhere within the cavity and as such less susceptible to pulse breakups and distortion due to high intensities.

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Chapter 6

Mode-locking of ceramic Nd: YAG with carbon based nano-materials

Carbon based nano-materials such as single-walled carbon nanotubes (SWCNTs) and graphene have been demonstrated to exhibit saturable absorption properties [1-2] and as such is thought to be able to function as SAs in lasers systems. Set *et. al.* [3] demonstrated the usage of a mixed sample of SWCNTs containing multiple species of semiconducting and metallic nanotubes in a transmission setup in which he called “saturable absorber incorporating nano-tube” or (SAINT) to mode-lock fiber lasers. In 2009 Bao *et. al.* [4] demonstrated mode-locking of fiber lasers at 1.55 μm by doping a polymer matrix with graphene in a transmission setup. This early works is indicative that both SWCNTs and graphene are competent mode-locks and can be used in laser systems as SAs. The author shall demonstrate in this chapter that this idea can be extended to SSLs and that both SWCNTs and graphene can potentially replace SESAMs in SSL as the SA to initiate mode-locking.

6.1.: Mode-locking of ceramic Nd: YAG with (7, 5) SWCNT

SESAMs are the current state of the art technology used for achieving mode locking in diode pumped solid SSL systems [5]. In recent years however, SWCNTs has emerged as an alternative to the SESAM due to their excellent non-linear absorption properties [1, 6], ease and lower costs of fabrication as compared to SESAMs. The operation/absorption wavelengths of SWCNT SAs depended on the diameter and chirality of the nanotubes present in the sample. Only semiconducting nanotubes and nanotubes with diameters that resonated with the laser absorption/operating wavelength will take part in the saturation absorption process [7]. Non-resonate and metallic nanotubes serve only as scattering sites as they have lengths comparable to typical lasing

wavelengths. In addition, poorly dispersed SWCNTs can form bundles which can result in even more optical losses via scattering. The removal or reduction of non-resonant and metallic nanotubes would lead to reduced losses and improved laser performance. Work reported thus far in the literature for both fiber laser [8-9] and SSL [7-9] systems used multiple species SWCNTs which consisted of both semiconducting and metallic nanotubes. This was because of difficulties in nanotube filtration techniques. Recently, advances made in purification techniques have made it possible to obtain highly purified samples of semiconducting SWCNTs which can contain a high percentage of single species semiconducting SWCNTs. This can be done either through the usage of polymer [13] or deoxyribonucleic acid (DNA) [14] extraction methods. These techniques enable one to selectively choose nanotubes that are both semiconducting and at the same time has an absorption that is tailored to the desired operating laser wavelength.

Although a single species of nanotubes would have a limited saturable absorption bandwidth, which limits its application as a broadband mode locker, it would be possible to use a variety of polymers or DNA to extract single species of semiconducting nanotubes. By mixing these nanotubes together, one is able to extend the operation wavelengths of the as fabricated SWCNT SA. For example, for mode locking a Yb based crystal, the (6,5), (7,5), (10,2) and (8,4) semiconducting nanotubes can be extracted and mixed together such that the operating wavelength spans between 1000 to 1100 nm [14]. By doing so, one can tailor the SWCNT SA to absorb at the wavelength of interest while avoiding non-resonant SWCNT species that could contribute to additional losses. Cho *et. al.* [15] mixed several single species SWCNTs with peak absorption at several different wavelengths together. The fabricated SA had an operating wavelength that spanned more than 500 nm and could be used to mode-lock SSLs which operated in the range of 1 μm to 1.5 μm . The author shall demonstrate in this chapter the possibility of using a polymer extraction method to extract a single species of semiconducting (7, 5) nanotube to mode lock a ceramic Nd: YAG SSL at 1064 nm.

Fig. 6.1.1.: (a) Dispersed mixture of unpurified SWCNTs in PFO/toluene mixture. (b) Purified and wrapped SWCNTs float to the top of the mixture while unwrapped SWCNTs drop to the bottom of the mixture after centrifugation.

SWCNT SA fabrication

The SWCNTs used to fabricate the SA was purified using the method suggested by Chen *et. al.* [13] in which he demonstrated that poly (9, 9-dioctylfluoreny-2,7-diyl) or PFO polymer has an affinity to wrap itself around semiconducting (7, 5) SWCNTs and as such forms the basis of a filtration method capable of extracting a highly purified sample of this particular species of nanotube. To carry out this filtration process, 10 mg of PFO was first dissolved in 20 ml of toluene. 5 mg of unpurified SWCNTs powder was then dispersed in the PFO/toluene mixture. The solution was then homogenized in an ultrasonic bath (MRC Ultrasonic Cleaner, Model: DC150) for an hour which was then sonicated by a probe sonicator (Sonics & Materials Inc, Model: VCX 130) for 40 minutes with on and off pulse of 1 second. The solution was centrifuged (Thermo-Electron Corporation, Model: Jouan B4i) at a speed of 10,000 rpm for 20 minutes. SWNTs which were not wrapped by PFO sank to the bottom while wrapped nanotubes floated to the top of the solution. The wrapped SWCNTs were then extracted. Fig. 6.1.1 (a) and (b) shows the SWCNT mixture before and after centrifugation respectively. PFO was found to have the ability to wrap itself around nanotubes of certain diameters especially (7, 5) semiconducting nanotubes [13]. This resulted in the solution extracted to consist mainly of (7, 5) semiconducting nanotubes. (7, 5) nanotubes have absorption at 1064 nm [14] and as such can operate as a SA for mode locking ceramic Nd: YAG lasers. Fig. 6.1.2 shows the UV-VIS-IR measurements carried out on UV-VIS-IR spectrometer (Perkin-

Elmer, Lambda 900a). The shape peak at around 1040 nm is the signature absorption peak of (7, 5) nanotubes. The absence of strong peaks in the spectrum also demonstrates that the sample is highly purified. Although the absorption weakens towards 1064 nm, it nevertheless has absorption at that wavelength and as such is capable of functioning as a SA for ceramic Nd: YAG lasers.

Fig. 6.1.2: UV-VIS-IR absorption spectrum of the purified (7,5) semiconducting SWCNT solution. The vertical broken line marks the operating wavelength of ceramic Nd: YAG at 1064 nm.

To fabricate the SWCNT SA, the as fabricated solution was dropped casted onto the surface of commercially available HR dielectric mirrors placed on top of a hot plate. Throughout the drop casting process, the hotplate was maintained at 80 °C. These coated mirrors were used to mode lock the ceramic Nd: YAG laser. In addition, blank pieces of quartz were also coated using this process such that the saturable absorption properties of these SWCNT films can be measured using open aperture Z-scan. The film thickness on both the HR mirrors and quartz substrates were estimated to be about 500 nm thick.

Fig. 6.1.3.: Schematic of the experimental setup used to mode-lock ceramic Nd: YAG with SWCNTs. The SWCNT SA mirror/SESAM was placed at M4. F1 and F2 are beam reshaping lens. M1, M2 and M3 are concave mirrors while M5 is the output coupler.

Mode-locking experiments

The experimental setup is as shown in Fig 6.1.3. M4 was the SWCNT mirror. An X folded cavity was used in which the fundamental pulse repetition rate was 90 MHz. The laser crystal used was a piece of $3(\text{L}) \times 3(\text{B}) \times 4(\text{H}) \text{ mm}^3$ ceramic Nd: YAG (Konoshima). The gain medium used was a 0.5 at. % Nd: YAG doping. The crystal had HT coatings at the pump and laser wavelengths on both its surfaces. It was wrapped in a piece of indium foiled and placed within a water cooled housing that was maintained at 18°C . The piece of ceramic Nd: YAG was pumped by a single emitter (nLight, C-6.0-808) at 808 nm and was capable of delivering up to 6.4 W of pump power. The diode was attached to a large open heat sink. Active cooling of the diode was not employed. The diode used had a micro-lens to collimate the output beam in the fast axis direction such that it can be collimated by F1, placed at 75 mm away from it. F2 then focus the pump beam into a spot size of diameter of about $100 \mu\text{m}$ (fast axis) $\times$ $250 \mu\text{m}$ (slow axis) within the crystal. The output coupler used in had a transmission of 4 % at 1064 nm and had a 3° wedge to reduce back reflections into the cavity. This improves mode-locking stability. ABCD analysis of the cavity showed that the laser spot size had a diameter of about $140 \mu\text{m}$ within the crystal and on the SWCNT SA. The mode locked pulse trains

were measured using a low noise photoreceiver (New Focus 1611-FC-AC) connected to 1 GHz digital oscilloscope (Tektronix DP0714). The optical spectrum was obtained using a high resolution optical spectrum analyzer purchased from Ando (AQ-6315B) while the pulse width was measured using autocorrelation techniques (Femtochrome Research Inc., FR-103 XL). All optical powers were measured using power meters from Gentec-e (Solo PE) and Coherent (Powermax, PM30).

Fig. 6.1.4.: (a) Single pulsing at an output power of 130 mW (b) multi-pulsing at an output power of 275 mW.

The ceramic Nd: YAG laser initially emits CW laser and upon further increment of the pump power operated in Q-switched mode-locking (QML). Further increment of the pump power resulted in CW mode-locking and when the pump power was further increased the laser operated under multi-pulsing mode. Typical mode-locked pulse trains are shown in Fig. 6.1.4. Fig. 6.1.4 (a) was obtained when the laser emitted an output power of 130 mW. It shows the pulse separation obtained from the pulse trains was found to be about 11 ns which corresponded to the repetition rate of the cavity. This is an indication that the laser was mode locked at its fundamental frequency at this output

power. The shape of the pulse trains obtained resembles those that can be obtained with SESAMs. The pulses are clean, narrow and stable. The pulse to pulse stability unfortunately cannot be quantified as the author's oscilloscope lacked the required statistical software package to analyze the pulses. Under stable CW mode-locking, the laser produced a maximum output power of 130 mW. Stable CW mode-locking was observed from 3.37 W to 5.38 W of pump power. Stronger pumping resulted in a maximum output power of about 275 mW but multiple pulsing could not be suppressed. Fig. 6.1.4 (b) shows a pulse train obtained at an output power of 275 mW. It shows that within a time span of 11 ns, three pulses exist within the cavity as the pulse breaks up due to overdriving of the SWCNT SAs [16]. The slope efficiency curve of the laser is shown in Fig. 6.1.5. The SWCNT mode-locked laser has a $\eta_{\text{picosecond}}$ of close to 5 % under stable fundamental mode-locking. This was lower than that reported in ref. [17] in which a ceramic Nd: YAG was mode locked with a piece of SESAM and had a $\eta_{\text{picosecond}}$ of 25 %. This could be attributed to the higher non-saturable losses of the SWCNT fabricated. This was expected as SESAMs are commercial products and as such have lower non-saturable losses compared to SWCNTs SA fabricated in laboratories. To confirm this suspicion, the SWCNT SA was replaced by a commercial SESAM (BATOP) whose central wavelength was designed to be at 1064 nm with a ΔR of 0.6 %, a ΔR_{ns} of 0.4 % and a relaxation time of 500 fs was also used to mode-lock ceramic Nd: YAG. The SESAM mode-locked laser generated 1 W of mode-locked output power at maximum pump power of 6.4 W with a $\eta_{\text{picosecond}}$ of 20 %. This shows that by improving the SWCNTs SA fabrication process to reduce the non-saturable losses, the mode-locked slope efficiency of the laser can be enhanced. The advantage of SWCNTs over SESAMs however is that they can be fabricated much more easily and cheaply via solution processable methods.

The autocorrelation trace and its corresponding laser spectrum are shown in Fig 6.1.6. It has a FWHM of about 11.7 ps assuming that the mode locked pulses have a Gaussian intensity profile, this corresponded to a pulsewidth of about 8.3 ps. The optical spectrum of the pulses has a FWHM spectral bandwidth of about 0.21 nm, centered at 1064.4 nm. Therefore the TBP was found to be 0.46, indicating that they are near Fourier transform - limited. When mode-locked, with a SESAM, the laser generated had a Fourier transform-

limited pulse width of 25 ps. The SWCNT SA produced lesser power but much shorter pulses compared to the SESAM. The shorter pulses maybe attributed to a larger ΔR in the SWCNT SA. This shows that SWCNT SAs fabricated using PFO assisted purification are competent mode lockers.

Fig. 6.1.5.: Slope efficiency curve of ceramic Nd: YAG with SWCNT as the SA. The broken vertical lines indicate the different modes of operation the laser undergoes.

Fig. 6.1.6. (a) The autocorrelation trace of the mode locked pulses, where solid circles are experimental points while the solid line is a Gaussian fit. (b) The corresponding optical spectrum.

Fig. 6.1.7.: Schematic of the open aperture z scan setup used to measure the saturation absorption of SWCNT coated quartz substrates.

Z-scan of (7, 5) SWCNT SAs

Open aperture z scans were conducted to measure the saturable absorption properties of the SWCNT films fabricated. The SWCNT coated quartz substrates were placed into the Z-scan setup as shown in Fig. 6.1.7. The laser used to conduct the Z scan was from a home built laser emitting at 1040 nm with pulse widths of about 500 fs with a repetition rate of about 90 MHz. The output of the probing laser was controlled by rotating the optical axis of the $\frac{\lambda}{2}$ plate to change the polarization of the pump beam. The optical axis of the polarizer was fixed throughout the experiment. The polarizer used was a polarizing cube. The output power of the probing beam impinging on the sample was controlled to be at 1.42 mW and was focused down to a diameter of spot size of 11 μm by a converging lens at the focal point.

Fig 6.1.8.: Transmission of a SWCNT coated quartz substrate with increasing pump intensity. Solid circles are experimental points while the solid line is a mathematical fit obtained using (6.1.1).

As the SWCNTs SAs have semiconducting properties, the author assumed that its saturation properties were similar to that of a SESAM. A two level saturation absorption model similar to that adopted in ref. [18] was adapted such that the data obtained from the open aperture z scan of the SWCNT coated quartz substrates were fitted to:

$$T = A \text{Exp}\left(-\frac{B}{1+\frac{I}{I_{\text{sat}}}}\right) \quad (6.1.1)$$

where T is the absolute transmission, A is the normalizing constant, B is the ΔR of the SWCNT SA, I is the intensity and I_{sat} is the saturation intensity. Fitting of the Z-scan data resulted in Fig. 6.1.8 in which A was 0.945, B was 6 % and I_{sat} was 1.7 MW cm^{-2} . Fig 6.1.8 also shows that the ΔR_{ns} of the SWCNT SA were about 5.5 %. Compared with the mode locking performance of the laser mode locked with the SESAM, the lower average output power and shorter pulse width obtained with the SWCNTs can be attributed to the larger ΔR_{ns} and ΔR of the current SWCNT SA [19] respectively. Although drop casting is a simple method for thin film fabrication, it is a relatively crude way of fabricating thin films. In particular, the uniformity of the film is difficult to

control and usually leads to poor quality film which is most likely the origin of the non-saturable losses. It maybe better to use techniques such as spin coating to fabricate higher quality films such that the non-saturable losses can be further reduced.

The authors also note the work of Mart ín-Ror íguez [17] *et.al* in which they studied the conditions required in order to suppress QML in ceramic Nd: YAG. Although a SA with large ΔR would generally result in QML in ceramic Nd: YAG lasers but in the case of SWCNT SAs, the saturation intensity/energy required to induce transparency in the SA used is about seven times smaller than those of commercial SESAMs [20]. This offsets the larger ΔR as shown in equation (3.3.3) and allow SWCNTs to suppress QML [5].

Conclusions

In conclusion, a diode-pumped ceramic Nd: YAG laser mode locked with single species semiconducting (7, 5) SWCNT was demonstrated. The laser mode-locked with the as fabricated SWCNT SA produced an average output power of 130 mW with pulses as short as 8.3 ps in an uncompensated setup. A maximum output power of 275 mW could be obtained under higher pump powers but multi pulsing could not be suppressed. Z scan studies of SWCNT SA showed that it had a modulation depth of 6 %, a saturation intensity of 1.7 MW cm^{-2} and a non-saturable loss of about 5.5 %. The saturable losses in the sample was attributed to poor film quality due to the drop casting method which could be improved by using spin coating instead. The results presented is also the first demonstration of mode-locking using a single species of (7, 5) SWCNT to initiate mode-locking. This demonstration opens up two possibilities: (a) the selective usage of a particular species of SWCNT to tailor to the operating wavelength of the SWCNT SA and (b) to use a mixture of single species semiconducting SWCNTs to extend the operating range of SWCNT SAs.

Fig 6.2.1.:Saturable absorption process in graphene. (a) Excitation processes in graphene under low intensity illumination. (b) At high intensity illumination, Pauli blocking results in saturable absorption. [4]

6.2.: Mode-locking of ceramic Nd: YAG with graphene

Graphene is a two dimensional material that contains only a single layer of carbon atoms. Due to its unique electronic structure, graphene possesses many unique optical properties. It was found that a single layer of graphene absorbs about 2.3 % of the light incident on it and that each additional layer adds another 2.3 % to the overall absorption [21]. This coupled with the fact that graphene has no bandgap [22], implies that its absorption is independent of light wavelength. In addition, pump probe studies on epitaxial graphene revealed that it has saturable absorption with ultrafast recovery times [2]. The implication of these results is that graphene could be used as a wavelength insensitive SA. Indeed Zhang *et al* [23] and Bao *et al* [4] have recently demonstrated mode-locking of an erbium-doped fiber laser at about 1550 nm using graphene as a SA. When graphene is exposed to high intensity illumination, the states in both the conduction and valance bands near the Dirac point will be filled by the electrons and holes. Pauli blocking then prevents electrons in the valance bands to be further excited, thus achieving saturated absorption and high transparency [4] as shown in Fig. 6.2.1.

As mentioned in the previous section, the mode-locking of SSLs is currently dominated by SESAMs. Although SESAMs offers robust and good performances, these devices are usually difficult and expensive to fabricate [5]. In addition, SESAMs usually

have an operating range that is limited to tens of nanometers. To operate at a new wavelength would require the replacement of the whole device. On the other hand, SWCNTs could be easily and cheaply fabricated using wet chemistry methods making them an economical alternative to the SESAM. Although the operating wavelengths of SWCNTs maybe extended by mixing semiconducting SWCNTs of different, this method is only applicable if the particular species of SWCNT with the desired absorption wavelength exists [14]. Alternatively, one could also use graphene as a SA. Graphene can also be solution processed [24] like SWCNTs and coated onto different types of substrates, resulting in lower fabrication costs. In addition, the number of layers of graphene deposited onto a piece of substrate can be controlled thus varying the amount of light absorbed and the absorption modulation depth of the SAs. The number of layers of graphene can be controlled via controlling the time and number of passes in which the airbrush is swept across the substrate while dispensing graphene oxide solution. In addition, graphene being a zero bandgap material can function as a saturable absorber over a wide range of wavelengths without having to tune any material parameters. This would potentially make graphene a wavelength insensitive SA as it could function from the visible to far infrared. This in a sense would make the processing of graphene in a SA even easier than that of SWCNT. Finally, graphene being a two dimensional material does not form bundles like SWCNTs. Reduction of scattering losses is critical in SSL systems than in fiber lasers as the latter tend to have much larger gain to overcome losses introduced into the system.

Fig 6.2.2.: Flow chart showing the fabrication process of graphene SAs.

Fig 6.2.3. : (a) Optical microscopy and (b) AFM images ($10 \times 10 \mu\text{m}^2$) of the as fabricated graphene SAs.

Graphene SA Fabrication

The graphene SA used to mode-lock ceramic Nd: YAG was fabricated using wet chemistry and solution processable methods. Graphene oxide (GO) was first fabricated from graphite flakes using a variation of Hummer's method [25]. The graphene SAs were then fabricated by spray coating a piece of quartz heated on a hotplate with GO solution using an air brush method. The temperature of the hotplate was maintained at 80 °C during the coating process. The number of layers of graphene formed was controlled by the number of times that the quartz was coated using the air brush. It was found that three coatings of graphene oxide yielded the best mode locking results. To obtain graphene from GO, a reduction reaction was conducted. To initiate this reaction, the GO coated quartz was sealed in a bottle with 200 µl of hydrazine (98%) (N_2H_4) and heated at 50 °C for 24 hours. The reduction process was completed by transferring the quartz into a furnace and annealed at 400 °C for three hours. A 20% H_2/Ar laminar airflow was blown over the surface of the quartz during annealing to facilitate the reduction process. Fig. 6.2.2 shows a flow chart that summarizes the fabrication process. It should be noted that this method of fabrication a graphene SA is a variation of the method reported in ref. [24].

Fig. 6.2.3 shows the optical microscopy and the atomic force microscopy (AFM) images of the as fabricated graphene samples. It is clear that by reducing GO to form graphene, one could obtain graphene flakes that cover a wide area of the quartz substrates. However evident from Fig 6.2.3 is that this method of fabricating graphene results in non-uniform thickness samples to be obtained. In addition, some of the graphene flakes have dimensions that are micrometers in size and as such comparable to the laser wavelength. This would result in scattering losses. With recent progress made in graphene fabrication techniques, it is now possible to fabricate graphene flakes using wet chemistry/solution processable methods with much larger dimensions [25]. Larger dimension graphene samples would reduce scattering losses and improve laser performances. Unfortunately, these techniques for large dimension graphene were not available at the time the experiment was carried out. Fig. 6.2.3 also reveals that the graphene SAs were multi-layered. This shows that multi-layer graphene were used.

Fig. 6.2.4.: Raman signature of graphene obtained from reducing graphene oxide.

Fig. 6.2.5.: Linear absorption of graphene coated onto a piece of quartz substrate.

Raman and UV-VIS-IR spectroscopy of graphene samples

Raman spectroscopy was carried out on the as-fabricated samples to detect the graphene Raman signal. A typical Raman trace is shown in Fig 6.2.4. The D, G and 2D bands appeared at about 1550 cm^{-1} , 1600 cm^{-1} and 2700 cm^{-1} , respectively, which are the signature graphene Raman peaks. The detection of these signature peaks confirms the existence of graphene on the quartz substrates and that GO has indeed been reduced to form graphene. In addition, Fig. 6.2.5 shows the linear absorption of the graphene sample conducted using UV-VIS-IR spectroscopy. Absorption exists around the wavelength of 1064 nm. Due to the zero bandgap of graphene, the linear absorption is featureless and extends from the visible to the mid IR region. As such graphene can potentially serve as a SA for different lasers whose wavelength locates in this region. The kink observed at around 800 nm is due to the detector in the UV-VIS-IR spectrometer changing to one capable of detecting longer wavelengths.

Fig 6.2.6.: Mode-locked pulses trains at (a) nanosecond and (b) microsecond time scales.

Fig. 6.2.7: (a) Autocorrelation trace of the mode locked pulses. Closed circles are data points and the solid line is a Gaussian fit. (b) The corresponding optical spectrum.

Mode-locking experiments

The mode-locking of ceramic Nd: YAG with graphene was conducted using the same cavity as when ceramic Nd: YAG was mode-locked with SWCNTs and is shown in Fig. 6.1.3. The only difference was that M4 was replaced with a HR mirror at the laser wavelength and that L3 was 5.5 cm. The experimental conditions were kept the same as when ceramic Nd: YAG was mode-locked with SWCNTs. The graphene SA was attached onto a high precision three dimensional stage and carefully inserted along L3. ABCD analysis of the cavity showed that the laser spot size with the gain medium was 100 μm in diameter and the diameter of the spot size on the graphene SA was estimated to vary between 100 μm to 140 μm . Determination of mode locking was done by coupling laser light leaking out of M2 into a high speed detector (New Focus, 1611 IR 1GHz) connected to a 1 GHz bandwidth oscilloscope (Tektronix DPO 7104). The mode locked pulse duration was measured using a commercial non-collinear autocorrelator (Femtochrome Research, FR-103XL). The optical spectrum was obtained using a high resolution optical spectrum analyzer purchased from Ando (AQ-6315B) while the pulse width was measured using autocorrelation techniques (Femtochrome Research Inc., FR-103 XL). All optical powers were measured using power meters and detectors from Gentec-e (Solo PE)

Fig. 6.2.6 (a) shows the mode-locked pulse train at the nanosecond time scale. It clearly shows that the pulse to pulse separation is about 11 ns which corresponded to the cavity repetition rate. Hence fundamental mode-locking have been achieved in ceramic Nd: YAG using graphene as the SA. Fig. 6.3.6 (b) shows the mode-locked pulse at the

microsecond time scale. It shows that the mode-locked pulse train obtained was not that due to Q-switched mode-locking. Unfortunately, the author's oscilloscope lacked the required statistical software and as such the pulse to pulse stability could not be analyzed. Fig. 6.2.7 (a) and (b) shows the autocorrelation trace and mode-locked laser spectrum respectively. Assuming a Gaussian shaped pulse, the pulse width obtained was about 4 ps while the mode-locked laser spectrum was centered about 1064.4 nm and had a laser bandwidth of 0.42 nm. The TBP was about 0.442 indicating that the pulses obtained were near the transform limit. The results obtained demonstrate that graphene can be a competent mode-locker for SSL systems. When mode-locked, the laser generated 100 mW of output power. To the author's knowledge, the demonstration of the mode-locking of ceramic Nd: YAG with graphene is the first demonstration of a SSL being mode-locked by graphene. However the author would like to comment that prolonged operation of the graphene mode-locker results in the graphene to breakdown probably due to intense heat. To overcome this problem and improve the reliability of graphene mode-locker would require improvements in its fabrication process. However the purpose of this experiment was mainly a proof of concept to show that graphene can be used to mode-lock SSLs around the 1 μm regime.

The results obtained in this work complement that of Bao *et. al.* [4] and Zhang *et. al.* [23] who also used graphene SAs to mode-lock fiber lasers at 1.55 μm thus showing that graphene can be used as a broadband SA due to its zero bandgap and as such exhibit a featureless absorption curve as shown in Fig. 6.2.5. It is worth pointing out recently, Xu *et. al.* [26] and Sun *et. al.* [27] successfully demonstrated the mode-locking of Nd: GdVO₄ and Nd: YVO₄ respectively using graphene as the SA and producing 350 mW and 1 W of output power. The output powers demonstrated by these groups were very much higher than that obtained in this work. It is very likely that in these experiments, they were able to produce graphene SAs of higher quality which has lower non-saturable losses. This is most likely to be the case in the work of Xu *et. al.* [26] who was able to produce graphene flakes with dimensions of greater than 20 μm . Flakes of such dimensions are very much greater than the laser operating wavelength and as such would have lower scattering losses which lead to lower non-saturable losses. In addition, it is most likely that the graphene sample in their work has less lattice defects than the ones used in this

work. It is known that lattice defects in graphene are common in samples fabricated using wet chemistry methods.

Z-scan of graphene SAs

The as fabricated graphene SAs coated onto quartz substrates were subjected to open aperture Z-scans as in the case of (7, 5) SWCNT coated substrates. The setup used was the same as the one used in the open aperture Z-scan of the SWCNT substrates and as such the experimental setup is shown in Fig. 6.1.7 can be referred to. The probing laser used was also the same as the one used to probe the SWCNT coated substrates. The probing laser was a home built oscillator mode-locked at 1040 nm. Its output power was controlled using the $\frac{\lambda}{2}$ waveplate and the polarizing cube as shown in Fig. 6.1.7. The output power was controlled to be 1.14 mW. All output powers measured in the Z-scan of graphene SAs were conducted using a silicon based detector for fast response. It is important that a thermopile detector is not used for these measurements as the response times of such detectors are slow and the accuracy tends to be lower. The saturable absorption mechanism presented by Bao *et. al* [4] suggests that the saturable absorption properties of graphene can be modeled by a two level atom model such as in the case of SWCNTs. To that end, the Z-scan results obtained were also fitted with equation (6.1.1).

Fig. 6.2.8: Transmission of graphene with increasing pump intensity. Closed circles are data points while the solid line is the fitting obtained from equation (6.6.1).

Fitting of the Z-scan data with equation (6.1.1) resulted in Fig 6.2.8, which yielded the values of A , B and I_{sat} as 0.94, 11 % and 0.5 M Wcm^{-2} respectively. These results compare well with those of Bao *et. al.* [4]. The value of A implies that the ΔR_{ns} induced by the graphene sample were about 6 %. The ΔR_{ns} induced were attributed to multiple layers of graphene taking part in the saturable absorption process and scattering losses. The non-saturable losses should be reduced if better performance if desired with graphene SAs. This could be done growing graphene of better quality and larger dimensions as demonstrated by Xu *et. al.* [26]. One way of growing larger dimension graphene would be to use chemical vapor deposition (CVD) methods. Using this technique, Bae *et. al.* [29] grew graphene of thirty inches in dimension. Moreover, graphene grown by CVD method suffer from less lattice defects than those grown by wet chemistry methods and are expected to be of higher quality. However a more effective way to reducing the non-saturable losses would be to reduce the number of graphene taking part in the saturable absorption process as each layer can absorb up to 2.3 % of the incident light [22]. Ideally, a single layer of graphene should be used to initiate the mode-locking. Lee *et. al.* [28] showed that using a single layer of graphene it was also

possible to initiate mode-locking in SSL systems. The shorter pulse width obtained could be understood from the large modulation depth of 11 % for the graphene SA used in this work. A larger normalized modulation depth would result in shorter mode locked pulses [19]. As in the case with (7, 5) SWCNTs, QML is avoided due to the smaller saturation intensity/energy required to induce transparency and this offsets the higher ΔR_{ns} shown in equation (3.3.3).

Conclusion

The mode-locking of ceramic Nd: YAG with graphene as the SA was demonstrated for the first time. This is also the first SSL mode-locked using graphene. When mode-locked, the laser generated 4 ps pulses with an output power of about 100 mW. This demonstration shows that graphene can be a competent mode-locker for SSLs at the 1 μm regime.

6.3.: Concluding remarks

In conclusion, the mode locking of ceramic Nd: YAG have been demonstrated for the first time using single species semiconducting (7, 5) SWCNTs and graphene as the SA. To the author's knowledge, the work presented in this thesis this is also the first demonstration of mode-locking in SSLs using these type of novel SAs. In the case of semiconducting (7,5) SWCNTs SA, the demonstration of mode-locking demonstrates the possibility of using singles species SWCNTs to initiate mode-locking in a SSL and opens up the possibility of mixing different species of semiconducting SWCNTs to extend the operating wavelengths. In addition, graphene SA, the demonstration of mode-locking at 1064 nm demonstrates also that graphene has a wide operating wavelength range and can function as a wavelength insensitive mode-locker. Both novel SAs were subjected to open aperture Z-scans and were found to demonstrate saturable absorption properties. For better laser performances however, the ΔR_{ns} of the as fabricated SAs would have to be reduced by improving the fabrication process of the samples. Finally, for comparison purposes, the shortest pulse width possible for Nd: YAG is typically limited by the gain bandwidth which is about 0.6 nm and as such the shortest possible pulse width obtainable is about 2.7 ps.

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Chapter 7

Conclusions

7.1.: Thesis summary

The lasing performances of Nd: GdVO₄, Yb: SSO and Yb: CYA were investigated. These gain media were investigated using fiber coupled or free space diodes in V-shaped or X-folded cavities while mode-locking was initiated using commercial SESAMs. In addition the saturable absorption properties of semiconducting (7, 5) SWCNTs and graphene as potential replacement of the SESAM in SSLs were also investigated using ceramic Nd: YAG as the gain media in an X-folded cavity. Certain parameters related to their saturable absorption properties were measured using a Z-scan setup.

Dual-wavelength mode-locked Nd: GdVO₄

For the work presented using Nd: GdVO₄, the author demonstrated that by exploiting the birefringence and thermal lensing of the gain media, the mode-locked wavelengths can be selectively chosen. The author showed that it was possible for the laser to mode-lock at 1065 nm with σ polarization at low pump power. Upon increasing the pump power to moderate levels, the laser would simultaneously mode-locked at 1065 nm with σ polarization and at 1063 nm with π polarization. At high pump power, the laser would only mode-lock at 1065 nm. A simple explanation was provided in terms of mode-matching between the pump and lasing modes of 1063 nm and 1065 nm to explain the selective lasing of Nd: GdVO₄ at those wavelengths. To the author's knowledge, this is also the first demonstration of mode-locking of Nd: GdVO₄ at 1063 nm and simultaneously at 1063 nm and 1065 nm. Such a laser might find uses in laser interferometry, where a laser with dual wavelengths and orthogonal polarizations is required [1]. In addition, as the frequency separation for Nd: GdVO₄ is in the THz region, the laser scanning speeds in laser interferometers can be increased significantly [1].

Laser performances of Yb: SSO and Yb: CYA

The mode-locking of Yb: SSO and Yb: CYA in both the positive and negative dispersion regimes were investigated in this thesis. In the negative dispersion regime, solitary mode-locking of Yb: SSO was achieved. The mode-locking was initiated close to zero cavity dispersion at 1041 nm. This was confirmed via the observation of a satellite peak on the edge of the principal peak in the mode-locked spectrum as shown in the theoretical works of Herrmann *et. al.*[2] for KLM Ti: Sapphire laser. The satellite peak manifests due to TOD becoming more important when GVD is well compensated. Under this configuration, the laser emitted 145 fs pulses at 1041 nm with a spectral FWHM of 8 nm. The TBP was 0.32 and the laser emitted 40 mW of output power. Upon decreasing the negative dispersion in the system such that the laser operates even closer to zero dispersion, the laser generated 93 fs pulses at 1044 nm with a spectral FWHM of 12.2 nm. The TBP was 0.316 and the output power was about 20 mW. This mode-locked laser spectrum however exhibited two satellite peaks on either sides of the principal spectrum. Herrmann *et. al.*[2] showed that this was due to the FOD effects becoming dominant. To the author's knowledge, this is the first observation of FOD effects observed in any Yb³⁺ doped SSL. In the addition, the author also demonstrated the solitary mode-locking of Yb: SSO at 1060 nm and 1077 nm generating near transform limited pulse widths of 144 fs and 125 fs respectively. The output powers generated at 1060 nm and 1077 nm were 50 mW and 100 mW respectively. The solitary mode-locking of Yb: CYA was conducted at a large net negative dispersion with the crystal orientated in the π direction. This resulted in the laser to generate near transform limited pulses of 156 fs duration at 1045.3 nm with an output power of 740 mW. This laser is highly efficient for a femtosecond mode-locked laser and has a $\eta_{\text{femtosecond}}$ of 37 % and an η_{opt} of 20 %. To generate even shorter pulses using Yb: CYA, the crystal can be oriented in the σ direction to exploit the broadband plateau in the emission spectrum. Nevertheless, under solitary mode-locking conditions, both Yb: SSO and Yb: CYA demonstrated that they are excellent femtosecond light sources.

The mode-locking of Yb: SSO and Yb: CYA were also investigated under positive dispersion mode-locking. In the case of Yb: SSO, a highly chirped pulse was obtained in which the mode-locked spectrum exhibited very step edges that resemble that of a

rectangular. This deviates from the traditional Gaussian shaped spectrum expected from normal dispersion mode-locking. The mode-locked spectrum had a FWHM of 7.5 nm centered at 1063 nm while the pulses generated had a pulsewidth of 2.7 ps. This implies a TBP of 5.4 which was 17 times above the transform limit indicating that the pulses are highly chirped. The output powers of about 120 mW. Such observations provide strong evidence of the existence of DSs in Yb: SSO [3-5]. Similarly, in the positive dispersion mode-locking of Yb: CYA, DSs were also observed as the mode-locked spectra resemble that of a rectangular and were highly chirped. With $T = 0.8\%$, the Yb: CYA DS laser generated 7.4 ps pulses with a rectangular mode-locked spectrum centered at 1057 nm with a spectral FWHM of 13 nm. The corresponding TBP was 26 which was 80 times above the transform limit indicating that the pulse was highly chirped. The laser emitted a maximum mode-locked power of 0.29 W. When compressed with a pair of extra-cavity gratings, pulses as short as 340 fs were obtained. With $T = 1.6\%$, the Yb: CYA DS laser generated 8.5 ps pulses with a rectangular mode-locked spectrum centered at 1055 nm with a spectral FWHM of 8 nm. The corresponding TBP was 19 which was 60 times above the transform limit indicating that the pulse was highly chirped. The laser emitted a maximum mode-locked power of 1.5 W. When compressed with a pair of extra-cavity gratings, pulses as short as 380 fs were obtained. To the author's knowledge, the observation of DSs in Yb: SSO and Yb: CYA represent the first experimental observations of DSs in and Yb³⁺ doped SSL. The results obtained also indicate that DSs are an accessible operation regime of Yb³⁺ doped SSLs. By operating Yb³⁺ doped SSL lasers, one can obtain femtosecond pulses at higher output powers as demonstrated in the case of Yb: CYA. This is because the pulses are chirped everywhere within the cavity [5] and as such the threshold peak intensity required for pulse breakup and distortion is not reached. In addition, the pulses are already strongly chirped and as such do not require a stretcher when amplifying the pulses. This can reduce the cost and complexity of amplifier systems.

The CW performances of Yb: SSO and Yb: CYA were also investigated. Under CW configuration, the Yb: SSO laser generated a maximum output power of 2.7 W at an absorbed pump power 4.6 W. The η_{CW} was 70 %. Under CW configuration, Yb: CYA generated a maximum output power of 2.3 W under an absorbed pump power of 3.25 W.

The η_{CW} was 92 %. The CW results obtained demonstrate that both Yb: SSO and Yb: CYA are highly efficient gain media for SSLs.

Saturable absorption properties carbon based nano-materials

Finally, the saturable absorption properties of carbon based nano-materials such as semiconducting (7, 5) SWCNTs and graphene were investigated by using them in place of a SESAM and subjecting them to Z-scan measurements. Both carbon based nano-materials were used as SAs in an X-folded cavity to mode-lock a piece of commercial ceramic Nd: YAG.

When mode-locked with semiconducting (7, 5) SWCNTs, the laser generated 8.3 ps pulses at 1064.3 nm with a maximum output power of 130 mW. Z-scan investigations reveal that the sample had a non-saturable loss of 5.5 %, a modulation depth of 6 % and a saturation intensity of 1.7 M W cm^{-2} . When mode-locked with graphene, the laser generated 4 ps pulses at 1064.4 nm with a maximum output power of 100 mW. Z-scan investigations reveal that the sample had a non-saturable loss of 6 %, a modulation depth of 11 % and a saturation intensity of 0.6 M W cm^{-2} . The results obtained showed that both with semiconducting (7, 5) SWCNTs and graphene are competent mode-lockers for SSLs.

7.2.: Future work

Although some interesting results have been demonstrated in this thesis, there is still scope for further work and experiments for the improvement and development of the laser systems demonstrated. The recommendations for future work are as below:

- In the case of Yb: SSO, a comparison of the results obtained and the emission spectrum of the gain medium show that solitary mode-locking of the laser at longer wavelengths would be advantageous. This is because of the existence of an emission peak at 1087 nm which is relatively broad but more importantly, the curvature at the top of this peak is much smoother than those located at 1036 nm and 1062 nm. In addition, reabsorption is negligible close to the 1087 nm peak. It would be interesting to operate the laser near the vicinity of zero dispersion to

obtain the shortest possible pulsewidth in which the author speculates would be shorter than 93 fs. The laser can then be operated at a much larger net negative dispersion to obtain larger output powers at the expense of longer pulsewidths.

- In the case of Yb: CYA, it would be interesting to investigate the solitary mode-locking with the crystal orientated in the σ direction. This is done to exploit the broadband plateau observed in the polarized emission spectrum. As the emission is flat, it is likely that very short pulsewidths could be obtained. As in the case of Yb: SSO, it would be interesting to measure the pulsewidth when the laser is operated close to vicinity of zero dispersion and at a larger net negative dispersion. The author speculates that high output power with sub 100 fs pulsewidth maybe possible with the crystal orientated in the σ direction.
- Further work in the DS mode-locking of Yb: SSO and Yb: CYA should be conducted. Chong *et. al.* [4] showed that DSs in fiber lasers manifest due to the interplay GVD, SPM and filter bandwidth. Clearly in SSLs, the amount of GVD is different from that in fiber lasers by many orders of magnitude and that the magnitude of the SPM is very different as well. In fact the magnitude of the SPM in SSLs is very much dependent on cavity design and that the intracavity powers in SSLs are very much higher than in fiber systems. These effects in SSLs can be modeled using the CQGLE [4] to better understand pulse evolution and as such design SSLs that can better exploit the phenomena of DSs.
- More work can be done in the area of carbon based nano-materials SAs. Particularly these SA can be used to mode-lock Yb^{3+} doped gain media to generate femtosecond pulses. However, the non-saturable losses would have to be significantly reduced first. In the case of SWCNTs, this can be done by using spin coating techniques to fabricate uniform films. For graphene SAs, it would be better to grown large dimension single layer graphene on HR mirrors to reduce the non-saturable losses.
- Finally it would be interesting to use carbon based nano-materials to mode-lock SSLs operating at the 2 μm spectral region. For SWCNTs, this can be achieved by mixing semiconducting nanotubes with chiralities that absorbs strongly in this region. In the case of graphene, an as fabricated sample can be used directly to

mode-lock a laser whose operating wavelength locates at 2 μm without any additional modifications to the graphene SA.

7.3.: References

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