

Extending the wavelength coverage of frequency-doubled fibre lasers

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For Teddy and my parents

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Timothy Runcorn

Abstract

The unique coherence properties of laser light has revolutionised modern life through fields as diverse as materials processing, optical communications and scientific research. Despite the enormous engineering progress made in developing laser systems into tools used in a wide range of applications, there are still many spectral regions where suitable laser sources are demanded. This thesis describes progress made towards developing practical and efficient nonlinear frequency conversion techniques to access some of these spectral regions, whilst exploiting the inherent benefits that master oscillator power fibre amplifier (MOPFA) systems offer.

In particular, two of the developed sources were designed to satisfy the depletion beam requirements for stimulated emission depletion microscopy using the green fluorescent protein. The sources operated at 560 nm and were based on a frequency-doubled Raman amplifier that was pulse-pumped by an ytterbium MOPFA system. The first source developed proved that this architecture was effective at generating picosecond pulse trains at megahertz repetition rates with nanojoule pulse energy in a compact and alignment-free system. The second source improved on the design by adding temporal flexibility, increasing the efficiency and pulse energy and demonstrated that it was a completely turn-key solution.

The third source extended the frequency-doubled pulse-pumped Raman amplifier concept to operation around 590 nm by utilising cascaded Raman amplification, whilst maintaining similar output characteristics. The final source developed operated around 780 nm and was based on direct frequency-doubling of an erbium MOPFA system. It was designed for subsequent nonlinear frequency conversion and featured high average power with flexible wavelength and temporal characteristics. Watt-level average powers and pulse energies of hundreds of nanojoules were obtained at every wavelength region, demonstrating that the implemented nonlinear frequency conversion techniques are a viable method for extending the wavelength coverage of MOPFA systems.

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1 Introduction

We interpret the world around us predominantly from light in the visible spectral region, between 380–780 nm by the broadest definition [Sch87]. The unique coherence properties of lasers, first demonstrated in 1960 [Mai60], has led them to become indispensable tools for probing and modifying matter. It is unsurprising, therefore, that many different fields, as diverse as medicine, scientific research and materials processing, demand laser radiation in the visible spectrum. Every application has specific constraints on the wavelength, power and temporal characteristics of the laser source and in spite of the plethora of laser media, many applications are not currently satiated.

In contrast to the near-infrared (NIR) spectral region, there are few visible laser sources that are flexible in wavelength, power scalable and temporally versatile. Dye lasers were once thought the most promising candidate, displaying excellent wavelength tunability [Joh82], capability for extremely high average powers [Bas92] and femtosecond [Val85] to continuous-wave (CW) [Bav82] operation with high efficiencies. Arguably, their performance and flexibility are unrivalled amongst laser gain media and yet present-day deployment is restricted to a handful of niche applications, due to the high maintenance and expertise required to operate them. This serves to highlight that the practical aspects of laser sources - efficiency, compactness and easy of use - are as important as the performance properties of the laser medium if they are to be useful in a broader range of applications.

The desire to satisfy the application-driven demand for laser sources in the visible and near-visible spectral region has fueled the extensive research effort into the use of nonlinear propagation of light to convert the frequency of commercially mature NIR lasers. Since the first demonstration shortly after the laser [Fra61], the rapid progress of nonlinear frequency conversion has coincided directly with the pace of development of laser sources. The concomitant increase in nonlinear conversion efficiency from $\sim 10^{-7}$ to close to unity has enabled the properties of practical and versatile NIR laser sources to be enjoyed in the visible and near-visible. Fibre lasers are an ideal platform for nonlinear conversion due to their power scalability, excellent beam quality and temporal flexibility. A key technological problem is, therefore, extending the wavelength coverage of frequency-converted fibre lasers beyond existing systems using practical techniques.

1.1 Overview

This thesis is organised as follows: Chapter 2 discusses the applications of laser sources in the yellow-green (550–600 nm) spectral region and reviews the existing sources of direct laser emission. Stimulated emission depletion (STED) microscopy using green fluorescent protein (GFP) is then introduced, with a particular attention paid to the requirements of the depletion beam source. The concept of using Raman amplification to extend the wavelength coverage of ytterbium master oscillator power fibre amplifier (Yb-MOPFA) schemes and the design steps necessary to facilitate efficient frequency-doubling into the yellow-green spectral region are subsequently discussed. Following that, the development of a fibre-integrated source of picosecond pulses operating at 560 nm, based on a frequency-doubled, pulse-pumped Raman amplifier is detailed.

Chapter 3 seeks to improve on the system design introduced in Chapter 2 by examining potential seed sources to increase the flexibility in the 560 nm pulse parameters. Significant infrastructural improvements are implemented, which results in a highly efficient, turn-key source of duration-tunable pulses at 560 nm. In Chapter 4, the pulse-pumped Raman amplifier concept is extended to a longer wavelength region using cascaded Raman amplification. The performance of different cascaded Raman amplifier designs is contrasted and frequency-doubling is subsequently used to produce pulsed yellow radiation at 590 nm. In Chapter 5, direct frequency-doubling of a wavelength-tunable erbium master oscillator power fibre amplifier (Er-MOPFA) system is performed, resulting in high average power, wavelength and pulse-duration tunable radiation around 780 nm. Finally, Chapter 6 provides a brief overview of the systems described in this thesis and the potential future developments of these sources.

The rest of this introductory chapter provides an overview of the linear propagation of light in free-space and optical fibres, light amplification in rare-earth doped optical fibres and the nonlinear propagation of light in optical fibres and nonlinear crystals.

1.2 Electric field propagation in dielectric media

1.2.1 Linear propagation

Maxwell's equations govern the propagation of electric fields in materials. For a dielectric medium such as silica, which is non-magnetic and has no free charges or current, the wave equation for a net electromagnetic field $\mathbf{E}$ is given by:

$$\nabla^2 \mathbf{E} - \mu_0 \epsilon_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}}{\partial t^2} \quad (1.1)$$

where ϵ_0 and μ_0 are the permittivity and permeability of free space respectively and $\mathbf{P}$ is the induced polarisation. This equation shows that the electric field acts a driving force which induces time-varying dipole moments. These re-radiate, coupling into the wave equation, leading to propagation of the electromagnetic wave through the medium.

The induced polarisation, the dipole moment per unit volume, can be expanded as a Taylor series in terms of the applied electric field:

$$\mathbf{P} = \epsilon_0 \left[\chi^{(1)} \cdot \mathbf{E} + \chi^{(2)} : \mathbf{E}\mathbf{E} + \chi^{(3)} :: \mathbf{E}\mathbf{E}\mathbf{E} + \dots \right] \quad (1.2)$$

where $\chi^{(j)}$ is the j^{th} order susceptibility and is a tensor of rank $j + 1$. Most optical fields applied to dielectric media are significantly weaker than the potentials that bind the electrons to the nucleus of atoms and therefore, the polarisation response can be considered linear with the driving field, i.e:

$$\mathbf{P} = \epsilon_0 \chi^{(1)} \cdot \mathbf{E} \quad (1.3)$$

Inserting Eqn. 1.3 into the wave equation (Eqn. 1.1) results in:

$$\nabla^2 \mathbf{E} = [1 + \chi^{(1)}(\omega)] \mu_0 \epsilon_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} \quad (1.4)$$

which has the solution of a travelling plane wave that propagates with a phase velocity given by:

$$v = \frac{1}{\sqrt{\mu_0 \epsilon_0 [1 + \chi^{(1)}(\omega)]}} = \frac{c}{n_0(\omega)} \quad (1.5)$$

where $c = (\mu_0 \epsilon_0)^{-1/2}$ is the speed of light in a vacuum and $n_0(\omega) = [1 + \chi^{(1)}(\omega)]^{1/2}$ is the linear refractive index.

All real dielectric media have absorption resonances, therefore, the amplitude of the dipole oscillations depends on the frequency of the optical driving field. This frequency dependence of the electric susceptibility results in a frequency-dependent refractive index, as is seen from Eqn. 1.5. The frequency-dependent refractive index results in chromatic dispersion: optical waves of differing frequency propagate with different phase velocities. Absorption and the refractive index are simply two manifestations of the complex nature of the electrical susceptibility: absorption results from the imaginary part and the refractive index from the real part. Furthermore, the Kramers-Kronig relations show that the real and imaginary parts of a complex function, with conditions that the electric susceptibility satisfies, are not independent and that the function can be reconstructed given just one of the parts.

If a polychromatic driving field is applied to a dielectric, the beating between the frequencies result in the formation of a wave packet, whose envelope propagates with a

group velocity given by:

$$v_g = \frac{\partial \omega}{\partial k} = \frac{c}{n_g(\omega)} \quad (1.6)$$

where $n_g(\omega)$ is defined as the group refractive index.

1.2.2 Gaussian beams

So far, only one-dimensional propagation of electric fields has been considered to facilitate simple interpretation of light propagation in medium. All nonlinear processes are dependent on the intensity of the electric field, however, and thus the transverse profile of the field is of prime importance. Therefore, we wish to find ‘beam-like’ solutions to the wave equation (Eqn. 1.1), that propagate in one direction with a known transverse profile. In order to simplify the derivation of a solution, we assume a homogeneous, isotropic and linear medium so that Eqn. 1.3 is valid with $\chi^{(1)}$ reducing to a single component. In this case, the wave equation (Eqn. 1.1) reduces to the Helmholtz equation:

$$\nabla^2 \mathbf{E} + k^2 \frac{\partial^2 \mathbf{E}}{\partial t^2} = 0 \quad (1.7)$$

where $k = n\omega/c = 2\pi n/\lambda$ is the wave number. Assuming propagation along the z-axis, the electric field can be separated into a transverse profile that varies with propagation and a travelling wave:

$$\mathbf{E}(x, y, z) = \mathbf{u}(x, y, z) \exp[(\omega t - kz)] \quad (1.8)$$

Inserting this into the Helmholtz equation (Eqn. 1.7), we obtain:

$$(\nabla^2 + k^2) \mathbf{E} = \left[\nabla^2 \mathbf{u} - 2ik \frac{\partial \mathbf{u}}{\partial z} \right] \exp[(\omega t - kz)] = 0 \quad (1.9)$$

We now invoke the paraxial approximation, assuming that the variation of the transverse profile is slow compared with λ and the z-dependence of the transverse profile is slow compared with the transverse variations [Sie86]. Using this approximation, Eqn. 1.9 simplifies to the paraxial wave equation in Cartesian and polar coordinates:

$$\begin{aligned} \left[\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right] \mathbf{u}(x, y, z) - 2ik \frac{\partial \mathbf{u}(x, y, z)}{\partial z} &= 0 \\ \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2} \right] \mathbf{u}(r, \theta, z) - 2ik \frac{\partial \mathbf{u}(r, \theta, z)}{\partial z} &= 0 \end{aligned} \quad (1.10)$$

Since any optical beam can be thought as a superposition of plane waves, the physical implication of the paraxial approximation is that so long as most of the plane wave components of the beam propagate with an angle less than 30° to the optic axis, then

no significant corrections to the paraxial wave approximation are necessary. This angle corresponds to a $\sim 1 \mu\text{m}$ focal spot size for a wavelength $\lambda = 1 \mu\text{m}$, so the approximation is valid for all the focusing condition used in this thesis.

There are many solutions to the paraxial wave equation (Eqn. 1.10), but the most relevant one to laser physics is a Gaussian solution of the form:

$$u(r, z) \propto \exp \left[-i \left(P(z) + \frac{kr^2}{2q(z)} \right) \right] \quad (1.11)$$

where it has been assumed that the beam has cylindrical symmetry, such that there is no θ dependence. Inserting this trial solution into the paraxial equation gives:

$$q(z) = z + iz_R \quad \text{and} \quad P(z) = -\ln \left[1 - i \left(\frac{z}{z_R} \right) \right] \quad (1.12)$$

where it assumed that $q(z)$ is complex to give a finite beam energy and z_R is the Rayleigh range. Therefore, the Gaussian beam solution becomes:

$$u(r, z) = \frac{u_0}{\sqrt{1 + (z/z_R)^2}} \exp \left[-\frac{kz_R r^2}{2(z^2 + z_R^2)} \right] \exp \left[-i \frac{kz r^2}{2(z^2 + z_R^2)} \right] \exp \left[i \tan^{-1} \left(\frac{z}{z_R} \right) \right] \quad (1.13)$$

Squaring this amplitude results in the radial beam intensity profile, given by:

$$\begin{aligned} I(r, z) &= \frac{I_0}{1 + (z/z_R)^2} \exp \left[-\frac{kz_R r^2}{(z^2 + z_R^2)} \right] \\ &= I_0 \left(\frac{w_0}{w(z)} \right)^2 \exp \left[-\frac{2r^2}{w^2(z)} \right] \end{aligned} \quad (1.14)$$

where w_0 is the beam waist, defined as the beam radial position at $z = 0$ where the intensity has fallen to $1/e^2$ of the central ($r = 0$) maximum value (I_0):

$$\begin{aligned} I(r = w_0, z = 0) &= I_0 \exp \left[-\frac{kz_R w_0^2}{z_R^2} \right] = I_0 \exp[-2] \\ \Rightarrow w_0^2 &= \frac{2z_R}{k} = \frac{\lambda z_R}{\pi n} \end{aligned} \quad (1.15)$$

and the $1/e^2$ beam radius, $w(z)$, is defined as:

$$w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_R} \right)^2} \quad (1.16)$$

Figure 1.1 plots the $1/e^2$ beam radius for a Gaussian beam as a function of position along the beam from the waist position ($z = 0$). It is clear that the beam waist w_0 corresponds

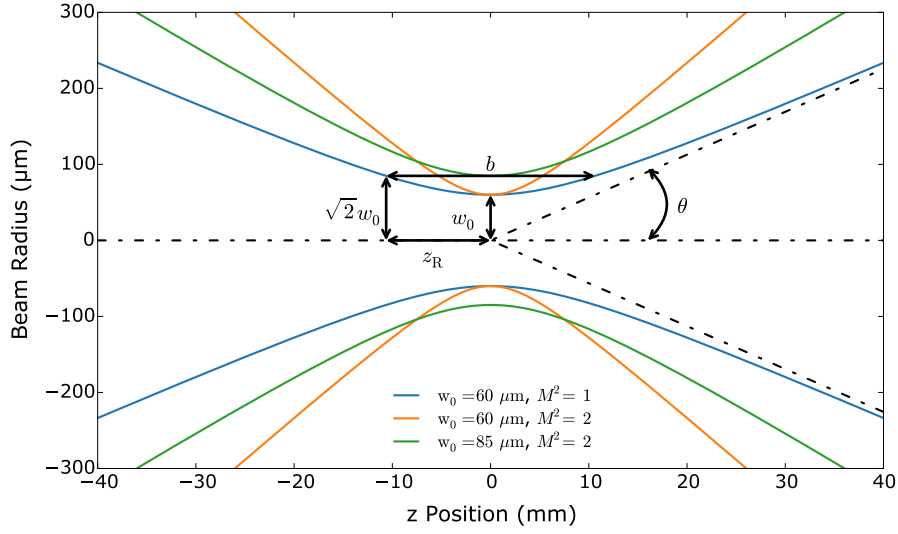


Figure 1.1: Beam radii of $\lambda = 1 \mu\text{m}$ Gaussian beams as a function of distance from the waist position ($z = 0$) with the beam waist radius w_0 , Rayleigh range z_R , confocal parameter b and far-field divergence θ labeled.

to the minimum beam radius and it is where the wavefronts have zero curvature. The Rayleigh range z_R corresponds to the distance from the waist position to where the beam radius has increased by a factor of $\sqrt{2}$ from the waist radius. The distance between the two points where the beam radius is $\sqrt{2}w_0$ is known as the confocal parameter and by definition is twice the Rayleigh range:

$$b = 2z_R = \frac{2\pi n w_0^2}{\lambda} \quad (1.17)$$

The confocal parameter is an important parameter when focusing Gaussian beams into nonlinear crystals, since it is the distance range where the intensity of the beam is approximately constant. When $z \gg z_R$ the beam radius tends to a linear increase with distance (Eqn. 1.16) and thus the intensity decreases with the square of the distance from the waist position. In the far-field, the divergence of the Gaussian beam (dotted line Fig. 1.1) is well approximated by a cone with a half-angle divergence given by:

$$\theta = \frac{\lambda}{\pi w_0} \quad (1.18)$$

The Gaussian beam solution (Eqn. 1.13) to the paraxial wave equation (Eqn. 1.10) is the lowest-order solution in an infinite family of higher-order solutions and is often denoted TEM_{00} . Higher-order modes have transverse intensity profiles that are polynomials with

terms up to $[1/w^2(z)]^{n+1}$, where n is the mode order. In systems that produce laser beams with Cartesian (rectangular) symmetry, the polynomials are Hermite polynomials and in systems with cylindrical symmetry they are Laguerre polynomials. In both cases, the n^{th} order mode has $(n + 1)$ intensity maxima in the transverse intensity profile.

The Hermite-Gaussian and Laguerre-Gaussian modes are vitally important in laser physics because they are also mode solutions of optical resonators that are used in bulk laser cavities. Due to its transverse intensity profile, the TEM_{00} (fundamental) mode has the lowest diffraction losses in idealised laser cavities and therefore is often the first to achieve laser oscillation [Kog66]. This is highly desirable since the TEM_{00} mode also has the smallest beam parameter product (BPP), the product of the waist radius and the far-field divergence. The BPP is important as it is directly related to the brightness (radiance) of a laser source – the power of a laser beam divided by the product of the mode area in the focus and the solid angle in the far-field.

Brightness is a key parameter of a laser source because it cannot be increased by passive optical systems or by combining incoherent beams. Due to this, almost all bulk lasers are designed to operate on the fundamental mode in order to maximise their brightness. It becomes increasingly more difficult, however, to achieve fundamental mode operation at higher laser output powers. When lasers emit on higher-order modes, the output beam quality degrades, which can be quantified by the M^2 dimensionless parameter:

$$M^2 = \text{BPP} \cdot \frac{\pi}{\lambda} = w_0 \cdot \theta \times \frac{\pi}{\lambda} \quad (1.19)$$

It is clear, using Eqn. 1.18, that $M^2 = 1$ for the fundamental mode; whereas, a higher-order Hermite-Gaussian mode TEM_{nm} , has an $M^2 = (2n + 1)$ and $M^2 = (2m + 1)$ in the x and y directions, respectively [Sie90].

At high output powers, real laser cavities have spatial inhomogeneities (e.g. due to thermal lensing) that mean the output beam rarely consists of only one higher-order mode. Instead, the output beam is often an incoherent superposition of several modes and is therefore referred to as multimoded. For such laser beams, it is not possible to use Eqn. 1.19 because the beam size, w_0 , cannot be defined as for the fundamental mode due to the complex transverse beam profile. Instead, the variance of the transverse profile in the spatial and spatial-frequency domain are used to quantify the beam width and far-field divergence [Sie90]:

$$\sigma_x^2(z) \equiv \frac{\iint (x - \bar{x})^2 I(x, y, z) dx dy}{\iint I(x, y, z) dx dy} \quad \text{and} \quad \sigma_{s_x}^2 \equiv \frac{\iint (s_x - \bar{s}_x)^2 \hat{I}(s_x, s_y) ds_x ds_y}{\iint \hat{I}(s_x, s_y) ds_x ds_y} \quad (1.20)$$

where $\bar{x}$ is the centroid of the beam profile in the x -direction, given by:

$$\bar{x} = \frac{\iint x I(x, y) dx dy}{\iint I(x, y) dx dy} \quad (1.21)$$

Equations 1.20 and 1.21 can similarly be defined for the y -axis.

For the TEM₀₀ fundamental mode, the intensity in the spatial and spatial-frequency (Fourier) domain are given by:

$$\begin{aligned} I(x, y, z) &= I_0 \exp \left[-\frac{2x^2}{w^2(z)} \right] \exp \left[-\frac{2y^2}{w^2(z)} \right] \\ \text{and} \quad \hat{I}(s_x, s_y) &= \hat{I}_0 \exp \left[-2\pi^2 w_0^2 s_x^2 \right] \exp \left[-2\pi^2 w_0^2 s_y^2 \right] \end{aligned} \quad (1.22)$$

where $s = \sin \theta / \lambda \simeq \theta / \lambda$ is the spatial frequency and thus $\hat{I}(s)$ is a measure of the far-field divergence of the Gaussian beam, as expected from Fourier diffraction theory. The variances for a Gaussian beam are, therefore, given by:

$$\sigma_x(z) = \sigma_y(z) = \frac{w(z)}{2} \quad \text{and} \quad \sigma_{s_x} = \sigma_{s_y} = \frac{1}{2\pi w_0} \quad (1.23)$$

A spatial-beamwidth product can be defined in each dimension as the product of the minimum beam variance and the spatial-frequency variance, which for a Gaussian beam is given by:

$$\sigma_x^{\min} \cdot \sigma_{s_x} = \sigma_y^{\min} \cdot \sigma_{s_y} = \frac{w_0}{2} \times \frac{1}{2\pi w_0} = \frac{1}{4\pi} \quad (1.24)$$

Analogous to the time-bandwidth product for Gaussian signal pulses, the spatial-beamwidth product is minimised for a Gaussian beam and any real laser beam has a product equal to or greater than $1/4\pi$. From this, a far more generalised M^2 definition can be obtained:

$$M_x^2 = 4\pi \sigma_x^{\min} \sigma_{s_x} \quad \text{and} \quad M_y^2 = 4\pi \sigma_y^{\min} \sigma_{s_y} \quad (1.25)$$

As before, a Gaussian beam has $M^2 = 1$ but now the M^2 of almost any laser beam, irrespective of its mode content, can be calculated by measuring the variance (Eqn. 1.20) of the beam profile at a number of positions along the propagation direction. Not only can the M^2 factor describe the beam quality of almost any laser beam profile in two numbers, it can also describe the propagation of non-diffraction-limited beams through optical systems. To do this, we define a beam width as $W_{x,y}(z) \equiv 2\sigma_{x,y}(z)$ and from this, it can be shown that the beam width propagates as:

$$W(z) = W_0 \sqrt{1 + \left(\frac{zM^2}{z_R} \right)^2} \quad (1.26)$$

where $W_0 \equiv 2\sigma^{\min}$ is the beam waist of the laser beam.

Comparing Eqn. 1.26 with 1.16, we see that the non-diffraction-limited beam can be thought of as propagating with an ‘embedded Gaussian beam’ that has the same waist position but has a waist a factor of $\sqrt{M^2}$ smaller. This is a hugely powerful concept as it enables the propagation of the real laser beam through optical components, such as lenses, to be calculated using the embedded Gaussian beam. The waist positions will occur in the same places and the beam radius will always be a factor of $\sqrt{M^2}$ larger than the embedded Gaussian beam.

Figure 1.1 shows the embedded Gaussian beam radii for beams that have an $M^2 = 2$ with waist radius of $60 \mu\text{m}$ (orange line) and $\sqrt{2} \cdot 60 \mu\text{m}$ (green) line. It is immediately obvious why diffraction-limited beam quality is highly desirable, especially for applications such as nonlinear frequency conversion in a bulk crystal. For the same waist radius, a non-diffraction-limited beam (orange line Fig. 1.1) has a Rayleigh range that is M^2 smaller and, therefore, average beam intensity is lower over a given length. Indeed, to achieve the same Rayleigh range, the non-diffraction-limited beam (green line Fig. 1.1) needs to have a waist radius that is $\sqrt{M^2}$ larger than the diffraction limited beam.

1.2.3 Nonlinear propagation

For sufficiently intense pump fields ($I > 10^4 \text{ W/cm}^2$), the approximation of a harmonic potential for the bound electrons of a medium is no longer valid. Thus the response of the dielectric medium can no longer be considered a linear relationship with the applied field and higher order terms in the polarisation must be considered. The second order term in the polarisation expansion ($\propto E^2$) has the next highest strength but only in non-centrosymmetric media.

Centrosymmetric media have inversion symmetry [$\mathbf{P}(-\mathbf{E}) = -\mathbf{P}(\mathbf{E})$] and since the even-order terms do not change sign when the field is reversed, they must be zero to remain consistent. This means that second-order nonlinear effects are not seen in any liquids or gases. In solids, of the 32 crystal classes, 21 are non-centrosymmetric and 18 of these are anisotropic, meaning that they exhibit birefringence, often used as a tool to achieve phase-matching in second-order nonlinear processes. The SiO_2 molecule is non-symmetric, indeed the first demonstration of second-harmonic generation (SHG) was in a quartz crystal [Fra61]. The amorphous nature of glass in fused silica fibres, however, results in an inversion symmetry that renders second-order effects negligible.

To illustrate the importance of the second order term, consider a one dimensional (thus the susceptibility tensors are reduced to a single component) monochromatic sinusoidal

driving field $E = A \cos(\omega t)$, the resulting polarisation is given by:

$$\begin{aligned} P &= \epsilon_0 [\chi^{(1)} A \cos(\omega t) + \chi^{(2)} A^2 \cos^2(\omega t)] \\ &= \epsilon_0 (\chi^{(1)} A \cos(\omega t) + \frac{1}{2} \chi^{(2)} A^2 [1 + \cos(2\omega t)]) \end{aligned} \quad (1.27)$$

It is immediately obvious that the inclusion of the second order term has enabled the creation of frequencies that were not present in the original driving field. In this case, the created frequency is at 2ω and therefore the process is known as SHG or equivocally frequency-doubling; this process is described in more detail in Section 1.4. With different optical and electrical driving fields other second-order nonlinear effects can be observed: sum-frequency generation, difference-frequency generation, the Pockels effect and optical rectification.

In a similar vein, the polarisation with a driving field of $E = A \cos(\omega t)$ including the third order term is given by:

$$\begin{aligned} P &= \epsilon_0 (\chi^{(1)} A \cos(\omega t) + \chi^{(2)} E^2 + \chi^{(3)} A^3 + \frac{1}{4} \chi^{(3)} [3A^3 \cos(\omega t) + \cos(3\omega t)] + \dots) \\ &= \epsilon_0 [\chi^{(1)} + \frac{3}{4} \chi^{(3)} A^2] A \cos(\omega t) + \epsilon_0 (\chi^{(2)} E^2 + \chi^{(3)} [A^3 + \frac{1}{4} \cos(3\omega t)] + \dots) \\ &= \epsilon_0 \chi_{\text{eff}}^{(1)} A \cos(\omega t) + \epsilon_0 (\chi^{(2)} E^2 + \chi^{(3)} [A^3 + \frac{1}{4} \cos(3\omega t)] + \dots) \end{aligned} \quad (1.28)$$

In this case, we see that in addition to a frequency component at 3ω (third-harmonic generation) being created, we see that the effective electric susceptibility has a term proportional to the pump intensity ($I \propto A^2$). This results in an intensity-dependent refractive index:

$$n(\omega, A^2) = \sqrt{1 + \chi_{\text{eff}}^{(1)}} = \sqrt{1 + \chi^{(1)} + \frac{3}{4} \chi^{(3)} A^2} = n_0 \sqrt{1 + \frac{3}{4n_0^2} \chi^{(3)} A^2} \quad (1.29)$$

and since $\chi^{(3)} \ll \chi^{(1)}$, Eqn. 1.29 can be approximated as:

$$n(\omega, I) \approx n_0 \left[1 + \frac{3}{8n_0^2} \chi^{(3)} A^2 \right] = n_0 \left[1 + \frac{3}{4n_0^3 c \epsilon_0} \chi^{(3)} I \right] = n_0(\omega) + n_2 I \quad (1.30)$$

where $I = \frac{1}{2} n_0 \epsilon_0 c A^2$ has been used, n_2 is the nonlinear refractive index and it is assumed that the optical field is linearly polarised, such that only one component of the $\chi^{(3)}$ tensor contributes to the nonlinear refractive index. For pulse durations > 10 fs, the electronic contribution to n_2 can be regarded as instantaneous. The intensity-dependent refractive index is also known as the optical Kerr effect, which is the mechanism behind self-phase modulation (SPM), cross-phase modulation (XPM), four-wave mixing (FWM) and self-focusing.

In common with the linear susceptibility, $\chi^{(3)}$ is also a complex parameter. Parametric

effects, where no energy is transferred to the medium, correspond to the real part of $\chi^{(3)}$, which is responsible for third-harmonic generation and the optical Kerr effect. The imaginary part of $\chi^{(3)}$ results in inelastic scattering, where optical energy is not conserved as observed in stimulated Raman scattering (SRS) and stimulated Brillouin scattering (SBS).

One might sensibly ask whether practical laser sources can deliver high enough intensities to observe $\chi^{(3)}$ effects with reasonable efficiencies, especially in silica that has an n_2 value ($2.74 \times 10^{-16} \text{ cm}^2/\text{W}$ at 1053 nm [Mil98]) that is up to two orders of magnitude smaller than most commonly used nonlinear materials. To answer this question, the product of the optical intensity and the nonlinear interaction length, a figure of merit for nonlinear processes, can be compared between a free-space Gaussian beam propagating through a nonlinear crystal and fundamental mode propagation in an optical fibre. In a nonlinear crystal of length L , the power length product can be evaluated as an integral of the Gaussian beam intensity over the length of the crystal:

$$A_{\text{nw}} = \int_{-L/2}^{L/2} \frac{P}{\pi w^2(z)} dz < \int_{-\infty}^{\infty} \frac{P}{\pi w^2(z)} dz = \frac{\pi n}{\lambda} P \quad (1.31)$$

where P is the peak-power of the laser and $w(z)$ is the beam radius as a function of distance from the beam waist, positioned at the centre of the crystal. The result from Eqn. 1.31 is that due to diffraction, the power length product is independent of the crystal length and only depends on the peak-power of the pump.

In contrast, the power length product for an optical fibre of length L is given by:

$$A_{\text{f}} = \frac{PL_{\text{eff}}}{\pi a^2} = \frac{P}{\pi a^2 \alpha} [1 - \exp(-\alpha L)] \quad (1.32)$$

where a is the mode-field radius, L_{eff} is the effective fibre length and α is the fibre attenuation coefficient. For short fibres ($\alpha L \ll 1$) attenuation is unimportant and so $L_{\text{eff}} = L$; whereas, for very long fibres ($L \rightarrow \infty$, $\alpha L \gg 1$) attenuation limits the effective fibre length to $L_{\text{eff}} \rightarrow 1/\alpha$. The ratio of the power length products for the optical fibre and nonlinear crystal is given by:

$$\frac{A_{\text{f}}}{A_{\text{nw}}} = \frac{\lambda}{\pi^2 n a} \cdot \frac{L_{\text{eff}}}{a} \quad (1.33)$$

For standard single mode fibres this ratio is $\sim 10^8$, as illustrated by SMF-28 operating at the loss minimum ($\lambda = 1.55 \mu\text{m}$) with a mode field radius of $a = 5.2 \mu\text{m}$ and a loss coefficient of $\alpha = 0.2 \text{ dB/km}$, resulting in an $L_{\text{eff}} = 22 \text{ km}$. The very large enhancement factor over a bulk crystal results from the high intensity of the fundamental mode being maintained for the potentially extremely long (in comparison to a nonlinear crystal) interaction length of the fibre. It is for this reason that, despite the low nonlinearity of

silica, nonlinear fibre optics is a vibrant research field [Agr07] and is the basis for much of the work presented in this thesis.

1.3 Optical fibres

All of the laser systems developed in this thesis use optical fibres as gain media, providing gain either via stimulated emission using rare-earth (Yb and Er) doped fibres or nonlinear gain using stimulated Raman scattering (SRS). This section introduces the linear and nonlinear propagation of light in optical fibres, the guidance mechanism, rare-earth doped and large mode-area (LMA) optical fibres.

1.3.1 Guidance mechanism

A multimode step-index fibre is an optical waveguide structure that guides light by having a core refractive index n_{co} that is higher than the surrounding cladding n_{cl} [Fig. 1.2 (a)]. To obtain the electromagnetic field profiles that are guided by this refractive index structure, one must solve the wave equation with the appropriate boundary conditions. To gain a more intuitive physical insight to the solutions, however, one can consider the following heuristic argument.

From Fourier theory, a wave that is guided by the optical fibre can be thought of as a superposition of a travelling plane wave and a transverse standing wave [Fig. 1.2 (b)]. It can be argued that the transverse field must be a standing wave if energy is to be guided down the fibre and not propagate away into the cladding. The travelling plane wave in the axial direction has a propagation constant $\beta = n_{\text{eff}}k_0$ in both the cladding and core, where n_{eff} is the effective refractive index of the propagating mode and $k_0 = 2\pi/\lambda$ is the vacuum wave number. The transverse standing wave has a propagation constant k_t that can only take on a restricted number of values: the optical path length (including phase shifts) between the interfaces of the core and the cladding has to be a half-integer number of wavelengths. The norm of the wave vector of the resulting electromagnetic field superposition has to equal k_0n , where n is the local refractive index. Hence, in the core, the two propagation constants are related by:

$$k_0^2 n_{co}^2 = \beta^2 + k_t^2 \quad (1.34)$$

and in the cladding, the relation is given by:

$$k_0^2 n_{cl}^2 = \beta^2 + k_t^2 \quad (1.35)$$

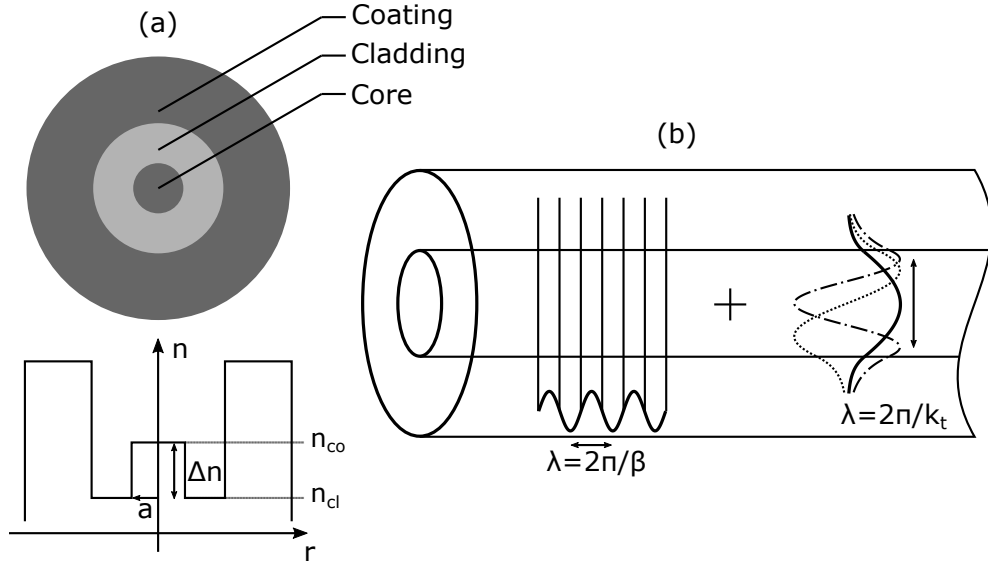


Figure 1.2: (a) Geometry of a multimode step index fibre with core radius a , core refractive index n_{co} , cladding refractive index n_{cl} and corresponding index contrast $\Delta n = n_{co} - n_{cl}$. (b) Illustration of the decomposition of a propagating wave into a travelling plane wave and transverse standing wave with an evanescent field in the cladding.

It can, therefore, be seen that if:

$$n_{co}k_0 \geq \beta \geq n_{cl}k_0 \quad (1.36)$$

then k_t is a real value in the core and purely imaginary in the cladding, since $k_t^2 = k_0^2 n_{cl}^2 - \beta^2 < 0$. If the condition of Eqn. 1.36 is satisfied, the transverse field is an oscillating wave in the core (where the wave vector is real) and an exponentially decaying wave (evanescent wave) in the cladding (where the wave vector is purely imaginary). Since the evanescent wave carries no energy away from the boundary of the core and the cladding, this corresponds to the physical process of total internal reflection. Hence, step-index optical fibres achieve waveguiding through the process of total internal reflection.

From the earlier argument that k_t can only take on restricted values, it is clear from Eqns. 1.34 and 1.35 that β can also only take on restricted values: discrete modes of propagation exist. The number of propagation modes is determined by the wavelength, the core radius and the refractive indices of the core and the cladding. In a more rigorous analysis, the β values and corresponding transverse field distribution are eigenvalues and eigenvectors of the fibre propagation equation [Sny83].

For a circularly symmetric step-index fibre, the number of propagating modes is de-

terminated the dimensionless V-parameter, given by:

$$V = k_0 a \sqrt{n_{co}^2 - n_{cl}^2} = \frac{2\pi a}{\lambda} \text{NA} \quad (1.37)$$

where a is the fibre core radius and NA is the numerical aperture of the fibre. If the geometry of the fibre is such that $V < 2.405$ for wavelengths greater than a specific value, there is only one possible value of β and only one pair of degenerate (two polarisations) modes can propagate. The fibre is thus said to be single-mode and the specific wavelength below which the guidance becomes multi-mode is named the cut-off wavelength. The most important aspect of single mode guidance is that the transverse intensity profile from the fibre output, which is close to Gaussian for a step-index fibre, is independent of the intensity profile of the input beam. Only the fraction of the input light that is transmitted decreases when the input profile does not match the mode profile of the fibre.

In contrast, for step-index multimode fibres with a large V parameter, the number of guided modes is well approximated by $N = \frac{V^2}{2}$ [Glo71b]. If all of the modes are assumed to be equally excited and incoherent, the output beam quality is well approximated by $M^2 = \frac{\pi}{12} + \frac{V}{3}$ [Zer14]. It is clear, therefore, that single-mode fibres can achieve significantly higher beam brightness than multimode fibres. Optical fibres are typically protected by a polymer coating [Fig. 1.2 (a)] that increases the mechanical robustness, allowing them to be wound and coiled to reasonably (tens of mm's) small radius whilst maintaining their guidance properties.

1.3.2 Attenuation

The optical fibre waveguide described in the proceeding section could theoretically be constructed of any two dielectrics with the correct dimensions and refractive index contrast. In all but a few specialist applications, however, optical fibres are based on silica glasses. Indeed, the vast success of optical fibres underpinning the entire worldwide telecommunications network is primarily down to the superior chemical, physical and optical properties of fused silica.

In comparison to other glasses that could be drawn into optical fibres, fused silica is mechanically more robust, chemically inert and not hygroscopic. Additionally, the abundance of silicon and the centuries refining glass processing techniques allows very pure fused silica to be produced at relatively low cost. Finally, the optical fibre fabrication process is made relatively simple by the fact that the refractive index of silica can be altered using dopants that do not significantly affect its advantageous properties. Fused silica can be doped using vapour phase deposition with elements such as GeO_2 and P_2O_5 to increase the refractive index or B and F to reduce the refractive index, with extremely high

degrees of precision (e.g. state-of-the-art LMA fibres have $\Delta n = 5 \times 10^{-4}$ [Jai15]). Once a preform of the fibre structure has been fabricated with the correct dimensions and index contrast, the slow variation of viscosity with temperature and large glass temperature range enable kilometre long optical fibres to be drawn with extremely tight dimension tolerances and uniformity.

Of course, the superior physical properties of fused silica for the fabrication of optical fibres would be irrelevant were it not for its excellent optical properties as well. Fused silica is generally considered to be transparent between 0.3–2.5 μm , with the short wavelength edge determined by electronic resonances of the SiO_2 (and GeO_2) molecules and the long wavelength edge from atomic and molecular vibrational resonances. Additional to the intrinsic absorption loss from fused silica, impurities can give absorption features, most notably the OH^- ion has an overtone at 1.38 μm . With modern fabrication processes, however, the OH^- concentration can be reduced below 1 ppm, resulting in the lowest optical attenuation of any solid material. Indeed, it is density fluctuations over a length scale of $< \lambda/10$ in the amorphous glass structure that result in Rayleigh scattering, which is the dominant contribution to optical fibre attenuation. The attenuation from Rayleigh scattering scales as λ^{-4} and so the loss minimum (~ 0.2 dB/km) of optical fibres occurs around 1.55 μm .

The vast progress in optical fibre fabrication has enabled the loss to be decreased from the >1000 dB/km value for early optical fibres [Hop54] to the 0.2 dB/km value of present day fibres [Miy79]. This has facilitated the deployment of optical fibres for trans-continental telecommunications networks and, as discussed in Section 1.2.3, has enabled fused silica optical fibres to be a practical nonlinear medium, in spite of their low nonlinearity.

1.3.3 Dispersion

The absorption resonances of fused silica used in optical fibres in the UV and infrared result in anomalous chromatic dispersion ($dn/d\lambda > 0$) in the immediate vicinity of these features. Optical fibres are used in the transmission window between these features, where the chromatic dispersion is normal ($dn/d\lambda < 0$) and the refractive index monotonically decreases with increasing wavelength. Around 1.27 μm [Mal65], there is a non-stationary point of inflection in the refractive index of fused silica and hence $d^2n/d\lambda^2 = 0$ at this point, the zero-dispersion wavelength (ZDW).

In an optical fibre, the overall dispersion results from the combination of the material dispersion and a contribution from the waveguide. The waveguide contribution can be understood from the fact that the propagation constant of each mode is wavelength dependent [i.e. $\beta = n_{\text{eff}}(\omega)k_0$]. In addition, if the fibre is multimode then the modes will

have different propagation constants, causing inter-modal dispersion, but this is not considered further here.

To mathematically describe the dispersion in an optical fibre, a Taylor series expansion of the mode-propagation constant, β , about the centre frequency, ω_0 , of the pulse spectrum, is used:

$$\beta(\omega) = n_0(\omega) \frac{\omega}{c} = \beta_0 + \beta_1(\omega - \omega_0) + \frac{1}{2}\beta_2(\omega - \omega_0)^2 + \dots \quad (1.38)$$

where the parameters β_1 and β_2 are related to the refractive index $n_0(\omega)$ by:

$$\beta_1 = \frac{1}{v_g} = \frac{n_g(\omega)}{c} = \frac{1}{c} \left(n_0(\omega) + \omega \frac{dn_0(\omega)}{d\omega} \right) \quad \text{and} \quad \beta_2 = \frac{1}{c} \left(2 \frac{dn_0(\omega)}{d\omega} + \omega \frac{d^2 n_0(\omega)}{d\omega^2} \right) \quad (1.39)$$

β_2 is the intra-pulse dispersion, otherwise known as group-velocity dispersion (GVD). GVD is responsible for pulse broadening and most often described by the dispersion parameter, D , in units of $\text{ps nm}^{-1} \text{ km}^{-1}$:

$$D = \frac{d\beta_1}{d\lambda} = -\frac{2\pi c}{\lambda^2} \beta_2 = -\frac{\lambda}{c} \frac{d^2 n_0(\omega)}{d\lambda^2} \quad (1.40)$$

If the GVD of the fibre is positive ($\beta_2 > 0$, $D < 0$), higher frequency components of the pulse travel slower than lower frequency components and an unchirped (transform limited) pulse acquires positive chirp (up chirp). Conversely, if the GVD is negative ($\beta_2 < 0$, $D > 0$) the lower frequency components will travel slower and an unchirped pulse acquires negative chirp (down chirp). It is common practice to call positive ($\beta_2 > 0$) GVD ‘normal’ and negative ($\beta_2 < 0$) GVD ‘anomalous’ and to refer to GVD simply as dispersion. Although this risks confusing the origin of the dispersion [either $n_0(\omega)$ or $n_g(\omega)$], in most situations where dispersion is important in fibre optics it relates to GVD of pulses, therefore, this will be the convention used in this thesis.

The fact that the optical fibre GVD has a contribution from the waveguide dispersion, which is dependent on the refractive index profile, opens the possibility to engineer the fibre’s dispersion profile. In standard step-index fibres, however, the scope to alter the dispersion is quite limited because the waveguiding is said to be weak due to the small index contrast between the core and the cladding. This results in the waveguide contribution to the GVD being significantly smaller than the material dispersion, apart from in the region where the material GVD is close to zero. If single-mode guidance is required, the refractive index profile is very constrained as it must give $1.5 < V < 2.405$ for suitable power confinement of the fundamental mode. It can be shown [Glo71a] that for a weakly-guiding step-index fibre, the waveguide dispersion is normal beyond the material ZDW of fused silica. Therefore, in a standard telecommunications single-mode fiber (e.g.

SMF-28) the ZDW is shifted around 40 nm from the fused silica ZDW to around 1.31 μm .

The ZDW of a conventional fibre can be shifted to longer wavelengths by increasing the waveguide dispersion through increasing the index contrast and correspondingly reducing the core radius to maintain single-mode guidance. In practice, to prevent the attenuation from increasing dramatically and to enable low splice losses to standard fibres, triangular core index profiles are used in dispersion-shifted fibres, which have ZDW around the loss minimum at 1.55 μm [Ain82]. Depressed cladding or W-shaped refractive index profiles [Miy81] and other multiple cladding designs [Coh82] can be used to produce dispersion flattened fibres and dispersion-compensating fibres. Dispersion-flattened fibres feature low dispersion over the range 1.3-1.6 μm and often have two ZDWs; whereas dispersion-compensating fibres typically feature normal dispersion and a negative dispersion slope around 1.55 μm , designed to compensate the dispersion of transmission fibres.

The main issue for dispersion engineering in conventional single-mode optical fibres is the limited achievable index contrast ($\Delta n \leq 0.03$), therefore, it is not possible to obtain anomalous waveguide dispersion in combination with single-mode guidance [Mog98]. Photonic crystal fibres (PCFs) [Kni96] can overcome this limitation by using a periodic array of air-holes as the cladding around a solid core, typically fabricated from fused silica. The celebrated physical properties of fused silica are arguably even more significant in the fabrication of PCFs due to the intricacy of the pre-forms that are required to form the microstructured optical fibres. The great benefit of the microstructured cladding is that the effective refractive index of the periodic structure can approach that of air, whilst preserving most of the mechanical advantages of the optical fibre geometry. This facilitates far stronger waveguiding because the index contrast can be much larger than in step-index fibres.

The periodic cladding of PCFs allows a greater number of degrees of freedom to engineer the dispersion profile whilst maintaining single-mode guidance in comparison to conventional optical fibres. Crucially, it enables the ZDW of a single-mode PCF to be shifted to wavelengths as short as 0.57 μm [Kni00], which is not possible in step-index fibres. The shifting of the ZDW close to the emission wavelength of Ti:Sapphire and ytterbium fibre (Yb-fibre) lasers has led to the PCF becoming the dominant platform for supercontinuum generation [Dud10]. In addition, PCFs can be engineered to be single-mode for any wavelength, so called endlessly-single-mode PCFs [Bir97], again something that is not possible in conventional fibres. The ability to tailor the dispersion profile of PCFs whilst maintaining single-mode guidance in conjunction with tight confinement and modest attenuation (typically a few dB/km) makes PCFs an excellent platform for nonlinear optics.

1.3.4 Nonlinear effects

The amorphous nature of the glass matrix in fused silica optical fibres results in an inversion symmetry that renders $\chi^{(2)}$ nonlinear effects negligible. Third-order nonlinear effects can be efficiently excited in optical fibres despite the low nonlinearity of fused silica, however, due to the tight confinement of the waveguiding and the long interaction lengths. These third-order effects can broadly be separated into parametric effects relating to the real component of $\chi^{(3)}$ and inelastic scattering processes related to the imaginary component of $\chi^{(3)}$. The parametric third-order effects typically observed in fibres originate from the intensity-dependent refractive index (Section 1.2.3), since it is very difficult to achieve phase matching for third-harmonic generation in silica fibres. Different manifestations of the optical Kerr effect, relating to different input electromagnetic fields, are named as separate nonlinear effects despite their common origin. This section describes these third-order parametric and inelastic nonlinear effects.

Self-phase modulation

Self-phase modulation (SPM) describes the local modulation of the refractive index via the Kerr effect due to the variation in instantaneous intensity across an optical pulse. The phase accumulated by an optical wave propagating down a fibre of length L is given by:

$$\Delta\phi = n(\omega, I)k_0L = [n_0(\omega) + n_2I]k_0L \quad (1.41)$$

where the definition of the nonlinear refractive index (Eqn. 1.30) has been used. The instantaneous optical frequency is given by the derivative of the phase, hence, the pulse will obtain a frequency shift from the central frequency across the temporal profile, given by:

$$\Delta\omega = -\frac{d(\Delta\phi)}{dt} = -n_2k_0L\frac{dI}{dt} \quad (1.42)$$

The change in instantaneous optical frequency across the pulse is referred to as chirp, as introduced in the context of GVD (Section 1.3.3). Clearly, from Eqn. 1.42, the pulse shape will dictate the variation in the chirp, hence, depending on the chirp of the input pulse, SPM can lead to a broadening or narrowing of the optical spectrum of the pulse.

For unchirped Gaussian or sech^2 pulses, SPM always leads to spectral broadening and spectral modulation. The broadening is a result of the creation of new frequencies from the chirp and the modulation is a consequence of interference between the same frequency shift that occurs twice within the temporal profile (since the pulse is symmetric about the centre). A pulse with an initial positive linear chirp will undergo more significant spectral broadening, however, the oscillatory modulation structure is less pronounced. A

pulse with an initial negative linear chirp can undergo spectral narrowing because the SPM induced chirp has the opposite sign around the centre of the pulse, which allows for pulse spectral compression [Obe93].

A pulse propagating in an optical fibre is also, of course, subject to GVD in addition to SPM. As noted in Section 1.3.3, GVD also results in an optical pulse acquiring a frequency chirp, whose sign is dependent on whether the dispersion is normal or anomalous. The extent of the interplay between GVD and SPM depends on the relative scale of the dispersion length, L_D and the nonlinear length, L_{NL} , given by:

$$L_D = \frac{\tau_0^2}{|\beta_2|} \quad \text{and} \quad L_{NL} = \frac{1}{\gamma P_0} \quad (1.43)$$

where τ_0 is the input pulse duration, P_0 is the pulse peak-power and γ is the nonlinear parameter, given by:

$$\gamma = \frac{n_2(\omega)\omega}{c A_{\text{eff}}} \quad (1.44)$$

where A_{eff} is the effective area of the propagating mode. If L_D is comparable to L_{NL} and of the order of the fibre length then the interplay between GVD and SPM can give rise to notable effects. In the normal-dispersion regime, SPM acts to enhance the rate of pulse broadening due to the dispersion of the newly generated frequency components. For Gaussian and sech^2 pulses, the resulting chirp is linear and positive across a significant proportion of the central part of the pulse. This linear chirp can be dechirped by a linear dispersive delay, for example a pair diffraction gratings, and therefore the SPM generated bandwidth enables pulse durations shorter than the initial pulse to be obtained [Sha82].

In the anomalous dispersion regime with sech^2 pulses, the SPM generated chirp can exactly cancel the GVD chirp, resulting in the propagation of optical solitons. As a soliton propagates through a lossless fibre, its intensity profile remains unchanged and only a constant phase is added to the field envelope. Optical solitons remain unchirped (transform limited) as they propagate and therefore do not experience temporal broadening. To balance the SPM induced chirp with GVD, the peak-power of a fundamental soliton is given by:

$$P_0 = \frac{|\beta_2|}{\gamma \tau_0^2} \quad (1.45)$$

A higher-order soliton forms when the peak-power is an integer multiple of the square of the fundamental soliton's peak-power and it undergoes periodic temporal and spectral changes over the soliton period, given by:

$$z_s = \frac{\pi \tau_0^2}{|\beta_2|} \quad (1.46)$$

One might expect that solitons would be unstable in the presence of fibre attenuation and dispersion changes, however, provided that the changes are small over the soliton period then the soliton will adjust its duration to compensate without losing energy (adiabatically) [Mam91]. Furthermore, the formation of a fundamental or higher-order soliton is rather insensitive to the input pulse parameters. If the peak-power is sufficient, a range of input pulse shapes and durations will evolve into a soliton of the closest integer order by shedding energy in the form of dispersive waves [Blo88]. This enables them to be observed experimentally rather easily [Has81, Gou89] and the long distance propagation without temporal broadening is highly desirable in telecommunications applications.

Cross-phase modulation

Cross-phase modulation (XPM) describes the local modulation of the refractive index of one optical pulse via the Kerr effect due to the intensity profile of a copropagating pulse with a different frequency. In effect, self-phase modulation (SPM) is the degenerate case of cross-phase modulation (XPM), with both waves having the same frequency and polarisation. When two pulses with different wavelengths propagate down an optical fibre where the nonlinear length, L_{NL} , is of the order of the fibre length, then XPM and SPM will always occur simultaneously.

In common with SPM, the intensity profiles of the pulses result in a frequency chirp but in XPM it is proportional to the sum of the pulse intensities. This similarly results in spectral broadening and modulation of both pulses, but in XPM if the nonlinear lengths of the two pulses are not equal, the broadening is asymmetric. If the dispersion length L_D is comparable to the nonlinear length, L_{NL} , then asymmetric temporal changes can also occur. XPM can be used to temporally compress a signal pulse by chirping and creating additional bandwidth using a pump pulse and then dechirping using a dispersive delay line, in a similar manner to using SPM but with the advantage that a high peak-power signal pulse is not required [Dia88].

Four-wave mixing

Third-order parametric nonlinear processes couple four interacting waves via the real part of $\chi^{(3)}$. The previous examples of SPM and XPM are essentially four-wave interactions with different degrees of degeneracy. In a general parametric four-wave interaction, energy must be conserved and therefore, the four interacting frequencies are related by:

$$\omega_4 = \omega_1 + \omega_2 + \omega_3 \quad (1.47)$$

Of course, the photon momentum must also be conserved and so assuming single mode propagation in the same direction along a fibre, this gives the relation:

$$\Delta k = \beta(\omega_4) - \beta(\omega_1) - \beta(\omega_2) - \beta(\omega_3) = 0 \quad (1.48)$$

which is also known as phase matching. In SPM, all four interacting waves have the same frequency or the negative ($\omega = \omega - \omega + \omega$) and in XPM two of the four waves have different frequencies ($\omega_1 = \omega_1 + \omega_2 - \omega_2$ or $\omega_2 = \omega_1 + \omega_2 - \omega_1$). It is clear, therefore, that both SPM and XPM are automatically phase matched because the propagation constants sum to zero (if $\omega = -\omega$ then $\beta = -\beta$).

In general, one has to work quite hard to achieve phase matching and so the automatic phase matching of SPM and XPM might, at first, sound advantageous. In most cases, however, SPM and XPM cause spectral broadening that reduces the power spectral density of a pulsed signal. This is an issue for subsequent conversion using second-order parametric processes, as these processes can only be phase matched for a finite range of input frequencies. Therefore, the reduction in power spectral density as a result of SPM and XPM can compromise the conversion efficiency of parametric second-order processes. This is a particularly pertinent issue when using second-order nonlinear crystals to frequency convert the output of fibre amplifiers, since a high peak-power spectral density is required to achieve high conversion efficiencies. The compromise between achieving a high peak-power and a high power spectral density from a fibre amplifier is a recurring theme in the experimental chapters of this thesis.

Four-wave mixing (FWM) refers to the more general case of a four-wave interaction when two pump frequencies (ω_1 and ω_2) mix, generating two new frequencies (ω_3 and ω_4) due to the Kerr effect. In the degenerate case of FWM, the two pump photons have the same frequency ($\omega_1 = \omega_2 \equiv \omega_p$) and therefore a single pump beam generates two frequency sidebands that are symmetrically spaced by Ω from the pump frequency:

$$\Omega = \omega_p - \omega_3 = \omega_4 - \omega_p \quad (1.49)$$

Traditionally, the low-frequency sideband is called the Stokes wave ($\omega_3 \equiv \omega_s$) and the high-frequency component the anti-Stokes ($\omega_4 \equiv \omega_a$), after the pioneering work of Sir George Stokes on the frequency downshift of the fluorescence emitted by excited matter [Sto52]. The phase-mismatch of the four interacting waves in degenerate FWM is therefore given by:

$$\Delta k = \beta(\omega_a) + \beta(\omega_s) - 2\beta(\omega_p) \quad (1.50)$$

In addition to this linear phase-mismatch, the intensity dependent refractive index con-

tributes a nonlinear term resulting in an effective phase-mismatch given by:

$$\kappa = \Delta k + 2\gamma P_0 \quad (1.51)$$

where P_0 is the peak-power of the pump wave.

In the case where all four waves have the same polarisation (scalar FWM) the parametric gain, g , is given by:

$$g = \sqrt{(\gamma P_0)^2 - (\kappa/2)^2} \quad (1.52)$$

Clearly, the parametric gain is maximised when the effective phase-mismatch is zero, resulting in a gain determined by:

$$g_{\max} = \frac{g_P P_0}{A_{\text{eff}}} = \frac{2\pi n_2}{\lambda_p} \frac{P_0}{A_{\text{eff}}} \quad (1.53)$$

For a pump wavelength of $1 \mu\text{m}$ in a silica fibre, the parametric gain coefficient is slightly higher than the peak polarised Raman gain coefficient ($g_P \sim 1.6 \times 10^{-13} \text{ m/W}$ cf. $g_R \sim 1.2 \times 10^{-13} \text{ m/W}$). The key difference, however, is that the SRS process is automatically phase-matched, whereas the parametric gain is highly sensitive to the phase-matching condition.

For standard single mode fibres pumped around $1 \mu\text{m}$, the linear phase-mismatch is positive, therefore, the effective phase-mismatch cannot be reduced to zero. Significant FWM can still occur, however, if the length of the fibre is shorter than the coherence length associated with the effective phase-mismatch. Away from the ZDW of the fibre, the linear phase-mismatch can be approximated as $\Delta k \approx \beta_2 \Omega^2$ by assuming that higher than second order terms in the Taylor series expansions of the mode-propagation constants (Eqn. 1.38) are negligible for small frequency shifts. Using this approximation, the coherence length of the degenerate scalar FWM is given by:

$$L_{\text{coh}} = \frac{2\pi}{\kappa} = \frac{2\pi}{\beta_2 \Omega^2 + 2\gamma P_0} \quad (1.54)$$

A standard single-mode fibre (SMF) for $1 \mu\text{m}$ operation (e.g. Corning HI 1060) has $\beta_2 = 23 \text{ ps}^2/\text{km}$ and $\gamma = 5.4 \text{ W}^{-1}\text{km}^{-1}$ at 1060 nm . Therefore, for a typical pump peak-power of 1 kW , $L_{\text{coh}} \leq 0.6 \text{ m}$ and so significant FWM is not expected to occur in the fibre lengths used in this thesis. An important point to note, however, is that the coherence length will not be constant in fibres where the pump field experiences gain. The non-constant coherence length can result in significant FWM occurring in fibres (e.g. ytterbium-fibre amplifiers) where the linear phase-mismatch is not conducive to efficient FWM [Fèv07].

Efficient FWM in standard SMF pumped around $1 \mu\text{m}$ can also occur using polarisation-

maintaining (PM) fibres, by polarising the interactive fields such that the fibre's birefringence reduces the effective phase-mismatch to zero. There are five permutations of the interacting fields' polarisations in degenerate FWM in birefringent fibres. Of these, Type-I phase matching, where the pump power is split equally between the fast and the slow axis of the fibre, is most readily observed in standard fibres for a 1 μm pump. In this case, the Stokes and anti-Stokes fields propagate on different axes and the linear phase-mismatch is given by:

$$\Delta k = \beta_2 \Omega^2 - \delta n \Omega c^{-1} \quad (1.55)$$

where δn is the fibre birefringence. This enables the dispersion of PM fibres to be approximated from the frequency spacing of the FWM sidebands using $|\beta_2| = \delta n / c\Omega$.

The tailorable dispersion of PCFs enables degenerate FWM to be used as a practical and efficient method for shifting the spectral output of pulsed near-infrared (NIR) lasers to the visible, near-visible and the mid-infrared [Nod09, Lav10, Jau12]. Degenerate FWM can also be used to parametrically amplify a signal injected at either the anti-Stokes or Stokes phase matched wavelengths, which can provide gain for a parametric oscillator [Mur12, Lam13] or enable tunable narrow linewidth radiation to be generated [Lef12].

Stimulated Raman scattering

The Raman effect describes the small fraction ($\sim 10^{-6}$) of light that is red-shifted when it is scattered by a molecular medium, first observed by K. R. Ramanathan in 1923 [Ram23a, Ram23b] and later explained by C. V. Raman in 1928 [Ram28]. When the spectra of the downshifted scattered light was observed, Raman and colleagues showed that the frequency downshift only occurred at discrete (quantised) frequencies, which corresponded to the vibrational modes of the molecular media. The discovery was significant because only the elastic scattering of light (Rayleigh scattering) had previously been predicted and observed. Indeed, Ramanathan's observations occurred in the same year that the inelastic scattering of X-rays (Compton scattering) was published [Com23], which was part of the growing body of work demonstrating the quantum nature of electromagnetic radiation.

The inherently quantum nature of Raman scattering leads it to be described as a pump photon exciting a molecule into a virtual energy state, from which it relaxes to a higher-order vibrational state, leading to the emission of a frequency downshifted photon. The downshifted photons are known as the Stokes wave and the frequency downshift occurs at discrete frequencies due to the quantised nature of the vibrational states. The quanta of vibrational energy involved in the Raman scattering process is referred to as an optical phonon. It is also possible that the molecule could be in a higher-order vibrational

state when the pump photon is incident, therefore, when it relaxes from the virtual energy state a frequency upshifted photon is emitted (anti-Stokes wave). This anti-Stokes Raman scattering is rarely observed in molecular medium at room temperature because Boltzmann statistics predict that the vast majority of molecules will be in the vibrational ground state.

It was accidentally discovered in 1962 that the spontaneous Raman scattering process could be stimulated using a high intensity pump beam [Woo62]. This was evidenced in the output of a Ruby laser, Q-switched with a nitrobenzene Kerr cell, by the appearance of laser oscillation at a second red-shifted wavelength (765.8 nm) in addition to the standard emission wavelength (693.4 nm). The frequency difference between the two wavelengths corresponded exactly to a vibrational resonance of nitrobenzene (40.3 THz) and thus the term Raman laser was coined. The stimulated Raman scattering (SRS) process occurs due to a coupling of the pump and the Stokes waves via the imaginary part of $\chi^{(3)}$, which enables energy to be transferred from the pump to the Stokes wave. The imaginary part of $\chi^{(3)}$ results from the vibrational resonance of the molecule, analogous to the imaginary component of $\chi^{(1)}$ resulting from absorption resonances. Indeed, the resonance associated with SRS need not be vibrational, they can be rotational or electronic but the frequency shifts related to vibrational resonances in media transparent to laser radiation are readily observable. As the medium plays an active role in SRS, phase matching is automatically achieved in both the forward and backward directions.

Since SRS is related to the vibrational resonances of molecules, the Raman gain coefficient is a function of the frequency separation between the pump and the Stokes waves [i.e. $g_R = g_R(\Omega)$, where $\Omega = \omega_p - \omega_s$]. In most molecular media this results in a gain spectrum with multiple narrow peaks, however, in optical fibres, the amorphous nature of the glass matrix results in significant inhomogeneous broadening. Figure 1.3 (a) shows the calculated Raman gain as a function of frequency separation from the pump for a pure silica-core optical fibre. A multiple damped oscillator model was used to calculate the Raman response function of a fused silica fibre [Hol02]. The electronic contribution to $\chi^{(3)}$, which results in the real part, is normally considered instantaneous but the Raman response is delayed by approximately 100 fs [Sto89]. It is this delayed Raman response that results in the imaginary component of $\chi^{(3)}$, the Fourier transform of which gives the Raman gain spectrum shown in Fig. 1.3 (a).

From Fig. 1.3 (a) it is clear that the Raman gain curve for a pure silica-core fibre is very broadband, extending over 40 THz with a broad (~ 8 THz) peak at 13.2 THz and a secondary peak at 14.7 THz. When the cores of silica-based fibres are doped with elements (e.g. germanium and phosphorous) to vary the refractive index for waveguiding, the Raman gain spectrum changes due to the differing vibrational resonances of the

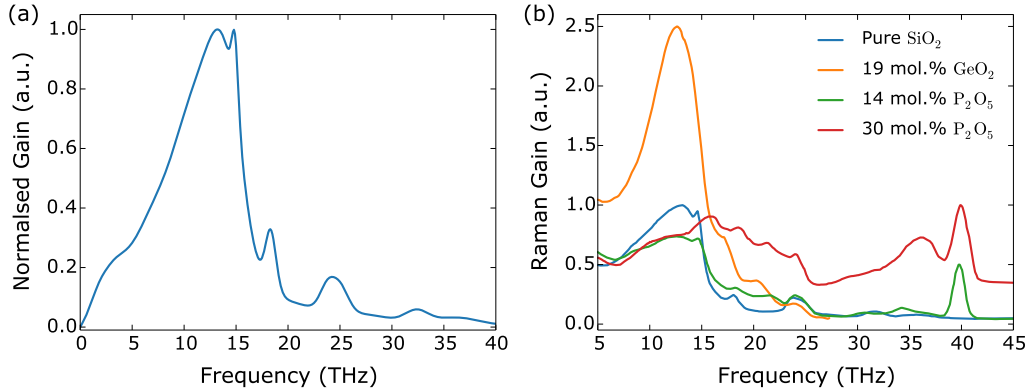


Figure 1.3: (a) Calculated Raman gain curve for a pure silica-core optical fibre, based on the multiple damped oscillator model [Hol02]. (b) After [Shi81]. Raman gain curves of doped silica-core optical fibres.

molecular bonds [Fig. 1.3 (b)]. Indeed, it was shown by Galeener et al. [Gal78] that vitreous GeO₂, P₂O₅ and B₂O₃ all have higher Raman gain coefficients than pure silica (SiO₂). The position and relative intensity of the Raman peaks of doped-silica fibres depends on the molecular concentration of the dopant and the positions can shift significantly from the pure vitreous samples [Shi81].

Of the dopants, GeO₂ is most notable since the peak of its Raman gain at 12.6 THz downshift is 9.2 times greater than silica [Gal78] and the position overlaps with the broad peak of pure silica. The magnitude of the Raman gain peak of a germanium-doped silica fibre [orange curve Fig. 1.3 (b)] is therefore proportional to the molecular concentration of the germanium [Dav89]. Germanium-doping of the core is commonly used in dispersion shifted or flattened fibres to increase the strength of the waveguiding (due to the increased index contrast) and thus increase the waveguide contribution to the GVD of the fibre (cf. Section 1.3.3). Since these fibres also have reduced core diameters to maintain single-mode guidance, they feature Raman gain efficiencies (C_R) that can be 15 times higher than pure silica-core fibres [Bro02]. Phosphorous-doped silica fibres are also interesting due to the larger (40 THz) Raman shift [green and red traces in Fig. 1.3 (b)], which enables Raman lasers operating around 1.45 μm (for amplification of 1.55 μm signals) to have fewer Raman cascades using Yb-fibre pump lasers [Kur00].

The stimulated growth of the Stokes wave with propagation distance, z , and the corresponding decay of the pump wave due to the $\chi^{(3)}$ coupling is given by [Agr07]:

$$\frac{\partial I_s}{\partial z} = g_R I_p I_s - \alpha_s I_s \quad (1.56)$$

$$\frac{\partial I_p}{\partial z} = -\frac{\omega_p}{\omega_s} g_R I_p I_s - \alpha_p I_p \quad (1.57)$$

where g_R is the Raman gain coefficient, α_p , α_s is the attenuation coefficient at the Stokes and pump wavelengths and I_p , I_s are the intensities of the pump and Stokes waves, respectively. The Raman gain coefficient is heavily polarisation dependent: it is maximum when the pump and Stokes waves are co-polarised and an order of magnitude smaller when the polarisations are orthogonal [Hel75]. If the energy transfer from the pump to the Stokes wave is small then pump-depletion (first term on the RHS of Eqn. 1.57) can be neglected and therefore the growth of the Stokes wave is given by:

$$\frac{\partial I_s}{\partial z} = g_R I_p(0) \exp(-\alpha_p z) I_s - \alpha_s I_s \quad (1.58)$$

This has the straightforward solution for a fibre of length L :

$$I_s(L) = I_s(0) \exp[g_R I_p(0) L_{\text{eff}} - \alpha_s L] \quad (1.59)$$

where $L_{\text{eff}} = \frac{1}{\alpha_p} [1 - \exp(-\alpha_p L)]$ is the effective fibre length. Hence the gain of the Stokes wave is exponential with the pump intensity. Therefore, in the small-signal regime the on-off Raman gain in decibels is given by:

$$G_R = \frac{10 g_R P_p L_{\text{eff}}}{A_{\text{eff}} \ln(10)} = \frac{10 C_R P_p L_{\text{eff}}}{\ln(10)} \quad (1.60)$$

where P_p is the pump peak-power, A_{eff} is the effective mode area and $C_R = g_R / A_{\text{eff}}$ is the Raman gain efficiency. The gain for the Stokes wave can be so high that spontaneously Raman scattered photons are amplified to a level comparable to or greater than the pump, forming Raman amplified spontaneous emission (ASE). It can be shown [Smi72] that if the on-off Raman gain is approximately 70 dB then the amplified spontaneous Raman scattering, approximated as injecting one fictitious photon per mode into the fibre, will be of equal power to the pump at the end of the fibre, assuming the Raman gain spectrum is a single Lorentzian. Despite the rather crude approximations in calculating this critical Raman threshold condition, it is found to be a reasonably accurate measure of when significant Raman ASE will be observed.

Stimulated Brillouin scattering

The process of electrostriction describes the nonlinear effect where an optical field induces stress and strain in a medium, which results in the propagation of an acoustic wave. This acoustic wave travels in the same direction as the optical pump at the acoustic velocity in the medium and results in a periodic modulation of the refractive index due to the

photoelastic effect, since stress and strain cause density changes. The period of the index modulation will be the same as the optical pump and therefore the acoustic wave acts as a travelling reflective Bragg grating. Due to the velocity of the acoustic wave, the backscattered light from the Bragg grating will be frequency downshifted by the Doppler shift associated with the acoustic velocity. Analogously to SRS, the imaginary part of $\chi^{(3)}$ couples the optical pump and the backscattered wave (Stokes wave), enabling energy to be transferred from pump to Stokes wave in the process known as stimulated Brillouin scattering (SBS).

Indeed, the physical process of SBS closely parallels SRS but the the main differences arise from the quanta of molecular energy that mediate the interaction: in SBS it is acoustic phonons rather than optical phonons in the case of SRS. Acoustic phonons have orders of magnitude smaller frequency than optical phonons, and the frequency downshift, Ω , of the Stokes photons is related to the acoustic velocity by:

$$\Omega = \frac{2n_0(\omega_p)v_A}{\lambda_p} \quad (1.61)$$

where v_A is the acoustic velocity, which in fused silica is ~ 6 km/s [Agr07]. Hence, the frequency shift is dependent on the pump wavelength and is ~ 16 GHz for a $1 \mu\text{m}$ pump wavelength in fused silica, nearly three orders of magnitude smaller than the SRS frequency downshift (~ 13.2 THz). The Brillouin gain bandwidth is likewise significantly narrower (typically tens of MHz) than the Raman gain bandwidth as it is proportional to the acoustic phonon lifetime, which is approximately 10 ns in fused silica. From Eqn. 1.61, the Brillouin frequency shift is inversely proportional to the pump wavelength and therefore, the bandwidth of the Brillouin gain spectrum follows a λ_p^{-2} dependence. As a result of this, the peak value of the Brillouin gain is approximately independent of the pump wavelength, in contrast to SRS.

Although SBS can be used for amplification analogously to SRS, the small frequency shift and the fact that it is only phase matched for counter-propagating beams limits its uses, mainly to sensor applications. Indeed, SBS is generally considered a deleterious nonlinear effect because the Brillouin gain coefficient is three orders of magnitude greater than the Raman gain coefficient. This results in a very low peak-power threshold for significant power transfer to the backwards propagating wave: the threshold peak-power is ~ 2 mW for 50% power transfer with 20 km of SMF-28. Fortunately, there are a number of methods for significantly reducing the Brillouin gain coefficient, else high power MOPFA system would be nearly impossible to implement. The main strategy used is to ensure the spectrum of the pump beam is significantly broader than the Brillouin gain bandwidth: the Brillouin gain coefficient reduces by a factor of $1 + \Delta\nu_p/\Delta\nu_B$ for a

Lorentzian pump spectrum of width $\Delta\nu_p$ [Lic87]. Therefore, for pulse durations less than approximately 2 ns, the SBS threshold is raised higher than the SRS Raman threshold and other competing nonlinearities, so it can be neglected. For longer pulse durations or CW operation, if the application does not require high power spectral densities then broad bandwidth seed sources can be used to mitigate SBS (cf. Section 3.1). For applications that require high peak-power spectral densities (e.g. nonlinear frequency conversion) with long pulse durations, then the pump spectrum can be controllably broadened using a phase modulator [Yan96]. Otherwise, special fibre designs [Ver12] or external temperature [Ima93] and strain [Yos93] gradients can be applied to make the Brillouin shift frequency inhomogeneous along the fibre length, disrupting the exponential growth of the backwards propagating wave.

1.3.5 Fibre amplifiers

In the proceeding section, it has been shown that optical fibres are an excellent platform for nonlinear optics due to their waveguiding properties. These properties also make optical fibres an excellent laser gain medium when doped with rare-earth elements, primarily ytterbium (Yb), erbium (Er) and thulium (Tm). The waveguide nature of optical fibres solves two of the major issues encountered in bulk solid-state lasers. Firstly, the vastly larger surface area to volume ratio of an optical fibre in comparison to a laser crystal provides greater dissipation of the heat inherently generated due to the quantum defect. Secondly, the laser and pump beams are guided by the optical fibre structure, which eliminates the need to align the pump beam to the laser mode of a bulk cavity and with the correct geometry, the laser mode can output the fibre with diffraction-limited beam quality. The combination of these two factors has enabled single-mode Yb-fibre lasers to be scaled to 10 kW of average power [Sti09], the highest brightness achieved by a solid-state laser.

The fibre laser was first proposed by Snitzer in 1961 [Sni61], shortly after the demonstration of the first laser, and was followed a few years later by the demonstration of optical gain using a neodymium-doped glass fibre [Koe64]. As Snitzer pointed out in his seminal proposal, the main issue with a single-mode gain fibre is the ability to couple the pump light into the core: the pump laser has to have higher brightness than the output of the fibre laser. Indeed, despite the enormous progress made in increasing the brightness of semiconductor pump lasers [Kan15], single-mode diode lasers for pumping fibre lasers do not currently deliver more than 1 W of average power.

The issue of the pump laser brightness requirement of single-mode gain fibres was solved by the proposal of a double-clad gain fibre by Snitzer [Sni88a]. In a double-clad gain fibre, the signal is guided in a single-mode core that is doped with a rare-earth element

and the lower index cladding surrounding the core is used as a multimode waveguide for guiding the pump light. This is achieved by surrounding the first inner cladding with a second lower index outer cladding, thereby creating a waveguide structure. Typically, the outer cladding is a low index fluoropolymer which results in a cladding NA of ~ 0.46 but a glass cladding can be used to improve the thermal handling properties, at the expense of a reduced NA (~ 0.24).

The large NA, multimode pump waveguide is suitable for coupling in low brightness semiconductor pump laser diodes, whilst the signal retains all the benefits of single mode guidance (e.g. diffraction limited beam quality). Double-clad gain fibres can, therefore, be thought as highly efficient brightness converters: the brightness of the laser output can be 10^5 times greater than the pump laser [Ric10, Zer14]. This enables very high average powers from highly efficient semiconductor pump diodes to be converted into fundamental mode radiation, without suffering from the thermally induced beam quality degradation that plagues bulk solid-state lasers.

Of the rare-earth dopants, neodymium (Nd) was the first to gain traction, with the first gain fibre and single-mode fibre lasers demonstrated using Nd-doped fibres [Koe64, Mea85]. This can mainly be attributed to the prevalent use of Nd-doped media in bulk solid-state lasers and, correspondingly, the maturity of 808 nm pump laser diode technology. Focus then shifted toward Er-doped fibre amplifiers [Mea87], whose broad gain bandwidth around $1.5\ \mu\text{m}$, coinciding with the loss minimum of silica transmission fibres, led them to become the leading technology for optical repeaters in long-haul telecommunications networks.

At a time when all the attention was focused on Er-doped and, to a lesser extent, Nd-doped fibre amplifiers, Hanna et al. suggested that Yb-doped fibres offered a number of significant advantages over Nd-doped fibres [Han88]. The advantages of Yb-doping relate to its comparatively simple energy level structure [Fig. 1.4 (a)] consisting of only two manifolds in the energy range of NIR and visible photons: the ground manifold $^2F_{7/2}$ with four Stark levels and the excited manifold $^2F_{5/2}$ with three Stark levels [Pas95]. The lack of higher energy levels prevents excited state absorption and the large energy gap between the two manifolds suppresses nonradiative decays and quenching of the fluorescence due to energy transfer between closely spaced ions (concentration quenching). The suppressed concentration quenching enables Yb-doped fibres to have higher doping concentrations than, for example Er-doped fibres, which is beneficial for reducing nonlinearities in pulsed fibre amplifiers due to the reduced absorption length.

The absorption and emission cross-sections of a typical aluminosilicate Yb-doped fibre are shown in Fig. 1.4 (b). Cladding-pumped Yb-doped fibre lasers are typically pumped using 976 nm pump diodes, where the absorption cross section is the largest and,

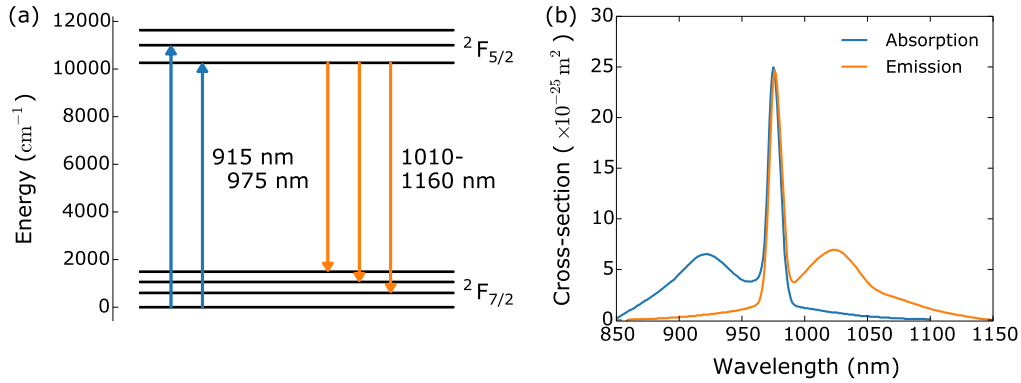


Figure 1.4: (a) After [Pas95]. Yb^{3+} energy level structure consisting of a ground manifold, $^2F_{7/2}$, and an excited manifold, $^2F_{5/2}$. (b) Absorption (blue) and emission (orange) cross-sections of ytterbium-doped aluminosilicate optical fibre as a function of wavelength.

correspondingly, the absorption length is the shortest. The narrow width of the absorption peak at 976 nm results in a tight tolerance on the pump diode central wavelength, which can be difficult to maintain across large temperature and current ranges. To relax this requirement, Yb-doped fibres can be pumped around the broad absorption peak at 915 nm. Pumping at 915 nm also allows for higher inversion levels, since at 976 nm only a 50% inversion level can be achieved as the absorption and emission cross-sections are equal. Of course, for laser emission around the 976 nm peak, the pump must be at a shorter wavelength.

Cladding pumped step-index Yb-doped fibres operate most efficiently between 1060–1100 nm, where the absorption cross-sections are small and therefore nearly four-level operation occurs [Kur07]. When pumped at 976 nm, the quantum defect is $>90\%$, enabling the optical-to-optical efficiencies of Yb-fibre lasers to exceed 80% [Jeo04]. Given that the semiconductor pump diodes can have electrical-to-optical efficiencies $>60\%$ at full power [Gap14], the wall-plug efficiency of commercial Yb-fibre lasers can exceed 40%. For high peak-power amplification, where the use of short lengths is necessary to mitigate nonlinearities, a signal wavelength close to the emission peak at 1030 nm is usually preferred to give the highest gain per unit length. Operation of Yb-doped fibre amplifiers below ~ 1040 nm is three-level [Pas97] and therefore higher pump intensities are required, which can have a detrimental impact on efficiency due to unabsorbed pump light.

To operate Yb-doped fibre lasers at wavelengths beyond ~ 1100 nm, the main issue is suppressing ASE and parasitic lasing occurring around the gain peak at 1030 nm, due to the much larger relative emission cross-section. In lasers, this can be achieved with high-

Q cavities that have very large losses for the higher gain shorter wavelengths [Ota06] or by heating the fibre to shift the absorption and emission cross-section to longer wavelengths [Kur07]. These techniques are not effective for Yb-doped fibre amplifiers operating >1100 nm, where photonic bandgap fibres are typically used; these fibres feature high transmission losses to the higher gain shorter wavelengths, enabling amplification of signal wavelength up to 1180 nm [Shi10]. Even so, only moderate gain can be extracted with long fibre lengths and the microstructured nature of the photonic bandgap fibres preclude the splicing and integration with standard fibres.

MOPFAs

Master oscillator power amplifier (MOPA) schemes, where a low power seed laser is amplified in a single amplifier or chain of amplifiers, are a ubiquitous technique for scaling the average output power of laser systems. By decoupling the signal generation from the amplification, it enables the design of the master oscillator to be optimised for the desired temporal and spectral characteristics without having to engineer it to generate significant average power. Indeed, in many cases it would be impossible to scale the output power of the seed lasers to the desired level due to constraints imposed by the gain media: thermal issues and nonlinear effects most notably. An additional advantage of the MOPA concept is that different gain media can be used for the oscillator and the amplifier(s), enabling the specific benefits of different technologies to be exploited in a single system.

MOPA schemes using fibre amplifiers, referred to as master oscillator power fibre amplifier (MOPFA) schemes herein, in many ways exemplify the benefits of the MOPA concept. As was seen in the previous section, fibre amplifiers can deliver kW saturated output average powers, whilst maintaining a constant near diffraction-limited mode profile that is independent of the output power. In contrast, high average power bulk solid-state amplifiers have to balance thermal lensing and aberrations, therefore, the beam profile is generally optimised for a specific output power and it is not possible to achieve as high beam quality [Goo06, Rus10]. Furthermore, the broad gain bandwidths of fibre amplifiers can support subpicosecond pulse durations or alternatively it allows flexibility in the central wavelength of the seed laser, covering the emission wavelengths of many different semiconductor and solid-state media.

A MOPFA scheme that is implemented multiple times in this thesis utilises a pulsed semiconductor laser diode as the master oscillator, which is then amplified to Watt-level average power using doped-fibre amplifiers. As reviewed in Section 3.1, pulsed semiconductor laser diodes are very flexible in pulse duration and repetition rate but they can only deliver limited average power and pulse energy. Therefore, the high gain and

high average power capability of fibre amplifiers is well suited to amplifying the pulsed semiconductor signals with high wall-plug efficiency. Furthermore, the semiconductor diodes and the optical components required between amplification stages (e.g. isolators, filters, polarisers etc.) are available in packages that are fibre pigtailed, thus the whole MOPFA chain can be fibre-integrated. This results in alignment-free sources, that can potentially be very compact and mechanically robust, broadening the number of environments that they could be deployed in. The power scalable, temporally flexible nature of such MOPFA schemes, combined with the excellent beam quality, make them ideal platforms for nonlinear frequency conversion. This enables the benefits of the MOPFA scheme to be realised in the many spectral regions that are not covered by rare-earth doped fibre amplifiers.

1.3.6 Large mode area fibres

The long interaction lengths of fibre amplifiers in comparison to bulk solid-state laser gain media, whilst beneficial for achieving high gain with good thermal management, poses a significant obstacle for obtaining high output peak-powers. Most of the nonlinear effects described in Section 1.3.4 (e.g. SPM, SRS and SBS) are automatically phase matched and, therefore, will occur if the peak-power length product is sufficiently high. Figure 1.5 illustrates the peak-power limitations imposed by the main nonlinear effects by plotting their average power and pulse energy thresholds as a function of the full width at half maximum (FWHM) pulse duration of transform-limited Gaussian pulses. All of the threshold calculations are based on a 3 m passive fibre with an effective mode-area $A_{\text{eff}} = 511 \mu\text{m}^2$, typical for a 30 μm core-diameter step-index fibre with an NA = 0.06 [Sch08], and a linearly polarised signal with a central wavelength of 1064 nm. The SRS and SBS limits are based on the equal power threshold conditions in [Smi72]. The SBS threshold assumes that the Brillouin gain coefficient is reduced by $\left(1 + \frac{\Delta\nu_s}{\Delta\nu_B}\right)$, where $\Delta\nu_s$ is the signal bandwidth including SPM broadening and $\Delta\nu_B = 100$ MHz is the Brillouin gain bandwidth. The SPM threshold is defined as the peak-power required to broaden the pulse spectrum to a FWHM of 0.25 nm and the self-focusing threshold is 4 MW at the signal wavelength, based on the calculations in [Far06a].

It is important to stress that these thresholds are quite arbitrary: all of the nonlinear effects bar critical self-focusing occur below these threshold values, it is dependent on the application to what extent the nonlinearities can be tolerated. For example, the pulse quality of an ultrafast linear chirped-pulse amplification (CPA) system is highly dependent on the total nonlinear phase that is accumulated by the signal and so even small amounts of SPM cannot be tolerated [Lim06]. In contrast, the quality of the temporal and spectral characteristics of high pulse energy fibre amplifier systems used for materials processing

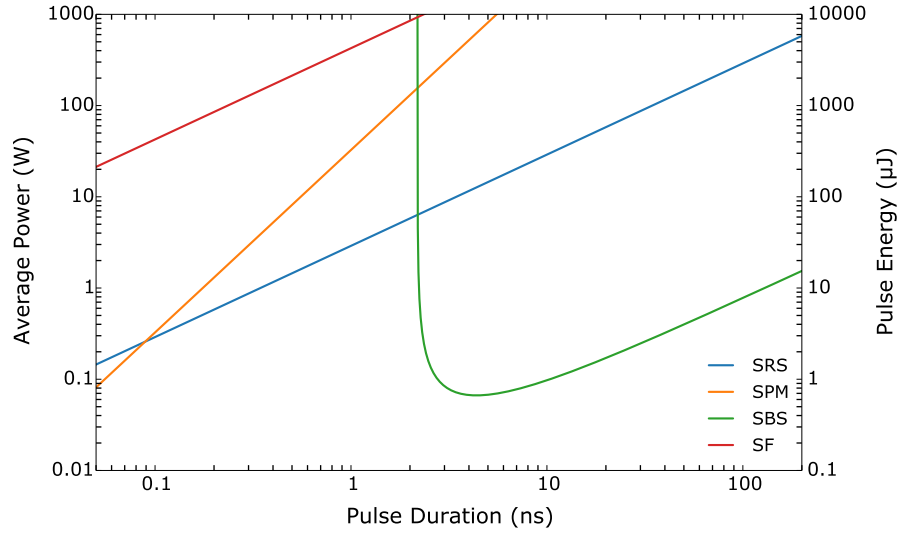


Figure 1.5: Peak-power thresholds of nonlinear effects in optical fibres as a function of the FWHM pulse duration of transform-limited Gaussian pulses, quoted as an average power and pulse energy for a repetition rate of 100 kHz. SF, self-focussing.

are less important; the main issue is the peak-power of the signal being diminished by energy transfer to other wavelengths (due to SRS and FWM) or backscattered light (due to SBS) [Vu06, Zer06].

Self-focusing (red line Fig. 1.5), where the intensity dependent refractive index causes a focusing of the Gaussian mode such that the intensity exceeds the catastrophic optical damage threshold of silica, is the only nonlinear effect that is independent of the mode-area of the fibre. As such, self-focusing represents the upper limit on the peak-power that can be achieved by a fibre amplifier, which is ~ 4 MW for silica fibres with a signal wavelength of $1 \mu\text{m}$ [Far06a]. The self-focusing critical peak-power is proportional to the square of the signal wavelength, therefore, moving to Tm-doped fibre amplifiers that operate around $2 \mu\text{m}$ is seen as a promising avenue for scaling the peak-power of MOPFA systems [Stu15].

The spectral broadening due to SPM is dependent on the power-length product and input bandwidth, for initially unchirped pulses [Agr07]. If a high power spectral density is required (e.g. for nonlinear frequency conversion), it is the limiting nonlinearity for pulse durations approximately < 100 ps (orange line Fig. 1.5). For pulse durations approximately > 2 ns, that have a spectral bandwidth of the order of the Brillouin gain bandwidth (50–100 MHz in silica fibres), SBS is the limiting nonlinearity. The SBS threshold increases sharply around 2 ns (green line Fig. 1.5) because the transform-limited bandwidth begins

to exceed the Brillouin gain bandwidth and the pulse bandwidth is further broadened by SPM. Additionally, the reduced overlap between the pump and the backscattered pulse decreases the Brillouin gain. For pulse durations in the range 0.1–2 ns or pulses with bandwidths exceeding the Brillouin gain bandwidth, SRS is the limiting nonlinearity (blue line Fig. 1.5). When the Raman gain is high enough, the Stokes wave grows from noise and once it reaches a reasonable intensity, power is rapidly transferred from the pump to the Stokes, diminishing the the pump peak-power (cf. Section 1.3.4).

To increase the threshold peak-power of deleterious nonlinear effects, the mode-field area of the gain fibre must be increased. Generally, to maintain the benefits of double-clad fibres as gain media, the core must guide in the fundamental mode for the signal wavelength, which in a step-index fibre can be guaranteed by ensuring that $V = 2\pi a \text{NA} / \lambda < 2.405$ (cf. Section 1.3.1). Hence, to increase the mode-field diameter by increasing the core diameter ($2a$), the NA must be correspondingly decreased to maintain single-mode guidance. These fibres are known as large mode-area (LMA) fibres and this technique has been successfully applied to step-index fibres but there are limitations on how small the NA can be reduced to [Li09, Ric10]. Firstly, it becomes increasingly more difficult to fabricate the fibres, as smaller NAs require the index contrast between the core and the inner cladding to be reduced. Indeed, only recently [Jai15] has a step-index fibre with a core NA=0.038 been demonstrated, which corresponds to a index contrast of only $\Delta n = 5 \times 10^{-4}$. Secondly, as the core NA is reduced, the waveguiding becomes weaker and, therefore, the minimum bend radius that doesn't induce significant bend losses becomes impractically large.

In practice, it is possible to obtain fundamental mode guidance in active fibres that support a few modes ($V \sim 10$) with the correct launch conditions [Fer98] and by coiling the fibre to a radius that induces substantial bend losses for the higher-order modes without attenuating the fundamental mode significantly [Kop00]. Commercially available double-clad gain fibres have NAs as low as 0.06 and using these techniques, near diffraction-limited beam quality can be obtained using fibres with a core diameter of 30 μm ($A_{\text{eff}} \sim 510 \mu\text{m}^2$) [Far06b].

The manufacturing issue of fabricating low NA fibres can be circumvented by the use of microstructured air-hole claddings in photonic crystal double-clad fibres [Lim04, Lim05a]. Indeed, core diameters up to 100 μm have been demonstrated [Bro06, Eid11] with fundamental mode guidance, but the effective core NA is so low that the 'fibres' must be kept straight. Further increases in the core diameter to 135 μm with effective single-mode guidance has been achieved in large-pitch fibres [Stu12, Lim12b], which delocalise the higher-order modes such that most of their energy is outside the doped core, making them difficult to excite and reducing their gain. All PCF fibres with core diameters

>50 μm have to be kept straight but they have the advantage of high cladding NAs, even with comparatively small pump claddings, resulting in very high pump absorption [Stu12]. These so called rod-type PCFs exhibit the largest mode-areas for active fibres (up to $A_{\text{eff}} \sim 8600 \mu\text{m}^2$) but they lack most of the practical benefits of the fibre geometry and are susceptible to mode instabilities at high average powers due to thermal issues [Jau13].

Many other LMA active fibre designs exist, for example: leakage channel [Bar12, Gu13]; chirally coupled core [Ma14]; single and multi-trench [Jai14a, Jai14b]; solid-core photonic bandgap [Dia09, Gu14] and parabolic [Fin13], representative of the vast research effort into scaling the mode-field area of active fibres. None of these designs currently, however, offer the performance of rod-type PCFs or are able to be manufactured on the mass scale of step-index doubled-clad fibres. Indeed, despite the smallest mode-field area, step-index double-clad fibres are the most widely used due to their cost effectiveness and their ability to be spliced to components such as fused pump combiners and fibre Bragg gratings.

1.4 SHG using nonlinear crystals

In the previous section, it was shown that optical fibres are a versatile platform for optical transmission, nonlinear optics and high-power lasers. Second-order nonlinear effects, however, are generally not observed in silica optical fibres as they are dipole forbidden by the inversion symmetry of the amorphous glass matrix. Electric quadrupole moments in conjunction with photoinduced changes in the fibre structure can, however, lead to reasonably significant SHG occurring [Öst86]. Indeed, SHG was observed in the pulse-pumped Raman amplifiers in Chapters 3 and 4 but the conversion efficiency did not exceed 1%. Second-order nonlinear processes are technologically significant for frequency conversion of well-developed NIR laser systems into the ultraviolet (UV), visible and mid-infrared (MIR). This section covers SHG in non-centrosymmetric media, with an emphasis on periodically poled nonlinear crystals that are used throughout this thesis.

1.4.1 Second-harmonic generation

Second-harmonic generation (SHG) was the first optically-driven nonlinear effect to be demonstrated [Fra61] and since then, it has become the most commercially significant nonlinear effect, primary for the frequency-doubling of NIR lasers into the visible. SHG describes the parametric $\chi^{(2)}$ effect where two pump photons are annihilated and a single photon of twice the frequency is created. In effect, SHG can be considered as a degenerate case of sum-frequency generation, where the two input frequencies are identical. Since it is a parametric effect, the energy and momentum of the three interacting photons must

be conserved, which gives the relation:

$$\hbar k(\omega) + \hbar k(\omega) = \hbar k(2\omega) \quad (1.62)$$

where $k(\omega) = \frac{n(\omega)\omega}{c}$ is the magnitude of the wave vector. Therefore, Eqn. 1.62 simplifies to:

$$n(\omega) = n(2\omega) \quad (1.63)$$

Due to dispersion, Eqn. 1.63 is not normally satisfied in nonlinear crystals unless special measures are taken. Therefore, there exists a phase-mismatch, given by:

$$\Delta k = k(2\omega) - 2k(\omega) = \frac{2\omega}{c} [n(2\omega) - n(\omega)] \neq 0 \quad (1.64)$$

which results in a periodic flow of power back and forth between the fundamental and second-harmonic due to the accumulated phase slip between the two waves. This occurs over a length scale known as the coherence length, $L_{\text{coh}} = \pi/|\Delta k|$. By solving the coupled wave equations for fundamental and second-harmonic plane waves, one can show that in the case of negligible pump depletion, the second-harmonic intensity at the end of a crystal of length L is given by:

$$I_{2\omega}(L) = \frac{2\omega^2 d_{\text{eff}}^2}{n(2\omega)n(\omega)^2 \epsilon_0 c^3} \text{sinc}^2\left(\frac{\Delta k L}{2}\right) L^2 I_{\omega}^2(0) \quad (1.65)$$

where d_{eff} is the effective nonlinear coefficient, which is related to which elements of the $\chi^{(2)}$ tensor are engaged in the interaction. Figure 1.6 shows the second harmonic intensity as a function of propagation distance in a nonlinear crystal, in units of the coherence length, for illustrative phase-matching cases. In the perfectly phase-matched case, $\Delta k = 0$, the second-harmonic intensity grows quadratically with increasing crystal length (blue line Fig. 1.6) and is proportional to the square of the pump intensity and the nonlinear coefficient. Whereas, if there exists a finite phase-mismatch due to dispersion, $\Delta k \neq 0$, the second-harmonic intensity varies sinusoidally with a period of $2L_{\text{coh}}$ (orange line Fig. 1.6). In this case, the maximum second-harmonic intensity occurs for a crystal length of L_{coh} and odd multiples of it.

1.4.2 Birefringent phase-matching

Perfect phase-matching ($\Delta k = 0$) can be achieved in the presence of dispersion by noting that 18 of the 21 non-centrosymmetric crystal classes are anisotropic and, therefore, they exhibit birefringence: different crystal axes have different refractive indices. In uniaxial birefringent crystals, only one axis has a different refractive index, referred to as the

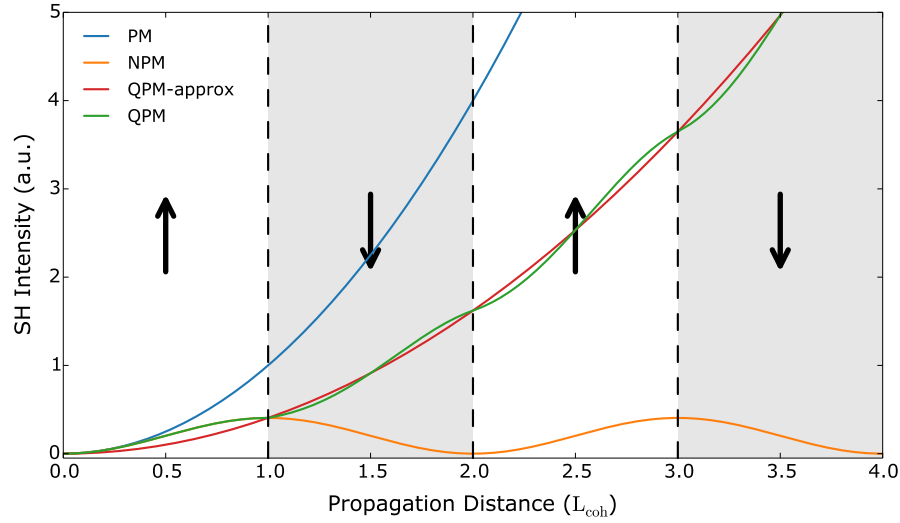


Figure 1.6: Second-harmonic (SH) intensity growth as a function of propagation distance for phase matched (PM), non-phase matched (NPM) and quasi-phase matched (QPM) cases. For QPM, the arrows denote the domain orientation. The QPM case can be approximated with a parabola by setting $d_{\text{QPM}} = \frac{2}{\pi} d_{\text{eff}}$ (QPM-approx).

extraordinary refractive index $[\bar{n}_e(\omega)]$. A beam propagating along the optic axis - the polarisation direction that experiences the extraordinary refractive index - will experience the ordinary refractive index $[n_o(\omega)]$ regardless of its polarisation. If, however, a beam propagates at angle θ to the optic axis, the polarisation perpendicular to the plane containing the wave vector will experience $n_o(\omega)$ (ordinary wave) and the orthogonal polarisation (extraordinary wave) will experience a refractive index between n_o and $\bar{n}_e$, dependent on the angle [i.e. $n_e(\omega, \theta)$]. In a negative uniaxial crystal, $n_e(\omega, \theta) < n_o(\theta)$, therefore, the normal dispersion that results in $n(2\omega) > n(\omega)$ can be counteracted by choosing θ such that $n_e(2\omega, \theta) = n_o(\omega)$. Hence, perfect phase-matching has been achieved, which is known as critical birefringent phase-matching (BPM) [Mak62, Gio62] because of the sensitivity to the propagation angle.

The critical BPM scheme described above is known as Type-I phase-matching, where the second-harmonic photons are polarised orthogonally to the two fundamental photons. Type-II phase-matching, where the two fundamental photons are polarised orthogonally can also be used to achieve BPM for certain interactions in uniaxial and biaxial crystals. The main drawback of critical BPM arises from the unusual propagation properties of the aptly-named extraordinary wave. The response of the medium is not parallel to the electric field of the extraordinary wave; therefore, the Poynting vector, which determines

the direction of energy flow of an electromagnetic wave, is not parallel to the wave vector of the extraordinary wave. The result is that, unlike waves propagating in homogeneous media, the energy flow of the extraordinary wave propagates at an angle to its wave vector. Since the wave vectors of the fundamental and second-harmonic are in the same direction in SHG, this results in the extraordinary wave diverging away from the ordinary wave. After some propagation distance, this angular divergence results in spatial walk-off of the two beams, resulting in the double-refraction that give birefringent crystals their name. The spatial walk-off is an issue for SHG since it limits the effective length of the interaction and can result in an elliptical second-harmonic beam.

Spatial walk-off can be avoided by propagating perpendicular to the optic axis [$n_e(\omega, \theta = 90^\circ) = \bar{n}_e(\omega)$], where the Poynting vector of the extraordinary wave is parallel to its wave vector. To achieve phase-matching, the fact that $\frac{\partial}{\partial T} n_o \neq \frac{\partial}{\partial T} \bar{n}_e$ can be used such that at a certain temperature, $\bar{n}_e(2\omega, T) = n_o(\omega, T)$. Since $\frac{\partial}{\partial T} (n_o - \bar{n}_e) \sim 10^{-6}/\text{K}$, the phase-mismatch must be relatively small at room temperature to enable correction within the temperature range that a nonlinear crystal can feasibly be held at ($\sim 15\text{--}250^\circ\text{C}$). This type of phase-matching is referred to as non-critical phase-matching (NCPM), since the angle tolerance of the SHG conversion efficiency is significantly wider than for critical phase-matching. Lithium triborate (LBO) is the most prevalent nonlinear crystal used for NCPM since it can be phase-matched for SHG between 1064–1585 nm in the temperature range 25–150°C. The additional advantage of NCPM is that the spectral acceptance bandwidth is broader than in critical phase-matching, for example the spectral acceptance bandwidth of SHG of 1064 nm in LBO using NCPM is $\Delta\lambda = 3.83 \text{ nm} \cdot \text{cm}$.

1.4.3 Quasi phase-matching

The main hindrance to implementing BPM is that nonlinear crystals with specific birefringent properties are required for different second-order interactions and for different fundamental wavelengths. Additionally, the crystal must also have: a high nonlinearity, high optical quality, high damage threshold, be able to be grown to sufficient aperture and length and generally have good physical and chemical properties. These severe restrictions mean that despite the extraordinarily large number of crystalline solids, typically only a few crystals are suitable for certain interactions and for many interactions no options exist.

Phase-matching is necessary in order to obtain coherent growth of the second-harmonic, resulting in a quadratic build up of the second-harmonic intensity with propagation distance (blue line Fig. 1.6). An alternative strategy to obtain quadratic growth of the second-harmonic in the presence of a finite phase-mismatch ($\Delta k \neq 0$) is to invert the orientation of the crystal structure every coherence length. The sign reversal of the non-

linear coefficient after the first coherence length results in the power continuing to flow from the fundamental to the second-harmonic, rather than flowing from the second-harmonic back to the fundamental as usually occurs (orange line Fig. 1.6). The resulting second-harmonic growth (green line Fig. 1.6) is well approximated by a quadratic (red line Fig. 1.6), thus achieving so called quasi phase-matching (QPM) [Arm62]. From Fig. 1.6, it is clear that the gradient of the approximate quadratic for QPM is reduced compared with the perfectly phase-matched case, which corresponds to a reduction in the effective nonlinearity of $d_{\text{eff}}^{\text{QPM}} = \frac{2}{\pi} d_{\text{eff}}$.

At first, it might seem counterproductive to propose implementing QPM by sectioning an optical crystal on a length scale that is typically a few microns, to achieve a lower effective nonlinearity than in an un-sectioned crystal using BPM. The real power in the QPM technique, however, is that potentially any second-order nonlinear interaction within the transparency window of the crystal can be phase-matched, so long as the correct inversion period ($2L_{\text{coh}}$) can be achieved. Indeed, the complete freedom from birefringence means that crystals with much higher nonlinear coefficients can be used for interactions that they would otherwise be unable to phase-match. Furthermore, since the use of birefringence isn't necessary, one is free to choose the polarisations of the interacting waves, which allows spatial walk-off to be avoided. Usually, Type-0 phase-matching is implemented, where all three waves have the same polarisation, a combination that is not possible with BPM. This has the great advantage that the diagonal $\chi^{(2)}$ tensor elements, which are often the largest, can be engaged by the interaction, something that isn't possible with BPM.

For first-order QPM, which has the highest effective nonlinear coefficient, the period of the crystal domain inversion, Λ , needed to compensate the phase-mismatch is given by:

$$\Lambda = 2L_{\text{coh}} = \frac{2\pi}{|\Delta k|} \quad (1.66)$$

Hence the momentum conservation relation becomes:

$$\Delta k = 4\pi \left(\frac{n(2\omega)}{\lambda} - \frac{n(\omega)}{\lambda} - \frac{1}{2\Lambda} \right) = 0 \quad (1.67)$$

where λ is the fundamental wavelength with frequency ω . Effectively, the wave vector of the domain inversion grating compensates the phase-mismatch caused by dispersion.

The main disadvantage of QPM is that the SHG conversion efficiency typically rolls off more quickly with deviations in the fundamental wavelength or the crystal temperature in comparison to BPM. From Eqn. 1.65, the SHG conversion efficiency will fall to 50% of the maximum value when the phase-mismatch is $|\Delta k| = 2 \times 0.4429\pi/L$, such that $\text{sinc}^2(\Delta k L/2) = 1/2$. The FWHM spectral acceptance bandwidth is defined as the total

deviation either side of the phase-matched fundamental wavelength that causes the SHG conversion efficiency to fall to 50% of the maximum, given by:

$$\Delta\lambda \approx \frac{4 \times 0.4429\pi}{L} \left(\frac{\partial \Delta k}{\partial \lambda} \right)^{-1} = \frac{0.4429\lambda}{L} [n_g(2\omega) - n_g(\omega)]^{-1} \quad (1.68)$$

where L is the crystal length, n_g is the group refractive index. As an example, the spectral acceptance bandwidth of a 20 mm stoichiometric periodically poled lithium tantalate (PPLT) crystal for SHG of 1064 nm at a phase-matching temperature of 130°C (7.8 μm grating) is 0.12 nm. This corresponds to transform-limited sech^2 pulse duration of 10 ps and imposes a severe limitation on the amount of SPM spectral broadening that can be tolerated in a MOPFA system.

When the temperature of a QPM crystal is varied, the phase-mismatch changes due to both the change in refractive index with temperature and additionally from the change in the grating period due to thermal expansion. Therefore, the FWHM temperature bandwidth is given by:

$$\Delta T \approx \frac{4 \times 0.4429\pi}{L} \left(\frac{\partial \Delta k}{\partial T} \right)^{-1} = \frac{0.4429\lambda}{L} \left[\frac{\partial \Delta n}{\partial T} + \alpha \Delta n \right]^{-1} \quad (1.69)$$

where $\Delta n = n(2\omega) - n(\omega)$ and α is the linear thermal expansion coefficient. For the example crystal used to illustrate the spectral acceptance bandwidth, the temperature bandwidth is 1.2°C. Whilst this is significantly broader than the temperature precision of most crystal ovens (typically 0.1°C), small variations in the crystal temperature lead to power fluctuations in the second-harmonic, even if the fundamental power is constant.

1.4.4 Materials for QPM

The first demonstration of QPM using domain inversion involved frequency-doubling a CO_2 laser using an alternating stack of Brewster angled, non-birefringent cadmium telluride (CdTe) plates, each with an optical path length of 950 μm ($5L_{\text{coh}}$) [Pil76]. For SHG of light around 1 μm , the coherence length is typically under 10 μm , making it unfeasible to have physically separated inverted crystal domains. Due to this, using QPM for SHG did not gain traction until the post-growth periodic polling of lithium niobate (LiNbO_3) using photolithographic phase masks was first reported in 1989 [Lim89].

LiNbO_3 belongs to the family of ferroelectric crystals that exhibit an electric dipole moment even in the absence of an external electric field, analogous to the permanent magnetic moment of ferromagnetic materials, despite the fact that most ferroelectric crystals do not contain iron. The most important aspect of the ferroelectricity is that the spontaneous dipole moment can be permanently reversed by applying a large static

electric field, referred to as the coercive field. Photolithographic techniques can be used to pattern a uniform periodic electrode on the surface of a ferroelectric crystal, enabling a periodical alternating static electric field to be applied; thereby permanently inducing a periodic reversal of the crystal domains [Mat91]. The maturity of the photolithographic process, inherited from semiconductor manufacturing, enabled the micron period, highly uniform periodically poled structures that are required for visible SHG to be implemented.

LiNbO_3 is the most widely used periodically poled ferroelectric crystal for QPM of second-order nonlinear interactions due to its very high nonlinearity and wide transparency between 0.4–5 μm . QPM interactions in LiNbO_3 typically use the Type-0 scheme, with propagation in the x-axis and all the waves extraordinarily polarised in the z-axis, since this couples to the largest component of the susceptibility tensor $d_{33} = 25 \text{ pm/V}$ at 1 μm [Sho97]. After the $2/\pi$ reduction in the nonlinear coefficient due to QPM and further reductions from polling imperfections the nonlinear coefficient is typically $d_{\text{eff}}^{\text{QPM}} \sim 16 \text{ pm/V}$. This is more than three times greater than the nonlinear coefficient used in Type-I BPM of SHG at 1 μm ($d_{31} = 4.6 \text{ pm/V}$ [Sho97]). For the same crystal length, this results in a twelve-fold increase in the SHG conversion efficiency using QPM as opposed to BPM, which enables laser sources with significantly lower peak-power (e.g. CW lasers) to be frequency-doubled with reasonable efficiency [Mil97].

The main drawback of LiNbO_3 is that it has a low resistance to photorefractive damage (PRD), which is caused by the release of free electrons into the conduction band due to photoexcitation from impurity energy levels that lie between the valance and conduction bands of LiNbO_3 . The photogenerated electrons diffuse from the intense part of the beam that created them to regions of lower optical intensity, where they recombine with holes and return to an impurity level, thereby becoming trapped. The electric field (space charge field) that results from the holes left in the intense region causes a spatially varying refractive index due to the electro-optic (Pockels) effect. This refractive index grating distorts the wavefronts of the optical beam and, therefore, leads to beam quality degradation. Furthermore, at sufficiently high optical intensities, the self-diffraction of the second-harmonic beam can lead to catastrophic optical damage.

Photorefractive is a particular issue for visible light generation using SHG because the rate of photoexcitation of free electrons is far higher for visible photons in comparison to NIR photons and also high intensities are desired to achieve high SHG conversion efficiencies. A two-centre model of photorefractive is currently accepted [Jer93], where a defect (normally $\text{Fe}^{2+}/\text{Fe}^{3+}$) acts as the photovoltaic centre creating the free electrons and trapping them and a second defect (normally a $\text{Nb}_{\text{Li}}^{4+}/\text{Nb}_{\text{Li}}^{5+}$ antisite) contributes to the electron transport. It has been shown that the photorefractive can be significantly suppressed by doping LiNbO_3 with MgO [Bry84] and obtaining a closer to stoichiomet-

Property	Material		
	PPLN	PPLT	PPKTP
Transparency	0.32-5.0 μm	0.28-5.5 μm	0.35-4.5 μm
d_{33} at 1 μm	25.0 pm/V	13.8 pm/V	14.6 pm/V
Typical $d_{\text{eff}}^{\text{QPM}}$	16 pm/V	9 pm/V	10 pm/V
α at 1 μm	0.003 cm^{-1}	0.002 cm^{-1}	0.003 cm^{-1}
α at 0.5 μm	0.02 cm^{-1}	0.02 cm^{-1}	0.05 cm^{-1}
κ at 300 K	3.9 W/mK	8.5 W/mK	3 W/mK
I_{PRD} at 0.5 μm	$\sim 10 \text{ MW/cm}^2$	$> 100 \text{ MW/cm}^2$	$> 100 \text{ MW/cm}^2$

Table 1.1: After [Nik05]. Comparison of periodically poled ferroelectric crystals. PPLN, 5 mol.% MgO-doped congruent periodically poled lithium niobate (LiNbO_3); PPLT, periodically poled stoichiometric lithium tantalate (LiTaO_3); PPKTP, flux-grown periodically poled potassium titanyl phosphate (KTiOPO_4).

ric composition (50:50 ratio of $\text{Li}_2\text{O}:\text{Nb}_2\text{O}_5$ oxide concentrations) [Ang72] due to the reduction in the number of Nb_{Li} antisite defects and the associated increase in photoconductivity.

PRD limits the use of LiNbO_3 for applications that require high peak-power visible radiation to be generated, since the PRD threshold of 5 mol.% MgO-doped periodically poled lithium niobate (PPLN) is generally considered to be approximately 10 MW/cm^2 for green light [Kie12]. The two most widely used periodically poled ferroelectric crystal alternatives to PPLN are periodically poled lithium tantalate (PPLT) and periodically poled potassium titanyl phosphate (PPKTP), whose material properties are summarised alongside PPLN in Table 1.1. Lithium tantalate (LiTaO_3) is closely related to LiNbO_3 and whilst it has a lower nonlinearity ($d_{33} = 14 \text{ pm/V}$ [Sho97]), its PRD threshold is an order of magnitude higher at $> 100 \text{ MW/cm}^2$. In the same manner as LiNbO_3 , the PRD threshold of LiTaO_3 can be significantly increased by either doping with MgO or by increasing the stoichiometry [Nak02]. Potassium titanyl phosphate (KTiOPO_4) has a very similar nonlinearity to LiTaO_3 but has the advantage of a low susceptibility to photorefractive and high surface damage thresholds.

The PRD thresholds of PPLT and especially PPKTP are high enough that generally they are not a limiting factor in high peak-power visible generation. Instead, photochromic effects, where the high intensity visible light causes a nonlinear increase in absorption, can lead to a disruption of the QPM, reducing the SHG conversion efficiency. In PPLT, the predominant photochromic effect is green-induced infrared absorption (GRIIRA), where

high intensity visible light increases the absorption of the fundamental infrared radiation. In common with PRD, the level of GRIIRA is related to the number of Ta_{Li} antisite defects and is thought to result from a small polaron absorption band in the NIR [Fur01].

In addition to GRIIRA, PPKTP also suffers from photochromic damage known as grey tracking, due to the visible dark tracks that form when a crystal has been irradiated with high intensity visible light. Gray tracking has a similar mechanism to PRD: high intensity light causes free electrons to be released due to photoexcitation from Fe^{3+} impurity levels, which are trapped by the Ti^{4+} ion, resulting in the formation of a Ti^{3+} colour centres [Wan04]. The Ti^{3+} colour centres have a broad absorption feature around 500 nm [Scr95], which can lead to catastrophic optical damage if the high intensity visible light continues to irradiate the crystal.

1.4.5 SHG conversion efficiency

The conversion efficiency of the SHG process is the most important parameter for determining whether it is feasible for it to be used as a practical method for nonlinear frequency conversion. In the low conversion limit, where pump depletion can be neglected, it was already shown that an analytic solution to the coupled-wave equations exists for a plane-wave interaction (Eqn. 1.65). Using this, the SHG conversion efficiency for a crystal of length L , in the case of plane-waves in the low conversion limit, is given by:

$$\eta = \frac{I_{2\omega}(L)}{I_{\omega}(0)} = \frac{8\pi^2 d_{\text{eff}}^2}{n(2\omega)n(\omega)^2 \epsilon_0 c \lambda^2} \text{sinc}^2\left(\frac{\Delta k L}{2}\right) L^2 I_{\omega}(0) \quad (1.70)$$

where λ is the wavelength of the fundamental. Clearly, the SHG conversion efficiency is proportional to the fundamental beam intensity, therefore, in almost all practical implementations of SHG, the fundamental beam is focused into the nonlinear crystal. In the case of a focused fundamental beam, the plane-wave assumption is clearly no longer valid. The SHG conversion efficiency in the case of focused Gaussian beams, typical of the output of most lasers (cf. Section 1.2.2), was derived by Boyd and Kleinman in the low conversion limit [Boy68]:

$$\eta = \frac{16\pi^2 d_{\text{eff}}^2}{n(2\omega)n(\omega)\epsilon_0 c \lambda^3} L P_{\omega} h(\kappa, B, \mu, \xi, \sigma) \quad (1.71)$$

where P_{ω} is the fundamental peak-power, $h(\kappa, B, \mu, \xi, \sigma)$ is the Boyd-Kleinman h-factor with κ related to the crystal absorption of the fundamental and second-harmonic, B is the Poynting vector walk-off, μ is the position of the focus, $\xi = L/b$ is the focusing parameter (where b is the confocal parameter) and $\sigma = b\Delta k/2$ is the phase-mismatch.

The functional dependence of the h-factor can be significantly simplified in the case of

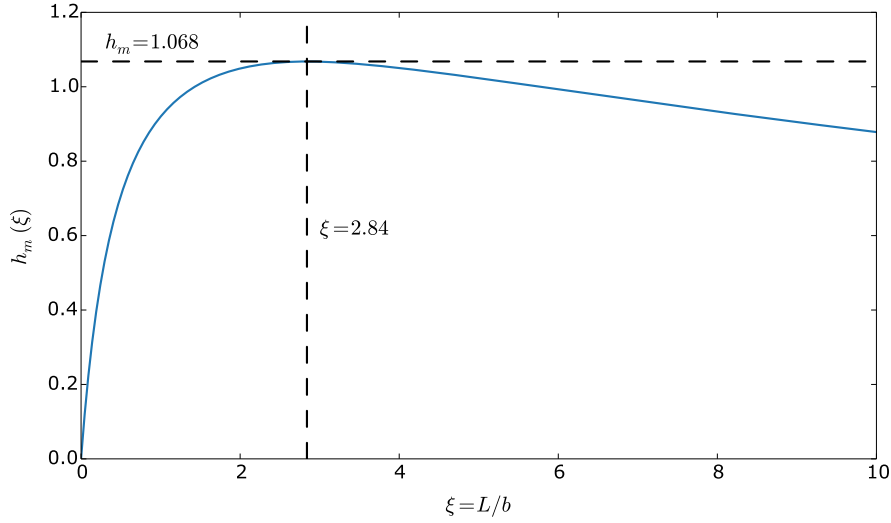


Figure 1.7: Boyd-Kleinman h-factor with optimal phase-mismatch as a function of focusing parameter, ξ , in the case of zero Poynting vector walk-off. The focusing parameter is defined as the ratio of the crystal length L to the confocal parameter b of the focused Gaussian beam in the crystal. This analytic function, derived in [Che03], is an approximation of the numerical solution.

obtaining maximal SHG conversion efficiency using NCPM, which is normally true for SHG using periodically poled crystals. In this case, it can be assumed that: the crystal absorption is negligible ($\kappa = 0$); there is no Poynting vector walk-off ($B = 0$); the focus is at the centre of the crystal ($\mu = 0$) and σ is optimised such that $h_m(\xi) = \max[h(\sigma, \xi)]_\sigma$. The function $h_m(\xi)$ is a two-dimensional integral that has no analytic solution and must, therefore, be solved numerically. It was shown in [Che03], however, that the solution is well-approximated by an analytic Lorentzian line-shape function. This Lorentzian is plotted in Fig. 1.7 in the case of zero Poynting vector walk-off ($B = 0$), which is the case when using NCPM. From Fig. 1.7, $h_m(\xi)$ has a single maximum value of 1.068 for a focusing parameter $\xi = 2.84$, the so-called Body-Kleinman focusing condition, which gives the largest theoretical SHG conversion efficiency.

It is worth noting that, in contrast to the plane-wave case, the optimum phase-mismatch when using a focused fundamental beam is positive and non-zero. In practice, however, this is rarely noticed since the temperature of the periodically poled crystal is adjusted to give the optimum SHG conversion efficiency. The shift in the optimum phase-matching temperature from the theoretical plane-wave optimum is difficult to observe due to the poor calibration of the absolute crystal temperature as recorded by the temperature controller.

The most important implication of the SHG conversion efficiency expression using focused Gaussian beams (Eqn. 1.71) in comparison to the plane-wave result (Eqn. 1.70) is that the dependence on the crystal length is linear rather than quadratic. This is due to the fact that for a Gaussian beam, the intensity is inversely proportional to its confocal parameter, the length range over which the peak intensity is maintained. Therefore, tighter focusing results in a higher peak intensity but this intensity is maintained over a shorter length scale, so the nonlinear power-length product is independent of the crystal length (Eqn. 1.31).

In order for SHG to be a practical technique for converting the output of NIR laser into the visible, one hopes to exceed the low conversion limit ($\eta > \sim 10\%$). In this case, the coupled-wave equations that lead to the SHG conversion efficiency must be solved numerically rather than the analytically, as in the low conversion limit. It was shown in [Whi70], however, that the SHG conversion efficiency with pump depletion and optimum phase-mismatch is well approximated by:

$$\eta = \tanh^2 \left[\sqrt{\frac{16\pi^2 d_{\text{eff}}^2}{n(2\omega)n(\omega)\epsilon_0 c \lambda^3}} P_{\omega} L h \right] \quad (1.72)$$

Using the typical $d_{\text{eff}} = 16 \text{ pm/V}$ for PPLN, Eqn. 1.72 predicts a conversion efficiency of 50% for a peak-power of only $\sim 5 \text{ W}$ using a 50 mm long crystal. This enables efficient single-pass frequency-doubling of CW sources [Mil97, Kum11, Chi12b], which is only possible due to the high nonlinear coefficient that QPM can access. Indeed, periodically poled crystals are ideally suited for high-efficiency SHG of pulsed MOPFA systems using very simple and robust single-pass optical setups. The very high nonlinear coefficient of periodically poled crystals relaxes the peak-power requirements of the pulsed MOPFA, which would otherwise be challenging to scale to the level required to obtain comparable conversion efficiencies in non-poled crystals (without using external resonators) due to the high intensity-length products of fibre amplifiers (cf. Section 1.3.6). Using periodically poled crystals, SHG conversion efficiencies of $>80\%$ are routinely obtained with pulsed MOPFAs (cf. Chapter 5, [Tav98]), providing a practical and power-scalable method for spectral conversion.

The average power of SHG using periodically poled crystals has been scaled to around the 40 W level [Vya05, Chi12b], further power-scaling is limited by a process known as thermal dephasing [Lia04]. Although SHG is a parametric process, heat is deposited in the nonlinear crystal due to linear absorption of the fundamental and second-harmonic and additionally due to nonlinear absorption of the fundamental caused by the second-harmonic. The use of a focused fundamental beam and the nature of the growth of the second-harmonic leads to a spatially inhomogeneous heat load in the nonlinear crystal

due to absorption. The relatively poor thermal conductivity of ferroelectric crystals results in the inhomogeneous heat load forming local temperature variations that disrupt the QPM, reducing the SHG conversion efficiency. Whilst the focusing conditions [Sab15] and thermal mounting of the crystal [Sab14] can be optimised to reduce thermal dephasing, it is largely determined by the crystal properties: namely the absorption coefficient at the second-harmonic wavelength (normally an order of magnitude larger than at the fundamental wavelength) and the thermal conductivity. In this regard, PPLT has a significant advantage over PPLN and PPKTP since its thermal conductivity is nearly twice as large [Yu04].

2 Fibre-integrated 560 nm source

This chapter describes the development of a fibre-integrated source of picosecond pulses operating at 560 nm to satisfy the requirements of a depletion beam source for stimulated emission depletion (STED) microscopy using green fluorescent protein (GFP). It begins with a brief introduction to applications that demand radiation in the yellow-green spectral region and laser media that directly emit in this region. Following that, STED microscopy using GFP is introduced, with a specific focus on the requirements of the depletion beam source and the existing solutions. The concept of using stimulated Raman scattering (SRS) in conjunction with second-harmonic generation (SHG) to shift the spectral output of a ytterbium master oscillator power fibre amplifier (Yb-MOPFA) into the yellow-green region is subsequently discussed. Based on this concept, the implementation of a narrow linewidth seeded Raman amplifier, pulse-pumped by a Yb-MOPFA system is then detailed. Following that, the frequency-doubling of the Raman amplifier output using a fibre-coupled periodically poled lithium niobate (PPLN) crystal module is described. Finally, a short summary will conclude this chapter.

2.1 Yellow-green applications and sources

The yellow-green spectral region (550-600 nm) in many ways exemplifies the technological challenges of developing wavelength versatile, high power and temporally flexible visible lasers. It is a region with many existing demands for laser sources but it also has great potential for new applications that are not catered for by current technology. Several of the existing application areas with specific examples are listed below:

- Fundamental research – resonances of atoms such as barium [Pap80] and ytterbium [Kuw99] lie in this region, thus tunable lasers are in demand for spectroscopy and laser cooling. The D_2 line of the sodium doublet line is resonant at 589.0 nm and so lasers can be used to excite sodium atoms in the mesosphere to provide an ‘artificial star’ as a wavefront reference for adaptive optics systems in astronomical telescopes [Hol10].
- Medicine – pulsed yellow lasers have been used in ophthalmology for photocoagulation of the retina [Mai86] and in dermatology for the treatment of vascular

lesions [Tan03], due to the preferential absorption of oxyhaemoglobin over melanin and reduced haemoglobin around 580 nm.

- Biophotonics – many key fluorescent proteins that are used in fluorescence microscopy [Lic05] and flow cytometry [Gal12] such as mCherry, the red fluorescent protein, DsRed and fluorescent dyes such as CY3 and Atto565 have absorption features in this region. GFP and the yellow fluorescent protein (YFP) have emission features, which can be utilised in STED microscopy [Wil06].

The applications of yellow-green laser sources listed above each have unique requirements on the central wavelength, average power or pulse energy and temporal duration. For example, laser trapping of ytterbium atoms requires an extremely narrow linewidth, relatively low average power, continuous-wave (CW) laser source that can be precisely tuned and locked to the atomic transition frequency [Uet08]. In contrast, the wavelength requirements for dermatological laser sources are far more relaxed but very high pulse energy at low repetition rates are required for effective vascular destruction [Mor86]. The available laser sources that directly emit in the yellow-green spectral region and their emission characteristics are detailed in the paragraphs that follow.

Semiconductor lasers are often the first option considered in many applications due to their low cost, high efficiency, compact size and ease of use. Unfortunately, 520 nm is the longest wavelength emitted by efficient InGaN diodes [Miy09] and 630 nm is the shortest wavelength emitted by efficient AlGaInP diodes [Blu13], leaving the so-called ‘green gap’ between 520–630 nm, where laser diodes are not available.

Solid-state lasers based on the trivalent praseodymium ion (Pr^{3+}) have multiple emission lines in the visible and subpicosecond mode-locked operation has been demonstrated [Sut96]. Lithium yttrium fluoride (YLF) has been the most successful Pr^{3+} host, with up to 2.9 W CW average power at 523 nm with an optical-to-optical efficiency of 67% having been demonstrated [Met14]. The rapid increase in the output power of multimode InGaN laser diodes operating around the Pr^{3+} absorption peak at 444 nm in recent years is very promising for the development of high-power visible solid-state lasers. Pr^{3+} does not, however, have any emission lines between 523–604 nm, in the yellow-green region.

Krypton ion gas lasers have an emission line at 568.2 nm that can produce multiple-Watt CW average powers and mode-locking of this line has generated 55 ps pulses at 80 MHz repetition rate with 420 mW of average power [Ste80]. The large-frame ion laser used in this demonstration (Spectra Physics 171) had a 2 m laser head, weighed 200 kg and required 38 kW of electrical power, which consequently required a 13 l/min water flow rate to provide cooling. The extremely low efficiencies of argon and krypton ion lasers and the concomitant large size and high power requirements is the primary reason for

their decline in use in research laboratories.

Copper vapour lasers are significantly more efficient (typically 1-3%) than argon and krypton ion lasers and operate on two lines at 510.6 nm and 578.2 nm, with the power typically split in a 2:1 ratio. They can be scaled to very high average powers, with up to 211 W having been demonstrated from a single oscillator [Kim94], 650 W from a single amplifier [War91] and 2.4 kW from a chain of four amplifiers [Kim00]. Laser action in copper vapour lasers is self-terminating, therefore, they emit pulse trains with a pulse duration between 5-60 ns at a repetition rate between 2-100 kHz. Their power scalability has been used to pump wavelength tunable dye lasers, generating average powers up to 1.5 kW for laser isotope separation [Bas92]. The main drawback of copper vapour lasers is the limited scope to vary the temporal properties, for example CW operation is not possible. Additionally, the laser heads are very large (>1 m) for high average power output and the maintenance requirements are high in comparison to solid-state lasers.

Dye lasers based on rhodamine 6G (R6G) can be operated across the whole of the yellow-green spectral region. Examples include 546–592 nm operation with up to 35 mJ pulse energy using a frequency-doubled Nd:YAG pump [Har78] and continuous tuning from 560–650 nm with up to 33 W average power using a multi-line argon ion CW pump laser [Bav82]. In addition to the extreme wavelength flexibility, pulses as short as 27 fs have been obtained directly from a mode-locked oscillator [Val85] and up to 400 J of pulse energy in 10 μ s pulses has been obtained using flashlamp pumping [Bal74].

The versatility of dye lasers is unparalleled amongst laser gain media, however, the dye solution is also the biggest hindrance to wide-scale adoption. To avoid triplet-state absorption and chemical degradation of the laser dye, the solution has to be re-circulated and flowed, either as a thin jet or through a dye cell, and then periodically replaced. Many laser dyes are toxic and some are mutagenic or carcinogenic [Wue85] and the solvents used are often highly flammable, sometimes toxic and can aid the absorption of dyes through skin. Thus, the requirement for frequent handling of the laser dye solutions has significant safety implications and the high level of expertise required to run and maintain dye lasers restricts their use to specialist applications.

It has been shown that there is a limited number of laser media that directly emit effectively in the yellow-green spectral region, namely krypton ion, copper vapour and dye lasers. The gas lasers only operate on specific narrow-band laser lines, are temporally inflexible and not practical due to their large power and high maintenance requirements. Dye lasers are highly versatile but, likewise, are not practical systems to operate in non-specialist laboratories due to the difficulties in handling the degradable dye solutions. The large number of applications that require practical laser sources in the yellow-green has fuelled the intense research effort into using nonlinear optical techniques to convert the

spectral output near-infrared (NIR) semiconductor, solid-state and fibre-based sources into this region [Fed14, Wan12, Uet08].

2.2 STED microscopy using GFP

Confocal laser scanning fluorescence microscopy (CLSM) is the de facto standard for the noninvasive imaging of cells in three dimensions due to the enhanced selectivity over standard confocal microscopy [Lic05]. The lateral (radial) resolution of CLSM is restricted by the Abbe diffraction limit to $\Delta r = 0.44\lambda/\text{NA} \sim 200$ nm, where λ is the wavelength of light and NA is the numerical aperture of the objective lens. This resolution is detailed enough to image cells but in order to probe the sub-cellular level, towards the molecular level, this diffraction limit must be overcome.

There are several techniques used to image beyond the diffraction limit, which can be broadly separated into techniques that use statistical analysis to build a super-resolved image from an ensemble (e.g. PALM and STORM) and direct time-sequential imaging using reversible saturable optically linear fluorescence (RESOLFT) [Hel07]. The main benefit of RESOLFT techniques is the dramatically faster acquisition time, which can be critical for in-vivo imaging applications.

STED microscopy is the most widely utilised RESOLFT technique, where a doughnut-shaped depletion beam with a central minima is spatially overlapped with a Gaussian excitation beam at the sample [Hel94]. The depletion beam depopulates the excited state through stimulated emission, inhibiting the fluorescence in the periphery of the spot. When the pulse fluence of the depletion beam is increased past the saturation fluence of the fluorophore, the area of the fluorescence inhibition increases, decreasing the area of the fluorescence at the focal point. Hence, despite the fact that both the excitation and depletion beams are diffraction-limited, the saturation of the fluorescence inhibition can reduce the effective point spread function of the fluorescence that is detected to below the diffraction limit. It follows that the resolution improvement over CLSM scales with the depletion beam pulse fluence according to:

$$\Delta r = \frac{0.44\lambda}{\text{NA}\sqrt{1 + F_{\text{STED}}/F_s}} \quad (2.1)$$

where F_{STED} is the pulse fluence of the depletion beam and F_s is the saturation fluence of the fluorophore, the fluence at which the rates of stimulated and spontaneous emission are equal. Resolutions down to ~ 20 nm have been demonstrated in biological samples [Wes05, Don06, Wil09] and as small as 6 nm imaging nitrogen vacancies in diamond, which can tolerate far higher pulse fluences [Rit09].

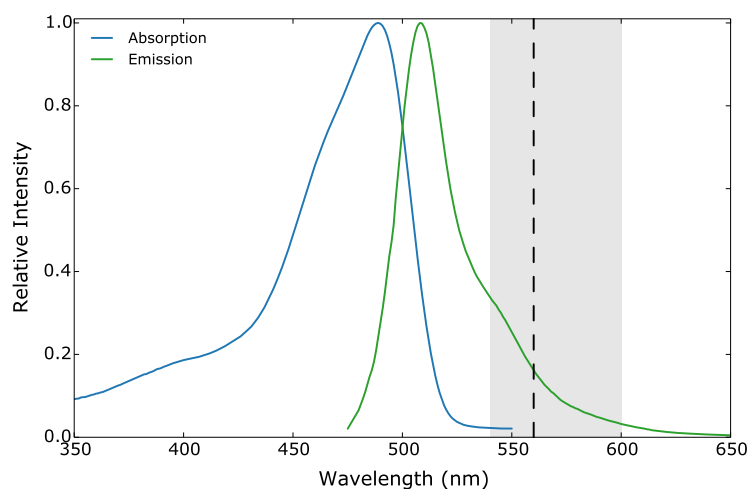


Figure 2.1: Absorption (blue) and emission (green) spectrum of emerald green fluorescent protein (Life Technologies). The shaded region indicates the STED microscopy depletion wavelength range and the dashed line indicates the optimum 560 nm depletion wavelength.

There are many fluorophores that can be used for fluorescent microscopy but of these, endogenous fluorophores can have issues with contrast between target and background behaviour and many extrinsic fluorophores are phototoxic so cannot be used for in-vivo imaging. GFP is a gene that can be introduced and expressed in a wide variety of biological samples, enabling intrinsic labelling of specific cells in living organisms. The absorption and emission spectrum of a typical GFP is shown in Fig. 2.1, which has an absorption peak at 490 nm and an emission peak at 509 nm. For efficient fluorescence inhibition, the depletion beam wavelength needs to be in the long wavelength tail of the emission spectrum (shaded region Fig. 2.1). The exact wavelength is a compromise between having a small enough absorption cross section to avoid re-excitation and having an appreciable stimulated emission cross section, whilst enabling the fluorescence to be filtered and detected. For GFP, it has been demonstrated that a depletion beam wavelength of 560 nm (dotted line in Fig. 2.1) is the optimum, although a reasonably broad workable wavelength range exists [Ran11].

In the temporal domain, the optimal pulse duration for the depletion beam should be longer than the non-radiative decay time that depopulates the lower stimulated emission state but shorter than the upper state lifetime, to ensure efficient suppression of the fluorescence. For typical fluorophores, this would indicate an optimal pulse duration of 20–30 ps but in practice durations between 100–150 ps are found to be more effective [Leu10]. This is most likely due to the reduction in polarisation effects due to fluorophore rota-

tion [Wes05] and a reduction in photobleaching from multi-photon absorption [Dyb03]. Moreover, STED has been demonstrated using CW beams [Wil07] and increasing the depletion beam pulse duration towards the upper state lifetime (~ 2 ns for GFP [Sol07]) enables a reduction in the repetition rate whilst maintaining a constant peak-power. This reduces photodegradation from triplet state build-up, due to the increased time between pulses, without causing photobleaching due to excessive peak-power [Wil08].

In summary, the ideal attributes of a depletion beam source for STED using GFP are as follows:

- Central wavelength close to 560 nm for efficient stimulated emission without re-excitation.
- High pulse energy to yield a good resolution improvement – GFP has a saturation fluence of $\sim 10^{17}$ photons/cm², so for a depletion wavelength of 560 nm and a typical doughnut area of $\sim 10^{-9}$ cm², the corresponding saturation energy is ~ 35 pJ.
- 0.1–2 ns pulse duration for effective fluorescence depletion.
- Few megahertz repetition rate as a compromise between image acquisition times and avoidance of photodegradation effects.
- Diffraction-limited beam quality with high polarisation purity.
- Compact, efficient and turn-key.

The only source of direct laser emission that can satisfy these criteria is a dye laser, however, their footprint is too large and the maintenance requirements too great to be considered for most imaging setups. Using nonlinear frequency conversion of NIR lasers is, therefore, the only practical option for providing the depletion beam. In the paragraphs that follow, the existing solutions for providing the depletion beam for a STED microscope using GFP are discussed.

Frequency-doubled pulsed semiconductor diodes are compact, efficient and turn-key but have limited average output powers even at tens of megahertz repetition rates, resulting in a maximum pulse energy of ~ 35 pJ at 560 nm [Kal15]. This pulse energy is too low to offer a significant resolution improvement, especially considering fibre-coupling is required to spatially clean the beam, reducing the available power further due to coupling losses (typically $\sim 60\%$).

The original reports of STED microscopy using GFP utilised the frequency-doubled output of an optical parametric oscillator (OPO), synchronously pumped by an ultrafast Ti:Sapphire oscillator, to provide the depletion beam [Wil06]. These systems typically

produce 200 fs pulses that are wavelength tunable over the range 505–740 nm with a maximum average power of ~ 700 mW. The standard ~ 80 MHz repetition rate of the ultrafast oscillator pump results in a pulse energy of ~ 10 nJ. Whilst these systems satisfy most of the depletion beam criteria, they have several drawbacks: the combined Ti:Sapphire and OPO is very costly, has a large footprint and the output pulses need to be stretched in duration to be suitable for STED. In addition, the overall optical-to-optical efficiency from the green Ti:Sapphire pump laser (18 W at 532 nm) is around 4% and the pump lasers themselves typically have wall-plug efficiencies of $< 5\%$.

More recently, supercontinuum sources based on extreme spectral broadening in photonic crystal fibres (PCFs) [Dud10] of picosecond Yb-MOPFA schemes have been used to provide the excitation and depletion beams for STED microscopy [Wil08]. The spectral output of the source used spanned 450–1750 nm with 4 W of total average power at a repetition rate of 1 MHz [Alm09]. After filtering the visible part of the supercontinuum, an energy spectral density of > 1 nJ/nm was available between 500–700 nm. To provide the depletion beam, a prism and a variable-width aperture was used to select a 20 nm bandwidth centred on the depletion wavelength. A portion of the visible supercontinuum was split for the excitation beam using interference filters, since the spectral extinction of the excitation beam is less critical. Following the spectral filtering and the optics required to spatially clean and polarise the beam, a depletion beam pulse energy of ~ 2 nJ was available at the sample with a pulse duration of ~ 80 ps. Supercontinuum sources are compact, maintenance-free and cost effective but the requirement of having to spectrally filter the output leads to a low optical-to-optical efficiency, which limits the ability to scale the pulse energy in the visible.

2.3 Raman conversion of ytterbium MOPFA schemes

Master oscillator power fibre amplifier (MOPFA) schemes are an ideal platform for nonlinear frequency conversion due to the power scalability, excellent beam quality and flexible temporal characteristics. In addition, they are very attractive from a practical perspective: they are compact, have high wall-plug efficiencies and are very cost effective. The frequency-doubling of Yb-MOPFA schemes gained traction in the late 90's [Gus98, Pop00] and since then the average power has been scaled to 700 W quasi-continuous-wave (QCW) average power, with a wall-plug efficiency of 23% [Avd15]. This single-mode green average power is significantly higher than what can be obtained using traditional rod-type diode pumped solid-state lasers, which are challenging to scale > 100 W due to thermal degradation of the mode profile [McD07, Mei10, Sin11]. Indeed, only highly complex multi-pass thin-disk amplifiers can produce greater single-mode green power [Neg15], highlighting

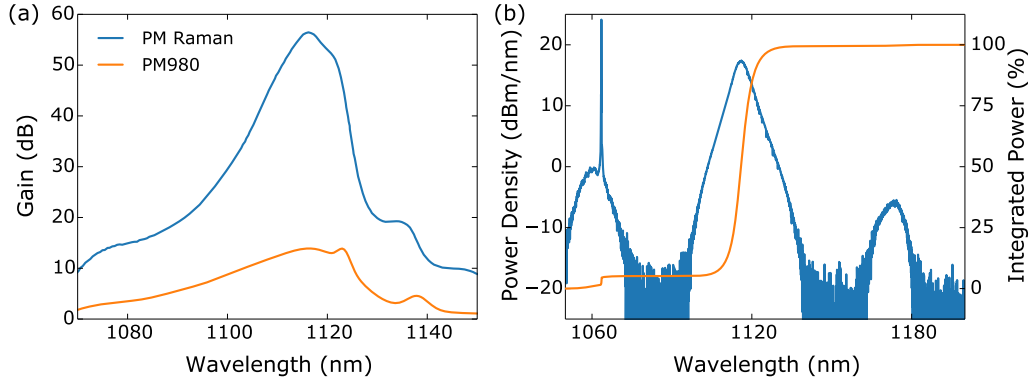


Figure 2.2: (a) Calculated small-signal on-off Raman gain spectrum of a 1 m length of PM Raman fibre (blue) and PM980 (orange) when pumped with a polarised source at 1064 nm with 1 kW of peak-power. (b) Optical spectrum of a 15 m length of PM Raman fibre pumped with polarised 1064 nm pulses with 160 W of peak-power.

the benefits of the fibre architecture for power scaling.

The frequency-doubling of pulsed Yb-MOPFA schemes is an established technique for generating high pulse energies in the green spectral region [Liu05]. To extend this technique to the yellow-green spectral region, it requires the Yb-MOPFA to be operated at wavelengths longer than 1100 nm. Whilst pulsed emission of ytterbium fibre (Yb-fibre) lasers has been demonstrated at 1120 nm [Wan13], the larger emission cross section at shorter wavelengths can lead to excessive amplified spontaneous emission (ASE) and parasitic lasing. It is, therefore, technologically challenging to implement high-gain Yb-MOPFA schemes at this wavelength [Gro03, Zha13] and standard fibre components and amplifiers cannot be utilised.

Stimulated Raman scattering (SRS) can be used to provide gain in spectral regions where rare-earth doped fibres do not have emission bands, since the central wavelength of the gain is determined by the pump wavelength and the frequency shift corresponding to the medium's molecular vibrational modes (cf. Section 1.3.4). Figure 2.2 (a) shows the calculated small-signal on-off Raman gain spectrum for a meter length of polarisation-maintaining (PM) Raman fibre (OFS Fitel, blue trace) and PM980 (Nufern, orange trace) pumped by a polarised source with 1 kW of peak-power at 1064 nm. In the case of the PM Raman fibre, the Raman gain spectrum is based on manufacturer supplied data, whereas, a multiple damped oscillator model [Hol02] is used for the PM980 fibre. Clearly, large signal gains can be extracted over the range of 1100–1130 nm, with the peak gain at 1116 nm for both fibres, corresponding to a frequency downshift of 13.2 THz from the pump.

The Raman gain is high enough with kW·m power-length products in passive fibres that spontaneous Raman scattering is amplified forming Raman ASE [Sto72, Sto84]. Once a threshold peak-power has been exceeded, highly efficient conversion from the pump to the Stokes pulse occurs. This can be seen in Fig. 2.2 (b), where integrating the spectral output of a 15 m length of PM Raman fibre pumped with 160 W of peak-power (orange line) reveals that the Stokes pulse centred on 1116 nm contains 94% of the total power. When the Stokes pulse has sufficient peak-power, it can amplify spontaneous Raman scattering at the next Stokes shift, which is evident in Fig. 2.2 (b) as the peak forming around 1173 nm. In this manner, an efficient cascade can form, where the dominant Stokes output from the passive fibre is determined by the input peak-power and fibre length [Sto84]. Passive fibres can, therefore, be used as ‘Raman converters’, a highly-efficient, fibre-integrated method for frequency downshifting the output of fibre lasers [Rak08, Say11].

An important difference between amplification using SRS in comparison to rare-earth doped fibre amplifiers is the fact there is no energy storage in the gain medium. This can be understood by the fact that the energy states in Raman amplification are virtual, therefore, if a pulse-pump source is used, Raman gain is only available in the window of the pump pulses. If pump and Stokes wavelengths lie in the normal dispersion region of the passive fibre, the Raman ASE will have similar temporal properties to the pump [Lin77, Sto86]. In contrast, if the Stokes wavelength lies in the anomalous dispersion region, femtosecond Raman solitons can form [Gou87].

The frequency-doubling of Raman converted Yb-fibre lasers can be used to access the yellow-green spectral region and due to the fibre-integrated nature and high efficiency of the process, there is little penalty in terms of the peak-power of the fundamental. The efficiency of the SHG process can be severely compromised, however, by the fact that the Raman gain bandwidth is usually significantly larger than the spectral acceptance bandwidth of the frequency-doubling crystal [Fen04, Che05b, Avd15]. This is illustrated in Fig. 2.2 (b), where the 3 dB spectral bandwidth of the 1116 nm Stokes component is 6.4 nm, compared to spectral acceptance bandwidths of 1.9 nm for lithium triborate (LBO) and 0.13 nm for PPLN using non-critical phase-matching (NCPM) with 20 mm long crystals.

To overcome the issue of the SHG spectral acceptance bandwidth, it is possible to first frequency-double the output of a Yb-MOPFA and then use a passive fibre to Raman shift the green output into the yellow and red spectral regions [Ran09, Lin11]. The main drawback of this approach is the need to couple the frequency-doubled light into passive fibre – the alignment is far more sensitive than for SHG and optical fibres are subject to photodarkening under green light irradiation – which greatly restricts the power scalability.

Alternatively, the spectral bandwidth of the Stokes output can be significantly reduced

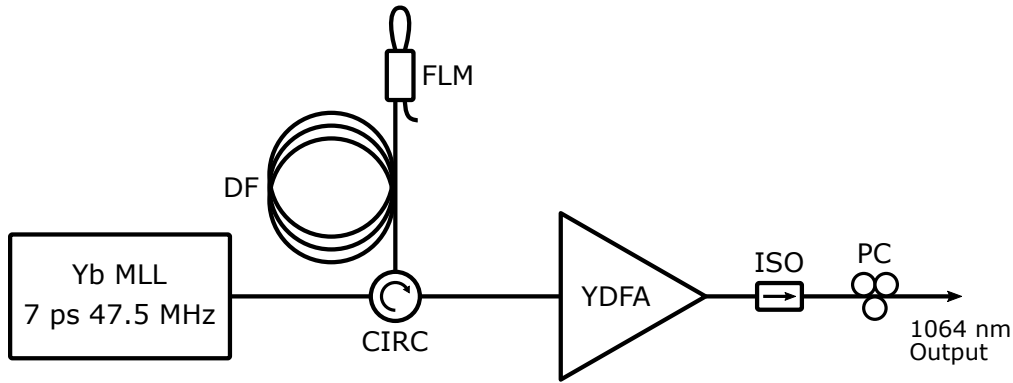


Figure 2.3: Schematic of the picosecond Yb-MOPFA. Yb MLL, ytterbium mode-locked laser; DF, dispersive fibre; FLM, fibre loop mirror; CIRC, circulator; YDFA, ytterbium-doped fibre amplifier; ISO, isolator; PC, polarisation controller.

by providing a narrow linewidth signal at the Stokes wavelength, rather than it forming from broadband spontaneous Raman emission. Effectively, this is using a passive fibre as a Raman amplifier rather than a Raman ASE source. The virtual nature of the energy states in Raman amplification result in the temporal properties of the amplifier output being determined by the pump pulses; whereas, the spectral properties are predominantly determined by the seed signal. This is an extremely powerful concept as it enables the output of pulsed Yb-MOPFA schemes to be spectrally downshifted to arbitrary wavelengths, whilst possessing a narrow spectral bandwidth suitable for efficient SHG.

The technique of CW seeding a pulse-pumped Raman amplifier to spectrally shift the output of Yb-MOPFA has been demonstrated in combined ytterbium-Raman (Yb-Raman) amplifiers [Dup06a, Row13, Row14, Kar15]. In these systems, a CW seed signal at the Stokes-shifted wavelength is introduced using a wavelength division multiplexer (WDM) before the final power amplification stage in a Yb-MOPFA. The pulsed signal in the Yb-fibre gain band is amplified by stimulated emission to a high enough peak-power that Raman amplification of the CW seed occurs in the power amplifier. At the operating point, the length and peak-power need to be optimised such that the pulse energy is efficiently shifted to the Stokes wavelength at the output of the amplifier.

This is a very elegant approach as the output can be directly frequency-doubled to the yellow-green, which avoids any additional losses of a Raman conversion stage. The Yb-fibre amplifier design (length, core and cladding diameters and pump parameters), however, is severely constrained by the need to achieve efficient stimulated emission amplification and the correct amount of nonlinearity for efficient Raman conversion. Additionally, since the pump and Stokes signals are amplified simultaneously, strong four-wave interactions [four-wave mixing (FWM) and cross-phase modulation (XPM)]

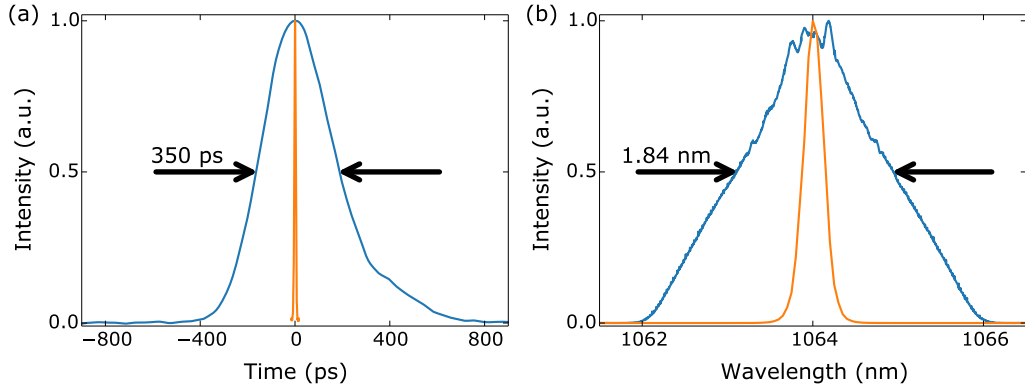


Figure 2.4: (a) Sampling optical oscilloscope trace of the stretched mode-locked oscillator pulse (blue) and deconvolved intensity autocorrelation trace of the original oscillator output (orange). (b) Optical spectrum of the stretched (blue) and original (orange) oscillator pulses.

can occur, which can make it difficult to achieve a high power spectral density, necessary for efficient SHG [Dup06a, Row14].

2.4 Picosecond Raman amplifier at 1120 nm

A source was developed to satisfy the criteria of a depletion beam source for STED using GFP that used two nonlinear frequency conversion stages to shift the spectral output of a pulsed Yb-MOPFA to 560 nm. A discreet Raman amplifier, seeded by a CW narrow linewidth signal, was used to frequency downshift the output of a Yb-MOPFA with a high power spectral density. The Raman amplifier output was frequency-doubled in a fibre-coupled PPLN module, resulting in a compact, turn-key and maintenance-free fibre-integrated source.

2.4.1 Ytterbium MOPFA

A commercial passively mode-locked Yb-fibre laser (Fianium) was used as the master oscillator for the Yb-MOPFA (Fig. 2.3), which provided a reliable turn-key pulse train to seed the fibre amplifier. The oscillator produced 7 ps, near transform-limited, sech^2 pulses at a repetition rate of 47.5 MHz with a central wavelength of 1064 nm and an average power of 2 mW. For the pulses to be a suitable duration for STED and to enable amplification to high average powers, the oscillator's pulses were stretched to 350 ps [Fig. 2.4 (a)] by double-passing a 1.25 km length of normally dispersive fibre using an optical circulator and fibre-loop mirror. Due to the high nonlinearity of the normally dispersive fibre (OFS

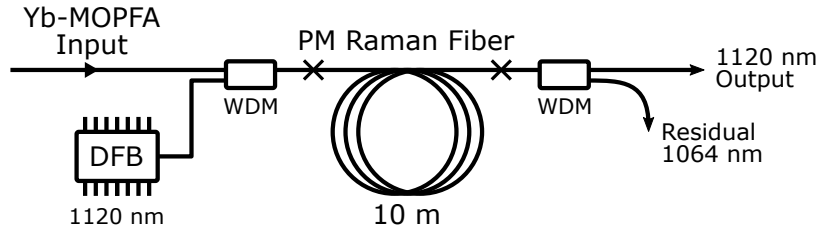


Figure 2.5: Schematic of the Raman fibre amplifier. Yb-MOPFA, ytterbium master oscillator power fibre amplifier; DFB, distributed feedback laser diode; WDM, wavelength division multiplexer.

Raman fibre), the oscillator's spectrum was broadened through self-phase modulation (SPM) [Fig. 2.4 (b)], which enhanced the rate of dispersive broadening of the pulses.

The temporally-stretched oscillator pulses, reduced to -5 dBm average power from the 8 dB insertion loss of the stretcher, were directly amplified in a commercial multi-stage Yb-fibre amplifier (IPG Photonics) with a gain of 45 dB and a saturated output power of 10 W. An optical isolator was spliced to the output of the amplifier, to protect it from backwards propagating Raman ASE generated by the Raman amplifier. Since the Yb-MOPFA was constructed from isotropic fibre, a three-paddle polarisation controller (PC) was spliced to the isolator to provide complete control over the randomly polarised output of the amplifier.

2.4.2 Raman amplifier

The pulsed 1064 nm pump radiation from the Yb-MOPFA was spectrally combined with a CW narrow linewidth 1120 nm signal from a fibre-pigtailed distributed feedback (DFB) laser diode (QD Laser) in a PM WDM (Fig. 2.5). The laser diode delivered an average output power of 40 mW in a single longitudinal mode, corresponding to a spectral linewidth of <10 MHz. A 10 m length of PM Raman fibre (OFS Fitel) was used as the Raman gain medium and a second PM WDM was spliced to the output of the Raman fibre to filter residual Yb-MOPFA pump light. The PM Raman fibre was chosen over a standard single-mode fibre (SMF) due to its higher Raman gain efficiency ($C_R \sim 13 \text{ W}^{-1}\text{km}^{-1}$ cf. $C_R \sim 3 \text{ W}^{-1}\text{km}^{-1}$ for PM980 with a polarised pump at 1064 nm). The increased Raman gain efficiency is primarily due the reduced effective area of the Raman fibre ($A_{\text{eff}} \sim 11 \mu\text{m}^2$ cf. $A_{\text{eff}} \sim 44 \mu\text{m}^2$ for PM980 at 1120 nm) but also the higher germanium content in the core increases the polarised Raman gain coefficient ($g_R \sim 1.4 \times 10^{-13} \text{ m/W}$ cf. $g_R \sim 1.2 \times 10^{-13} \text{ m/W}$ for PM980 at 1120 nm), which reduces the relative impact of linewidth broadening nonlinear effects in comparison to SMF. In addition, the stronger waveguiding of the PM Raman fibre ($\text{NA}=0.23$ cf. $\text{NA}=0.12$ for PM980) and germanium doping increases the positive disper-

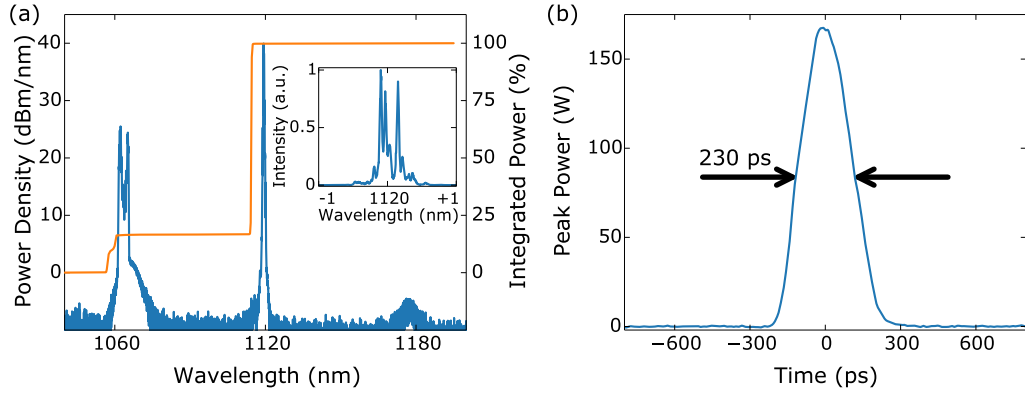


Figure 2.6: (a) Optical spectrum of the Raman amplifier output with the integrated power offset by 5 nm and linear spectrum of the peak (inset). (b) Sampling optical oscilloscope trace of the filtered 1120 nm component.

sion around 1 μm , which discourages FWM due to the increased linear phase-mismatch. A comparison of the performance of the two fibres as the gain medium in a pulsed-pump Raman amplifier is presented in Section 4.3.

Figure 2.6 (a) shows the optical spectrum of the Raman amplifier output for an average power of 2.25 W, which was the most efficient operating point for the subsequent frequency-doubling. The integrated spectral power revealed that 83% of the output power was contained in the 1120 nm signal, equivalent to 1.87 W. The corresponding pump power from the Yb-MOPFA was 5.1 W (270 W peak-power), resulting in an amplifier efficiency (signal output power less the signal input power over the pump power) of 36%. Based on spectral integration of the output of the Raman fibre before the second WDM, the efficiency of the Raman conversion is 81%, with the seed power taken into account. The amplifier efficiency was significantly lower than the Raman conversion efficiency due to the excess loss of the two WDMs and the two splice losses to the small-core Raman fibre.

The 3 dB spectral linewidth of the 1120 nm signal in Fig. 2.6 (a) (inset) is 0.28 nm, which was significantly broader than the seed signal linewidth due to SPM in the excessive length of Raman fibre. This effect could be mitigated by performing a cut-back on the Raman fibre to optimise the peak-power spectral density of the 1120 nm signal. The 1120 nm signal had a pulse duration of 230 ps [Fig. 2.6 (b)] at the pump repetition rate of 47.5 MHz, with no observable pedestal on a sampling optical oscilloscope with a dynamic range of 30 dB (Hamamatsu OOS-01). The reduction in duration from the Yb-MOPFA pump pulses was expected due to the nonlinear nature of the Raman amplification.

In spite of the fact that there were no polarisation discriminating elements in the Yb-

MOPFA, the use of a polarised 1120 nm seed laser and PM fibre in the Raman amplifier enabled the Raman conversion to be optimised using the PC of the Yb-MOPFA. The conversion was very sensitive to the PC position and the filtered 1120 nm output had a polarisation extinction ratio (PER) of 14 dB, implying that the Yb-MOPFA had a reasonable degree of polarisation. When the PC was adjusted away from the optimal position, significant peaks were seen in the Raman amplifier spectrum at 1045 nm and 1084 nm, which were due to degenerate FWM of the 1064 nm pump, phase-matched by the birefringence of the PM Raman fibre.

2.5 Fibre-coupled frequency-doubling

To frequency-double the output of the Raman amplifier to 560 nm, the optimum depletion wavelength for STED microscopy using GFP, simple single-pass SHG in PPLN was chosen. PPLN has the highest effective nonlinear coefficient of commonly used SHG crystals, which enables a high SHG conversion efficiency to be obtained with the relatively modest peak-power of the Raman amplifier output (~ 170 W), without the need for complex external resonators.

To decrease the footprint of the source and make the entire system alignment-free, the frequency-doubling optical setup was packaged by our industrial collaborator, Gooch and Housego, into a module measuring just $120 \times 40 \times 26.5$ mm (Fig. 2.7). Inside the module, a 15 mm long 5% MgO-doped congruent lithium niobate crystal was held in a metal housing, that was bonded to a thermoelectric cooler (TEC). The crystal had a thickness of 1 mm, a width of 3 mm and a manufacturer quoted effective nonlinear coefficient of >14 pm/V. The crystal was poled with a single $8.06 \mu\text{m}$ pitch grating, resulting in a phase-matching bandwidth of 0.176 nm for the fundamental wavelength of 1120 nm. The crystal was anti-reflection (AR) coated on the input and output facets for both the fundamental and second-harmonic (SH) wavelengths.

The PM980 input fibre to the module was focused to a waist radius ($1/e^2$) of $30 \mu\text{m}$ at the center of the crystal using an $f = 2.95$ mm aspheric lens. The estimated confocal parameter in the crystal was 11 mm ($n = 2.16$), corresponding to a focusing parameter ($\xi = L/b$) of 1.4. This was a looser focus in comparison to the theoretical optimum Boyd-Kleinman focusing condition of 2.84 [Boy68]. The slow-axis of the input fibre was aligned to the extraordinary axis of the crystal to maximize the SHG efficiency. After the crystal, the residual 1120 nm pump power was directed out of the module through an AR-coated window using an orthogonal pair of 45° dichroic mirrors. The generated second-harmonic was collimated to a diameter of 1.1 mm using an $f = -10$ mm and $f = 20$ mm telescopic pair of aspheric lenses.

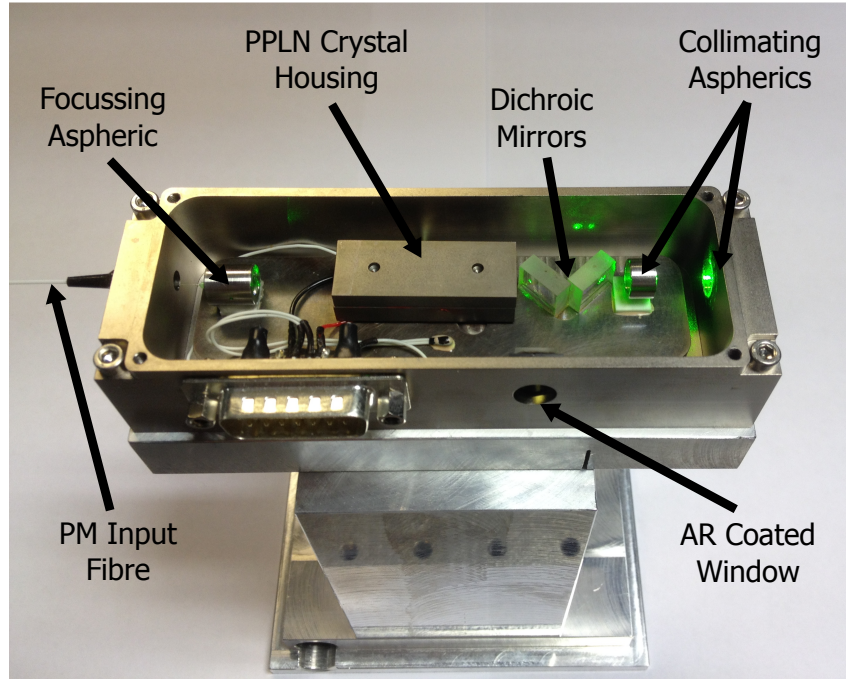


Figure 2.7: Labelled photograph of the fibre-coupled frequency-doubling module.

All of the optical components and the TEC controlled crystal housing were bonded to a baseplate that was temperature controlled using three TECs in series. The individual temperature control of the crystal and the baseplate ensured the phase-matching temperature could be maintained with a precision of ± 0.1 °C without perturbing the alignment of the optical setup. The calculated full-width half-maximum temperature acceptance bandwidth of the crystal was 1.8 °C, which with the precision of the temperature control sets an upper limit on the SHG power stability of <1%.

The Raman fibre amplifier described in the preceding section was fusion spliced to the input fibre of the frequency-doubling module. A maximum average power of 480 mW of 560 nm was generated for 1.87 W of 1120 nm, corresponding to a conversion efficiency of 26% at the optimum phase-matching temperature of 74.4°C. The conversion efficiency was predominantly limited due to the fact that the 1120 nm fundamental bandwidth (0.28 nm) exceeded the spectral acceptance bandwidth of the crystal (0.176 nm). Integrating the fundamental spectrum over the sinc^2 spectral acceptance function of the PPLN crystal revealed that only 45% of the 1120 nm power was within the spectral acceptance bandwidth. This was equivalent to 0.85 W, resulting in an effective 57% SHG conversion efficiency. Increasing the Yb-MOPFA pump power did not result in an increase in the 560 nm average power as both the Raman conversion efficiency and the power contained

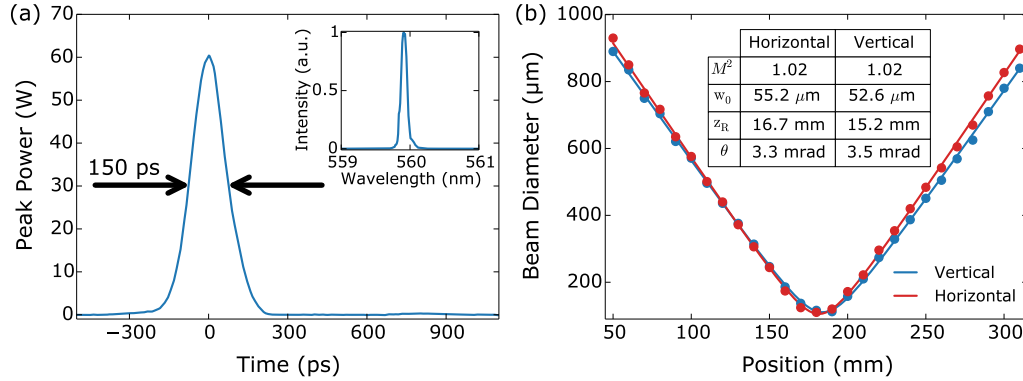


Figure 2.8: (a) Sampling optical oscilloscope trace and optical spectrum (inset) of the 560 nm module output. (b) Measured beam diameter (points) of the collimated module output focused by a $f = 200$ mm lens with Gaussian beam caustic fit (lines).

in the crystal bandwidth decreased.

The SH power was strongly dependent on the position of the PC on the output of the Yb-MOPFA, confirming that it had a reasonable degree of polarisation. Whilst the short-term stability of the source was very good, due to long-term thermal and mechanical perturbations of the Yb-MOPFA fibre, the SH output power would steadily decrease over a period of minutes but could be restored to the maximum power by adjustment of the Yb-MOPFA PC.

The 560 nm pulse duration was 150 ps at a repetition rate of 47.5 MHz, corresponding to a peak-power of 60 W and a pulse energy of 10 nJ [Fig. 2.8 (a)]. This pulse energy is nearly 300 times the saturation energy of GFP for a typical depletion beam doughnut area and is equivalent to the output of a ultrafast-pumped OPO but with double the optical-to-optical efficiency of 9%. The spectral linewidth of the 560 nm signal was measured to be <0.1 nm, limited by the resolution of the optical spectrum analyser [Fig. 2.8 (a) inset]. A CCD camera was used to measure the beam diameter of the collimated module output focused by an $f = 200$ mm lens [points Fig. 2.8 (b)]. A Gaussian fit of the beam caustic [lines Fig. 2.8 (b)] revealed a near-diffraction limited beam, with an $M^2 = 1.02$ and an ellipticity of 0.95.

The maximum peak intensity of the generated 560 nm signal was estimated to be ~ 4.2 MW/cm², which was approaching the photorefractive damage (PRD) threshold limit of 5% MgO-doped PPLN [Kie12]. Therefore, despite the fact that significantly more 1120 nm power in the crystal bandwidth could have been obtained by optimising the PM Raman fibre length in the Raman amplifier, it would have required the module to be

redesigned with a looser focus, so as not to exceed the PRD threshold.

2.6 Summary

This chapter has described the demand for convenient laser sources in the yellow-green spectral region and in particular looked at satisfying the criteria for a depletion beam source for STED microscopy using GFP. It was discussed that nonlinear frequency conversion of NIR lasers is more practical than sources of direct laser emission in this wavelength region. Fibre lasers display excellent characteristics for both nonlinear frequency conversion and their practical aspects, such as size, cost effectiveness and maintenance-free operation. It was shown that simulated Raman scattering can be used to extend the wavelength coverage of rare-earth MOPFA schemes with high efficiency using fibre-integrated setups but it requires seeding with a narrow linewidth signal in order to have an output bandwidth that can be frequency-doubled efficiently.

Using a discreet Raman amplifier, seeded with a narrow linewidth CW signal, the pulsed output of a Yb-MOPFA operating at 1064 nm was shifted to 1120 nm. The Yb-MOPFA utilised a passively mode-locked oscillator, whose 7 ps pulses were stretched to 350 ps to be suitable for STED microscopy applications. The pulse-pumped Raman amplifier produced 1.87 W of 1120 nm power for 5.1 W of 1064 nm pump power with a Raman conversion efficiency of 81%. The 1120 nm output had a pulse duration of 230 ps at the pump repetition rate of 47.5 MHz and had a 3 dB spectral bandwidth of 0.28 nm, which was significantly broadened by SPM due to the excessive length of the Raman gain fibre.

A fibre-coupled frequency-doubling module, based on a 15 mm long PPLN crystal was developed in collaboration with Gooch and Housego, to dramatically reduce the footprint of the source and extend the alignment-free benefits of fibre systems to the frequency-doubling stage. Using this module, a maximum 560 nm average power of 480 mW was obtained, corresponding to a conversion efficiency of 26%, which was severely limited by the 0.176 nm spectral acceptance bandwidth of the PPLN crystal.

The 560 nm pulse duration was 150 ps, which is ideally suited to STED microscopy applications. Despite the restricted SHG conversion efficiency, the 10 nJ pulse energy is equivalent to the output of an ultrafast-pumped OPO but with a higher optical-to-optical efficiency of 9% in a far more compact and cost-effective package. The diffraction-limited, circular output beam with a high polarisation purity is suitable for direct integration into a microscopy setup. The only limiting factor that prevents this source from being a compact, turn-key tool for use in imaging setups is the use of a randomly polarised Yb-MOPFA, an issue that is addressed in the next chapter.

3 Duration-tunable 560 nm source

In the previous chapter, it was shown that pulse-pumped Raman amplifiers are a highly efficient technique for shifting the spectral output of ytterbium master oscillator power fibre amplifier (Yb-MOPFA) systems. Seeding the amplifier with a narrow linewidth continuous-wave (CW) signal results in a high power spectral density, facilitating efficient frequency-doubling into the yellow-green (550–600 nm) spectral region. A fibre-integrated source was developed to provide the depletion beam for stimulated emission depletion (STED) microscopy using green fluorescent protein (GFP) which was compact, alignment-free and more efficient than existing depletion beam sources. It could not, however, be used as a practical source in an imaging setup due to the requirement of having to adjust the polarisation of the Yb-MOPFA pump in order to maintain the 560 nm output power. In this chapter, several key improvements to the design of the 560 nm source are implemented, which make the source more efficient, more powerful and, most importantly, completely turn-key.

The chapter begins with an overview of the master oscillator options for seeding Yb-MOPFA schemes, to ascertain which have the most suitable temporal characteristics for a depletion beam source. Following that, the performance of a frequency-doubled, pulse-pumped Raman amplifier is compared using two different master oscillators to seed the pulsed Yb-MOPFA pump system. The first master oscillator was designed to give the best performance and greatest flexibility in the 560 nm pulse duration and was based on a Mach-Zehnder amplitude modulator (MZAM). The second master oscillator was designed to be simpler and more cost effective and was based on a gain-switched distributed feedback (DFB) laser diode. The performance of the complete system using the two different master oscillators is described, followed by a summary that concludes the chapter.

3.1 MOPFA seed sources

The temporal characteristics of the CW-seeded, pulse-pumped Raman amplifier is predominantly determined by the pulsed Yb-MOPFA pump system, which in turn is mainly dictated by the master oscillator seed source. In the previous chapter, a passively mode-locked (PML) fibre laser was used as the master oscillator for the Yb-MOPFA system, which

provided a reliable, turn-key source of high-quality optical pulses. The main drawback of using a PML master oscillator is that the pulse duration and repetition rate are fixed by the cavity parameters and therefore, cannot be adjusted during operation.

One of the key benefits of rare-earth doped master oscillator power fibre amplifier (MOPFA) schemes is the flexibility in the temporal and spectral properties they offer, which enables a large number of different master oscillator technologies to be used. Therefore, to increase the temporal flexibility of the 560 nm source, the alternative options of master oscillator seed sources were investigated to ascertain which were the most suitable for a depletion beam source. The following parameters are key to determining the suitability of a MOPFA seed source for a specific application:

- Pulse duration – many applications have a small range of pulse durations that give optimal performance, for example, the depletion beam for STED microscopy should have a duration between 0.1–2 ns for the highest efficiency [Wil08]. Engineering a source to have a tunable pulse duration enables the optimal pulse duration for a number of different applications to be satisfied.
- Spectral bandwidth – if the application requires ultrashort pulses there must be sufficient bandwidth to support them. Most nonlinear frequency conversion processes benefit from high power spectral density and some require single longitudinal mode operation (e.g. external cavity SHG).
- Pulse quality – a high extinction ratio between the pulse and any background radiation is desirable to maximise the pulse energy after amplification. Some applications require specific pulse shapes or well defined peak-powers.
- Wavelength tunability – the ability to target specific absorption or emission features of atoms or molecules is vitally important for many applications (e.g. spectroscopy) and the ability to cover a broad range of wavelengths greatly increases the flexibility of the source.
- Synchronisation – some applications require the emission of pulses to be triggered by an external clock signal, often at an arbitrary repetition rate. The pulse-to-pulse jitter can be important in applications requiring synchronised pulse trains or time-gated detection.

In the following sub-sections, the most common types of master oscillator are described, highlighting their relative merits. A sub-section summarising and contrasting the attributes of master oscillator options concludes this section.

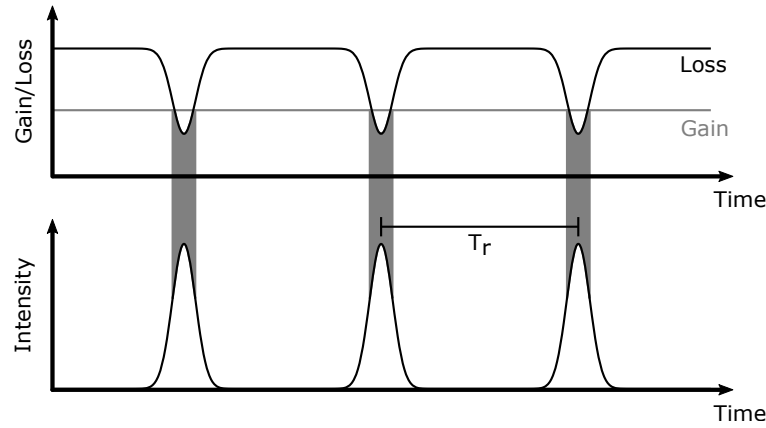


Figure 3.1: Illustration of the pulse formation mechanism in passive mode-locking using a fast saturable absorber. The optical pulse modulates the cavity loss via the saturable absorber, giving a window of net gain (shaded grey region) that leads to pulse shortening. After many round-trips, a steady-state pulse duration is obtained, spaced by the cavity round-trip time (T_r).

3.1.1 Mode-locked fibre lasers

For subpicosecond operation, PML lasers are the only feasible option as no other sources have sufficiently broad coherent bandwidths to support ultrafast pulses. In a PML laser, multiple cavity longitudinal modes are forced to oscillate with a fixed phase relationship by a saturable absorber that exhibits an intensity-dependent loss (Fig. 3.1). Interference between the oscillating modes results in the formation of a pulse train, whose pulse duration is determined by the number of oscillating modes, spaced by the cavity roundtrip time. PML was first demonstrated using a Q-switched Ruby laser with a liquid saturable filter in the cavity [Moc65].

The first ultrafast PML fibre laser was reported in 1990 [Dul91] and since then, they have become the natural seed for ultrafast MOPFA schemes due to the compatibility of the emission bandwidths and the alignment-free benefits of a monolithic system. De-chirped pulse durations < 30 fs have been obtained from ytterbium fibre (Yb-fibre) oscillators [Zho08, Kur12, Chi12a] and temporally stretched (using dispersive fibres) Yb-fibre oscillators are routinely used to seed monolithic chirped-pulse amplification (CPA) schemes [Rue10, Fer11, Mor12].

Even for picosecond pulses, where several other options exist, PML fibre lasers are often used because of their reliability, high pulse quality, low jitter and ease of integration. The main drawback of PML fibre lasers is the inflexibility caused by the fact that the pulse duration and repetition rate are fixed by the laser cavity parameters. Fine adjustment of the cavity length can be introduced by using piezo-actuated fibre stretchers [Was04] but

these only allow for a <10 ppm adjustment of the repetition rate. PML fibre lasers typically operate at megahertz repetition rates, therefore, pulse-pickers based on acousto-optic modulators (AOMs) or electro-optic modulators (EOMs) are frequently used to reduce the repetition rate to achieve high pulse energies after amplification. This affords some flexibility of the repetition rate but only factors of the original oscillator repetition rate can be selected and the emission of pulses cannot be electronically synchronised to an external clock signal.

The issue of external synchronisation can be somewhat alleviated by actively mode-locked (AML) fibre lasers, which pre-date their passive counterparts [Alc86a, Dul88, Kaf89]. Due to the finite electrical bandwidth of the EOMs used in the fibre laser cavity, subpicosecond pulse durations cannot generally be obtained from AML fibre lasers. This, combined with the complex electronics needed to drive the modulator and stabilise the repetition rate, means their use is far less widespread than PML fibre lasers. The ability to drive the intracavity modulator at high harmonics of the fundamental round-trip time, however, enables picosecond pulse trains with repetition rates of tens of gigahertz to be obtained. Using harmonic mode-locking in combination with cavity length adjustment, continuous tuning of the repetition rate can be achieved over a broad range (e.g. from 5–20 GHz) and the emission can be synchronised to an external clock. These high repetition rate sources are very useful for high bit-rate telecommunications applications but their very low pulse energy is not suitable for seeding high energy MOPFA systems.

3.1.2 Q-switched fibre lasers

For applications requiring high pulse energy, most notably in materials processing, Q-switched fibre lasers are often used as a simple seed for MOPFA schemes. Q-switching involves modulating the cavity loss, such that a large population inversion builds when the resonator loss is high. When the resonator loss is switched to a low value, spontaneous emission is rapidly amplified depleting the stored energy until the laser action terminates, resulting in the emission of a highly energetic pulse (Fig. 3.2).

The Q-switching of fibre lasers was reported just prior to active mode-locking in 1986 [Alc86b], using an almost identical cavity but with a AOM in place of the acousto-optic mode-locker to modulate the cavity loss. The maximum extractable energy from a fibre amplifier is approximately ten times the saturation energy [Ran01]. Since the saturation fluence for ytterbium-doped glass is $\sim 0.3 \mu\text{J}/\mu\text{m}^2$, the extractable energy simply scales with the mode-field area (MFA) of the fibre. Scaling the MFA of active fibres is more commonly discussed in the context of mitigating nonlinearities (cf. Section 1.3.6), but the challenge remains the same: maintaining single-mode guidance whilst increasing the MFA. Rod-type photonic crystal fibres (PCFs) have demonstrated the largest MFAs whilst

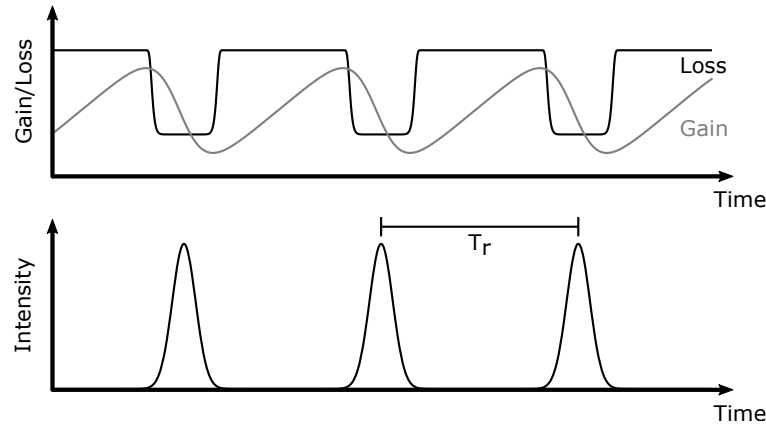


Figure 3.2: Illustration of the pulse formation mechanism in active Q-switching. The resonator loss is held at high value to allow a large population inversion to build, such that a highly energetic pulse is emitted when the loss is switched to a low value. The loss switching occurs periodically, leading to the formation of a pulse train with the modulation period T_r .

maintaining diffraction-limited beam quality [Lim05a, Lim12b]; using these fibres, the pulse energy of an Q-switched oscillator has been scaled to 2 mJ [Sch07] and up to 26 mJ has been achieved in a MOPFA configuration [Stu12]. Whilst this pulse energy is orders of magnitude smaller than other laser media can achieve, the average power of 130 W with diffraction-limited beam quality using only passive cooling is difficult to match with Q-switched solid-state systems [Hon99].

Whilst rod-type PCF systems offer the best performance figures, most of the benefits of fibre systems are lost due to the ~ 1 m rigid length of the amplifiers and the bulk-coupling of the pump and signal radiation. Monolithic Yb-MOPFA schemes using Q-switched fibre laser seeds are a compact, robust and cost effective solution for industrial applications such as marking and precision machining. Pulse energies of a few millijoules can be obtained with close to diffraction-limited beam quality [Pip04, Shi14] and ~ 10 mJ is achievable with multimode output [Mar06, Fan13]. The pulse duration of Q-switched fibre oscillators is proportional to the inversion ratio and cavity length, therefore, special fibre designs with very high pump absorption are required to obtain pulse duration < 10 ns [Lim05b]. The operational range of the repetition rate of Q-switched fibre lasers is generally restricted to 1–100 kHz, the lower limit is set by issues with amplified spontaneous emission (ASE) and the upper limit is set by the time required to build a sufficient inversion level. The lack of flexibility of the pulse duration, pulse shape and repetition rate restricts the applicability of Q-switched seeded MOPFAs, primarily to the fields of materials processing and remote sensing (e.g. LIDAR).

3.1.3 Q-switched microchip lasers

To obtain shorter pulse durations from Q-switched oscillators, solid-state microchip lasers [Zay89] can be used. In a microchip laser, the laser crystal (e.g. Nd:YAG) is monolithically bonded to a saturable absorber (e.g. Cr:YAG) with dielectric mirror coatings at each end forming a planar Fabry-Pérot cavity. Due to the short cavity length, nanosecond pulse durations at ~ 100 kHz repetition rates are most commonly obtained, although pulse durations as short as 37 ps and repetition rates up to 8 MHz have been demonstrated [Spü99]. A feature of passively Q-switched microchip lasers is that the pulse duration and pulse energy are constant so as the pump power is increased, the repetition rate and average power increase proportionally. Microchip lasers can suffer from high pulse-to-pulse jitter and after pulsing if there is competition between multiple longitudinal modes, therefore, most are designed to operate purely or close to single-frequency. Even so, small fluctuations in the pump power, temperature and the fact that the stochastic process of stimulated emission creates the first photon of the oscillation mode, can lead to timing jitters that are a few percent of the pulse period. Although there are passive methods to reduce the jitter to $\sim 10^{-5}$ [Ste10], modulating the pump diode current can reduce the jitter to 10^{-6} and enables triggered operation whilst preserving the simple, compact design [Man96, Lai01].

Passively Q-switched microchip lasers are compact and cost-efficient seed sources for nanosecond Yb-MOPFAs. Compared with other seed sources, their higher pulse energy results in fewer subsequent amplification stages. In addition, the 10–100 kHz repetition rate is very well suited to extracting high pulse energies from fibre amplifiers without being too low as to introduce issues with ASE. Their single-frequency operation is ideal for generating high power spectral densities, especially considering the ~ 1 ns pulse duration effectively mitigates stimulated Brillouin scattering (SBS), which is the main limiting factor on scaling the peak-power of narrow linewidth MOPFAs (cf. Section 1.3.4).

Using coiled step-index large mode-area (LMA) fibres or flexible PCFs, diffraction-limited beam quality has been obtained with peak-powers >1 MW in conjunction with millijoule pulse energies and narrow spectral linewidths using microchip-seeded Yb-MOPFA schemes [Bro05, Far06b, Sch08]. Further scaling of the peak-power to 4.5 MW with a pulse energy of 4.3 mJ has been achieved using a 100 μm core rod-type PCF, with diffraction-limited beam quality and a spectral linewidth of 20 GHz [Bro06]. This peak-power exceeds the theoretical self-focusing limit of ~ 4 MW for 1 μm operation in fused silica, which sets the upper limit on the achievable peak-power in optical fibres as it is independent of the mode area (cf. Section 1.3.6). In common with Q-switched fibre lasers, the seeding of MOPFAs with Q-switched microchip lasers, whilst demonstrating exemplary performance, allows no flexibility in the pulse parameters and operational repetition rate ranges are fairly limited.

3.1.4 Directly modulated semiconductor laser diodes

The previously introduced MOPFA seed sources, based on active or passive modulation of the laser cavity gain or loss, suffer from the same drawback: the pulse characteristics are largely determined by the cavity parameters and the repetition rate is either fixed or operates in a range that is only suitable for certain applications. A prime example of this hindrance is found in materials processing, where Q-switched fibre laser seeded Yb-MOPFA schemes were the dominant technology due to the ability to obtain high pulse energies in very simple and robust monolithic setups. The demand for higher average powers (requiring higher repetition rates) to increase throughput and the desirability of adaptive pulse shaping, however, has led to directly modulated semiconductor laser diode seeds replacing Q-switched fibre lasers in many materials processing applications [Zer06].

The most cost-effective and easily fabricated semiconductor diodes are of the Fabry-Pérot (FP) type, where the laser cavity is formed by a high-reflection (HR) and partial-reflection dielectric coatings on facets of the semiconductor gain chip. To directly modulate a semiconductor laser diode, current pulses of a few Amps, much greater than the 10's of milliamps threshold currents, with high duty factors are applied to the diode. Using FP laser diodes with direct modulation, the optical pulse width closely follows the electrical pulse width and is generally tunable from >100 – 1 ns with peak-powers of ~ 1 W.

The current pulse supplied to the diode can be driven by an arbitrary waveform generator (AWG), which enables the optical pulses to be shaped with nanosecond resolution. This can be used to compensate for pulse shaping effects due to gain saturation in subsequent amplification stages [Vu06] or to create 'designer' pulses for specific micromachining applications [Gay09, Bar15]. Monolithic Yb-MOPFA schemes seeded by directly modulated FP diodes have generated shaped pulses of tunable duration (0.5–500 ns) with 265 W of average power and 10.6 mJ of pulse energy [Mal13], equivalent to Q-switched seeded Yb-MOPFA schemes but with far greater flexibility.

The main drawback of FP diodes is that due to the broad bandwidth of both the gain and the feedback, the spectral output consists of a broad (few nanometres) envelope comprised of several oscillating longitudinal modes (~ 0.2 nm mode spacing at 1064 nm). The linewidth of each mode is of the order of ~ 10 MHz, which is narrower than the SBS bandwidth of silica fibres (10's MHz). Therefore, each mode can efficiently excite SBS and hence the SBS threshold is determined by the number of mode oscillating [Lic87]. Due to mode competition, the spectral output of a directly modulated FP laser diode can sporadically become dominated by a single mode, dramatically reducing the SBS threshold. Since the backward propagating SBS pulses can lead to catastrophic damage, fibre Bragg gratings (FBGs) are frequently spliced to the output of FP laser diodes to elongate the cavity and hence decrease the mode spacing. A reasonably broad bandwidth

(~0.3 nm) FBG is usually chosen, such that a large number of modes oscillate, increasing the SBS threshold beyond the competing nonlinearities [Mor09, Mal09, Jol14]. In addition, the FBG fixes the central wavelength of the diode with a much smaller wavelength shift with temperature than that of the free-running FP diode, effectively stabilising the spectral emission. The highest reported pulse energy obtained from a Yb-MOPFA utilised a FBG-stabilised directly modulated FP laser diode as a seed [Che05a]. Up to 82 mJ was obtained using a 200 μm core diameter step-index fibre, which supports >200 modes but by coiling the gain fibre to introduce mode-filtering, the number of modes in the output was reduced to ≤ 20 modes, corresponding to an $M^2 = 6.5$.

An alternative method for avoiding potential issues with SBS when directly modulating semiconductor laser diodes is to use pulsed superluminescent laser diodes (SLDs). SLDs have structures (e.g. tilted or curved gain strips) and coatings that suppress any feedback, leading to emission of broadband (10's of nanometres) ASE that does not have any mode structure. Thus SBS is mitigated due to the broad spectral linewidth that is independent of the pulse parameters, in contrast to directly modulated FP laser diodes [Nod11]. Using a directly modulated SLD, a Yb-MOPFA with 670 kW of peak-power and a pulse energy of 3 mJ has been reported [Men14]. The power spectral density of an SLD seeded Yb-MOPFA is typically very low, however, this is not an issue for an application such as materials processing; whereas, increasing the SBS threshold is vital to enable long pulse durations to be used. The spectral power density can be increased with the use of narrow bandwidth filters, at the expense of a greater number of amplification stages being required [Ber15].

3.1.5 Gain-switched semiconductor laser diodes

To obtain subnanosecond pulses from semiconductor diodes, the technique of gain-switching (a subset of Q-switching) is frequently used. In gain-switching, the diode is driven by a short electrical pulse that takes it from below threshold to gain inversion and thus the diode undergoes relaxation oscillations. If the driving pulse is sufficiently short in duration, the laser action that terminates after the first relaxation oscillation will not be initiated until the next driving pulse. In this manner, a single relaxation oscillation spike is emitted each electrical driving pulse period (Fig. 3.3). Although the conceptual implementation is identical to direct modulation – applying a current pulse offset with a bias current – in practice because the pulse formation mechanism is different, the pulse characteristics and drive conditions are quite dissimilar. Lower peak current (~200 mA), shorter pulse duration (~100's ps) driving pulses are typically used. The output pulse duration is fixed by the laser diode cavity roundtrip time and the magnitude of the driving pulse, and it is usually considerably shorter than the electrical pulse (10–100 ps) [Ito79, Lin80]. Since the output pulse does not follow the electrical driving pulse

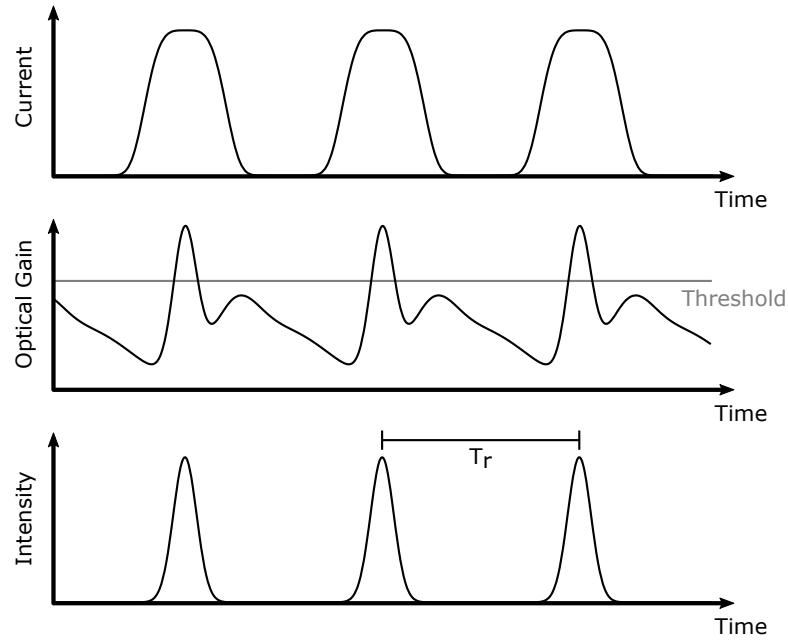


Figure 3.3: Illustration of the pulse formation mechanism in a gain-switched laser diode. The current pulse creates gain inversion, resulting in the emission of a relaxation oscillation that depletes the gain, terminating the laser action. The duration of the current pulse is sufficiently short, such that only one relaxation oscillation is excited for every electrical pulse period T_r .

shape, more simple sinusoidal modulation can be implemented, however, AWGs cannot be used to shape the optical pulses.

Gain-switching can be applied to FP laser diodes to produce subnanosecond pulses, however, the spectral output is broadened from a few nanometres to tens of nanometres [Zie79, Koc84, Lin83]. This is due to the frequency chirp imparted by refractive index change at the rising and falling edges of the current pulse, caused by the large change in carrier densities. To obtain single longitudinal mode (SLM) operation that is desirable for subsequent nonlinear frequency conversion [Raf01] or temporal compression [Tak89], the diode can be injection seeded by a narrow linewidth source [Kob80]. The injected seed can either be an external CW laser [Mat97] or self-seeded by a wavelength-selective reflection, usually from a (fibre) grating [Lun83, Zha97]. The obvious drawback of these techniques is the more complicated and costly optical setup, which detracts from the greatest benefits of semiconductor lasers – their simplicity and low cost. Furthermore, in the case of self-injection seeding, the optical delay between the wavelength-selective feedback (e.g. an FBG) and the laser diode has to be close to but slightly shorter than the electrical driving pulse period such that the reflected SLM pulse seeds the formation of the next relaxation oscillation. This removes the ability to vary the repetition rate

arbitrarily, another key benefit of gain-switching, however, it does significantly improve the pulse-to-pulse jitter [Sch94]. External seeding [Dup06b] and self-seeding [Che10] has been successfully used to control the spectral output of gain-switched FP laser diodes for the seeding of fibre-integrated Yb-MOPFAs.

DFB and DBR laser diodes

An alternative method for obtaining controlled spectral emission from a gain-switched laser diode is to use a gain chip that incorporates wavelength selective feedback to obtain SLM operation. Two gain chip structures exist to achieve this, distributed feedback (DFB) and distributed Bragg reflector (DBR). A DFB laser diode has a low-index-contrast, weakly-reflecting Bragg reflector distributed along the entire length of the gain ridge, buried in a high-index layer which then has a low-index cladding layer regrown over the top to provide waveguiding. In contrast, a DBR laser diode uses a high-index-contrast, strongly reflecting Bragg reflector etched into a passive ridge section at one end of the device. In a DFB laser diode, the cavity is formed by a HR coating on one facet and the distributed feedback from the grating, the output facet is anti-reflection (AR) coated to outcouple the light and destroy the symmetry of the device. In a DBR laser diode, the cavity is formed by the Bragg grating at one end and a partially reflective coating on the output facet. In both cases, due to the small reflection bandwidth of the Bragg reflector and the large free-spectral range of the short (mm) cavity, SLM operation is achieved.

The main difference between the two structures is the fact that when the active regions heat up due to current injection, in a DFB laser diode the grating also heats up; whereas, in a DBR laser diode, the grating is in a passive section so it remains at the temperature of the substrate. This results in a DBR laser having a smaller wavelength shift with drive current (~ 2 pm/mA cf. ~ 8 pm/mA for DFB) and as the drive current is increased, mode-hops occur when the emission of the active region red-shifts beyond the gain bandwidth of the grating. In contrast, in a DFB laser diode, as the drive current increases the reflection wavelength of the grating red-shifts in tandem with the gain, resulting in mode-hop-free operation. By varying the device temperature, both laser diode types can be tuned over ~ 2 nm at a rate of ~ 80 pm/°C. DBR laser diodes can provide higher average powers than DFB laser diodes since the feedback is decoupled from the active region but the latter is normally favoured for gain-switched operation due to the slightly shorter cavity length (giving shorter pulse durations) and more stable spectral characteristics. Both device structures exhibit CW linewidths of a few MHz, which is mainly constrained by the short cavity length as described by the Henry modified version of the Schawlow-Townes equation [Sch58, Hen82].

Gain-switched DFB laser diodes are a highly practical seed source for fibre-integrated

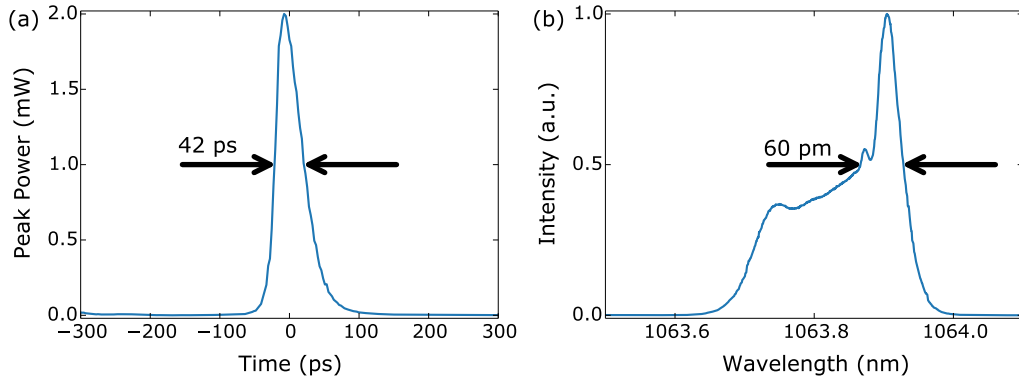


Figure 3.4: (a) Sampling optical oscilloscope trace and (b) optical spectrum of a gain-switched DFB semiconductor laser diode.

Yb-MOPFA schemes: they generate high-quality 10's of picosecond pulses using simple drive electronics that can be triggered at arbitrary repetition rates over a large range (single shot to several GHz). Figure 3.4 (a) shows 40 ps pulses generated by a gain-switched DFB laser diode that is used later in this chapter, it is clear from the spectrum [Fig. 3.4 (b)] that the CW linewidth (<10 MHz) is significantly chirped by the pulsed operation, with a time-bandwidth product of 0.67 (the minimum time-bandwidth product for a Gaussian pulse is 0.44). The main drawback of gain-switched operation of semiconductor diodes is, in common with other Q-switched sources, the fact that the pulse duration is mainly dictated by the cavity parameters. Therefore, although the duration can be tuned somewhat by adjusting the electrical drive parameters (duration, peak current), it is difficult to obtain tunable subnanosecond pulses without a transient being present, caused by the first relaxation oscillation [cf. Fig. 3.13 (a)].

3.1.6 Actively mode-locked semiconductor laser diodes

Some of the shortcomings of gain-switched semiconductor lasers can be overcome by mode-locking; in particular, shorter pulses with less frequency chirp and lower jitter can be produced. In contrast to most other laser architectures, since laser diodes are electrically pumped, active mode-locking [Ho78] is generally easier to implement than passive. Active mode-locking of FP laser diodes would require very high modulation frequencies (100's GHz) due to the short cavity length, which makes the drive electronics very challenging to implement. The active mode-locking of laser diodes is, therefore, usually performed in external cavities [Vas95]. Inscripting an FBG into the output fibre pigtail to form the laser cavity [Bur88, Mor92] is the most convenient configuration, as it does not impact on the practical benefits of semiconductor diode lasers as a seed source for MOPFA schemes.

The only difference between this cavity and the one frequently used for FBG-stabilised FP laser diodes is the use of a high-quality AR coating on the output facet of the diode, to prevent deleterious parasitic FP effects. By modulating the current supplied to the diode at the same frequency as the round-trip time (or a harmonic of) mode-locking is achieved using electronics that are almost identical to those used for gain-switching. The use of an FBG has the additional benefit of controlling the bandwidth of the laser oscillation, such that time-bandwidth limited (chirp-free) pulses can be generated [Mor92].

Most of the research effort on actively mode-locked semiconductor laser diodes has been focused in the erbium gain band, to satiate the demand for transform-limited picosecond pulses for telecommunications applications. More recently, actively mode-locked external FBG laser diodes have been investigated for the seeding of Yb-MOPFA schemes: a source generating transform-limited 18 ps pulses has been demonstrated that exhibited significantly less jitter than in gain-switched operation [Teh14]. A Yb-MOPFA scheme seeded with a similar source was reported with a polarised average power of 200 W, diffraction-limited beam quality and a peak-power of 100 kW [Teh13]. Whilst the pulse quality (jitter, chirp, profile etc.) of AML semiconductor diode lasers is superior to gain-switched DFB laser diodes, the ability to trigger at arbitrary repetition rates is lost, in common with all mode-locked lasers.

3.1.7 External modulation

In the proceeding sections, methods for generating pulsed emission from a variety of laser sources was discussed. An alternative approach for generating pulses to seed MOPFA schemes is to modulate the output of a CW (or macro-pulsed) laser source. The greatest benefit of this approach is the fact that the spectral characteristics are predominantly determined by the laser source, whereas, the temporal properties are determined by the modulator. This decoupling of the spectral and temporal properties is in stark contrast to most pulsed lasers sources, where the two are inexorably linked and tied to the gain medium used. The two main types of modulator, acousto-optic modulators (AOMs) and electro-optic modulators (EOMs), have broadband operation which allows spectral flexibility, including the possibility of extremely narrow linewidth operation.

Acousto-optic modulators

AOMs work on the principle that when a sound wave is applied to a material, typically tellurium dioxide (TeO_2) for 1 μm operation, the dynamic strain causes density variations that forms a periodic refractive index grating. A piezoelectric transducer (typically lithium niobate) is used to apply a travelling sinusoidal sound wave to the material in the transverse direction to the incoming light from an electrical radio-frequency (RF) signal

(typically ~ 100 MHz). The incoming light is diffracted at an angle determined by the Bragg condition and the RF power determines the proportion of light that is diffracted. Since the light is diffracted by a travelling acoustic wave, the optical carrier frequency is shifted by the acoustic drive frequency. By spatially isolating the diffracted beam from the non-diffracted beam, modulation of the sinusoidal RF signal results in optical intensity modulation.

Fibre-coupled AOMs are arranged such that the first diffracted order is coupled into the output fibre pigtail, whereas, the non-diffracted beam is dumped inside the casing. This results in a very high (>50 dB) optical extinction ratio between the on state (RF signal applied, beam diffracted) and the off state (RF signal off, beam not diffracted). With proper design, up to 85–90% diffraction efficiency is achievable giving a typical fibre-to-fibre insertion loss in the on state of ~ 2 dB. The low insertion loss and high damage threshold of the acousto-optic material result in average power handling capabilities of ~ 1 W for fibre-coupled devices and far higher (>100 W) for bulk devices based on fused silica.

The main drawback of AOMs is their relatively slow rise time, determined by the time taken for the acoustic wave to cross the laser beam. The transit time is proportional to the beam diameter and the inverse of the acoustic velocity, therefore, the incident beam needs to be focused to obtain rise times < 150 ns. There is a compromise, however, as the diffraction efficiency decreases with reducing spot size and additionally the peak intensity handling of the acousto-optic crystal must be taken into consideration. Furthermore, the RF driver rise time, which is proportional to the drive frequency, also contributes to the optical modulation rise time, resulting in a minimum rise time of ~ 5 ns for fibre-coupled AOMs.

AOMs can be used to generate pulse trains from very narrow linewidth (kHz) sources, such as DFB fibre lasers and external cavity semiconductor lasers, which would otherwise be difficult to obtain pulsed emission from [Phi04, Zhu12]. The relatively long (~ 5 ns) minimum pulse duration does not suppress SBS, therefore, to amplify to high peak-powers, such sources must be controllably spectrally broadened using a phase modulator. Conversely, AOMs have also been used to modulate filtered ASE sources to prevent SBS in MOPFA schemes [Mel14], highlighting their spectral versatility. Due to their high extinction ratio and high power handling capabilities, AOMs are more commonly used in high-gain MOPFA schemes for time-gated in-band ASE suppression, isolation and pulse picking or shaping [Che05a, Mal13].

Electro-optic modulators

Electro-optic modulators (EOMs) are used for external modulation applications that require higher modulation bandwidths than can be achieved by AOMs. EOMs utilise

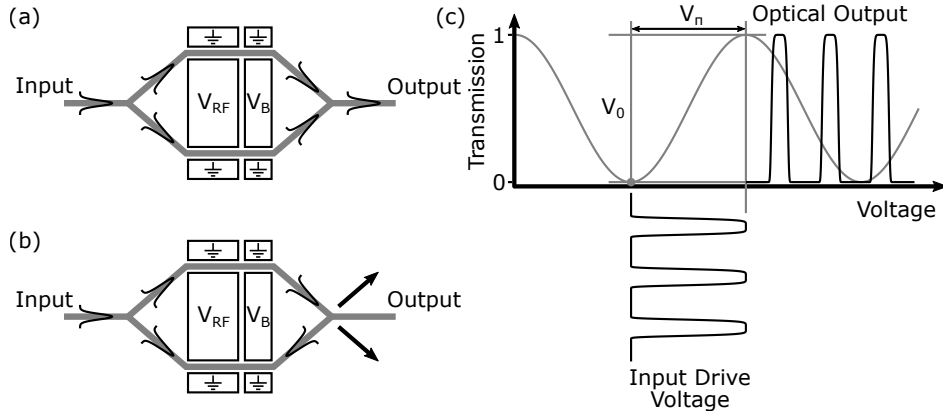


Figure 3.5: Illustration of an MZAM operated at the maximum (a) and minimum transmission points (b). (c) Illustration of an MZAM transfer function, showing how input electrical pulses form an optical pulse train when the modulator is biased at the minimum transmission point.

the Pockels effect, where an externally applied electric field (constant or slowly varying compared with the incident optical frequency) causes a linear change in the refractive index of non-centrosymmetric media. In this manner, an electrically-variable phase delay can be introduced to a light field propagating through an electro-optic crystal by applying a transverse or longitudinal electric field. In the case of bulk optic EOMs, a birefringent electro-optic crystal is used and the incident light is polarised such that the phase delay causes a change in the polarisation state, which is transformed into an amplitude modulation using a polariser on the output. The main drawback of bulk EOMs is the large voltage ($\sim$ kV) required to impart a π phase shift (half-wave voltage), which makes the engineering of very fast rise time electrical drivers very challenging.

To develop compact, low-power and very broad bandwidth optical modulators, lithium niobate (LiNbO_3) waveguide Mach-Zehnder amplitude modulators (MZAMs) were introduced [Ohm75]. A channel waveguide circuit is used in an MZAM, such that the incoming light is split into two paths and then recombined using two Y-junctions, forming a Mach-Zehnder interferometer [Fig. 3.5 (a),(b)]. Applying a voltage to either the RF or DC electrode causes a proportional phase-shift that leads to constructive or destructive interference at the output Y-junction. In the case of constructive interference [Fig. 3.5 (a)], the recombined optical field profile matches the mode of the waveguide and thus the field propagates past the junction. In the case of destructive interference [Fig. 3.5 (b)], the recombined field possesses odd symmetry about the midplane of the waveguide and thus it will excite non-confined radiation modes rather than the fundamental mode. This results in the field not propagating beyond the output Y-junction, therefore, the voltage

applied to the electrodes results in an amplitude modulation that is sinusoidal with the applied voltage [Fig. 3.5 (c)].

To obtain an optical pulse train from a CW laser source, an MZAM is biased using the DC electrode to operate at the minimum transmission point [V_0 in Fig. 3.5 (c)] and an electrical pulse train is applied to the RF electrode to obtain the desired modulation [Fig. 3.5 (c)]. The amplitude of the electrical pulses is empirically adjusted to be equal to the half-wave voltage (V_π) in order to maximise the optical pulse amplitude. In addition, the DC bias voltage needs constant adjustment in operation to maintain the optimum pulse contrast due to temperature fluctuations that shift the operating point.

The main issue of using MZAMs to generate pulse trains is that the modulator transmission is never completely zero – some radiation ‘leaks’ through at the minimum transmission point. This performance is quantified by the static extinction ratio, which is the ratio of the maximum to the minimum transmission power, measured by varying the DC bias between the minimum and maximum transmission points. The static extinction ratio is typically ~ 30 dB, which implies that the pulse energy is equal to the CW background energy in a pulse period when the duty factor is ~ 1000 . Clearly, the static extinction ratio of the modulator determines the maximum pulse energy that can be achieved after amplification of an MZAM modulated CW seed. To achieve very high pulse energy with the temporal flexibility that MZAMs afford, two modulators can be cascaded to obtain static extinction ratios > 50 dB, at the expense of greater cost and complexity.

LiNbO_3 optical waveguide MZAM circuits can be easily integrated into compact fibre-coupled packages, resulting in alignment-free operation with a typical insertion loss of ~ 4 dB at the maximum transmission point. The small dimensions of the electrodes allow very large modulation bandwidths to be obtained (> 40 GHz) with small half-wave voltages (few Volts). Whilst offering vastly superior modulation speeds in comparison to AOMs, the waveguide nature of MZAMs limits the average power handling to ≤ 100 mW. To amplify MZAM modulated seeds to high average power, therefore, requires a large gain factor in the subsequent amplifier stages.

Due to their limited contrast ratio, MZAMs are typically used for high temporal resolution shaping of macro-pulses produced by AOM modulated CW sources [Lag10] or pulsed semiconductor diodes [Mal09]. Using these techniques also reduces the impact of the limited average power handling as the loss from the duty factor reduction is removed from the MZAM stage, reducing the gain requirement for subsequent amplification. The unrivalled spectral and temporal flexibility of combined AOM and MZAM external modulation has lead it to become the front-end laser seed for several of the ultra-high intensity laser facilities around the world [Jol03, Wis04, Lag11].

Parameter	Fibre Laser		Semiconductor			Solid State	External	
	PML	QS	DFB	FP+FBG	AML	Microchip	AOM	EOM
Pulse duration	fs-ps	ns	ps-CW	ns-CW	ps	ns	ns-CW	ps-CW
Duration tunable	No	No	Yes	Yes	No	No	Yes	Yes
Pulse Shaping	No	No	Yes	Yes	No	No	Yes	Yes
Jitter	Low	Mid	Mid	Mid	Low	High	Mid	Mid
Triggerable	No	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Spectral bandwidth	TL	SLM	<0.1 nm	>0.1 nm	TL	SLM	SLM	SLM
Wavelength tuning	Large	Large	Mid	Small	Large	None	Large	Large
Extinction ratio	High	Mid	Mid	Mid	High	Mid	High	Low

Table 3.1: Comparison of frequently used seed sources for MOPFA schemes. PML, passively mode-locked; QS, Q-switched; DFB, distributed feedback laser diode; FP+FBG, fibre Bragg grating (FBG) stabilized Fabry-Pérot (FP) laser diode; AML, actively mode-locked; AOM, acousto-optic modulator; EOM electro-optic modulator; TL, transform-limited; SLM, single longitudinal mode.

3.1.8 Summary

This section has provided an overview of the most commonly used, practical seed sources for MOPFA schemes. The aim was to highlight the tremendous flexibility that the MOPFA architecture affords, allowing high average power and pulse energy to be generated in a conceptually identical manner whilst using seed sources that are tailored to the applications needs. Table 3.1 summarises the typically obtained parameters from the most frequently used MOPFA seed sources. It is worth noting that most of the sources can be engineered to operate outside these parameters but the most commonly obtained values are quoted. The spectral bandwidth of the sources is quoted as the minimum obtainable from a source, which in the case of external modulation will depend on the spectral bandwidth of the seed laser.

Comparing the requirements for a depletion beam source for STED microscopy using GFP (cf. Section 2.2) with the reviewed seed options for Yb-MOPFAs, it is clear that some are not compatible, whilst others are not well suited. The repetition rates of Q-switched fibre and microchip lasers are too low for workable image acquisition times and the pulse duration of Q-switched fibre lasers and AOM modulated sources are too long for effective depletion. Conversely, the pulse durations of actively mode-locked semiconductor lasers are too short to provide effective depletion. In the previous chapter it was shown that a mode-locked fibre laser was a reasonably effective source after the pulse duration was

stretched. It offered no flexibility, however, in either the pulse duration or repetition rate and it could not be triggered by an external clock. Clearly, pulsed semiconductor diodes or external modulation with an EOM, or a combination of the two, presents the best option for developing a temporally flexible depletion beam source. The two following sections of this chapter describe the development of an improved 560 nm source and the comparison of the performance of the system using either an MZAM-based or a gain-switched DFB laser diode as the master oscillator.

3.2 Intensity modulated 560 nm source

3.2.1 Ytterbium MOPFA

The first master oscillator scheme that was implemented was designed for temporal flexibility and performance and was based on an DFB laser diode modulated by an MZAM (Fig. 3.6). A picosecond pulse board (PPB) that incorporated a DFB laser diode, picosecond electronic driver and temperature controller on an integrated circuit board (QD Laser Inc.) was used as a versatile self-contained optical source with a compact footprint ($100 \times 100 \times 20$ mm). The DFB laser diode had a central wavelength of 1064 nm and delivered a CW average power of 50 mW in a SLM, corresponding to a spectral linewidth of <10 MHz. The incorporated picosecond pulse generator (PPG) was electronically programmable in peak current, pulse duration and bias current and had a internal clock source that could be set from 12 kHz–250 MHz or triggered externally from single shot up to a 250 MHz repetition rate. This enabled the PPB to be operated either CW or in gain-switched mode with tunable duration and repetition rate.

The output of the PPB was modulated by an MZAM (Photline Technologies) that had a modulation bandwidth of 10 GHz and a static extinction ratio of 30 dB. It was electrically driven by a PPG (Highland Technologies) that could generate pulses tunable in duration from 100 ps–25 ns with a 60 ps rise time, corresponding to a bandwidth of 5.7 GHz. The PPG was externally triggered and could be operated from single-shot up to a repetition rate of 125 MHz. The 750 mV amplitude electrical pulses of the PPG were amplified to the half-wave voltage of the MZAM using a 12.5 GHz bandwidth RF amplifier (Picosecond Pulse Labs) that ensured the rise time of the electrical pulse was preserved. A static voltage was applied to the DC electrode of the MZAM to bias it at the minimum transmission point, which was continually adjusted during operation to maximise the extinction ratio of the pulse train. The optical pulses generated by the MZAM followed the electrical input pulses, maintaining their 60 ps rise time.

The first stage of the power fibre amplifier chain was a 90 cm double-clad polarisation-maintaining (PM) Yb-fibre pre-amplifier, counter-pumped by a 5 W multimode 960 nm

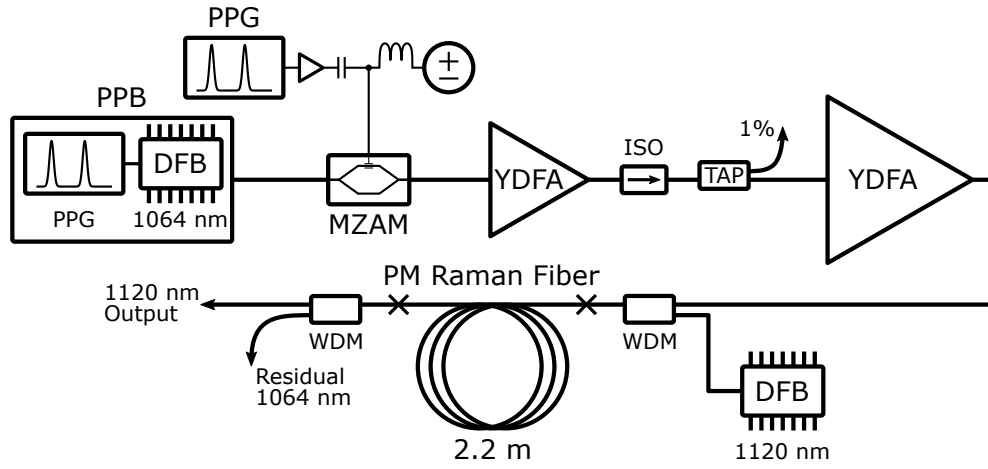


Figure 3.6: Schematic of the Raman amplifier pumped by an amplified intensity modulated diode. PPB, picosecond pulse board; DFB, distributed feedback laser diode; PPG, picosecond pulse generator; MZAM, Mach-Zehnder amplitude modulator; YDFA, ytterbium-doped fibre amplifier; ISO, isolator; WDM, wavelength division multiplexer.

diode. The gain of the pre-amplifier was adjusted by varying the pump power to emit 1 mW of average power irrespective of the input power. An optical isolator was spliced to the output of the pre-amplifier to prevent backwards propagating radiation being amplified and potentially damaging the DFB seed laser. A 1% tap coupler followed the isolator, which enabled the input pulse train to be monitored and facilitated the optimisation of the MZAM bias level. The 99% port of the tap coupler was amplified to a saturated output power of 6 W in a multi-stage commercial PM Yb-fibre amplifier (IPG Photonics).

Stimulated Raman scattering (SRS) had to be avoided in the Yb-MOPFA output, since its presence would lead to a deterioration in the signal-to-noise ratio of the pulse-pumped Raman amplifier. It was determined using 100 ps duration MZAM shaped PPB pulses that the peak-power of the Yb-MOPFA output was restricted to ≤ 3 kW in order to avoid significant SRS ($>0.1\%$ of the spectral power). Due to the 30 dB static extinction ratio of the MZAM, the duty factor of the modulation had to be <250 with the PPB operated in CW mode in order to avoid a significant CW component in the Yb-MOPFA output. This restriction meant that the peak-power limit imposed by SRS could not be achieved using the PPB in CW mode. To overcome this limitation, the PPB could be operated in pulsed mode to create macro-pulses, which were then shaped by the MZAM, enabling the 3 kW peak-power limit imposed by SRS to be achieved.

3.2.2 Raman amplifier

In common with the system described in the preceding chapter, a Raman amplifier was used to spectrally convert the pulsed output of the Yb-MOPFA to a narrow linewidth pulse train at 1120 nm. The Raman amplifier utilised the same DFB laser diode seed, that had a central wavelength of 1120 nm and delivered a CW average power of 40 mW in a SLM, corresponding to a spectral linewidth of <10 MHz. The seed signal was spectrally combined with the pulsed output of the Yb-MOPFA using a PM wavelength division multiplexer (WDM). PM Raman fibre (OFS Fitel) was again used as the Raman gain medium owing to its increased Raman gain coefficient over PM980. In contrast to the previous chapter, the length of the Raman fibre was carefully optimised in order to maximise the peak-power obtained within the spectral acceptance bandwidth of the second-harmonic generation (SHG) crystal. In the previous chapter, the length of the Raman fibre was excessive, which led to the 1120 nm signal being spectrally broadened by self-phase modulation (SPM) beyond the acceptance bandwidth of the crystal, leading to a reduction in SHG conversion efficiency. In addition, the excessive length of Raman fibre imposed a peak-power limitation on the 1120 nm signal, beyond which the power was efficiently transferred to the second Stokes order.

When the length of the Raman fibre was reduced, higher pump peak-powers were required to maintain the gain of the Raman amplifier and the amplifier efficiency, which resulted in the 1120 nm signal having greater peak-power. It was, therefore, expected that the optimal length of the Raman fibre would correspond to the case where the Raman conversion efficiency was optimal with the peak-power of the pulsed pump at its maximum (limited by the onset of SRS in the Yb-fibre power amplifier). It was found, however, that for Raman fibre lengths <2 m, four-wave mixing (FWM) was increasingly favoured over SRS as the Raman fibre length was reduced. Figure 3.7 shows a comparison of the spectral output of the Raman amplifier, pumped by 200 ps MZAM shaped PPB pulses at a repetition rate of 12 MHz, for two different lengths of PM Raman fibre. In both cases, the pump peak-power was optimised to achieve the highest 1120 nm peak-power within the spectral acceptance bandwidth of the SHG crystal (0.15 nm).

It can be seen in Fig. 3.7 that degenerate FWM of the 1120 nm signal resulted in the formation of a pedestal around the base of the signal peak. This process was not expected to be phase-matched, since the high normal dispersion in this region in addition to the nonlinear contribution are both positive leading to an overall effective phase-mismatch (Eqn. 1.51). The high gain of the 1120 nm signal, however, resulted in an effective phase-mismatch that had a nonlinear evolution over the Raman fibre length, which enabled the FWM signal to grow to a significant level [Fèv07].

From Fig. 3.7, it is clear that the degenerate FWM was significantly more prominent

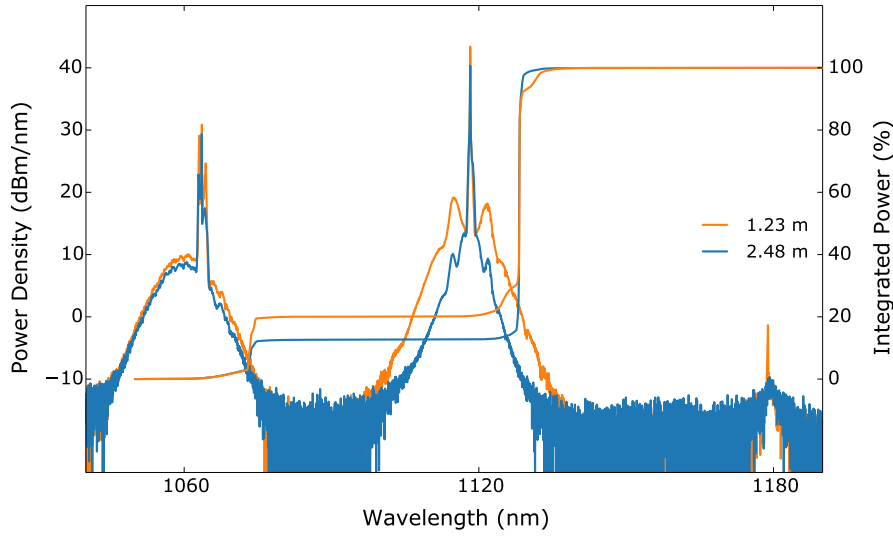


Figure 3.7: Optical spectrum of the Raman amplifier pumped by 200 ps MZAM shaped PPB pulses with a peak-power of 1.1 kW for the 2.48 m length (blue) and 1.5 kW for the 1.23 m length (orange). The integrated power has been offset by 10 nm.

when using the 1.23 m PM Raman fibre length (orange trace) in comparison to the 2.48 m length (blue trace). This was due to the fact that the shorter fibre length and concomitant higher pump peak-power increased the efficiency of the non-phase-matched FWM processes. This was corroborated by the appearance of a sideband at ~ 1179 nm when using the 1.23 m length, created by degenerate FWM of the 1120 nm signal and seeded by the residual 1064 nm pump signal. The larger phase-mismatch of this interaction due to the greater frequency spacing clearly indicate that longer Raman fibre lengths reduce the efficiency of FWM processes, at the expense of a lower optimal pump peak-power for the Raman conversion. It was found that a Raman fibre length of 2.2 m provided the best compromise between the Raman conversion efficiency and the power spectral density of the 1120 nm signal, important for maximising the efficiency of the subsequent SHG stage.

Figure 3.8 (a) shows the spectral output of the 2.2 m Raman amplifier when pumped with 200 ps MZAM shaped PPB pulses at a repetition rate of 21 MHz with 1 kW of peak-power [Fig. 3.8 (a) inset] and seeded with 30 mW CW average power at 1120 nm. Integrating the spectral power revealed that 85% was contained in the 1120 nm signal, which increased to 99.7% after the second WDM was spliced to the output to filter the residual pump power. The highest 1120 nm average power of 3.1 W was obtained from the Raman amplifier using 2.8 ns MZAM shaped PPB pump pulses at a repetition rate of 2 MHz [Fig. 3.8 (b) inset]. This corresponds to an amplifier efficiency (signal output power less the signal input

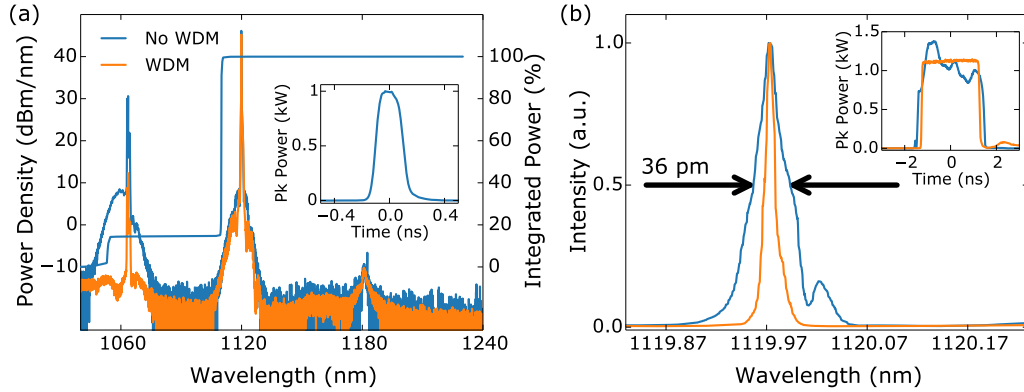


Figure 3.8: (a) Optical spectrum of the Raman amplifier before (blue) and after (orange) the second WDM, pumped with 200 ps MZAM shaped PPB pulses with 1 kW of peak-power (inset). The integrated power has been offset by 10 nm. (b) Linear optical spectrum of the 1120 nm amplified signal when the PPB was operated in pulsed (blue) and CW (orange) modes. Inset: sampling optical oscilloscope traces of the corresponding pump pulses.

power over the pump power) of 51%, considerably higher than the efficiency achieved in the previous chapter due to an increase in the Raman conversion efficiency and a reduction in the insertion losses of the amplifier.

The 3 dB spectral linewidth of the 1120 nm signal was 36 pm [blue trace Fig. 3.8 (b)], which was broadened from the initial seed linewidth due to SPM. The spectral linewidth could be reduced to below the 10 pm resolution limit of the optical spectrum analyser (OSA) [orange trace Fig. 3.8 (b)] by operating the PPB in CW mode and using the MZAM only to shape 2.5 ns pump pulses at a repetition rate of 2 MHz (Fig. 3.8 (b) inset orange trace). In this configuration, the 1120 nm average power was reduced to 2.71 W due to the CW component that was present in the Yb-MOPFA, caused by the limited extinction ratio of the MZAM.

The discrepancy between the Raman conversion efficiency obtained from the spectral integration (85%) and the figure obtained from the average power conversion (51%) was due to the attenuation of the pump and signal powers. The Yb-MOPFA pump power was attenuated by the splice to the WDM (0.6 dB), the insertion loss of the WDM (0.3 dB) and the splice to the Raman fibre (0.5 dB). The amplified 1120 nm power was attenuated by the splice to the output WDM (0.5 dB), the insertion loss of the output WDM (0.3 dB) and the Fresnel reflection from the angle-cleaved output fibre (0.15 dB). When these losses are taken in account, the conversion efficiency agrees very closely with the figure obtained from the spectral integration.

In contrast to the previous chapter, all of the amplifiers used in the Yb-MOPFA were

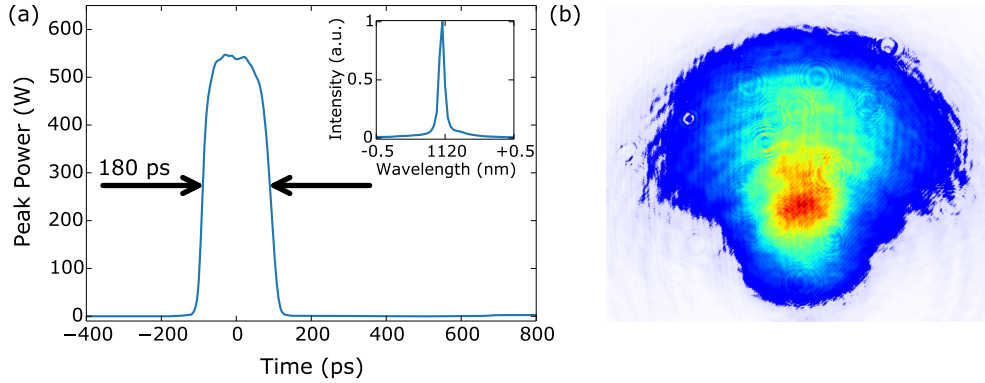


Figure 3.9: (a) Sampling optical oscilloscope trace of the 1120 nm Raman amplifier output and linear optical spectrum (inset) used to pump the fibre-coupled frequency-doubling module. (b) CCD image of the fibre-coupled frequency-doubling module output beam, exhibiting distortion caused by photorefractive damage.

PM, which combined with the use of a polarised seed and in-line polarisers before each amplifier resulted in the output of the Yb-MOPFA having a very high polarisation purity (polarisation extinction ratio (PER) of >30 dB). The 1120 nm output of the Raman amplifier had a PER of >20 dB, which was primarily limited by the PER of the output WDM. The use of a polarised pulsed pump system and PM fibre in the Raman amplifier resulted in a 1120 nm output power that was unaffected by environmental perturbations to the fibres.

3.2.3 Frequency-doubling

The output of the Raman amplifier was spliced to the fibre-coupled frequency doubling module described in Section 2.5. A maximum 560 nm average power of 880 mW was generated for 2.0 W of 1120 nm average power, with a pulse duration 180 ps at a repetition rate of 21 MHz and a spectral linewidth <20 pm, limited by the resolution of the OSA [Fig. 3.9 (a)]. To scale the 560 nm average power, the repetition rate was increased to 28 MHz but this led to catastrophic photorefractive damage (PRD): the output power of the module dropped to 140 mW and the output beam became severely distorted [Fig. 3.9 (b)]. The exact PRD threshold is specific to an individual crystal but for green light, it is generally considered to be approximately 10 MW/cm^2 for 5% MgO-doped periodically poled lithium niobate (PPLN) [Kie12]. Since the fundamental beam was focused to a waist radius of $30 \mu\text{m}$ in the PPLN crystal and the second-harmonic peak-power was estimated to be 240 W, the peak intensity of the second-harmonic was $\sim 17 \text{ MW/cm}^2$. It is, therefore, unsurprising that PRD was observed and that the onset was almost instantaneous as fundamental

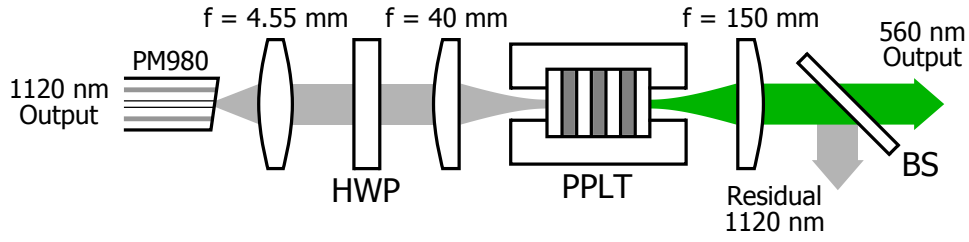


Figure 3.10: Schematic of the 560 nm second-harmonic generation stage. HWP, half wave plate; PPLT, periodically poled lithium tantalate; BS, beam splitter.

peak-power was increased. In mild cases of PRD, the refractive index grating formed by the photorefractive effect can be erased by annealing the crystal at a temperature $>170^{\circ}\text{C}$ for several hours, to allow the charge carriers re-diffuse [Ash66]. To this end, a hot-plate was used to hold the damaged PPLN crystal at 200°C for 4 hours but it was found that the PRD was irreversible.

In order to scale the peak-power of the 560 nm source, a frequency-doubling crystal with a higher PRD threshold was required. Since the maximum available 1120 nm peak-power was reasonably modest ($\sim 500\text{ W}$), only periodically poled crystals were considered as a non-poled crystal would not give reasonable conversion efficiencies with the achievable peak intensities. The two most widely used periodically poled crystals after PPLN are periodically poled lithium tantalate (PPLT) and periodically poled potassium titanyl phosphate (PPKTP), whose physical and optical properties are summarised in Table. 1.1. PPLT was chosen over PPKTP since the higher thermal conductivity and lower green absorption result in a greater potential to scale the 560 nm average power due to the reduced issues with thermal dephasing (cf. Section 1.4.5) [Kum09].

The PPLT crystal used had a length of 20 mm and was poled with a single $9.11\text{ }\mu\text{m}$ pitch grating (HC Photonics), resulting in a phase-matching bandwidth of 0.15 nm for a fundamental wavelength of 1120 nm. To further increase the PRD threshold, the crystal was in the stoichiometric form and was doped with MgO [Nak02]. The PPLT crystal was mounted in a resistive-heated copper oven to maintain the temperature at the quasi-phase-matching optimum of 123.6°C for the 1120 nm fundamental wavelength. The calculated temperature acceptance bandwidth of the PPLT crystal was 1.4°C , which set a theoretical upper limit on the SHG power stability of $<1.5\%$ considering the $\pm 0.1^{\circ}\text{C}$ precision of the temperature control.

The SHG was performed using a simple, single-pass optical setup, depicted in Fig. 3.10. The fiber output of the Raman amplifier was angle-cleaved to suppress feedback and quasi-collimated using an $f = 4.55\text{ mm}$ aspheric lens. It was found after empirical optimisation that an $f = 40\text{ mm}$ (plano-convex) lens gave the maximum SHG efficiency at the highest

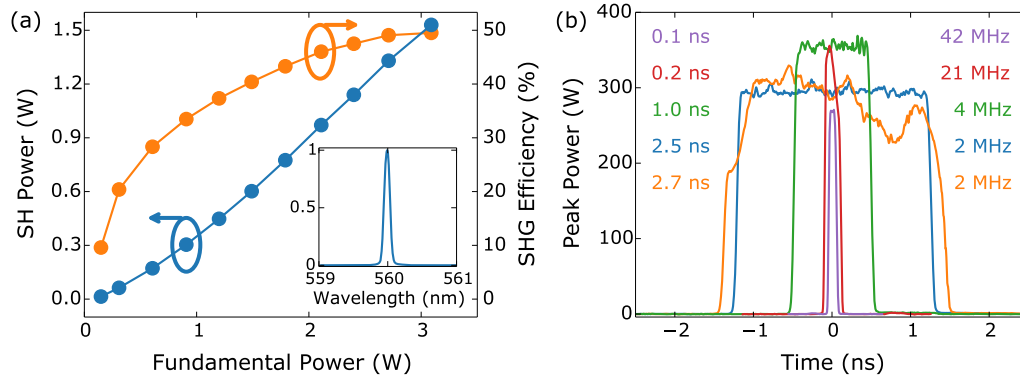


Figure 3.11: (a) Second-harmonic (SH) power and conversion efficiency as a function of fundamental average power and optical spectrum of the SH (inset). (b) Sampling optical oscilloscope traces of optimised 560 nm pulse durations at a variety of repetition rates when the PPB was operated pulsed (orange, red, purple) and CW (blue, green).

fundamental peak-power. The back focus of the Raman amplifier output collimating lens was also optimised in conjunction with the crystal position as a fine control of the spot size in the crystal. Using a scanning slit beam profiler, the optimum $1/e^2$ waist diameter was measured to be 42 μm , with the focus at the centre of the PPLT crystal. The estimated confocal parameter in the crystal was 2.5 mm ($n = 2.13$), corresponding to a focusing parameter ($\xi = L/b$) of 3.8, slightly over-focused in comparison to the theoretical optimum Boyd-Kleinman focusing condition of 2.84. The increased SHG conversion efficiency using over-focusing was most likely a result of the increased spectral acceptance bandwidth associated with the reduction in the effective crystal length. The generated second-harmonic (SH) was collimated by an $f = 150$ mm lens and the residual fundamental was separated using a beam splitter with an attenuation of >30 dB at 1120 nm. The input and output facets of the PPLT crystal were AR coated for the fundamental and SH and all of the lenses used had appropriate AR coatings.

The maximum obtained 560 nm average power was 1.53 W, generated from 3.09 W of 1120 nm fundamental [Fig. 3.11 (a)] using MZAM shaped PPB 1064 nm pump pulses of 2.8 ns duration at a repetition rate of 2 MHz [blue trace Fig. 3.8 (b) inset]. This corresponded to a conversion efficiency of 50% and a pulse energy of 765 nJ, close to two orders of magnitude higher than the system described in the previous chapter. The 560 nm output pulse duration was 2.7 ns with a peak-power of 330 W, it is clearly evident from Fig. 3.11 (b) (orange trace) that the output pulse had significant intensity modulations that originate from the PPB pulses [blue trace inset Fig. 3.8 (b)]. These modulations could be eliminated by operating the PPB in CW mode and only using the MZAM to generate the pulses, the

Pulse Source	Average Power	Repetition Rate	1120 nm Power	Conv. Eff.	Pulse Energy	Pulse Duration	Peak Power
PPB + MZAM	1.53 W	2 MHz	3.09 W	50%	765 nJ	2.7 ns	330 W
MZAM	1.46 W	2 MHz	2.71 W	54%	730 nJ	2.5 ns	300 W
MZAM	1.40 W	4 MHz	2.72 W	51%	350 nJ	980 ps	360 W
PPB + MZAM	1.40 W	21 MHz	3.01 W	47%	67 nJ	200 ps	350 W
PPB + MZAM	1.15 W	42 MHz	2.80 W	41%	27 nJ	100 ps	270 W
PPB + MZAM	0.91 W	58 MHz	2.45 W	37%	16 nJ	56 ps	280 W

Table 3.2: Summary of 560 nm output parameters using the MZAM-based master oscillator. The pulse duration was optimised at each repetition rate to obtain the highest 560 nm average power.

optimum pulse durations for 2 MHz and 4 MHz repetition rates are shown by the blue and green traces, respectively, in Fig. 3.11 (b).

The 560 nm pulse duration was continuously tunable over the range 56 ps–2.7 ns, a summary of the 560 nm output parameters for a selection of pulse repetition rates with the pulse duration optimised to give the maximum 560 nm power is shown in Table 3.2. To obtain the maximal 560 nm average power, the peak-power of 1064 nm pump of the Raman amplifier was effectively constrained: it had to be high enough to obtain efficient Raman conversion but low enough to avoid a reduction in the 1120 nm power spectral density due to competing nonlinear effects and excitation of the second Stokes order. Since the SHG conversion efficiency is proportional to the peak-power within the spectral acceptance bandwidth of the crystal, optimisation of the Raman amplifier length was critical to achieving a good conversion efficiency from the Yb-MOPFA pump to the 560 nm signal. A result of the constrained Yb-MOPFA pump peak-power was that the peak-power of the optimised 560 nm pulse was effectively constant, since the focusing conditions remained the same for each repetition rate. The slight variations in the achieved 560 nm peak-power (10% variance about the 315 W mean) were due to the differences in Raman and SHG conversion efficiencies and the pulse profiles [Fig. 3.11 (b)].

Watt-level 560 nm average power was achieved across the pulse duration tuning range for all but the shortest duration pulses. The reduction in average power at the higher repetition rates was due to the decrease in the Raman conversion efficiency, as evidenced by the decline in the 1120 nm Raman amplifier output power. This reduced the peak-power available for SHG, thus decreasing the conversion efficiency of the process. When the PPB was operated in CW mode and only the MZAM was used to generate the pulses, the 1120 nm average power was reduced due to the presence of the CW component in the

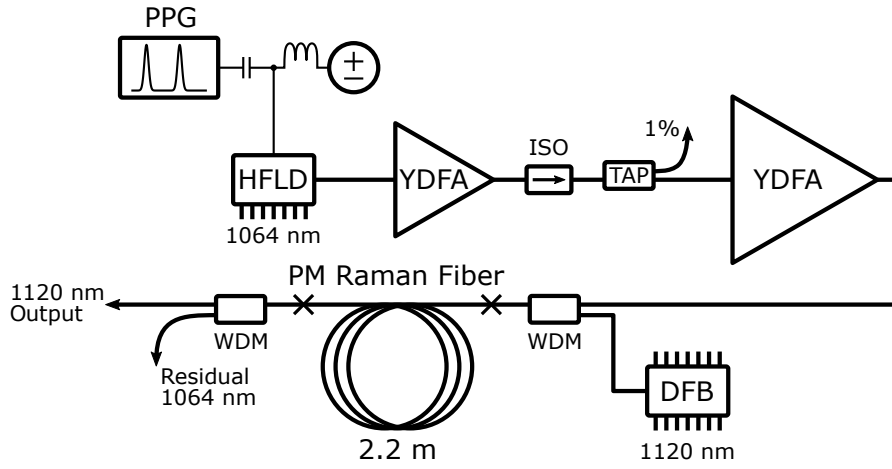


Figure 3.12: Schematic of the Raman amplifier pumped by an amplified gain-switched diode. PPG, picosecond pulse generator; HFLD, high frequency response laser diode; YDFA, ytterbium-doped fibre amplifier; ISO, isolator; DFB, distributed feedback laser diode; WDM, wavelength division multiplexer.

Yb-MOPFA output. The SHG conversion efficiency increased in this case, however, due to a reduction in the spectral linewidth of the 1120 nm fundamental [Fig. 3.8 (b)].

3.3 Gain-switched 560 nm source

The second master oscillator scheme that was implemented was designed for reduced complexity, and was based on a gain-switched DFB laser diode. The gain-switched DFB laser diode directly seeded the Yb-MOPFA, removing the MZAM and its associated drive electronics, significantly simplifying the system and increasing the cost-effectiveness (Fig. 3.12). It was found that the PPB used in the previous section could not be used to efficiently generate 560 nm radiation without the additional use of the MZAM to shape the PPB pulses. It was thought that this was due to the poor quality of the pulses that the PPB emitted when it was operated using long duration current drive pulses. The orange trace in Fig. 3.13 (a) shows the output of the PPB when it was driven with 2.85 ns electrical pulses of 110 mA peak current. It is clear that the short transient at the leading edge of the pulse has a significantly higher peak-power than the tail, which additionally suffers from intensity modulations. In the previous section, the MZAM was used to effectively suppress the transient (blue trace Fig. 3.8 inset) and it was found that using the MZAM to shape the PPB pulses led to higher conversion efficiencies than using the MZAM only to generate the pulses from CW operation of the PPB.

To remove the necessity for the MZAM pulse shaping, a high frequency response laser diode (HFLD) was used to seed the Yb-MOPFA. The HFLD (QD Laser Inc.) was

specially packaged to be impedance matched with a 50 Ω high-frequency SMPM RF connector [Yok14]. The packaging had a twofold advantage over conventional 14-pin butterfly packages that are typically used to house DFB laser diodes. Firstly, the improved frequency response reduces the period of the relaxation oscillations, therefore, a constant amplitude in the pulse tail is achieved more quickly. Secondly, the 50 Ω impedance is compatible with high-speed RF electronics that are far more prevalent than drivers designed for the few Ohm impedance of conventional butterfly DFB laser diodes. This allows for a much wider choice of electrical drive electronics, enabling high-quality, tailorable electrical pulses to be supplied to the laser diode. Indeed, it was later found that the PPB could produce higher quality pulses when the peak current of the electrical drive pulses was increased to 200 mA [cf. Fig. 4.2 (a)]. This was not discovered earlier due to the lack of ability to adjust the PPB electrical drive parameters (bias current, pulse current and pulse duration) during operation. On-the-fly adjustment is vital for quickly obtaining the optimal drive parameters since the efficiency of nonlinear processes give a definitive metric to optimise for, which would otherwise be difficult to deduce from the pulse profile.

Figure 3.13 (a) shows the output of the HFLD driven by 2 ns square electrical pulses with a peak current of 100 mA and a bias current of 8 mA at a repetition rate of 2.5 MHz. In comparison to the PPB pulses, the transient at the leading edge of the pulse settled to the steady state sooner and at a higher amplitude, additionally, the tail had a more constant amplitude. The manufacturer quoted modulation bandwidth of 8.4 GHz agreed well with the measured 200 ps period of the relaxation oscillations ($f \approx \sqrt{3}/T_R$). These pulses were used to seed the Yb-MOPFA and pump the Raman amplifier, both of which were unchanged from the previous section. When seeded with 30 mW of CW power at 1120 nm, the Raman amplifier produced 2.83 W of average power with a pulse duration of 2.3 ns and a peak-power of 570 W at the pump repetition rate of 2.5 MHz [Fig. 3.13 (b)].

Using the same SHG setup as the previous section (Fig. 3.10), a maximum 560 nm average power of 1.33 W was generated from 2.83 W of 1120 nm fundamental power, corresponding to a conversion efficiency of 47%. The 560 nm pulse duration was 2.0 ns at a repetition rate of 2.5 MHz, corresponding to a pulse energy of 532 nJ and a peak-power of 270 W [Fig. 3.13 (c)]. These figures are comparable to those obtained with the MZAM-based seed (Table 3.2), which was surprising given the transient present in the 1064 nm pump pulses. It is clear from Fig. 3.13 (b), however, that the transient was suppressed in the Raman conversion process, which was believed to be due to the fact that the transient peak-power significantly exceeds the optimal ~ 1 kW pump power for the Raman amplifier. In contrast, the tail of the pulse in Fig. 3.13 (a) had a peak-power of approximately 1 kW and was therefore expected to be converted efficiently.

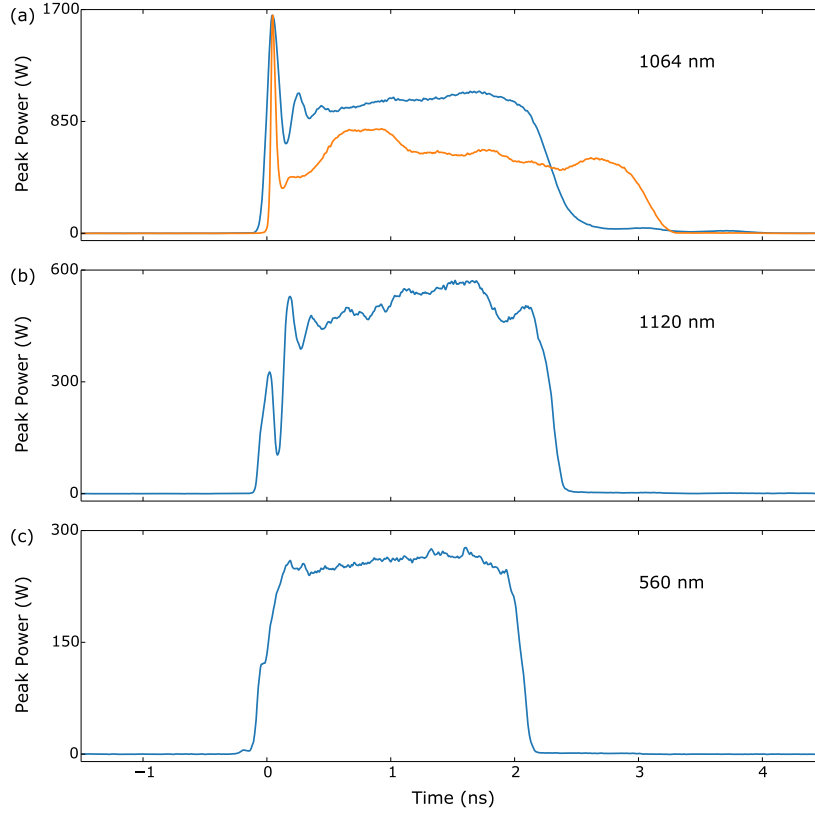


Figure 3.13: Sampling optical oscilloscope traces of the HFLD (blue) and PPB (orange) gain-switched pulses (a) the 1120 nm output of the Raman amplifier (b) and the 560 nm frequency-doubled output (c).

The intended application of the 560 nm source was to provide the depletion beam in a STED microscopy setup. To achieve sub-diffraction limited STED images, the beam quality of the depletion source is required to be as close to diffraction-limited as possible. It was found that the using the SHG setup depicted in Fig. 3.10, the 560 nm beam had an $M^2 \leq 1.15$ irrespective of whether the MZAM or HFLD master oscillator schemes were used. The 560 nm beam had an unacceptably high ellipticity of 0.8, however, which was attributed to the angled-cleaved output fibre of the Raman amplifier. To rectify this, an AR coated FC/PC connector based on PM980 fibre was spliced to the output WDM of the Raman amplifier in place of the angle-cleaved output. The AR coating had a reflectivity of 0.1% at 1120 nm, which was sufficient to avoid any issues due to feedback from the planar connector.

The output of the AR coated connector was collimated using an AR coated triplet lens collimator, which provides superior beam quality in comparison to a single aspheric lens. The $f = 40$ mm focussing lens used in the angle-cleaved SHG setup had to be replaced

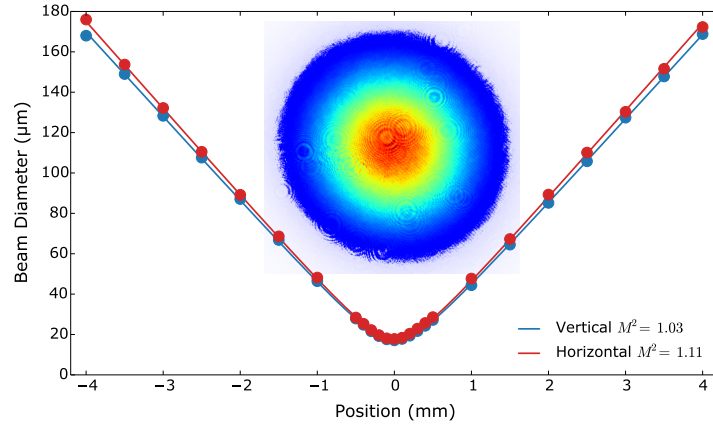


Figure 3.14: Beam diameter (points) of the collimated 560 nm beam focused by an $f = 50$ mm lens with Gaussian beam caustic fit (lines). Inset: CCD camera image of the collimated beam.

by an $f = 50$ mm lens in order to have the focus at the centre of the PPLT crystal whilst avoiding the focussing lens clashing with the crystal mount. This was not an issue when using the angle-cleaved output because the quasi-collimation led to a longer front focal length. The weaker $f = 50$ mm focusing lens used with the triplet lens collimator resulted in an increased measured $1/e^2$ waist diameter of $67 \mu\text{m}$, which corresponds to an estimated confocal parameter of 13 mm ($n = 2.13$). This corresponds to a focusing parameter ($\xi = L/b$) of 1.5 , looser focusing than the Boyd-Kleinman focusing condition of 2.84 .

The generated 560 nm beam was re-collimated to a diameter of 2.1 mm using an $f = 150 \text{ mm}$ plano-convex lens, resulting in a low divergence output beam. The beam quality was characterised using a scanning-slit profiler to measure the beam diameter through the focus of an $f = 50 \text{ mm}$ plano-convex lens (points Fig. 3.14). A Gaussian fit of the beam caustic (lines Fig. 3.14) revealed a near diffraction limited $M^2 = 1.03$ in the vertical axis and $M^2 = 1.11$ in the horizontal axis, with no measurable astigmatism. The collimated beam had an ellipticity of 0.95 (Fig. 3.14 inset) and the PER was measured to be $> 30 \text{ dB}$, demonstrating excellent polarization purity.

The electrical pulses supplied to the HFLD by the PPG (Geozondas) were tunable in duration from $0.38\text{--}2 \text{ ns}$. As the electrical pulse width was reduced, the duration of the tail in the optical pulse reduced accordingly until only a single gain-switched peak of $\sim 40 \text{ ps}$ remained [Fig. 3.4 (a)]. Table 3.3 lists the output parameters obtained by the system with the pulse duration optimised to give the maximum 560 nm average power at a variety of repetition rates. For comparison, the first row includes the parameters when the angle-cleaved SHG setup was used, whereas, the other rows used the triplet lens collimator setup. The decrease in the SHG conversion efficiency when using the weaker

Repetition Rate	Average Power	1120 nm Power	Conversion Efficiency	Pulse Energy	Pulse Duration	Peak Power
2.5 MHz	1.33 W	2.83 W	47%	532 nJ	2.0 ns	270 W
2.5 MHz	1.06 W	2.81 W	38%	424 nJ	1.9 ns	237 W
5 MHz	800 mW	2.43 W	33%	160 nJ	900 ps	183 W
10 MHz	540 mW	2.15 W	25%	54 nJ	430 ps	136 W
25 MHz	120 mW	1.55 W	8%	4.8 nJ	41 ps	96 W
50 MHz	65 mW	1.27 W	5%	1.3 nJ	37 ps	32 W

Table 3.3: Summary of 560 nm output parameters at various repetition rates. In each case, the pulse duration was optimised to obtain the highest 560 nm average power. The separated row utilised the angle-cleaved SHG setup, whereas the other rows used the triplet lens collimator SHG setup.

focusing triplet lens collimator setup in comparison to using the stronger focusing with the angle-cleaved setup was due to the reduction in the Boyd-Kleinman h-factor (Fig. 1.7).

The 560 nm output pulse duration could be tuned over the range 40 ps–2 ns [Fig. 3.15 (a)], a similar range to that obtained when using the MZAM based master oscillator. It is clear from Table 3.3 and Fig. 3.15 (b), however, that the average and peak-power of the 560 nm pulse drops dramatically as the duration decreases. This is in stark contrast to the near-constant value obtained by the MZAM based master oscillator (Table 3.2). This can be explained by the fact that when using the MZAM based seed, the Yb-MOPFA pump pulse duration was entirely controllable down to the minimum obtainable width (~ 90 ps); whereas, the HFLD pulses consisted of a fixed-duration transient and only the tail could be adjusted in duration. The complete pulse-duration control when using the MZAM based seed enabled the peak-power of the Yb-MOPFA pump pulses to be maintained close to the optimal ~ 1 kW value as the pulse energy was decreased when the repetition rate was increased. In contrast, when using the HFLD the objective was to maintain the peak-power of the tail at ~ 1 kW, which was similarly achieved by decreasing the duration of the tail as the pulse energy decreased at higher repetition rates. When the width of the tail was reduced, however, the poorly converted transient became a larger proportion of the pump pulses and therefore the Raman conversion efficiency decreased rapidly.

The fixed-duration transient also led to the pulse duration not being continuously tunable for durations < 300 ps. Indeed, at the highest tested repetition rate of 50 MHz, the HFLD seed pulse consisted of only the first relaxation oscillation with a duration of ~ 40 ps [Fig. 3.4 (a)]. This corresponded to a Yb-MOPFA pump peak-power of ~ 2.4 kW, well above

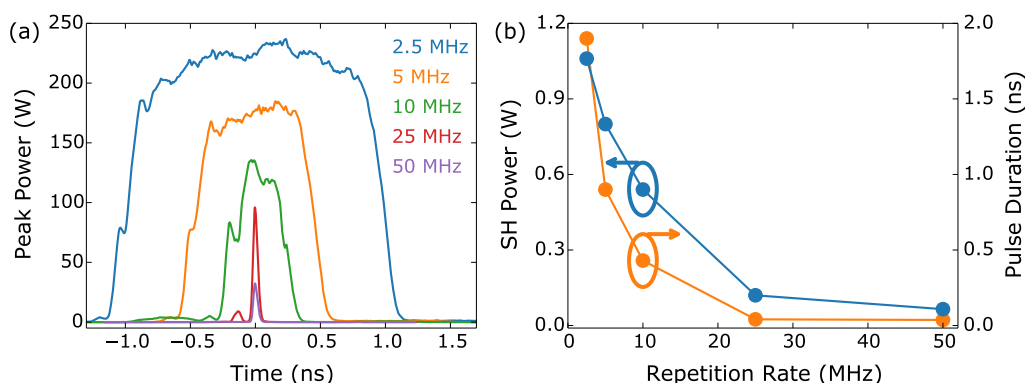


Figure 3.15: (a) Sampling optical oscilloscope traces of the optimised 560 nm pulse duration at different repetition rates. (b) Second-harmonic (SH) average power (blue points) and optimal pulse duration (orange points) as a function of pump pulse repetition rate.

the optimal value for efficient Raman conversion. It is possible that a better conversion efficiency could have been obtained by operating at higher repetition rates, however, this was not attempted as 50 MHz was the maximum repetition rate of the PPG that was used.

It was observed that the Raman conversion efficiency decreased when using shorter duration pulses, even when the peak-power of the pump was maintained at an optimal level using the MZAM based seed (cf. Table 3.3). It is thought that this could be a result of constant rise and fall time (~ 60 ps) of the MZAM pulses, which meant that at long pulse durations the pulses were effectively square but as the duration was reduced they tended towards a Gaussian pulse shape. Square pulses are advantageous in the Raman conversion process since the optimal peak-power is maintained across the whole duration of the pulse. In contrast, for a Gaussian pulse the optimum peak-power is only obtained at a single point, which results in a reduction in the conversion efficiency.

3.4 Summary

This chapter has detailed improvements in the design and implementation of the 560 nm source that was introduced and described in Chapter 2. The options for seeding MOPFA schemes were discussed, with a view to increasing the temporal flexibility of the pulse-pumped Raman amplifier concept. It was concluded that pulsed DFB laser diodes and external modulation using an MZAM were the most appropriate methods for obtaining the required pulse parameters for a STED microscopy depletion beam source. The performance of the 560 nm source, based on frequency-doubling the output of a CW narrow linewidth seeded, pulse-pumped Raman amplifier, was compared using two distinct

master oscillator schemes. The first scheme was designed for maximum performance and temporal flexibility and was based on modulating the output a DFB laser diode using an MZAM. The second scheme was designed for reduced complexity and was based on a gain-switched DFB laser diode. The same Yb-fibre amplifier chain was used with both master oscillators and its polarised output enabled the efficiency of the Raman amplifier to be maintained in the presence of environmental perturbations.

Using the MZAM-based master oscillator scheme, a maximum 1120 nm average power of 3.1 W was generated by the Raman amplifier for a Yb-MOPFA pump power of 6 W, corresponding to an amplifier efficiency of 51%. It was discussed that the major limiting factor on the amplifier efficiency was the pump and signal attenuation caused by the insertion losses of the components and splices in the Raman amplifier. Indeed, it was found that the Raman conversion efficiency from the Yb-MOPFA pump pulses to the pulsed 1120 nm signal was 85%. The output of the Raman amplifier was used to pump the fibre-coupled frequency-doubling module described in Section 2.5, however, the PPLN crystal suffered irreversible PRD due to the $\sim 17 \text{ MW/cm}^2$ peak intensity of the generated 560 nm radiation.

For subsequent frequency-doubling of the 1120 nm Raman amplifier, a 20 mm long PPLT crystal was used, owing to its higher PRD threshold. A maximum 560 nm average power of 1.53 W was generated from 3.1 W of 1120 nm fundamental using the PPLT crystal, corresponding to a conversion efficiency of 50%. The 560 nm output had a pulse duration of 2.7 ns at a repetition rate 2 MHz, corresponding to a pulse energy of 765 nJ and a peak-power of 330 W. This pulse energy was nearly two orders of magnitude greater than the system described in the previous chapter and significantly higher than alternative depletion beam sources. In addition, the 25% optical-to-optical efficiency from the Yb-fibre pump system to the 560 nm far exceeds that of fibre-based supercontinuum sources ($<1\%$) and ultrafast-pumped optical parametric oscillators (OPOs) ($\sim 14\%$), the two most commonly used depletion beam sources.

The 560 nm output pulse duration was continuously tunable over the range 56 ps–2.7 ns by adjusting the MZAM pulse width and scaling the repetition rate of the pulse train to maintain a Yb-MOPFA pump peak-power of $\sim 1 \text{ kW}$. It was found that the MZAM could be used to modulate a CW output from a DFB laser diode, however, better performance was obtained by using the MZAM to shape pulses emitted by the DFB laser diode. This was due to the limited extinction ratio of the MZAM, that led to a non-negligible CW component in the Yb-MOPFA output at high duty factors that reduced the conversion efficiency of the Raman amplifier. Watt-level 560 nm average power was achieved across the pulse duration tuning range, but it declined at higher repetition rates due to a fall in the Raman conversion efficiency.

The MZAM based system offered complete flexibility in the pulse duration across the entire tuning range that applies to efficient stimulated emission depletion in typically used fluorophores, whilst maintaining a nearly constant ~ 300 W of peak-power. MZAM-based master oscillators are, however, not cost-effective due to the drive electronics and the requirement for a separate laser source. This is especially true in the case where an MZAM is used to shape the pulses of a semiconductor diode. The second master oscillator scheme, therefore, used a gain-switched DFB laser diode to directly seed the Yb-MOPFA, thus removing the need for the MZAM and its associated drive electronics.

Gain-switched laser diodes are not normally considered to be able to produce duration-tunable pulses due to the presence of a short (~ 50 ps) transient at the leading edge of the pulse, corresponding to the first relaxation oscillation. Using a specially packaged DFB laser diode that had an improved frequency response over a standard butterfly package, it was found that the short transient that was present in the emitted pulses was suppressed in the Raman conversion process. This enabled square pulses at 1120 nm with up to 2.8 W of average power at a repetition rate of 2.5 MHz to be obtained. These were frequency-doubled using the same setup as for the MZAM based master oscillator, generating 1.33 W of 560 nm average power with a pulse duration of 2.0 ns, a peak-power of 270 W and a pulse energy of 532 nJ.

The performance of the 560 nm source using the gain-switched DFB laser diode master oscillator was comparable to the MZAM based master oscillator at low (~ 2 MHz) pulse repetition rates. This was unexpected due to the presence of the transient in the gain-switched DFB laser diode seed pulses but highly desirable due to the reduced complexity of the seed source and the associated increase in cost effectiveness. Furthermore, the 560 nm output pulse duration could be tuned over the range of 37 ps–2.0 ns, however, the tuning range was not continuous for durations < 300 ps. In contrast to the MZAM based seed, the 560 nm average and peak-power significantly declined as the pulse duration was reduced. This was due to the poorly-converted fixed-duration transient becoming a larger proportion of the gain-switched pulse as the duration of the tail was reduced.

When a triplet lens collimator was used in combination with an AR coated connector on the output of the Raman amplifier, the 560 nm beam had near diffraction-limited beam quality with an $M^2 \leq 1.11$. The circular, high polarisation purity output beam was ideally suited for direct integration into a microscopy system. This, combined with the unprecedented pulse energy, tunable duration and excellent wall-plug efficiency make the source an ideal turn-key tool for fluorescence-based imaging applications.

4 Cascaded Raman 590 nm source

A key advantage of using stimulated Raman scattering (SRS) to provide gain is that the central wavelength of the gain bandwidth is determined by the pump wavelength and the fixed frequency downshift corresponding to the molecule's vibrational modes. The operational wavelength of the amplifier is, therefore, not restricted to the energy transitions of rare-earth ions doped in optical fibres. In the preceding two chapters, pulse-pumped Raman amplifiers were used to efficiently downshift the pulsed output of a ytterbium master oscillator power fibre amplifier (Yb-MOPFA) system to the first Stokes order, thereby enabling frequency-doubling to 560 nm.

In this chapter, the pulse-pumped Raman amplifier concept is extended to operation at the second Stokes order around 1180 nm, which is subsequently frequency-doubled to 590 nm, a strategic wavelength in a diverse number of fields. The chapter begins with a brief overview of cascaded Raman amplification and then an unseeded cascaded Raman amplifier operating at 1180 nm is detailed. Following that, it is shown how the shortcomings of the unseeded cascaded Raman amplifier can be resolved by seeding the first Stokes order in the cascaded Raman amplifier. The frequency-doubling to 590 nm of the seeded cascaded Raman amplifier output is then described and a summary concludes this chapter.

4.1 Cascaded Raman amplification

Optical fibres can be used as highly efficient Raman converters to frequency downshift the spectral output of pulsed fibre lasers [Rak08, Say11, Yin13]. Once a critical peak-power threshold is exceeded, power transfers from the pump to the Stokes wavelength [Smi72]. If the Stokes wavelength has sufficient peak-power, it can pump the next Stokes order, and this can continue until the peak power of the final Stokes order is depleted below the threshold, forming a cascade [Lin77, Coh78, Ros82]. The main issue for subsequent frequency-doubling of the downshifted radiation is the low spectral power density of the Stokes signal due to the broadband nature of Raman spontaneous emission that is amplified. In the previous two chapters, Raman amplifiers were seeded with narrow linewidth signals at the Stokes wavelength in order to increase the power spectral density of the output. This enabled high second-harmonic generation (SHG) conversion effi-

ciencies to be obtained with relatively modest peak-powers by using periodically poled frequency-doubling crystals.

To extend the wavelength coverage of Raman amplifiers whilst maintaining the practical benefits of using a Yb-MOPFA pump system, cascaded Raman amplification can be used. In cascaded Raman amplification, the pump is frequency downshifted to an intermediate Stokes order, which is then used to provide Raman gain to a signal at a higher Stokes order. The intermediate Stokes orders can be seeded by a signal or, more commonly, originate from amplified Raman spontaneous emission (unseeded). As an example, a pump at $1.064\text{ }\mu\text{m}$ can be spontaneously Raman downshifted by three Stokes orders ($1.12\text{ }\mu\text{m}$, $1.18\text{ }\mu\text{m}$ and $1.24\text{ }\mu\text{m}$) to provide Raman gain for a signal at $1.3\text{ }\mu\text{m}$ [Nak84]. Since the effective Raman absorption bandwidth is as broad as the gain bandwidth, the low power spectral density of the unseeded intermediate Stokes orders is not an issue, although the pump peak-power is depleted with each Stokes shift by incomplete conversion and the quantum defect.

The benefit of unseeded cascaded Raman amplification is the fact that signals at the intermediate Stokes orders do not need to be provided, which greatly simplifies the setup. To this end, it was proposed that a pulsed Yb-MOPFA pump system could be spontaneously downshifted to $\sim 1120\text{ nm}$ to provide Raman gain to a narrow linewidth continuous-wave (CW) signal at 1180 nm in the window of the pump pulses. The resulting narrow spectral linewidth pulse train at 1180 nm could then be frequency-doubled in a periodically poled nonlinear crystal in the same manner as the previous two chapters to provide pulsed radiation at 590 nm .

4.2 Unseeded cascaded Raman amplification

The preceding chapter concluded that the best Raman amplifier and SHG conversion efficiencies were obtained using an intensity modulated master oscillator scheme. Therefore, the same Mach-Zehnder amplitude modulator (MZAM) modulated distributed feedback (DFB) laser diode seed system as used in Section 3.2 was utilised for the cascaded Raman amplification systems (Fig. 4.1). The ytterbium fibre (Yb-fibre) amplifier chain, however, was upgraded to increase the average power of the pulsed Yb-MOPFA pump system.

In the Yb-fibre amplifier chain, the pre-amplifier was replaced with a 60 cm double-clad polarisation-maintaining (PM) Yb-fibre amplifier that was co-pumped with a 5 W multimode 960 nm diode. To increase the optical efficiency of the pre-amplifier, the unabsorbed pump light was recirculated by splicing a multimode fibre between a pump combiner on the amplifier output and the $(6 + 1) \times 1$ input pump combiner. The power amplifier was replaced with a similar commercial multi-stage Yb-fibre amplifier (IPG

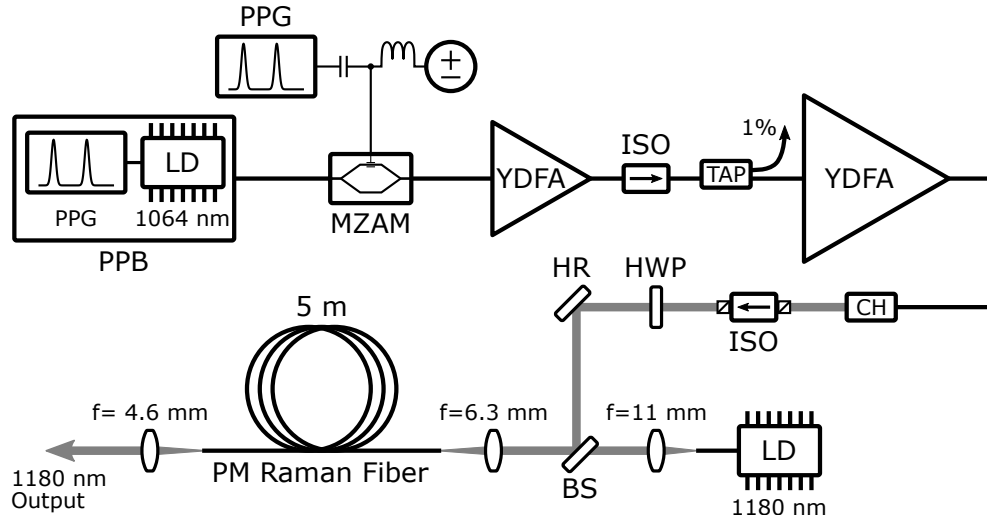


Figure 4.1: Schematic of the unseeded cascaded Raman amplifier. PPB, picosecond pulse board; LD, laser diode; PPG, picosecond pulse generator; MZAM, Mach-Zehnder amplitude modulator; YDFA, ytterbium-doped fibre amplifier; CH, collimator head; ISO, isolator; HWP, half wave plate; HR, high reflector; BS, beam splitter.

Photonics) that had a higher saturated output power of 15.5 W and featured a free-space collimated output beam. To protect the amplifier from backwards propagating Raman amplified spontaneous emission (ASE), a bulk optical isolator followed the collimator head, in contrast to the in-line fibre isolator that was present in the previous power amplifier. The resulting reduction in the length of the passive delivery fibre on the output of the amplifier increased the peak-power at which SRS was first observed ($<0.1\%$ of spectral power) to ~ 5 kW.

A DFB laser diode was used to provide the narrow linewidth signal at 1180 nm, which delivered a CW average power of 57 mW in a single longitudinal mode (SLM), corresponding to a spectral linewidth of <10 MHz. The output of the 1180 nm seed diode was collimated using an $f = 11.21$ mm aspheric lens and combined with the free-space output of the Yb-MOPFA using a dichroic beam splitter which was highly reflective to the pump wavelength ($R > 99\%$) and transmissive to the signal wavelength ($T \sim 80\%$). The combined pump and signal beam was coupled into a 5 m length of PM Raman fibre using an $f = 6.29$ mm aspheric lens with a coupling efficiency of 80% for the pump and 50% for the signal. The orientation of the flat-cleaved input end of the PM Raman fibre was rotated such that its principal axis was aligned with the polarisation axis of the 1180 nm signal and a half wave plate was used to rotate the pump polarisation onto the same axis. The rotation alignment was optimised by matching the pump polarisation to the signal

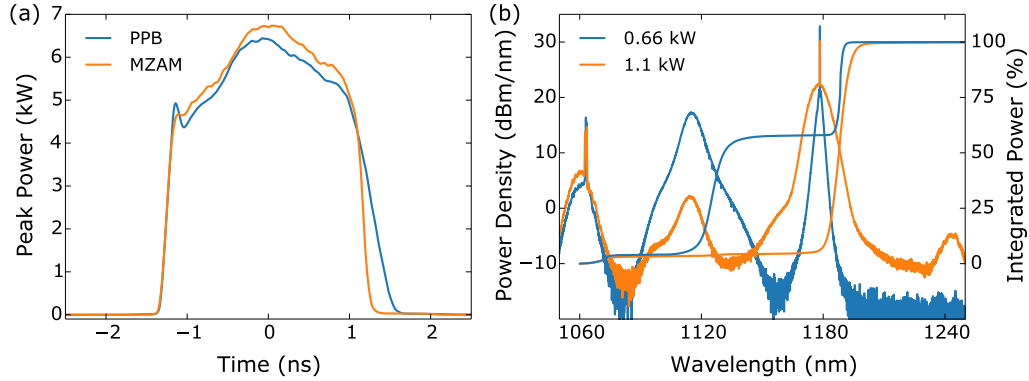


Figure 4.2: (a) Sampling optical oscilloscope traces of a PPB pulse (blue) modulated by the MZAM (orange). (b) Optical spectra of the output of a 5 m length of PM Raman fibre when pumped with a peak-power of 0.66 kW (blue) and 1.1 kW (orange) with an 1180 nm signal power of 20 mW. Integrated power has been offset by 10 nm.

and then rotating the axis of the PM Raman fibre to maximise the spontaneous Raman signal without the 1180 nm signal present.

The picosecond pulse board (PPB) used in the previous chapter was operated in pulsed mode with an electrical pulse width setting of 3 ns and a peak current setting of 200 mA, which generated 2.4 ns optical pulses [blue trace Fig. 4.2 (a)]. It was noted that the transient that was present in the previously obtained PPB pulses [cf. Fig. 3.13 (a)] was strongly suppressed, which was due to changes in the electrical driving pulse parameters caused by the increased peak current setting. The MZAM was used to further suppress the transient and increase the steepness of the falling edge of the pulse [orange trace Fig. 4.2 (a)], which was beneficial for the Raman conversion as the peak-power was greater and more constant across the pulse.

Figure 4.2 (b) shows the output spectrum of the 5 m length of PM Raman fibre when pumped with the MZAM modulated PPB pulses and seeded with 20 mW (after coupling losses) of CW 1180 nm signal. The narrow linewidth 1178.4 nm signal had an on-off gain of approximately 3 dB for a pump peak-power of 0.66 kW (2.4 ns duration pulses at 1 MHz repetition rate), however, increasing the peak-power further led to a decrease in the power spectral density. From the integrated spectral power, it is clear that for the 0.66 kW pump peak-power, the Raman conversion to the 1180 nm signal was poor: only 42% of the power was contained within a 60 nm bandwidth about the 1180 nm peak. In contrast, for the 1.1 kW pump peak-power, 95% of the spectral power was contained in the second Stokes signal. Most of the power of the second Stokes signal, however, was contained within the pedestal, which became broader and smoother as the pump-peak power was increased.

Fibre Type	Fibre Length	1180 nm Coupling	Pump Power	Raman Conversion	Crystal Power	1180 nm Power
PMRF	1.1 m	Bulk	4.5 kW	70%	0.5%	20 W
PMRF	5.1 m	Bulk	530 W	42%	8.6%	50 W
PMRF	15.3 m	WDM	220 W	83%	27%	60 W
PM980	5.8 m	WDM	2.1 kW	54%	3.4%	70 W
PM980	20.6 m	WDM	690 W	59%	17%	120 W

Table 4.1: Summary of the unseeded cascaded Raman amplification results at the pump peak-power that gives the greatest power within the 0.16 nm spectral acceptance bandwidth of the SHG crystal. To aid comparison, the pump peak-power takes into account the coupling losses. PMRF, polarisation maintaining Raman fibre; WDM, wavelength division multiplexer.

At lower pump peak-powers, clear peaks either side of the 1180 nm signal peak were visible and the constant frequency spacing of the peaks indicated that they were due to degenerate four-wave mixing (FWM) of the 1180 nm signal. As the pump peak-power was increased, an increased number of four-wave interactions occurred, including self-phase modulation (SPM) and cross-phase modulation (XPM), distributing the power amongst many modes that appeared as a smooth, broad continuum. At 1.1 kW of pump peak-power, a feature at the next Stokes order (1240 nm) was visible, indicating that a further increase in the pump peak-power would lead to power transfer to this Stokes order.

The most important metric for subsequent frequency-doubling of the Raman amplifier output is the peak-power contained within the spectral acceptance bandwidth of the SHG crystal. The SHG crystal that was available for frequency-doubling the 1180 nm signal was a 20 mm long periodically poled lithium niobate (PPLN) crystal with a single 9.27 μm pitch grating. This corresponds to a 3 dB spectral acceptance bandwidth of 0.16 nm for an 1178 nm signal at the quasi-phase-matching temperature of $\sim 130^\circ\text{C}$.

Integrating the spectra shown in Fig. 4.2 (b) over the sinc^2 spectral acceptance function of the PPLN crystal revealed that only 8.6% and 2.8% of the spectral power was within the crystal bandwidth for pump peak-powers of 0.66 kW and 1.1 kW, respectively. Therefore, for the 5.1 m length of PM Raman fibre, the maximum peak-power of the 1180 nm signal within the spectral acceptance bandwidth of the PPLN crystal was ~ 50 W. This was an order of magnitude smaller than the peak-power obtained by the 1120 nm Raman amplifier system described in the previous chapter, which would severely limit the SHG conversion efficiency that could be achieved.

To ascertain whether this was a fundamental limitation, or simply a result of the par-

ticular configuration, a number of different lengths of PM Raman fibre and PM980 were investigated and the results summarised in Table 4.1. The Yb-MOPFA pump system and the 1180 nm seed diode were the same in all the configurations, however, some used a dichroic beam splitter to bulk couple the 1180 nm signal into the Raman gain fibre; whereas, other configurations used a PM fused fibre 1064/1180 nm wavelength division multiplexer (WDM). In each case, the pump peak-power, quoted after coupling losses, was optimised to maximise the spectral power within the PPLN crystal's acceptance bandwidth.

From Table 4.1, it is clear that the power within the spectral acceptance bandwidth of the frequency-doubling crystal was considerably lower than what was achieved in the non-cascaded 1120 nm Raman amplifier system, which was up to 66% for the same crystal bandwidth. It was believed that the reason behind the more significant spectral broadening encountered in the unseeded cascaded Raman amplifier was due to the higher critical power threshold for conversion to the first Stokes order. In an unseeded Raman amplifier, the gain (Eqn. 1.60) has to be approximately 70 dB for the spontaneous Raman scattering to be amplified to a level equal to the pump power [Smi72]. By seeding a Raman amplifier, the gain requirement to reach the same Stokes power is reduced by the factor that the input power is increased, therefore, a lower pump peak-power is required to obtain the same signal output power. The higher pump peak-power requirements of unseeded cascaded Raman amplification increases the deleterious spectral broadening caused by competing nonlinear effects, which reduces the power spectral density of the amplified signal. In addition, the broadband nature of the amplified Raman ASE increases the number of FWM interactions that can occur, leading to a broad parametric pedestal around the 1180 nm signal, which further decreases the achievable power spectral density.

4.3 Seeded cascaded Raman amplification

In the previous section, it was shown that it is difficult to achieve a high power spectral density of the amplified signal using an unseeded cascaded Raman amplifier. This is a particularly pertinent issue for subsequent frequency-doubling of the output using a periodically poled crystal, since their narrow spectral acceptance bandwidth requires a high power spectral density to achieve a good SHG conversion efficiency. In order to avoid the issues encountered with the unseeded cascaded Raman amplification, a different approach was used where the intermediate Stokes order around 1120 nm was seeded with a signal in order to reduce the pump peak-power requirement.

Figure 4.3 shows the schematic of the seeded cascaded Raman amplifier system. The pulsed Yb-MOPFA pump system was identical to the unseeded case but in this configuration, the free-space pump beam was coupled into a PM fused fibre 1064/1120 nm

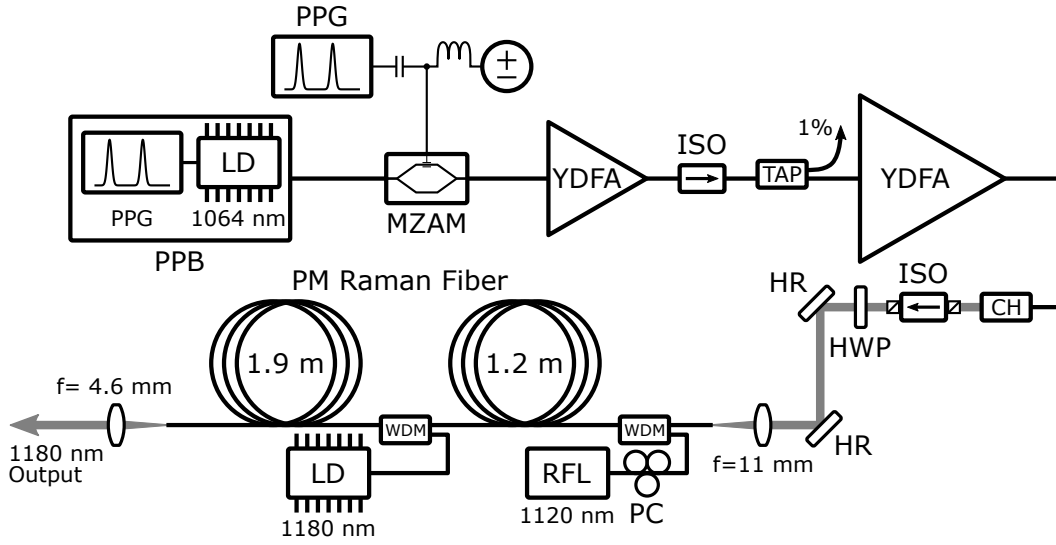


Figure 4.3: Schematic of the seeded cascaded Raman amplifier. PPB, picosecond pulse board; LD, laser diode; PPG, picosecond pulse generator; MZAM, Mach-Zehnder amplitude modulator; YDFA, ytterbium-doped fibre amplifier; CH, collimator head; ISO, isolator; HWP, half wave plate; HR, high reflector; PC, polarisation controller; RFL, Raman fibre laser; WDM, wavelength division multiplexer.

WDM using an $f = 11$ mm aspheric lens. Initially, a fibre Bragg grating (FBG) stabilised Fabry-Pérot (FP) laser diode, which had a central wavelength of 1118.3 nm and delivered a maximum CW average power of 200 mW with a resolution limited spectral linewidth of <10 pm, was used as the first Stokes seed signal. In an attempt to eliminate the splice loss associated with using the PM Raman fibre, a 3.3 m length of PM980 (including the WDM output pigtail) was used as the gain medium.

The output spectrum of the 3.3 m PM980 Raman amplifier is shown in the blue trace of Fig. 4.4 (a) when it was pumped with the pulses in Fig. 4.2 (a) at a repetition rate of 1 MHz and seeded with 40 mW of 1118 nm signal. The pump peak-power that gave the highest on-off signal gain was 1.8 kW, taking into account the 85% coupling efficiency and 0.24 dB insertion loss of the WDM. Increasing the pump peak-power further resulted in a reduction in the power spectral density of the 1118 nm signal due to an increase in the power contained within the pedestal beneath the signal, that is visible in Fig. 4.4 (a). From the integrated spectral power, the conversion to the 1118 nm signal is only 39%, which was substantially lower than the 85% obtained in the proceeding chapter [cf. Fig. 3.8 (a)].

To increase the Raman conversion efficiency, the PM980 gain fibre was replaced with a 1.2 m length of PM Raman fibre. Owing to a failure of the 1118 nm FBG-stabilised diode, it was replaced with a similar unit that had a central wavelength of 1119.5 nm and

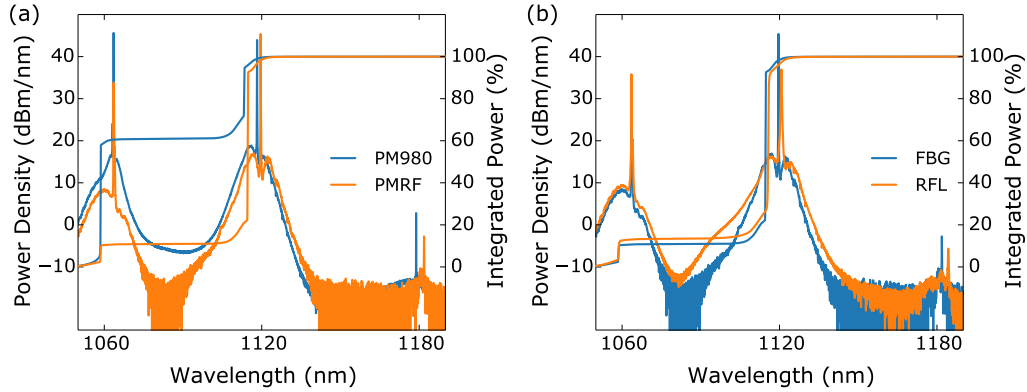


Figure 4.4: (a) Optical spectra of the output of a 3.3 m PM980 (blue) and 1.2 m PM Raman fibre (orange) Raman amplifier when pumped with 1.8 kW and 1.1 kW of peak-power and seeded with 30 mW and 40 mW of ~ 1120 nm signal, respectively. (b) Optical spectra of the 1.2 m PM Raman fibre amplifier seeded by an fibre Bragg grating (FBG) stabilised Fabry-Pérot (FP) laser diode (blue) and Raman fibre laser (RFL) (orange). The integrated spectral power has been offset by 5 nm in both plots.

delivered a maximum CW average power of 400 mW with a spectral linewidth of 26 pm. The output spectrum of the 1.2 m PM Raman fibre amplifier is shown in the orange trace of Fig. 4.4 (a), when seeded with 30 mW of CW average power at 1120 nm and pumped with a peak-power of 1.1 kW. From the integrated spectral power, the conversion efficiency to the 1120 nm signal is 89%, significantly higher than the conversion obtained with the PM980 gain fibre. It should be noted that similar Raman conversion efficiencies could be obtained when using the PM980 gain fibre by using significantly higher pump peak-power. The power spectral density of the 1120 nm signal, however, declined markedly due to degenerate FWM that resulted in the pedestal containing an increasing proportion of the spectral power. It was believed that the higher dispersion and larger Raman gain coefficient of the PM Raman fibre reduced the efficiency of the FWM processes, enabling a higher peak-power spectral density of the Raman amplified signal to be achieved.

A shorter length of PM Raman fibre was used in comparison to the optimum length used in previous chapter in order to obtain the maximum peak-power for the subsequent amplification of the 1180 nm signal. In contrast to SHG, the peak-power spectral density requirement is more relaxed due to the broadband nature of the Raman gain profile, therefore, greater spectral broadening can be tolerated. It is interesting to note the sharp spectral feature observed in the spectra of Fig. 4.4 (a) around 1180 nm. The peaks at 1178.8/1181.9 nm are caused by degenerate FWM of the 1118.2/1119.6 nm signals, seeded by the residual 1063.5 nm pump. It is possible to amplify this narrow linewidth FWM peak

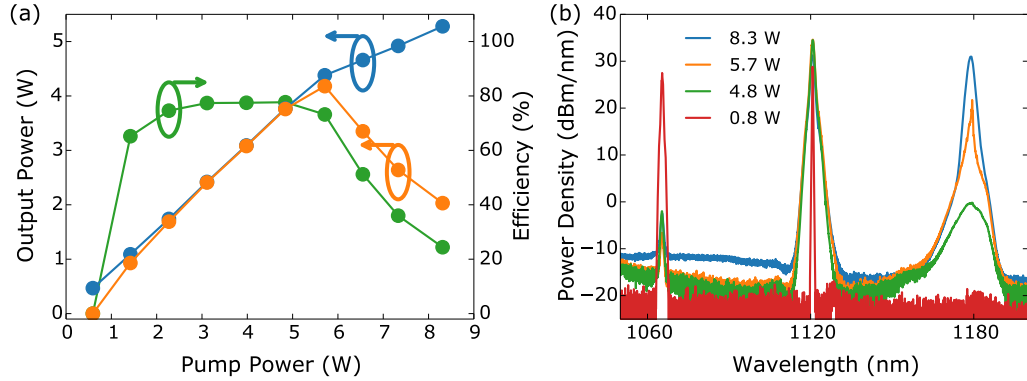


Figure 4.5: (a) Raman fibre laser total output power (blue points), 1120 nm output power (orange points) and 1120 nm efficiency (green points) as a function of 1065 nm pump power. Lines are guides for the eye. (b) Optical spectra of the Raman fibre laser output for various pump powers.

in a combined ytterbium-Raman amplifier, resulting in a pulsed signal at ~ 1180 nm with a high power spectral density [Kar15]. This was not attempted using our discreet Raman amplifier as the second FBG-stabilised laser diode seed also failed, thought to be due to inadequate isolation from the backwards propagating Raman ASE.

To replace the FBG-stabilised laser diode seed, a Raman fibre laser (RFL) was constructed to provide the ~ 1120 nm seed signal. The RFL was pumped by a CW Yb-fibre laser that delivered a maximum average power of 10 W, with a central wavelength of 1065.4 nm and a spectral width at the highest output power of 1.1 nm. The Raman gain was provided by a 390 m length of non-PM Raman fibre that was spliced (splice loss ~ 0.4 dB) to a standard single-mode fibre (SMF). The pump light was delivered through a 3-port circulator, to protect the pump laser from backwards propagating radiation, and a high-reflection (HR) ($R = 99.99\%$) FBG written in SMF fibre with a central wavelength of 1120.3 nm and a $R > 95\%$ reflection bandwidth of 1.1 nm. A second FBG with a reflectivity of 10%, a central wavelength of 1120.4 nm and a 0.60 nm 3 dB reflection bandwidth was spliced after the Raman fibre to complete the cavity.

The threshold of the RFL was ~ 0.7 W and the output power increases linearly with a slope efficiency of 78% until a pump power of ~ 5 W [blue points Fig. 4.5 (a)]. At this point, power begins to transfer to the second Stokes order at ~ 1180 nm [Fig. 4.5 (b)] and so the power in the 1120 nm signal [orange points in Fig. 4.5 (a)] decreases for pump powers > 6 W. A maximum 1120 nm average power of 4.2 W could be obtained for a pump power of 5.7 W (74% efficiency), however, the most efficient operating point gave an 1120 nm average power of 3.8 W for 4.8 W of pump power (78% efficiency). The spectral linewidth

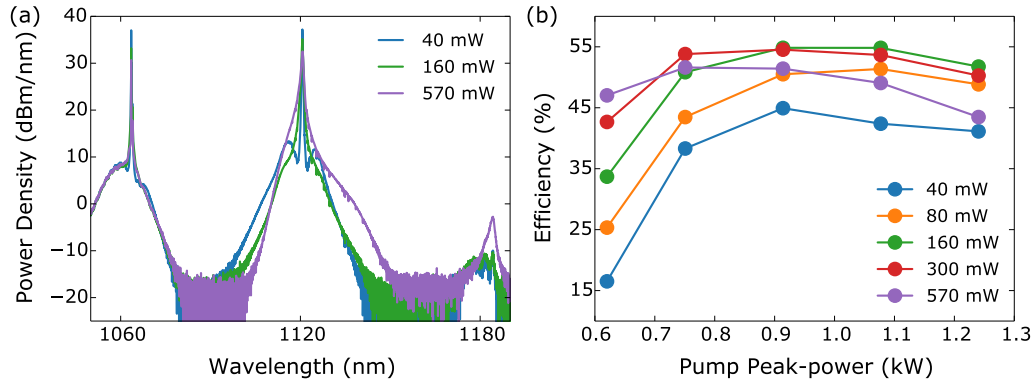


Figure 4.6: (a) Optical spectra of the 1.2 m PM Raman fibre amplifier pumped with 1.1 kW of peak-power at three RFL seed powers. (b) Efficiency of the 1.2 m Raman amplifier as a function of pump peak-power at the gain fibre (after coupling losses) for various RFL seed powers.

of the 1120 nm output monotonically increased with pump power from 0.1 nm just after threshold to 1.2 nm at the maximum 1120 nm output power. This broadening occurs due to the redistribution of power between the large number of oscillating longitudinal modes caused by FWM [Bou03, Bab06].

In comparison to the DFB and FBG-stabilised laser diode seeds, the RFL had a broader, power-dependent spectral linewidth but was capable of an order of magnitude higher average output power and more importantly was more robust against back reflections. Since the Yb-fibre pump laser had a randomly polarised output and the Raman cavity had no polarisation-discriminating elements, the output of the RFL was randomly polarised and had a varying degree of polarisation. Whilst a polarised output would have been preferable, it was not a critical issue due to the highly polarised Yb-MOPFA pump system (polarisation extinction ratio (PER) > 30 dB), the use of PM fibre for the Raman amplifier and the polarisation dependence of SRS gain.

When the RFL was operated at low output powers, there was significant residual pump radiation in the output [red trace in Fig. 4.4 (b)]. To remove this, an HR ($R > 99.99\%$) FBG with a central wavelength of 1065.75 nm and 3 dB reflection bandwidth of 4 nm was spliced to the output of the RFL, which resulted in an isolation between the 1120 nm output and the residual pump of >40 dB. Following the pump reflector, a three-paddle polarisation controller was spliced between the RFL output and the PM 1064/1120 nm WDM in order to optimise the RFL polarisation for the Raman amplification.

The output spectrum from the 1.2 m PM Raman fibre amplifier when seeded with the RFL delivering 40 mW of 1120 nm output power and pumped with 1.1 kW of peak-power

is shown in the orange trace in Fig. 4.4 (b). The broader spectral linewidth of the RFL in comparison to the FBG-stabilised laser diode seed results in a lower power spectral density, however, the conversion to the 1120 nm signal was 86%, which is comparable to the 89% achieved by when using the FBG-stabilised laser diode. It is possible that the polarisation of the RFL led to a decrease in the effective seed power that was co-polarised with the Yb-MOPFA pump. Indeed, by increasing the RFL output power, the conversion to the 1120 nm signal could be increased beyond the value obtained using the FBG-stabilised laser diode seed.

Figure 4.6 (a) shows the output spectra of the 1.2 m PM Raman fibre amplifier when pumped with 1.1 kW of peak-power for three different RFL seed powers. At the lowest seed power (40 mW, blue trace), the spectrum was similar to that obtained with the FBG-stabilised laser diode seed, with clear parametric sidebands either side of the 1120 nm peak, caused by FWM. As the seed power was increased (160 mW, green trace), the sidebands disappear and as a result, the width of the pedestal was reduced. At the highest seed power tested (570 mW, purple trace), the pedestal broadens asymmetrically and a parametric feature due to degenerate FWM of the 1120 nm signal appears around 1184 nm. The parametric sidebands about the 1120 nm peak were suppressed at higher RFL seed powers due to the increase in the spectral linewidth of the RFL output, which decreases the efficiency of the FWM process.

To evaluate the performance of the Raman amplifier, the efficiency (amplified signal power over the pump power) was measured as a function of RFL seed power and pump peak-power [Fig. 4.6 (b)]. The Raman signal output power was calculated by integrating the amplifier output spectrum over a 5 nm window centred on the 1120 nm signal peak. The window width was chosen to encompass the complete width of the RFL seed signal, without including significant contributions from the nonlinearly broadened pedestal. A maximum efficiency of 55% was obtained for a seed power of 160 mW and a Yb-MOPFA pump peak-power of 0.9 kW. This efficiency was calculated using the incident pump power, whereas, the pump peak-power was quoted after the 32% loss of the coupling, the WDM insertion loss and the splice loss to the Raman fibre were taken into account in order to aid comparison between setups. Figure 4.6 (b) shows that the efficiency remained close to the maximum value over a reasonable range of input seed powers and pump peak-powers.

The output of the 1.2 m PM Raman fibre amplifier was used as a pump for a second Raman amplifier in order to amplify the narrow linewidth 1180 nm signal (Fig. 4.3). To couple in the 1180 nm signal and filter any residual 1064 nm Yb-MOPFA pump radiation, a second PM fused fibre 1064/1120 nm WDM was spliced to the output of the 1.2 m PM Raman fibre (Fig. 4.3). The second Raman amplifier also utilised PM Raman fibre as the

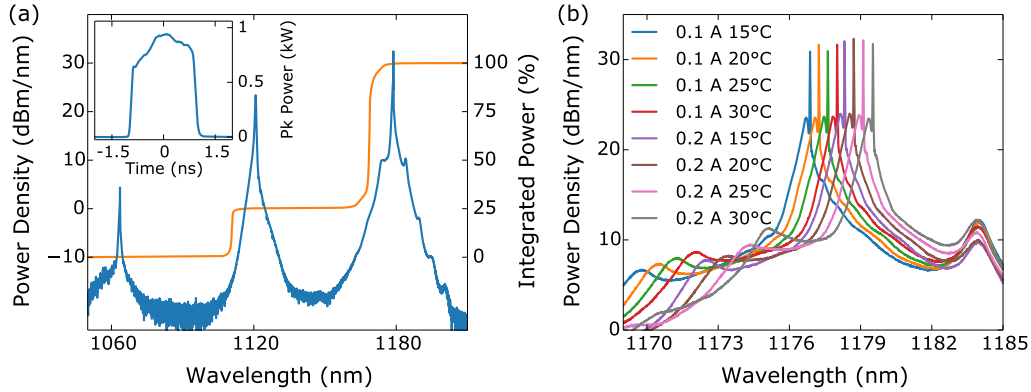


Figure 4.7: (a) Optical spectrum of the 1.9 m PM Raman fibre amplifier for an 1179 nm CW input signal of 40 mW, pumped by the output of the 1.2 m PM Raman amplifier. Inset: sampling optical oscilloscope trace of the Yb-MOPFA pump pulses. (b) Optical spectra of the 1.9 m PM Raman fibre amplifier for several signal wavelengths, pumped by the output of the 1.2 m PM Raman amplifier.

gain medium and a cut-back was performed to ascertain that a 1.9 m length resulted in the highest peak-power within the spectral acceptance bandwidth of the frequency-doubling crystal that was to be used (20 mm long PPLN, 0.16 nm bandwidth).

Figure 4.7 (a) shows the output spectrum of the 1.9 m PM Raman amplifier at an average output power of 1.3 W, using a 40 mW CW 1179 nm signal and pumped by 1.4 W of the filtered 1.2 m PM Raman fibre amplifier output. The 1.2 m Raman amplifier was seeded with 300 mW of 1120 nm signal from the RFL and pumped with 0.94 kW (after losses) of 1064 nm Yb-MOPFA peak-power, with a pulse duration of 1.84 ns at a repetition rate of 1 MHz [inset Fig. 4.7 (a)]. From the integrated spectral power, 75% of the power was contained in the 1179 nm signal which corresponded to an amplifier efficiency of 71%. Integrating the output spectrum over the sinc^2 spectral acceptance function of the PPLN crystal, however, revealed that only 17% of the power was within the acceptance bandwidth. In addition to the parametric pedestal that was routinely observed in the Raman amplification systems, the short-span spectrum [Fig. 4.7 (b)] revealed that considerable spectral power was contained within an asymmetric spectral feature whose power spectral density was only ~8 dB down on the 1178 nm peak.

To investigate the origin of this feature, the wavelength of the 1179 nm seed was coarsely varied by adjusting the bias current (either 100 mA or 200 mA) and finely varied by adjusting the gain chip temperature (5°C intervals between 15°C–30°C). From Fig. 4.7 (b), the spectral feature had a fixed ~40 GHz upshift from the ~1178 nm peak and an approximately constant extinction ratio to the peak. Therefore, the spectral feature cannot be attributed to Raman ASE and was most likely due to a four-wave interaction between the

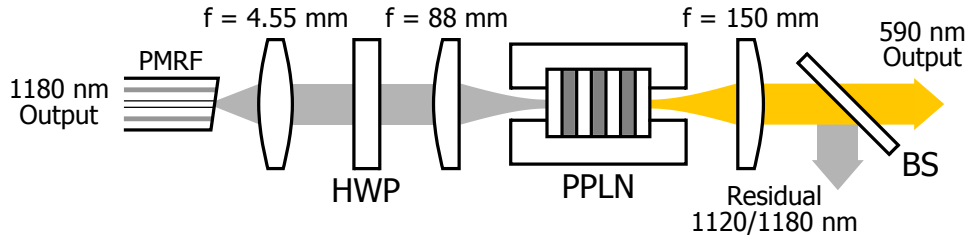


Figure 4.8: Schematic of the 590 nm second-harmonic generation stage. PMRF, polarisation maintaining Raman fibre; HWP, half wave plate; PPLN, periodically poled lithium niobate; BS, beam splitter.

~1178 nm signal and two frequency components in the 1120 nm pump line. This was evidenced by the fact that when the 1120 nm seed power to the first Raman amplifier was reduced, the frequency spacing of the upshifted feature was reduced to ~25 GHz, which can be attributed to the reduction in spectral linewidth of the 1120 nm pump signal associated with the reduced RFL output power.

It is clear from Fig. 4.7 that several different FWM processes occur simultaneously in both Raman amplifier stages, which make it difficult to achieve a high peak-power spectral density of the 1178 nm amplified signal. Indeed, it is these processes which impose a lower limit of the length of the Raman amplifier, since shorter gain fibre lengths favour FWM processes over SRS. Additionally, the FWM processes meant it was not possible to achieve high peak-power spectral densities in any configuration where the 1120 nm signal was amplified simultaneously with the 1178 nm signal, regardless of whether the 1120 nm signal was seeded or grew from spontaneous Raman scattering (unseeded).

4.4 Frequency-doubling in lithium niobate

In spite of the relatively poor power spectral density of the seeded cascaded 1178 nm Raman amplifier system in comparison to the single-shift 1120 nm system, the peak-power achieved was sufficient for subsequent frequency-doubling to 589 nm. Given the relatively modest peak-power, a PPLN crystal was used, which has the highest effective nonlinearity amongst commonly used frequency-doubling crystals. The 5 mol.% MgO-doped PPLN crystal had a length of 20 mm and was poled with a single 9.27 μm grating, which gave a 3 dB spectral acceptance bandwidth of 0.16 nm for an 1178 nm signal at the quasi phase-matching temperature of ~130°C.

The SHG setup, conceptually identical to one used in the previous chapter, is depicted in Fig. 4.8. The PM Raman fibre output of the 1.9 m 1180 nm Raman amplifier was angle-cleaved to suppress feedback and collimated using an $f = 4.55$ mm aspheric lens. A half-wave plate was used to rotate the polarisation axis of the PM Raman fibre output to align it

to the extraordinary axis of the PPLN, in order to maximise the SHG conversion efficiency. It was empirically found that an $f = 88$ mm (plano-convex) lens gave the maximum second-harmonic (SH) power for the given peak-power of the 1180 nm Raman amplifier output. The back focal length of the Raman amplifier output collimating lens was adjusted in conjunction with the crystal position relative to the focussing lens as a means of fine control of the spot size in the crystal.

The scanning-slit beam profiler previously used to measure the focused spot size was based on a silicon photodetector and therefore was not sensitive to the 1180 nm beam. In light of this, a knife-edge measurement was performed on the quasi-collimated Raman amplifier output, after the spot size in the crystal was optimised, at the position of the focusing lens. The measured beam diameter was 1.7 mm, which results in an approximated focused waist radius of 40 μm , assuming $M^2 = 1$ and that the wavefronts have no curvature at the focusing lens. The estimated confocal parameter in the crystal was 18 mm ($n = 2.176$), corresponding to a focusing parameter ($\xi = L/b$) of 1.1, close to confocal focusing. A dichroic beam splitter was used to separate the residual 1120 nm and 1180 nm light from the second-harmonic, with a measured attenuation of 19 dB at 1120 nm and 28 dB at 1180 nm and a transmission of $\sim 97\%$ for the 590 nm second-harmonic. The PPLN crystal was anti-reflection (AR) coated on the input facet but the output facet was uncoated, which results in a 14.6% loss for the 590 nm second-harmonic due to the Fresnel reflection.

Figure 4.9 shows the output 589 nm power, corrected for the Fresnel reflection, as a function of the total output power from the 1.9 m Raman amplifier using a 1178.7 nm CW signal power of 40 mW and pumped by the output of the 1.2 m Raman amplifier. The 1.2 m Raman amplifier was seeded with 210 mW of 1120 nm signal from the RFL and pumped with Yb-MOPFA pulses that were 1.84 ns in duration [inset Fig. 4.7 (a)] at a repetition rate of 6 MHz. From Fig. 4.9 (a), the growth in the second-harmonic was close to exponential, rather than the quadratic dependence that is theoretically expected, which was due to nonlinear dependence of the 1180 nm Raman gain with the 1120 nm pump power. Hence, the 1180 nm signal became a larger proportion of the total Raman amplifier output as the output power increased, leading to the rapid increase in the SHG conversion efficiency. At the maximum second-harmonic power of 1.13 W, the Raman amplifier output power was 6.52 W, which corresponds to a conversion efficiency of 17%. The proportion of the Raman amplifier output that was within the spectral acceptance bandwidth of the PPLN crystal, however, was only $\sim 20\%$ at the optimum pump peak-power. The corresponding SHG conversion efficiency of the 1179 nm signal was therefore $\sim 85\%$.

At the highest available Yb-MOPFA pump power (15.5 W), the output of the 1.9 m Raman amplifier was 7.50 W, using the same seed and pump parameters as described

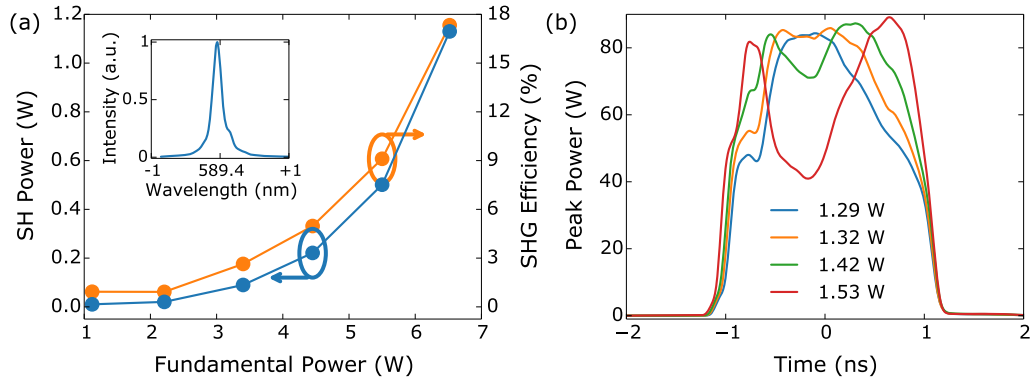


Figure 4.9: (a) Second-harmonic (SH) power and conversion efficiency as a function of fundamental average power at a repetition rate of 6 MHz. Inset: optical spectrum of the SH. (b) Sampling optical oscilloscope traces of the 590 nm pulses with increasing fundamental power at a repetition rate of 1 MHz.

for Fig. 4.9 (a). Pumping the PPLN crystal with this average power caused catastrophic damage to the output facet. It was believed that a previous cleaving of the facet without subsequent polishing may have led to a defect that, when exposed to the thermal load of the high average power beam, resulted in a fracture. The spectral and temporal properties and beam quality of the second-harmonic were taken before this fracture with the same seed and pulse settings but at a pulse repetition rate of 1 MHz, such that the average power was reduced by a factor of six. At this repetition rate, the maximum 589 nm average power obtained (corrected for the Fresnel reflection) was 150 mW for a Raman amplifier output of 1.42 W, corresponding to a conversion efficiency of 11%. The decrease in SHG conversion efficiency at the lower repetition rate was due to a reduction in the efficiency of the Raman amplification of the 1179 nm signal, which was likely due to the reduction in effective seed power from the decrease in the duty factor.

The 590 nm output pulses for several Raman amplifier output powers at a pulse repetition rate of 1 MHz are shown in Fig. 4.9 (b), where the pump and seed conditions are the same as previously described. It was observed that at the lowest fundamental power shown, the 590 nm pulse had the same features of the 1180 nm pulse but with an intensity that was dependent on the 1180 nm pulse intensity, as is expected for SHG. As the fundamental power was increased, the 590 nm peak-power saturated at ~86 W and as a result the top of the pulse broadened at the fundamental power of 1.32 W, which gave the highest SHG efficiency. Further increases in the fundamental power resulted in the 590 nm peak-power at the centre of the pulse, corresponding to the position of the highest instantaneous power of the fundamental, decreasing monotonically. This was evidence

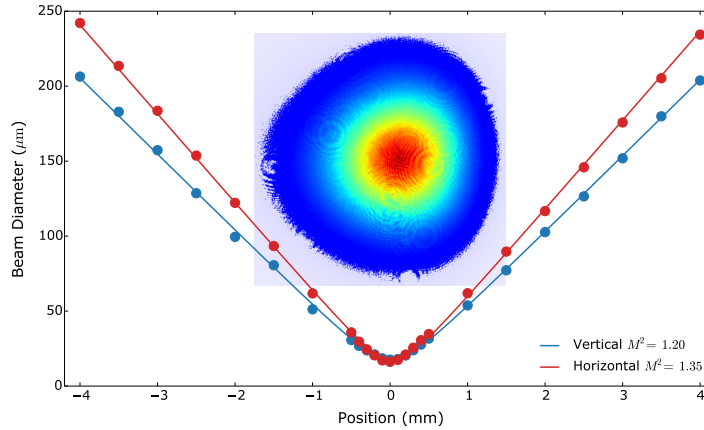


Figure 4.10: Beam diameter (points) of the collimated 590 nm beam focused by an $f = 50$ mm lens with Gaussian beam caustic fit (lines). Inset: CCD camera image of the collimated beam.

that the process of back-conversion, where power transfers from the SH back to the pump wavelength, was occurring. This indicates that the peak intensity of the fundamental in combination with the high nonlinearity of the PPLN crystal resulted in a nonlinear drive that was excessively high. It is for this reason that a flat-top pulse profile is preferred for nonlinear frequency conversion, such that the instantaneous power can be optimal across the whole duration of the pulse.

The frequency-doubled 590 nm output was collimated to a beam diameter of 1.6 mm using an uncoated $f = 150$ mm plano-convex lens, the collimated beam had an ellipticity of 0.97 but clearly suffered from distortion (inset Fig. 4.10). To quantify the beam quality, a scanning slit beam profiler was used to measure the beam diameter of the collimated beam focused by an AR coated $f = 50$ mm plano-convex lens (points Fig. 4.10), taken at the maximum second-harmonic power (150 mW) for a pump repetition rate of 1 MHz. A Gaussian fit of the beam caustics (lines Fig. 4.10) revealed an $M^2 = 1.20$ in the vertical axis and $M^2 = 1.35$ in the horizontal axis and no measurable astigmatism. The obtained M^2 values were significantly higher than what was obtained in the previous two chapters using both PPLN and periodically poled lithium tantalate (PPLT) crystals. This was evidence that the PPLN crystal was being operated close to its photorefractive damage (PRD) threshold, since the beam quality degradation suggests that significant photorefractive damage was occurring.

The operation of the PPLN crystal close to its PRD threshold was confirmed when the Yb-MOPFA pump system was operated at a repetition rate of 5 MHz. A maximum 590 nm average power of 1.09 W was obtained for a fundamental power of 6.13 W but the crystal suffered from catastrophic PRD after a few seconds due to the increased peak-power

of the fundamental. The maximum peak-power of the second-harmonic at the 5 MHz repetition rate was ~ 130 W, which using the approximated waist diameter corresponded to a peak-intensity of ~ 5 MW/cm².

4.5 Summary

This chapter has described the wavelength extension of the pulse-pumped Raman amplifier concept to the second Stokes order around 1180 nm, and the subsequent frequency-doubling of the output to generate pulsed yellow radiation around 590 nm. The simplest implementation of the cascaded Raman amplifier concept used amplified spontaneous Raman scattering at the intermediate Stokes order to provide gain to a 1180 nm signal. A DFB laser diode was used to provide the narrow linewidth CW signal at 1180 nm and a pulsed Yb-MOPFA system utilising an MZAM modulated master oscillator was used as the pump system. The pump and signal beams were combined in a passive length of fibre to act as the Raman gain medium.

Whilst on-off Raman gain was observed for the 1180 nm signal using this setup, it was found that achieving the high peak-power spectral density that was required for SHG in periodically poled crystals was not possible. This was due to the many four-wave interactions that occurred between the Yb-MOPFA pump, the broadband generated 1120 nm Raman ASE and the narrow linewidth 1180 nm signal. It was similarly found that even when the intermediate Stokes order was seeded to reduce the spectral bandwidth, the simultaneous amplification of the 1120 nm and 1180 nm in the same gain fibre resulted in insufficient peak-power spectral density due to FWM processes.

To overcome the limitations on the spectral power density of the combined cascaded systems, a two stage Raman amplifier system was developed. The first stage used 1.2 m of PM Raman fibre as the gain medium and a RFL was used to provide the 1120 nm seed signal. A maximum amplifier efficiency of 55% was obtained using a Yb-MOPFA pump peak-power (after coupling losses) of 0.9 kW and an 1120 nm seed power of 160 mW. The output of the 1.2 m Raman amplifier, after having been filtered using a WDM to remove the residual Yb-MOPFA pump, was used to pump a second Raman amplifier that had a 1.9 m length of PM Raman fibre as the gain medium. The WDM was additionally used to introduce a 40 mW CW narrow linewidth 1179 nm signal from a DFB laser diode into the second Raman amplifier.

The second Raman amplifier achieved an efficiency of 71% when seeded with 40 mW of CW average power and pumped by 1.4 W from the filtered output of the 1.2 m Raman amplifier. The 1.2 m Raman amplifier was seeded with 300 mW of CW average power from the RFL and pumped with 0.94 kW (after losses) of Yb-MOPFA pump peak-power,

with a pulse duration of 1.84 ns and a repetition rate of 1 MHz. Whilst the efficiency of the second Raman amplifier was excellent, integrating the spectral power over the sinc^2 spectral acceptance function of the PPLN crystal to be used for SHG revealed that only 17% of the 1.3 W output power was within the spectral acceptance bandwidth. It was found that a four-wave interaction between the 1179 nm signal and two frequency components in the 1120 nm pump signal caused a high power spectral density feature to appear on the short wavelength side of the 1179 nm signal. It is possible that using an 1120 nm seed source that has a smaller spectral bandwidth and fewer oscillating modes could eliminate this feature and correspondingly increase the power spectral density of the 1179 nm signal.

The amplified, pulsed 1179 nm signal was frequency-doubled using a 20 mm long, 5 mol.% MgO-doped PPLN crystal. A maximum 590 nm output power of 1.13 W, corrected for the Fresnel reflection of the uncoated output facet, was obtained for a Raman fibre amplifier output of 6.52 W. The repetition rate of the Yb-MOPFA pump pulses was 6 MHz, resulting in a 590 nm pulse energy of 190 nJ. The corresponding conversion efficiency was 17%, which was limited by the low proportion of the Raman amplifier output that was within the spectral acceptance bandwidth of the PPLN crystal. Pumping the crystal with higher Raman amplifier output power resulted in catastrophic failure of the output facet, believed to be caused by a defect.

The temporal profile of the 590 nm pulses at a repetition rate of 1 MHz displayed evidence that back-conversion was occurring, due to the saturation and eventual decrease in the peak instantaneous power observed as the fundamental peak-power was increased. The output beam quality at the maximum 590 nm average power of 150 mW for the repetition rate of 1 MHz had an $M^2 = 1.20$ in the vertical axis and $M^2 = 1.35$ in the horizontal axis, significantly worse than was previously obtained using either PPLN or PPLT SHG crystals. The beam quality degradation was due to photorefractive of the generated second-harmonic. The operation of the PPLN crystal close to the PRD threshold was corroborated by the fact that catastrophic PRD occurred when the Yb-MOPFA pump system was operated at a repetition rate of 5 MHz. A maximum 590 nm average power of 1.08 W was generated from 6.13 W of Raman amplifier output, corresponding to a 590 nm peak-power of 130 W and a peak intensity of $\sim 5 \text{ MW/cm}^2$ before catastrophic PRD occurred after a few seconds.

It is clear that in order to increase the SHG conversion efficiency, the power spectral density of the Raman amplifier must be improved by suppressing the FWM processes. The associated increase in the generated 590 nm peak-power would, however, require an SHG crystal with a higher PRD threshold, such as PPLT, to be used. In spite of the shortcomings, Watt-level average power at 590 nm with a pulse energy of 190 nJ and a

peak-power of 86 W was demonstrated. For applications in biomedical imaging, such as a depletion beam for stimulated emission depletion microscopy of the yellow fluorescent protein, such output parameters would be more than sufficient. In addition, the achieved optical-to-optical efficiency of 8% from the Yb-MOPFA pump and the seed signals to the 590 nm radiation is higher than many competing solutions.

5 Wavelength-tunable 780 nm source

This chapter describes the development of a wavelength and pulse duration tunable source around 780 nm that is designed for subsequent nonlinear frequency conversion in conjunction with a pulsed ytterbium master oscillator power fibre amplifier (Yb-MOPFA) system. The 780 nm source is based on direct frequency-doubling of a wavelength and pulse duration tunable erbium master oscillator power fibre amplifier (Er-MOPFA) system. The chapter begins with a brief introduction to erbium-doped fibre amplifiers (EDFAs) and a discussion of current sources of short-wavelength near-infrared (NIR) radiation and their applications. The wavelength-tunable, temporally flexible Er-MOPFA scheme is subsequently detailed, followed by the second-harmonic generation (SHG) in a periodically poled lithium niobate (PPLN) crystal. A short summary will then conclude this chapter.

5.1 Erbium-doped fibre amplifiers

EDFAs [Mea87, Des87] have revolutionised modern life through their use as optical amplifiers in the vast network of trans-continental undersea optical fibre links that enables the global internet to be possible. In comparison to ytterbium fibre (Yb-fibre) lasers, however, the use of EDFAs outside of telecommunications is far less widespread, primarily due to the less favourable energy level structure of the Er^{3+} ion. Comparing the energy level structure of the Er^{3+} ion [Fig. 5.1 (a)] to the Yb^{3+} ion [Fig. 1.4 (a)], it is immediately clear that there are significantly more Er^{3+} manifolds in the energy range of NIR and visible photons. This can lead to issues of excited state absorption but more importantly, the close proximity of the energy levels results in strong quenching effects, which limits the achievable Er^{3+} doping concentration.

The most relevant absorption bands of the Er^{3+} ion are the $^4\text{I}_{15/2} \rightarrow ^4\text{I}_{11/2}$ transition around 980 nm and in-band (tandem) pumping of the $^4\text{I}_{15/2} \rightarrow ^4\text{I}_{13/2}$ transition between 1.45–1.53 μm . The 980 nm band allows pumping with InGaAs-based laser diodes, which have been extensively developed and are commercially available with high average power and brightness. The main drawback of pumping at 980 nm, however, is the large quantum defect ($\sim 60\%$), which limits the efficiency of erbium fibre (Er-fibre) lasers to approximately 40%. In comparison to ytterbium (Yb)-doped fibres, the absorption per unit length at 980 nm is significantly smaller for erbium (Er)-doped fibres due to the smaller absorption

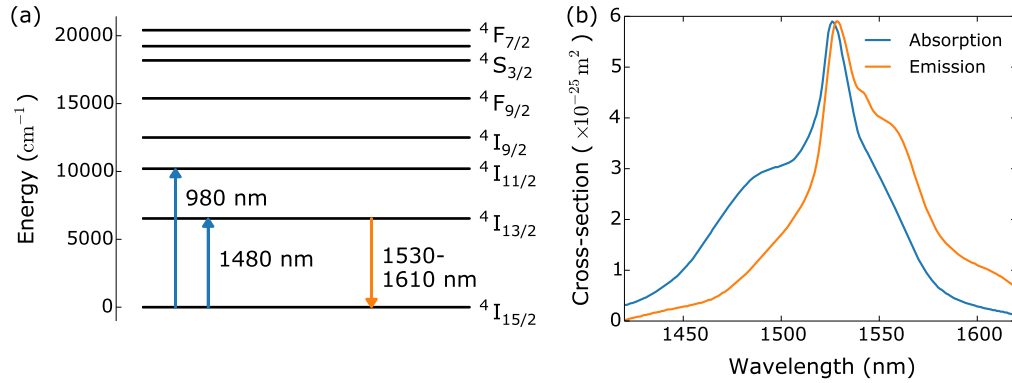


Figure 5.1: (a) After [Min91]. Er^{3+} energy level structure up to 21000 cm^{-1} with the primary absorption (blue) and emission (orange) wavelengths labeled. For clarity the label for level $^2\text{H}_{11/2}$ is missing. (b) Absorption (blue) and emission (orange) cross-sections of erbium-doped phosphosilicate optical fibre as a function of wavelength.

cross section of the Er^{3+} ion and the lower doping concentration that can be used. This is a particularly pertinent issue for cladding-pumped Er-doped fibre lasers and amplifiers, since the small overlap of the pump and core modes dramatically reduces the fibre absorption in comparison to core-pumped active fibres.

To overcome this issue, double-clad Er-doped fibres are often co-doped with high concentrations of Yb as a sensitizer [Sni88b]: the Yb^{3+} ions have strong absorption around 980 nm which excites them to the $^2\text{F}_{5/2}$ energy level, which coincides with the $^4\text{I}_{11/2}$ level in Er^{3+} ions. Cross-relaxation between the $^2\text{F}_{5/2}$ Yb^{3+} level and the $^4\text{I}_{11/2}$ Er^{3+} level can occur, efficiently transferring the energy to the Er^{3+} ion. The glass composition of the core in Er:Yb co-doped fibres needs to be optimised for efficient energy transfer, which introduces issues when scaling the mode-field diameter (MFD) due to the constraints on the refractive index profile [Kot15].

Er:Yb co-doped double-clad gain fibres have enabled cladding-pumped Er-fibre lasers to be scaled up to an average power of 297 W [Jeo07], significantly lower than the multiple kilowatts available from Yb-fibre lasers. The main issue with co-doping is avoiding or managing parasitic amplified spontaneous emission (ASE) and lasing around the Yb^{3+} emission peak at $\sim 1 \mu\text{m}$. This is due to the incomplete transfer of energy to the Er^{3+} ions, leading to a build-up of excited Yb^{3+} ions, that can lead to catastrophic optical damage through self-pulsation [Sob11].

An alternative approach to scaling the average power is to in-band pump (1.45–1.53 μm) Yb-free Er-doped fibres, which avoids issues with excited Yb^{3+} ions and has the advantage of reducing the quantum defect to a few percent. The disadvantage is that the average

power of multimode pump diodes around the absorption peak at ~ 1530 nm is fairly limited and their electrical-to-optical efficiency is significantly lower in comparison to 980 nm pump diodes. Nonetheless, in-band cladding pumped Yb-free Er-doped fibre lasers have been scaled to an average power of 103 W with an optical-to-optical efficiency of 75%, the highest efficiency reported for a cladding pumped Er-fibre laser [Jeb12]. Adding small amounts of Yb^{3+} to double-clad Er-doped fibres has been shown to reduce concentration quenching without the Yb^{3+} participating in the laser action, enabling in-band cladding pumped Er-fibre lasers to be scaled to 264 W with a slope efficiency of 74% [Jeb14].

The difficulties in scaling the average power and MFD of EDFAs has led to far less research effort into scaling the pulse energy and peak-power of Er-MOPFA schemes in comparison to Yb-MOPFA schemes. The ‘eye-safe’ nature of the $1.5\ \mu\text{m}$ radiation does, however, find applications for high-energy pulsed Er-MOPFA systems, most notably for remote sensing (e.g. LIDAR). The highest reported peak-power from an Er-MOPFA generated pulses with a peak-power of 1.2 MW and a pulse energy of 1.4 mJ using a multimode Er:Yb co-doped large mode-area (LMA) fibre [Des08]. The highest pulse energy achieved from a Er-MOPFA with a multimode output is 17 mJ pumped at 980 nm [Lal11] and 2.6 mJ with in-band pumping [Lim12a], both using Yb-free Er-doped fibre.

5.2 Short-wavelength NIR sources and applications

The frequency-doubling of Er-MOPFA schemes can access the 765–805 nm short-wavelength NIR spectral region, where in contrast to the visible spectral region, there are several mature laser technologies available. For example, unlike in the ‘green-gap’, AlGaAs-based semiconductor laser diodes directly emit in the 720–880 nm wavelength range. Indeed, AlGaAs laser diodes have been vital to the development of diode-pumped solid state lasers, most notably Nd^{3+} based solid state lasers (e.g. Nd:YAG, Nd:YVO₄) that are typically pumped around 808 nm. Multimode emitters have been scaled to the kilowatt level at this wavelength and hundreds of Watts are available across the wavelength range, for example at 793 nm for the pumping of thulium-doped double-clad gain fibres.

The scaling of the brightness of laser diodes is primary limited by catastrophic optical damage to the output facet (due to high optical intensity) and managing the heat generation and large carrier density at the semiconductor junction. One technique for mitigating these effects is to taper the gain region, such that the input end of the amplifier has waveguide dimensions for single mode operation (typically $1 \times 3\ \mu\text{m}$) whereas at the output the slow-axis has been expanded to hundreds of microns [Wal92]. Using tapered laser and amplifiers, average powers of a few Watts can be obtained with near diffraction

limited beam quality [Sum09]. Distributed feedback (DFB), distributed Bragg reflector (DBR) and external-cavity laser diode (ECLD) are commonly used as master oscillators in conjunction with tapered power amplifiers to achieve narrow linewidth operation for spectroscopic applications, for example probing the D_2 absorption line of rubidium at 780.0 nm.

Whilst semiconductor laser diodes are spectrally versatile, highly compact and efficient, their main drawback is the limited energy storage of the gain medium due to the small mode volume and short (typically <1 ns) upper state lifetime. Therefore, scaling the pulse energy is very challenging [Kim06], despite the considerable temporal flexibility in the duration (ps–ms) and repetition rate of pulsed semiconductor diodes (cf. Section 3.1). To obtain high pulse energies or ultrashort pulses in the short-wavelength NIR spectral region, solid-state lasers are typically used. There are several solid-state gain media available in this spectral region, but by far the most widespread in use is the titanium sapphire laser (Ti:Sapphire, $\text{Ti}^{3+}:\text{Al}_2\text{O}_3$), first demonstrated in 1982 [Mou86].

Ti:Sapphire has an extremely broad gain bandwidth, covering 670–1100 nm, which enables pulse durations as short as 5.5 fs to be generated directly from oscillators [Mor99] and widely-tunable narrow linewidth operation. Ultrafast Ti:Sapphire oscillators are commercially available delivering several Watts of average power at the gain peak around 800 nm producing ~ 100 fs pulses that are tunable central wavelength (680–1080 nm) with a typical pulse repetition rate of ~ 80 MHz. Alternatively, the whole gain bandwidth can be used to generate <10 fs pulses, with average powers of several hundred milliwatts. These ultrafast oscillators are the premier scientific lasers [Kle10], with applications in microscopy, nonlinear optics, optical metrology and the seeding of ultrafast chirped-pulse amplification (CPA) schemes. Widely-tunable continuous-wave (CW) Ti:Sapphire lasers are also commercially available delivering multi-Watt average power with kilohertz linewidths, for applications such as spectroscopy, atomic cooling and trapping.

Sapphire is an exemplary laser host material, with a very high heat conductivity and extremely high hardness, which facilitates high average power and high pulse energy operation of Ti:Sapphire lasers. In CW operation, up to 43 W average power has been reported [Erb91] and up to 37.4 J of pulse energy has been obtained in a CPA scheme, with a compressed pulse duration of 33 fs resulting in a peak-power of 0.85 PW [Aoy03]. The Ti^{3+} doping concentration and the length of the Ti:Sapphire crystal are limited by parasitic absorption of the laser radiation, which results in a fairly low single-pass gain despite the large emission cross-section of Ti^{3+} . To obtain large pulse energies, therefore, regenerative amplifiers are typically used, which restricts the pulse repetition rate to hundreds of kHz.

The main drawback of Ti:Sapphire is that its broad absorption band peaks at 490 nm, where high power semiconductor laser diodes have only recently surpassed the 1 W

level [Mas13]. In addition, the short upper state lifetime ($\sim 3 \mu\text{s}$) results in a high saturation intensity, which in conjunction with the relatively high parasitic losses mean that high brightness pump sources are required for efficient laser operation. Initially, argon ion lasers were used as pump sources but their very poor efficiency and concomitant large size and high power requirements led them to be replaced by frequency-doubled Nd^{3+} -based diode pumped solid state lasers [Har91]. More recently, frequency-doubled optically pumped semiconductor lasers [Chi07] and frequency-doubled ytterbium fibre lasers [Kum12] have emerged as alternative pump sources. The rapid development of green laser diodes for laser display applications has seen significant progress made in the power scaling of diode-pumped Ti:Sapphire lasers [Gür15].

The difficulties in diode-pumping Ti:Sapphire lasers had led to research into alternative solid-state gain media such as Cr:LiSAF ($\text{Cr}^{3+}:\text{LiSrAlF}_6$) that features a comparable gain bandwidth but can be directly pumped using red laser diodes [Fre93]. Diode-pumped Cr:LiSAF lasers have produced pulse durations as short as 10 fs [Uem00], have been scaled to 2.4 W of CW average power [Dem15] and are potentially tunable over the range 750–1150 nm. Other broad gain bandwidth trivalent chromium-doped solid-state lasers include Cr:LiCAF ($\text{Cr}^{3+}:\text{LiCaAlF}_6$), with similar properties to Cr:LiSAF [Wag03], and alexandrite ($\text{Cr}^{3+}:\text{BeAl}_2\text{O}_4$), which can also be pumped using red laser diodes and is tunable over the range 700–820 nm [Sch90, Tep14].

It is clear that due the range of available laser sources in the short-wavelength NIR spectral region, most application demands can be satisfied with lasers that directly emit in this region. There are still, however, several advantages that frequency-doubled Er-fibre lasers can offer over the direct emission sources. For example, the difficulties associated with pumping Ti:Sapphire lasers has led to the development of frequency-doubled ultrafast Er-fibre oscillators as extremely compact, highly efficient and cost effective alternatives. Furthermore, frequency-doubled Er-fibre sources have been scaled to average powers $>40 \text{ W}$ [Vya05, Chi12b], exceeding the diffraction-limited power of both Ti:Sapphire lasers and AlGaAs-based laser diodes.

The short-wavelength NIR 780 nm source detailed in this chapter was designed for subsequent sum-frequency generation (SFG) in a periodically poled crystal with a Yb-MOPFA system operating at 1064 nm to generate blue light around 450 nm. The Yb-MOPFA that was to be used is the same as described in Section 4.2, therefore, the 780 nm source was required to have matched and synchronised flat-top nanosecond pulse pulses at megahertz repetition rates for efficient SFG. To generate Watt-level average power blue radiation, the 780 nm source was also required to have high average power, since the shorter wavelength source in SFG requires a higher average power to balance the photon numbers. The pulse characteristics and average power requirements of the 780 nm source

are relatively straightforward to achieve using a frequency-doubled Er-MOPFA system but would otherwise be very challenging to implement using direct emission sources.

5.3 Erbium MOPFA

The Er-MOPFA system was designed to achieve the highest possible SHG conversion efficiency, such that maximum possible average power was available for subsequent SFG. In addition, the pulse characteristics of the frequency-doubled output needed to match that of the Yb-MOPFA system described in Section 4.2 to ensure a good temporal overlap to achieve efficient SFG. A narrow spectral linewidth was also required, to ensure a high conversion efficiency for both the SHG and more stringently the subsequent SFG using periodically poled crystals. Furthermore, wavelength tunability was desirable to give flexibility in the phase-matching of the SFG. In order to satisfy these requirements, external modulation of a CW source was chosen as the seed source for Er-MOPFA system. A Mach-Zehnder amplitude modulator (MZAM) was used to achieve the desired pulse characteristics with the spectral flexibility to offer tunable narrow linewidth operation.

A commercial ECLD system (Anritsu MG9638A) was used as the CW laser source, which offered both wavelength tunability and narrow linewidth operation (Fig. 5.2). The ECLD was tunable over the range 1.50–1.58 μm , delivering 5 mW of average power in a single longitudinal mode (SLM) with a typical linewidth of 700 KHz. A 10 GHz MZAM (Sumitomo) was used to modulate the output of the ECLD, which had a static extinction ratio of 36 dB for a signal wavelength of 1560 nm. The MZAM was driven by a picosecond pulse generator (PPG) (Geozondas) whose output pulse-duration was adjustable over the range 0.4–3 ns and could be externally or internally triggered at a repetition rate between 0–50 MHz. The PPG pulse peak-voltage was attenuated to be approximately equal to the half-wave voltage (~ 5 V) of the modulator and combined with a DC offset voltage to bias the MZAM at the minimum transmission point. A three-paddle polarisation controller (PC) was used to optimise the polarisation of the ECLD (polarisation extinction ratio (PER) >30 dB) to minimise the transmission loss through the polarisation sensitive MZAM.

After the MZAM, a 2% tap coupler was used to monitor the input pulses to the amplifier chain and also optimise the applied DC voltage to the MZAM in order to obtain the highest pulse extinction ratio. The 98% output of the tap coupler was amplified by EDFA-I, a core-pumped telecommunications-grade EDFA (Nortel), which delivered an output power of 2 mW irrespective of the input power level. The output of EDFA-I was further amplified in EDFA-II, a cladding-pumped EDFA that had a saturated output power of up to 1 W. EDFA-II had FC/APC connectors between its amplification stages, which enabled an interstage bandpass filter to be inserted. The bandpass filter used (Alnair Labs BVF-200CL) was

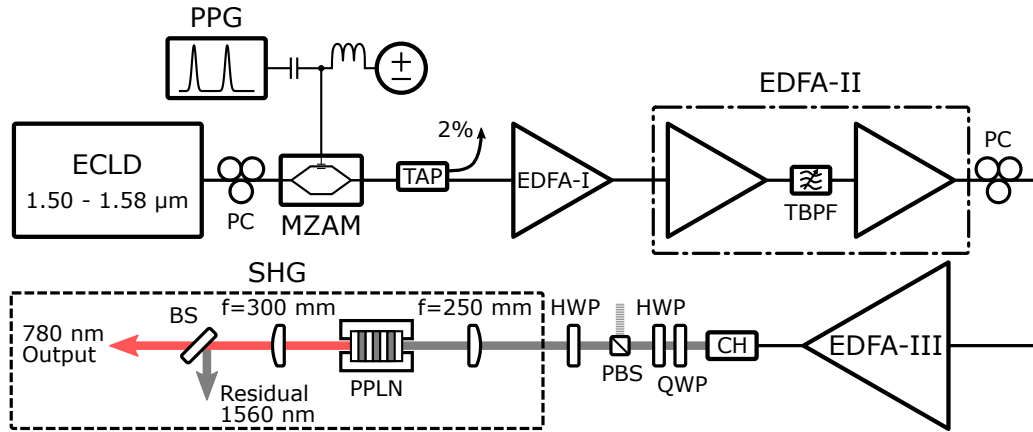


Figure 5.2: Schematic of the wavelength and duration tunable Er-MOPFA and second-harmonic generation (SHG) stage. ECLD, external-cavity laser diode; PC, polarisation controller; PPG, picosecond pulse generator; MZAM, Mach-Zehnder amplitude modulator; EDFA, erbium-doped fibre amplifier; TBPF, tunable band-pass filter; CH, collimator head; QWP, quarter wave plate; HWP, half wave plate; PBS, polarisation beam splitter; PPLN, periodically poled lithium niobate; BS, beam splitter.

tunable in central wavelength between 1.51–1.63 μm and in bandpass bandwidth between 0.1–13 nm (3 dB width). The out-of-band suppression of the tunable bandpass filter (TBPF) was >40 dB and the filter profile was flat-top with a sharp roll off of 150 dB/nm. EDFA-II was operated at a pump current such that the output power was 250 mW, enough to saturate the final power amplifier.

The final power amplifier (EDFA-III) was a multimode cladding-pumped EDFA that had a maximum saturated output power of 16 W. A three-paddle PC was used to optimise the input polarisation to EDFA-III in order to minimise the degradation of the seed PER. The output of the EDFA-III was an anti-reflection (AR) coated collimator head, with an output beam diameter ($1/e^2$) of 3.8 mm. The seed and amplifier chain was constructed entirely of isotropic fibre, therefore, a quarter and half wave plate were used on the output of the amplifier in order to linearise the output polarisation. A polarising beam splitter (PBS) was used to eliminate any non-linearly polarised component that would not contribute to the SHG to reduce the thermal load on the crystal.

Figure 5.3 (a) shows the MZAM modulated pulses after EDFA-I, which were tunable in duration over the range 0.45–2.9 ns by varying the pulse width of the PPG. The amplitude of the PPG drive pulses was adjusted to give the most constant peak-power across the pulse, in order to maximise the SHG conversion efficiency. Figure 5.3 (b) highlights the benefit of the interstage bandpass filter in EDFA-II. The output of EDFA-I [blue trace Fig. 5.3 (b)] had an extinction between the narrow linewidth signal at 1.56 μm and the

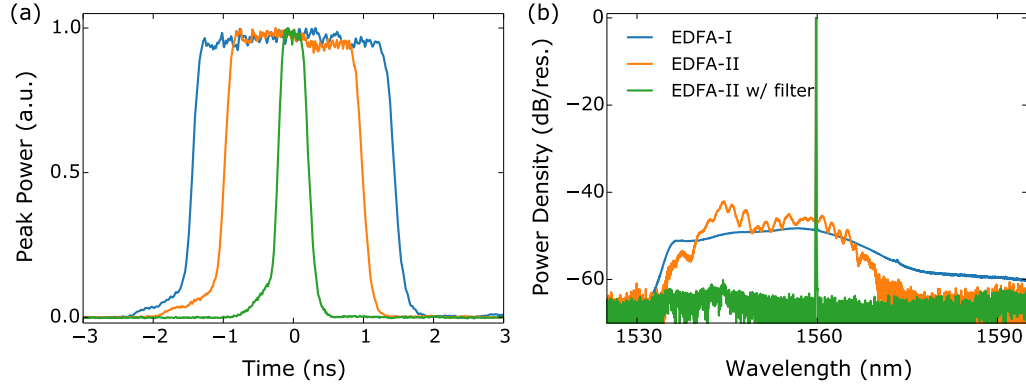


Figure 5.3: (a) Optical sampling oscilloscope traces of example 1560 nm MZAM modulated pulses. (b) Optical spectra of the output of EDFA-I (blue) and EDFA-II with (green) and without (orange) interstage filtering.

ASE background of approximately 50 dB. Without the interstage filter in EDFA-II, the ASE suppression of the output degraded to ~ 40 dB [orange trace Fig. 5.3 (b)] but with the interstage TBPF, the extinction was increased to >60 dB [green trace Fig. 5.3 (b)]. The suppression of the ASE was essential for maximising the pulse energy of the Er-MOPFA output, since the high gain of the amplifier chain could result in significant ASE between the pulses if it was left unfiltered.

Figure 5.4 shows the spectral output from EDFA-III at the maximum pump power as the wavelength of the ECLD was varied over the range 1.55–1.57 μm , with an optical pulse duration of 1 ns at a repetition rate of 50 MHz. The amplifier was specified to have an operational bandwidth from 1545–1565 nm, however, the short-wavelength operation could not be utilised for efficient SHG because the beam quality deteriorated due to an increase in the higher-order mode content. The long-wavelength operation was limited by the decrease in gain due to the reduction in the emission cross-section [Fig. 5.1 (b)] and the concomitant increase in ASE, which peaks around 1565 nm due to absorption of shorter wavelengths and re-emission. Indeed, it can be seen for the signal wavelengths of 1550 nm and 1555 nm, ASE is present around 1565 nm (Fig. 5.4), which is due to absorption of the signal and re-emission at longer wavelengths. In all the traces in Fig. 5.4, the spectral width of the signal peak is <10 pm, limited by the resolution of the optical spectrum analyser (OSA), demonstrating that there was negligible nonlinear spectral broadening.

In spite of the fact that the entire amplifier chain was constructed from isotropic fibre and only polarisation insensitive components were used after the MZAM, the output of the Er-MOPFA was reasonably polarised, with a typical PER of 14 dB. The polarisation

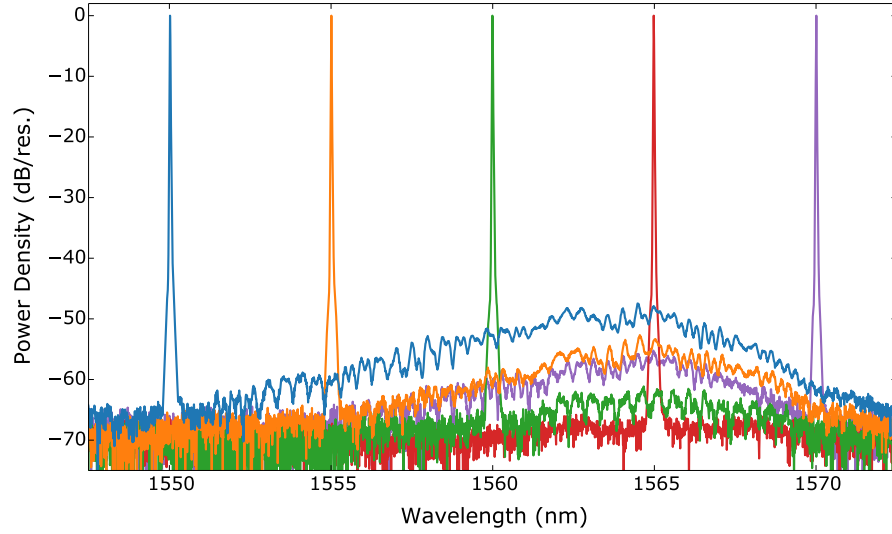


Figure 5.4: Optical spectra of the output of the EDFA-III as the seed wavelength is tuned from 1.55–1.57 μm .

state initially evolved on a fairly short timescale immediately after the power amplifier was turned on. After the temperature of the power amplifier had stabilised, a power stability of 1% for a polarised power of 15.8 W was obtained over a 15 minute duration without optimisation of the PCs. It was noted that whilst the PER of the Er-MOPFA was not always constant, the PC before the final power amplifier was vital for optimising the PER of the output.

5.4 Frequency-doubling in lithium niobate

In order to achieve the maximum SHG conversion efficiency of the Er-MOPFA, a 40 mm PPLN crystal was used to frequency-double the output. PPLN has the highest effective nonlinear coefficient of commonly used SHG crystals, which combined with the long crystal length ensures a high conversion efficiency even with relatively modest peak-power. The long crystal length has the additional advantage that the optimal focusing conditions (cf. Section 1.4.5) correspond to a larger spot size in the crystal in comparison to a shorter crystal. This is important for mitigating against photorefractive damage (PRD), which has the lowest intensity threshold of the potential optical damage mechanisms of PPLN. To further guard against PRD, the PPLN crystal was doped with 5 mol.% MgO and was held at elevated temperatures ($>150^\circ\text{C}$).

The domain inversion grating in a periodically poled crystal can only efficiently quasi phase-match a finite bandwidth around a specific SHG wavelength (Eqn 1.68). The

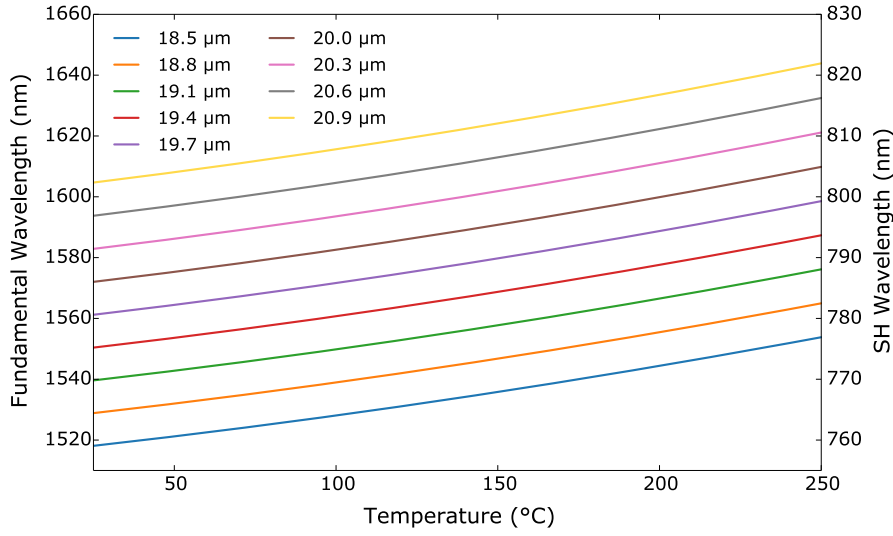


Figure 5.5: Calculated temperature phase-matching curves for the multiple grating period 5 mol.% MgO-doped congruent periodically poled lithium niobate crystal. The temperature-dependent refractive index was calculated using the Sellmeier equation from [Gay08].

grating period and the dispersive phase-mismatch are both temperature dependent due to linear thermal expansion and the thermo-optic effect, respectively. There is, therefore, a finite temperature bandwidth over which a specific wavelength is efficiently quasi phase-matched (Eqn. 1.69). Conversely, by tuning the temperature of a periodically poled crystal whilst simultaneously varying the wavelength of the fundamental, a wavelength-tunable frequency-doubled output can be achieved. The temperature range over which the crystal can practically be held due to the oven design is approximately 30–220°C, which is further restricted if elevated temperature is required to suppress PRD. To increase the SHG wavelength tuning range, therefore, periodically poled crystals are commonly poled with multiple gratings in the transverse direction, which is straightforward to implement using lithographic polling techniques.

The 40 mm, 5 mol.% MgO-doped congruent PPLN crystal used to frequency-double the Er-MOPFA system had a thickness of 0.5 mm, a width of 10 mm and was poled with nine gratings with polling periods between 18.5–20.9 μm in 0.3 μm intervals. Figure 5.5 shows the predicted SHG phase-matched wavelength as a function of crystal temperature for the nine grating periods. The SHG phase-matched wavelength is calculated by finding the roots of the phase-mismatch function (Eqn. 1.67), which is computed using the temperature-dependent refractive index and grating period. To obtain the temperature-dependent refractive index at an arbitrary wavelength, the empirical

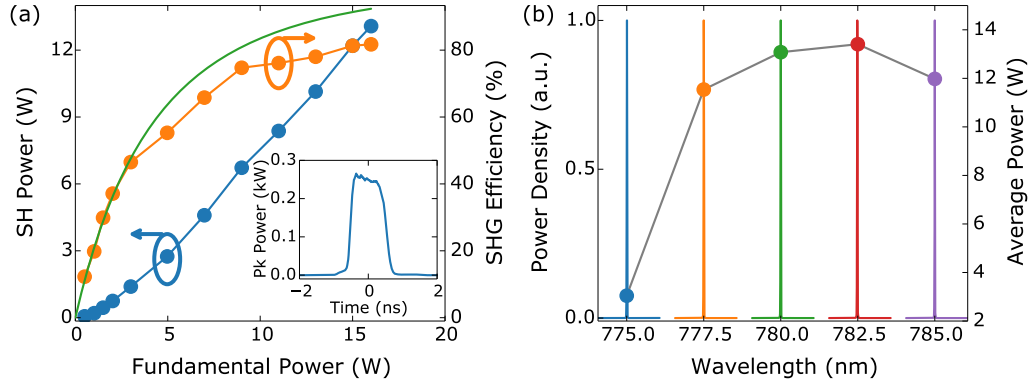


Figure 5.6: (a) 780 nm second-harmonic (SH) average power (blue points) and conversion efficiency (orange points), corrected for the beam splitter transmission, as a function of 1560 nm average power with $\tanh^2$ fit (green line). Inset: optical sampling oscilloscope trace of the 780 nm pulse. (b) Optical spectra and maximum average power (points) of the second-harmonic as the fundamental wavelength is tuned over the range 1.55–1.57 μm . Grey line is a guide for the eye.

temperature-dependent Sellmeier equation from [Gay08] was used. It is clear from Fig. 5.5 that the PPLN crystal can phase-match SHG over the entire 1.53–1.61 μm Er^{3+} gain bandwidth.

The setup for SHG of the Er-MOPFA system is depicted in the dashed box in Fig. 5.2. The collimated output after the PBS was focused to an estimated waist radius ($1/e^2$) of 65 μm at the centre of the PPLN crystal using an AR-coated $f = 250$ mm plano-convex lens. The PPLN crystal was held in a resistive-heated copper oven that was temperature controlled with a precision of $\pm 0.1^\circ\text{C}$. The generated second-harmonic was collimated using an AR-coated $f = 300$ mm plano-convex lens. A short-pass dichroic beam splitter, with a measured attenuation of 25 dB for 1560 nm and a transmission of 91% for 780 nm, was used to separate the residual fundamental from the second-harmonic.

It is worth noting that the transmission loss of the beam splitter could have been avoided by using two long-pass dichroic beam splitters to reflect the second-harmonic, since their combined reflection can be greater than 99%, whilst providing >30 dB attenuation to the fundamental. This configuration has the additional advantage that the second-harmonic does not pass through the dichroic substrate, thus avoiding attenuation and possible wavefront distortion.

Figure 5.6 (a) shows the obtained average power at 780 nm, corrected for the beam splitter transmission, for a signal wavelength of 1560 nm using an optimised fundamental pulse duration of 1.1 ns at a repetition rate of 50 MHz. A maximum 780 nm average power of 13.1 W (11.9 W uncorrected) was generated for 16.0 W of 1560 nm fundamental,

corresponding to a conversion efficiency of 82%. The 780 nm pulses had a slightly reduced duration of 1.0 ns with a corresponding peak-power of 265 W [inset Fig. 5.6 (a)]. In Fig. 5.6 (a), it is clear that at low average powers, the conversion efficiency agrees well with the expected $\tanh^2(\sqrt{P_{\omega}})$ relationship (Eqn. 1.72) but there is deviation from the fit for average powers >5 W. This is most likely due to thermal dephasing (cf. Section 1.4.5), where the absorption of the fundamental and particularly the generated second-harmonic leads to temperature variations that disrupt the quasi phase-matching (QPM). This is supported by the fact that at the full fundamental average power of 16 W, the optimum phase-matching temperature of the controller was reduced by $\sim 0.5^\circ\text{C}$, indicating that absorption had caused a heating of the crystal.

Figure 5.6 (a) also shows a significant saturation in the SHG conversion efficiency for fundamental average powers >10 W. The SHG conversion efficiency is ultimately limited by the process of back-conversion, whereby a second-harmonic photon undergoes degenerate difference frequency generation to form two photons at the fundamental wavelength. This is effectively the inverse process of SHG and, therefore, it is phase-matched necessarily. Back-conversion can be an issue due to the Gaussian radial intensity distribution of the fundamental beam, which results in a non-uniform nonlinear drive across the transverse extent of the beam. To avoid a significant loss in conversion efficiency due to the poorly-converted wings, the nonlinear drive at the peak in the centre of the beam needs to be significantly larger than is optimal to avoid back-conversion [Par02]. A similar argument would also apply to the time domain, explaining why a square pulse shape is the optimum for maximising the SHG conversion efficiency. One of the great advantages of the flexibility in the pulse duration and repetition rate of the Er-MOPFA system is that the peak-power of the fundamental can be optimised on-the-fly to maximise the SHG conversion efficiency, independent of the average power and spatial profile.

The wavelength of the second-harmonic was tuned over a range of 10 nm using the 18.8, 19.1 and 19.4 μm grating periods in the PPLN crystal and adjusting the crystal oven temperature accordingly to achieve QPM. Figure 5.6 (b) shows the second-harmonic output spectra as the fundamental wavelength was tuned over the range 1.55–1.57 μm , corresponding to the Er-MOPFA spectra shown in Fig. 5.4. All of the second-harmonic spectra have a 3 dB spectral bandwidth of <10 pm, limited by the resolution of the OSA.

The second-harmonic average power [points Fig. 5.6 (b)] was >11.5 W over the range 777.5–785.0 nm. A maximum second-harmonic average power of 13.4 W was obtained at 782.5 nm for a fundamental power of 16.4 W, corresponding to a conversion efficiency of 82%. The second-harmonic average power declined significantly for wavelengths <777.5 nm due to the fact that the higher order mode content of the Er-MOPFA output beam increased significantly as the signal wavelength was reduced. This was observed as a

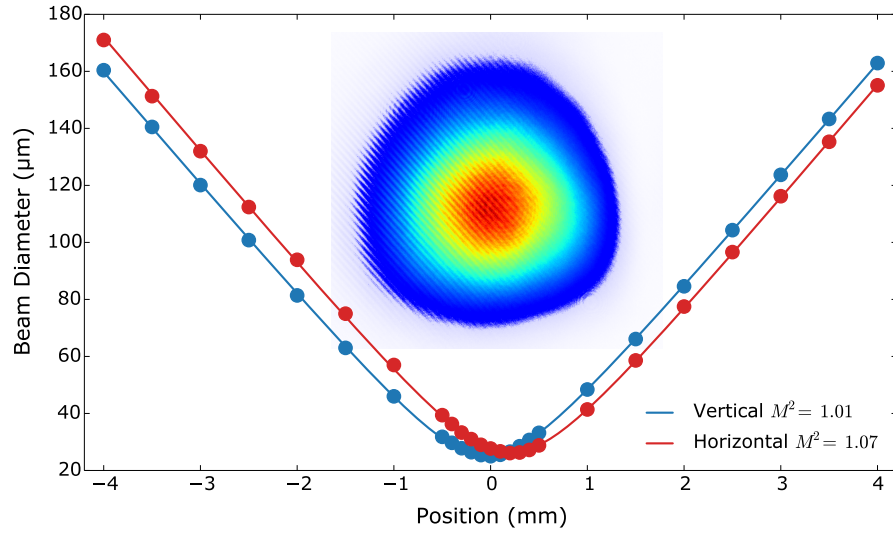


Figure 5.7: Beam diameter (points) of the collimated 780 nm beam focused by an $f = 50$ mm lens with Gaussian beam caustic fit (lines). Inset: CCD camera image of the collimated beam.

significant deterioration in the beam profile and average power of the second-harmonic. It was found that the beam quality deteriorated beyond a specific average power level, which decreased with signal wavelength. For a signal wavelength of $1.55 \mu\text{m}$, the maximum second-harmonic average power of 3.0 W was obtained for a fundamental power of 7.0 W, less than half of the maximum fundamental average power available. It is possible that the conversion efficiency at this signal wavelength could be significantly improved by adjusting the temporal parameters to increase the peak-power to a level equivalent of using 16 W of fundamental.

The second-harmonic average power declined at longer wavelengths due to the reduction in gain for longer signal wavelengths in the Er-MOPFA, which decreased the maximum fundamental average power and increased the ASE content in the Er-MOPFA output. In addition, the static extinction ratio of the MZAM had a maximum value of 46 dB at a signal wavelength of 1564 nm, which decreased significantly for shorter and longer wavelengths, which may have contributed to the decline in second-harmonic average power due to a decrease in the pulse energy.

The beam quality of the second-harmonic was measured for a signal wavelength of $1.56 \mu\text{m}$ using a scanning slit beam profiler to measure the beam diameter of the collimated beam focused by an AR-coated $f = 50$ mm plano-convex lens (points Fig. 5.7). A Gaussian fit of the beam caustic (lines Fig. 5.7) revealed an $M^2 = 1.01$ in the vertical axis and $M^2 = 1.07$ in the horizontal axis. There is noticeable astigmatism, apparent from the

position of the focus of the two axis not being coincident, which is most likely due to either tilt of the PPLN crystal or wavefront distortion from optics (e.g. beam splitter) after the collimating lens due to improper collimation. Experience from the previous chapters suggests that this astigmatism can be eliminated by the correct optical alignment of the frequency-doubling setup. The collimated second-harmonic beam has a beam diameter of 2.1 mm at the position of the $f = 50$ mm lens, with an ellipticity of 0.99 (inset Fig. 5.7).

5.5 Summary

This chapter detailed the development of a wavelength-tunable, temporally flexible source around 780 nm, designed for subsequent SFG with the Yb-MOPFA described in the previous chapter. The source was based on direct frequency-doubling of a wavelength and pulse duration tunable Er-MOPFA system using a multiple grating PPLN crystal. The seed source of the Er-MOPFA was a narrow linewidth, wavelength tunable ECLD system that was externally modulated using an MZAM. This seed source was tunable in wavelength over the entire gain bandwidth of the EDFAs used in the amplifier chain and was tunable in pulse duration over the range 0.45–2.9 ns at an arbitrary repetition rate. The seed source was amplified to a maximum polarised average power of 16 W in a chain of EDFAs that were designed to minimise ASE in order to maximise the pulse energy for a given repetition rate. Despite the amplifier chain being constructed entirely of isotropic fibre and only polarisation insensitive components used after the MZAM, the output of the Er-MOPFA had a typical PER of 14 dB. When the signal wavelength was tuned over the range 1.55–1.57 μm , the Er-MOPFA output maintained an ASE extinction of >50 dB and spectral broadening beyond the 10 pm resolution limit of the OSA was not observed.

The output of the Er-MOPFA was ideally suited for frequency-doubling due to its narrow linewidth, duration-tunable square pulses, good beam quality and reasonably high degree of polarisation. A 40 mm long, 5 mol.% MgO-doped PPLN was chosen to frequency-double the output of the Er-MOPFA to relax the peak-intensity required to obtain a high SHG conversion efficiency, due to the long length and high nonlinear coefficient. A maximum second-harmonic average power of 13.4 W was obtained at 782.5 nm for a fundamental power of 16.4 W using an optimised pulse duration of 1.1 ns at a repetition rate of 50 MHz. The corresponding conversion efficiency of 82% is comparable to some of the highest reported conversion efficiencies for single-pass SHG [Tav98] and external cavity SHG [Ou92].

The PPLN crystal had nine domain inversion gratings, which in combination with temperature tuning and varying the wavelength of the Er-MOPFA output, enabled the second-harmonic to be tuned over the range 775–785 nm. The obtained second-harmonic

average power was >11.5 W over the range 777.5–785 nm but it declined to 3.0 W for a wavelength of 775 nm. The sharp decline in second-harmonic average power was due to an increase in the higher order mode content of the Er-MOPFA output as the signal wavelength was reduced below 1555 nm. The increased high-order mode content restricted the maximum fundamental average power of the Er-MOPFA output to 7.0 W for a signal wavelength of 1555 nm, beyond which the second-harmonic average power declined. The beam quality of the 780 nm second-harmonic was near diffraction limited with a measured $M^2 = 1.01$ in the vertical axis and $M^2 = 1.07$ in the horizontal axis, with a collimated beam diameter of 2.1 mm and ellipticity of 0.99. The high average power, excellent beam quality, narrow linewidth and tailorable pulse characteristics of the source are ideal for obtaining a high SFG conversion efficiency in conjunction with a Yb-MOPFA system.

6 Conclusions and outlook

In this thesis it has been shown that the frequency-doubling of rare-earth doped master oscillator power fibre amplifier (MOPFA) schemes using periodically poled nonlinear crystals is a power scalable, wavelength versatile and temporally flexible technique for accessing spectral regions where specific laser sources are demanded. It was demonstrated that the wavelength coverage of frequency-doubled MOPFA schemes can be extended by both exploiting the large gain bandwidth of rare-earth doped fibre amplifiers and by using nonlinear wavelength conversion in optical fibres.

The thesis was organised such that each experimental chapter described a source that had been developed. In this chapter, there will be a review of the conclusions from the source implementation and the potential developments, organised by each wavelength covered by the sources. Finally, a brief outlook to contextualise the work in this thesis will conclude this chapter.

6.1 560 nm sources

In Chapter 2, it was identified that many applications from a diverse number of fields demand laser sources in the yellow-green spectral region (550–600 nm) but there is a lack of existing practical lasers that directly emit in this region with suitable power, wavelength and temporal characteristics for a great number of applications. In particular, it was discussed that a depletion beam source for stimulated emission depletion (STED) microscopy using green fluorescent protein (GFP) was required to emit 0.1–2 ns duration pulses at low megahertz repetition rates with high pulse energy and diffraction-limited beam quality at a central wavelength around 560 nm. Existing solutions to provide the depletion beam based on nonlinear frequency conversion of ultrafast Ti:Sapphire oscillators and ytterbium master oscillator power fibre amplifier (Yb-MOPFA) schemes were evaluated, with the conclusion that practicality and optical efficiency could be greatly improved upon.

It was discussed that frequency-doubled Yb-MOPFA schemes would be able to satisfy all of the depletion beam criteria, apart from the fact that operating with a fundamental wavelength of 1120 nm is technologically very challenging to achieve. It was subsequently demonstrated that using a pulse-pumped Raman amplifier that was seeded

with a narrow linewidth signal was a powerful concept for spectrally shifting the output of a Yb-MOPFA system, whilst maintaining a spectral bandwidth that is compatible with frequency-doubling in periodically poled nonlinear crystals.

A temporally-stretched mode-locked oscillator amplified in a ytterbium-doped fibre amplifier (YDFA) was used to pump a Raman amplifier that was seeded with a distributed feedback (DFB) laser diode operating at 1120 nm, generating 1.9 W of average power with a pulse duration of 230 ps at a repetition rate of 47.5 MHz. This output was frequency-doubled in periodically poled lithium niobate (PPLN) that was packaged by our industrial collaborators Gooch and Housego into a fibre-coupled module, resulting in an alignment-free fibre-integrated 560 nm pulse source. A maximum 560 nm average power of 480 mW was obtained with a pulse duration of 150 ps and a pulse energy of 10 nJ, which was primarily limited by spectral broadening of the 1120 nm fundamental in the Raman amplifier. The source satisfied all of the depletion beam criteria in a compact footprint but the turn-key nature of the source was severely compromised due to the randomly polarised Yb-MOPFA output.

Chapter 3 improved on the design of the 560 nm source described in Chapter 2 by using a polarised Yb-MOPFA pump system and by optimising the Raman amplifier length to give the highest 1120 nm peak-power spectral density. Furthermore, the temporal flexibility of the source was increased by using either external modulation with an Mach-Zehnder amplitude modulator (MZAM) or a gain-switched semiconductor laser diode as the master oscillator for the Yb-MOPFA, after a comprehensive review of the available options. The source was originally intended to be used with the fibre-coupled frequency-doubling module but the increased peak-power within the spectral acceptance bandwidth of the PPLN crystal caused catastrophic photorefractive damage (PRD). To rectify this issue, the output of the pulse-pumped Raman amplifier was frequency-doubled using a periodically poled lithium tantalate (PPLT) crystal, which has a significantly higher PRD damage threshold in comparison to PPLN.

The best performance was obtained using the the MZAM-based master oscillator, with a maximum 560 nm average power of 1.53 W and a pulse energy of 765 nJ being generated from 6 W of Yb-MOPFA pump power. The 560 nm pulse duration was tunable over the range 56 ps–2.7 ns with Watt-level average power and a nearly constant pulse peak-power of ~300 W. Using the simpler and more cost effective gain-switched laser diode master oscillator, a maximum 560 nm average power of 1.33 W with a pulse energy of 532 nJ was obtained for the same pump power. This result was unexpected due to the presence of a high peak-power transient at the leading edge of the gain-switched seed pulse but it was found that this was suppressed in the Raman conversion process with only a small reduction in the conversion efficiency. The 560 nm pulse duration was tunable over the

range 40 ps–2 ns, however, the average and peak-power declined significantly for the short pulse durations due to the poorly converted transient becoming a larger proportion of the Yb-MOPFA seed pulses.

The 560 nm source using either master oscillator could be directly integrated into a STED microscope due to its near diffraction-limited beam quality, high pulse energy and suitable temporal and spectral characteristics. Indeed, at the time of writing, the source is currently in use in the Biophotonics group at Imperial College London as the depletion beam source in a STED microscope using the GFP fluorophore. The average power of the 560 nm system is more than sufficient for STED microscopy, since the average power damage threshold of typically used fluorophores is quite limited. For applications that require greater average power, the source could be scaled to 10's of Watts by increasing the Yb-MOPFA pump power and adjusting the duty factor to maintain ~ 1 kW peak-power. The main limitation on scaling the 560 nm average power would be thermal dephasing in the periodically poled frequency-doubling crystal.

Scaling the pulse energy and correspondingly the peak-power of the 560 nm pulses, however, would be significantly more challenging. This is primarily due to the fact that the power spectral density of the Raman amplifier output cannot be maintained when pumping with higher peak-powers due to deleterious four-wave mixing (FWM) processes. To significantly scale the pulse energy, performing the Raman amplification in the power amplifier of a Yb-MOPFA system is preferable [Dup06a, Row14], with the additional benefit of eliminating the loss of the discrete Raman amplifier. Scaling the peak-power to multiple kilowatts would, however, preclude the use of periodically poled frequency-doubling crystals due to their low optical damage thresholds. Of the non-poled crystals, lithium triborate (LBO) is particularly attractive since non-critical phase-matching (NCPM) can be implemented over the wavelength range 1064–1585 nm by adjusting the crystal temperature over the range 25–150°C. The significantly lower effective nonlinear coefficient of LBO in comparison to periodically poled crystals, however, would make achieving a very high second-harmonic generation (SHG) conversion significantly more challenging.

6.2 590 nm sources

In Chapter 4, the pulse-pumped Raman amplifier concept was extended to 1180 nm by using cascaded Raman amplification, where Raman conversion is used to form an intermediate Stokes order that provides Raman gain to the signal wavelength. Initially, it was hoped that the intermediate Stokes order did not need to be seeded, such that amplified spontaneous Raman emission would provide the Raman gain to the 1180 nm signal. It was found, however, that whilst the unseeded cascaded Raman amplifier did

amplify the narrow linewidth 1180 nm signal, it could not be used for efficient frequency-doubling. This was due to the very poor power spectral density, caused by spectral broadening from competing four-wave interactions that were more prevalent due to the high peak-power required to form the Raman amplified spontaneous emission (ASE) and its broad spectral bandwidth.

To overcome this issue, a seeded pulse-pumped Raman amplifier operating at 1120 nm was used to pulse-pump the 1180 nm Raman amplifier to obtain narrow linewidth pulses at 1180 nm. These pulses were frequency-doubled in a PPLN crystal, generating a maximum 590 nm average power of 1.1 W with a pulse duration of 1.8 ns and a pulse energy of 190 nJ for a Raman amplifier output power of 6.5 W. The SHG conversion efficiency was limited by a spectral feature that was present in the 1180 nm signal that contained a significant amount of power outside the spectral acceptance bandwidth of the PPLN crystal. The experimental evidence suggested this spectral feature was due to FWM between two modes in the 1120 nm Raman fibre laser output and the 1180 nm signal. In spite of this, the generated 590 nm peak power was sufficient to cause catastrophic PRD at a pump repetition rate of 5 MHz, corresponding to a peak 590 nm intensity of $\sim 5 \text{ MW/cm}^2$. Work is ongoing to eliminate the spectral feature in the 1180 nm signal to improve the peak-power contained within the spectral acceptance bandwidth of the periodically poled frequency-doubling crystal. Additionally, the frequency-doubling will be performed in PPLT in order to increase the PRD threshold and, therefore, scale the peak-power of the 590 nm pulses.

A feature of the pulse-pumped Raman amplifier systems that has yet to be explored is the potential for significant wavelength-tuning, due to the lack of available seed sources and periodically poled crystals with the appropriate grating periods. The Raman gain bandwidth is approximately 6 THz for the PM Raman fibre, corresponding to $\sim 25 \text{ nm}$ at 1120 nm, therefore, significant wavelength tunability is expected. This will be explored in future experiments using a wavelength-tunable external-cavity laser diode (ECLD) source. To extend the wavelength tuning range of the pulse-pumped Raman amplifiers further, it would also be possible to simultaneously tune the wavelength of the Yb-MOPFA pump system. Furthermore, using pump and signal tuning in conjunction with cascaded Raman amplification, it should be possible to cover the entire yellow-green (550–600 nm) spectral region. To obtain longer wavelengths using fewer cascades, the 40 THz frequency shift Raman gain peak in phosphosilicate fibres could be used to generate pulses around 1240 nm (from 1064 nm) that can be frequency doubled into the red spectral region.

To significantly scale the pulse energy and peak-power of frequency-doubled Raman-shifted MOPFA systems, one potential avenue could be to use crystalline Raman gain media instead of optical fibres. Raman conversion of high peak-power solid state lasers

in crystalline materials such as diamond, barium tungstate (BaWO_4) or potassium gadolinium tungstate (KGW) is well established [Pip07]. The high ($\sim 1 \text{ GW/cm}^2$) peak intensity required to achieve efficient conversion due to the limited interaction length becomes an advantage for scaling the peak-power of the Raman-shifted source. Using confocal focusing, a pump peak-power of $\sim 50 \text{ kW}$ would be required to obtain the necessary intensity, therefore, given that conversion efficiencies as high as 85% have been demonstrated [Čer02], Raman-shifted peak-powers of 10's kW could be obtained. This pump peak-power is readily achievable using Yb-MOPFA schemes with large mode-area (LMA) active fibres [Ric10].

As an example, BaWO_4 has a Raman shift of 28 THz, which corresponds to a Stokes wavelength of 1180 nm for a 1064 nm pump. In addition, the Raman gain bandwidth of BaWO_4 is only 48 GHz (0.22 nm at 1180 nm), therefore, efficient frequency-doubling could be obtained using LBO without the need for narrow linewidth seed. NCPM can be implemented using LBO for SHG of both the 1064 nm pump and the 1180 nm Stokes and additionally sum-frequency generation (SFG) of the two wavelengths to generate 560 nm, simply by varying the crystal temperature. Using a LBO in conjunction with a BaWO_4 Raman-shifted wavelength-tunable Yb-MOPFA system could, therefore, cover the entire 515–600 nm wavelength range, only utilising a single Raman shift. The power scalability of Yb-MOPFA systems would facilitate the generation of unprecedented power levels across the entire yellow-green spectral region in a practical format.

6.3 780 nm sources

In Chapter 5, it was discussed that in contrast to the yellow-green spectral regions, there are many mature direct-emission laser sources in the short-wavelength near-infrared (NIR) spectral region around 800 nm. In spite of this, due to the very high conversion efficiencies that can be obtained using SHG, frequency-doubled erbium master oscillator power fibre amplifier (Er-MOPFA) systems offer a number of advantages over direct emission sources. Specifically, for SFG in conjunction with a Yb-MOPFA system to generate blue light, it was discussed that a frequency-doubled Er-MOPFA is more suitable than direct-emission sources due to the ease of obtaining temporally matched and synchronised pulse trains and the high average power available. In light of this, a frequency-doubled Er-MOPFA was designed for efficient, high average power SFG in conjunction with the Yb-MOPFA system used in Chapter 4. The key design features were temporal flexibility to match the Yb-MOPFA pulse train, narrow spectral linewidth to be compatible with the narrow spectral acceptance bandwidth of periodically poled crystals and wavelength flexibility to relax the SFG phase-matching requirements.

To match these design requirements, an Er-MOPFA was implemented using a wavelength tunable ECLD that was externally modulated by an MZAM. The Er-MOPFA system was tunable in wavelength over the range 1.55–1.57 μm , had a resolution limited spectral linewidth of <10 pm and was tunable in pulse duration over the range 0.45–2.9 ns. A 40 mm long multiple grating period PPLN crystal was used to frequency-double the output of the Er-MOPFA. A maximum second-harmonic average power of 13.4 W was obtained at 782.5 nm for a fundamental power of 16.4 W. The corresponding conversion efficiency of 82% is close to some of the highest reported values for SHG.

Wavelength tuning of the second-harmonic was demonstrated over the range 775–785 nm, with >11.5 W of average power generated for wavelengths over the range 777.5–785.0 nm. The second-harmonic average power declined significantly for wavelengths <777.5 nm due to an increase in higher-order mode content in the Er-MOPFA power amplifier output. The second-harmonic had a resolution limited linewidth of <10 pm and had near diffraction limited beam quality with an $M^2 \leq 1.07$. The high average power, excellent beam quality and narrow spectral linewidth of the second-harmonic is ideally suited for efficient SFG in conjunction with a temporally matched Yb-MOPFA system, which is the subject of future experimental work.

6.4 Outlook

Since the first demonstration of the laser in 1960, there has been remarkable progress in the translation of research on laser sources from laboratory curiosities to commercial turn-key laser solutions. Indeed, almost all aspects of modern life have been influenced by laser technology, from the fabrication of microprocessors to the vast optical telecommunications networks that are the backbone of the Internet. The progress in the field of nonlinear optics has concomitantly mirrored that of laser development, from initially being interesting on a fundamental physics level to becoming a practical method for shifting the spectral emission of laser sources. It is hoped that the nonlinear conversion techniques demonstrated in this thesis make a small contribution towards the large research effort into increasing the outreach of laser sources into applications and environments that currently do not fully benefit from the unique properties of laser light.

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List of Publications

Journal papers

1. T. H. Runcorn, R. T. Murray, E. J. R Kelleher, S. V. Popov, and J. R. Taylor, "Duration-tunable picosecond source at 560 nm with watt-level average power," *Optics Letters* **40**, 3085 (2015).
2. T. H. Runcorn, T. Legg, R. T. Murray, E. J. R. Kelleher, S. V. Popov, J. R. Taylor, "Fiber-integrated frequency-doubling of a picosecond Raman laser to 560 nm," *Optics Express* **23**, 15728 (2015).

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1. T. H. Runcorn, R. T. Murray, E. J. R Kelleher, S. V. Popov, and J. R. Taylor, "Watt-level, Duration-tunable Picosecond Source at 560 nm by Second-harmonic Generation of a Raman Fiber Laser," in *CLEO/Europe and EQEC 2015 Conference Digest*, CD-1.4 (2015).
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1. R. I. Woodward, E. J. R. Kelleher, T. H. Runcorn, S. Loranger, D. Popa, V. J. Wittwer, A. C. Ferrari, S. V. Popov, R. Kashyap, and J. R. Taylor, "Nanosecond to Picosecond Fiber Bragg Grating Compression of Giant-Chirped Pulses from an Ultra-Long Mode-Locked Fiber Laser," in *CLEO/Europe and EQEC 2015 Conference Digest*, CJ-9.6 (2015).
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 4. M. Zhang, E. J. Kelleher, T. H. Runcorn, D. Popa, F. Torrisi, A. C. Ferrari, S. V. Popov, and J. R. Taylor, “Synchronously coupled fiber lasers and sum frequency generation using graphene composites,” in *CLEO:2014*, STu1I.2 (2014).
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Acronyms

AML	Actively Mode-locked
AOM	Acousto-optic Modulator
AR	Anti-reflection
ASE	Amplified Spontaneous Emission
AWG	Arbitrary Waveform Generator
BPM	Birefringent Phase-matching
BPP	Beam Parameter Product
CLSM	Confocal Laser Scanning Fluorescence Microscopy
CPA	Chirped-pulse Amplification
CW	Continuous-wave
DBR	Distributed Bragg Reflector
DFB	Distributed feedback
ECLD	External-cavity Laser Diode
EDFA	Erbium-doped Fibre Amplifier
EOM	Electro-optic Modulator
Er	Erbium (Er)
Er-fibre	Erbium fibre
Er-MOPFA	Erbium Master Oscillator Power Fibre Amplifier
FBG	Fibre Bragg Grating

FP Fabry-Pérot

FWHM Full Width at Half Maximum

FWM Four-wave Mixing

GFP Green Fluorescent Protein

GRIIRA Green-induced Infrared Absorption

GVD Group-velocity Dispersion

HFLD High Frequency Response Laser Diode

HR High-Reflection

LBO Lithium triborate (LiB_3O_5)

LMA Large Mode-area

MFA Mode-field Area

MFD Mode-field Diameter

MIR Mid-infrared

MOPA Master Oscillator Power Amplifier

MOPFA Master Oscillator Power Fibre Amplifier

MZAM Mach-Zehnder Amplitude Modulator

NCPM Non-critical Phase-matching

Nd Neodymium (Nd)

NIR Near-infrared

OPO Optical Parametric Oscillator

OSA Optical Spectrum Analyser

PBS Polarising Beam Splitter

PC Polarisation Controller

PCF	Photonic Crystal Fibre
PER	Polarisation Extinction Ratio
PM	Polarisation-maintaining
PML	Passively Mode-locked
PPB	Picosecond Pulse Board
PPG	Picosecond Pulse Generator
PPKTP	Periodically Poled Potassium Titanyl Phosphate (KTiOPO_4)
PPLN	Periodically Poled Lithium Niobate (LiNbO_3)
PPLT	Periodically Poled Lithium Tantalate (LiTaO_3)
PRD	Photorefractive Damage
QCW	Quasi-continuous-wave
QPM	Quasi Phase-matching
R6G	Rhodamine 6G
RESOLFT	Reversible Saturable Optically Linear Fluorescence
RF	Radio-frequency
RFL	Raman Fibre Laser
RFP	Red Fluorescent Protein
SBS	Stimulated Brillouin Scattering
SFG	Sum-frequency Generation
SH	Second-harmonic
SHG	Second-harmonic Generation
SLD	Superluminescent Laser Diode
SLM	Single Longitudinal Mode
SMF	Single-mode Fibre

SPM Self-phase Modulation

SRS Stimulated Raman Scattering

STED Stimulated Emission Depletion

TBPF Tunable Bandpass Filter

TEC Thermoelectric Cooler

Tm Thulium (Tm)

UV Ultraviolet

WDM Wavelength Division Multiplexer

XPM Cross-phase Modulation

Yb Ytterbium (Yb)

Yb-fibre Ytterbium fibre

Yb-MOPFA Ytterbium Master Oscillator Power Fibre Amplifier

Yb-Raman Ytterbium-Raman

YDFA Ytterbium-doped Fibre Amplifier

YFP Yellow Fluorescent Protein

YLF Lithium Yttrium Fluoride

ZDW Zero-dispersion Wavelength