GAIN CHARACTERISTICS IN SINGLE-MODE PUMPED DIAMOND RAMAN LASERS

By

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Except where acknowledged in the customary manner, the material presented in this thesis is, to the best of my knowledge, original and has not been submitted in whole or part for a degree in any university.

_____________________________
Soumya Sarang
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Abstract

Single longitudinal mode (SLM) lasers are significant for applications in spectroscopy, precision measurements, and high-resolution interferometry. The techniques for achieving SLM in inversion lasers are well-developed, as are also frequency extension by frequency conversion using harmonic crystals and optical parametric oscillators. In contrast, Raman lasers, which are well-known for having advantages for diversifying wavelength range and producing high-quality output beams, are much more immature in terms of SLM operation.

This thesis presents an experimental approach to demonstrate SLM operation in a standing-wave crystalline Raman laser. Theoretical concepts are developed to elucidate the gain saturation mechanisms and mode competition in Raman lasers. It is shown that gain saturation is homogeneous for narrow linewidth pumping and that the absence of spatial hole burning in a Raman gain medium leads to the possibility of intrinsically stable SLM operation. This concept is implemented in narrow-linewidth pumped external cavity Raman lasers in the continuous wave regime. Two Raman crystals—diamond and potassium yttrium tungstate (KYW) are investigated to understand the effects of material properties on longitudinal mode structure.

In the case of KYW, it is seen that the laser output spectrum and efficiency were greatly influenced by a lesser-known low-frequency phonon mode (87 cm\(^{-1}\)), which is shown to have high gain coefficient higher than the mode currently accepted as the primary mode. Owing to the complexity of the laser output spectrum due to the participation of this low-frequency mode in the Raman frequency conversion process, investigation of the mode spectrum was challenging. However, the low-frequency mode at 87 cm\(^{-1}\) is advantageous for applications
requiring multi-wavelength laser sources. As the quantum defect is much lower than for the conventional phonon modes in Raman crystals, it could be used for beam enhancement and/or beam combination.

Diamond is seen as a much better option for investigating SLM as it has a "pure" single phonon mode, in addition to its high Raman gain and superior thermal conductivity. Despite having 35 longitudinal modes in the diamond gain bandwidth (45 GHz), SLM operation is demonstrated at 1240 nm up to 4 W of Stokes output power for the free-running diamond Raman laser. Moreover, this SLM demonstration is significant as the gain medium was positioned at the midpoint of the standing-wave cavity, which would normally be unsuited to SLM operation in the case of an inversion laser. The pumping level and output power of this SLM operation are higher than the reported SLM inversion lasers. The coupling between the intracavity Stokes power and the optical cavity length via the thermo-optic effect and thermal expansion of diamond was found to be responsible for the multimode operation at higher powers.

Extension to longer wavelengths through second Stokes generation was also investigated. The aggravated thermal effects in diamond and cascading effect on pump depletion led to the multimode operation at low output powers. As a result, a volume Bragg grating in a coupled cavity design was used to increase the SLM power. Up to 0.5 W SLM power at 1485 nm was demonstrated with a frequency stability of the order of the pump frequency fluctuations. The feasibility of the DRL as a LIDAR transmitter was investigated through a demonstration of environmental water vapour detection.

Finally, active stabilization of the cavity length was investigated to extend the range of power and to increase frequency stability. A novel variant of Hänsch-Couillaud (HC) technique for cavity locking was developed that is proposed to be based on polarisation dependent pump depletion. This successfully increased the maximum output power to 7.2 W, which is believed to be one of, if not the highest SLM power reported for a solid-state SLM oscillator. Stimulated Brillouin scattering is observed as parasitic effect but provides a novel platform for the future development of diamond Brillouin lasers.
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Introduction

The biggest news in the year 2016 in the scientific world was the first experimental observation of gravitational waves by the Advanced Laser Interferometer Gravitational-Wave Observatory (LIGO) [1]. To detect the gravitational waves, two beams split from a laser beam were passed through a Michelson interferometer with two 4 km long arms. The passage of the gravitational wave changed the length of two arms of the interferometer by about $10^{-18}$ m which is about three orders of magnitude less than the size of a proton. Detecting such a minuscule change is challenging and relies on a ultra-stable high-power single longitudinal mode (SLM) laser source with a narrow-linewidth.

In addition to gravitational wave sensing, there is a high demand for high power, narrow linewidth, SLM (or sometimes referred to as single frequency) laser sources for specific applications such as precision spectroscopy, atom interferometry, atom cooling [2], high-resolution spectroscopy and light detection and ranging (LIDAR) [3], optical metrology and interferometry [4], external enhancement resonators for frequency doubling [5] etc. All
these applications have stringent requirements such as single frequency operation, narrow linewidth, low-intensity noise and high frequency stability. In addition to the properties provided by SLM laser mentioned above, other properties such as high power, wavelength tunability, excellent spatial beam quality/low divergence are also critical, depending upon the specific needs of the applications. One of the major requirement for gravitational wave detection is high power SLM source (> 20 W) so that the signal-to-noise ratio of the detection signal is increased [6]. High-power SLM sources are also essential for applications such as LIDAR or atom cooling. Wavelength tunability is important for LIDAR and spectroscopic applications where a range of molecules having different absorption spectral peaks are to be probed. Applications, therefore, place a large range of requirements on SLM lasers which are not met by just one technology.

1.1 Inversion lasers

In 1958, Schawlow and Townes proposed an idea of extending maser technology to amplify infrared and visible light laying the foundation for the first laser, demonstrated two years later when Maiman developed a pulsed ruby laser operating at 694.3 nm [7, 8]. Subsequently, the first continuous-wave (CW) solid-state laser was demonstrated at Bell Labs in 1962 [9]. Resonating an optical field in a laser cavity requires the field to reproduce itself after a round trip. The phase reproduction results in longitudinal modes, which are separated by the free spectral range (FSR) or mode spacing, \( \Delta \nu_{\text{FSR}} = c/2nl \), where \( n \) is the refractive index of the medium and \( l \) is the cavity length. The theory for "optical masers" predicted that a SLM would oscillate in a cavity with high reflective walls, thereby producing a highly monochromatic and coherent radiation [7]. However, it soon became evident that free-running lasers usually generate multiple longitudinal modes [10].

Studies revealed that a simple standing-wave laser resonator is generally multimode because of an effect called "spatial hole burning" (described below) [11]. Hence, innovative techniques were incorporated into the cavity design to either avoid or eliminate this broadening mechanism and thus, enforce SLM.

The broadening mechanisms in the gain medium influencing the output spectrum and mode
competition of the inversion laser represent important concepts that underpin the ideas developed in this thesis. The following section describes the impact of spatial hole burning on the mode spectrum of the inversion lasers and the major techniques used to overcome spatial hole burning.

### 1.1.1 Gain spectrum broadening and mode competition

Within a homogeneously broadened gain profile, the probability of all the atoms to interact with a radiation field are equal. The broadening of the transition is due to the finite lifetime of the involved energy levels or the same average collision rate. The former is referred as to lifetime broadening and mainly occurs in crystals, whereas the latter is collisional broadening, typical in gases. The emission spectrum of a gain medium exhibiting this type of broadening is Lorentzian. For inhomogeneous broadening, all the atoms behave independently of each other and the probability of interaction with the field differs across the atomic ensemble. For example, Doppler shifts caused by different velocities of atoms or in disordered crystals where specific lattice locations experience varying local electric and magnetic fields. In these cases, the emission spectrum is broadened and in many cases, follows a Gaussian profile.

The longitudinal modes of a cavity compete for gain according to nature of gain broadening. In the case of homogeneous broadening, all modes compete equally for the same excited energy level. Since the gain at the centre of the profile is highest, the mode placed at this position attains threshold first and oscillates in the cavity. Saturation of the gain causes a reduction of gain available for other competing modes, thereby preventing their amplification in the cavity unless the mode can access the gain from another spatial region of the gain medium. Therefore, homogeneous broadening leads to strong mode competition resulting in SLM operation under ideal conditions.

In contrast, in an inhomogeneously broadened gain medium, the interaction of modes is stronger for a section of the inverted population than for others. Hence gain saturation and mode competition only occur most strongly for excited atoms in that section. To achieve an SLM operation well above threshold, it is inevitable to incorporate a frequency selective element in the laser cavity to eliminate other competing longitudinal modes.
1.1.2 Spatial hole burning

In a standing-wave cavity, the wave pattern of the longitudinal mode leads to a preferential saturation of the gain at the antinodes of the pattern. As a result, a periodic spatial modulation of gain develops in the cavity with a phase difference of $\pi/2$ with the intensity profile as shown in Fig. 1.1. Gain at the nodes is accessible by neighbouring modes and attain threshold generally for small increases in pump power above threshold. This effect prevails in many standing-wave lasers including those with homogeneously broadened gain medium [11, 12].

Figure 1.1: Schematic of spatial hole burning. The gain profile is $\pi/2$ out of phase with the longitudinal mode in a standing-wave cavity.

Since the longitudinal mode at the gain maximum achieves threshold first, a standing-wave cavity is SLM at pump powers marginally above the threshold [11]. The basic expressions for the threshold of the second mode for a cavity where the length of the medium is generally smaller than the mirror spacing, have been summarized in [13–15]. The function $f(r)$ defined in [15] determines the condition for SLM operation

$$f(r) < \frac{1 - e^{(-d/l)}}{2 - (d^2/l^2 + 2d/l + 2)e^{(-d/l)}} \frac{1}{\tau^2},$$  \hspace{1cm} (1.1)

where $d$ is the gain medium length, $l$ is the absorption depth. $\tau$ is defined as $\pi n l \Delta \lambda / \lambda_0^2$ where $n$, $\lambda_0$ and $\Delta \lambda$, are refractive index of the gain medium, central wavelength and transition
1.1 Inversion lasers

linewidth, respectively. Although the function does not hold any physical significance, the inverse of the function determines the required pump power above the single mode threshold for the second mode to oscillate. The inverse of \( f(r) \) is given as \( r(f) = \frac{1}{2}(f + 1)(f + 2) \).

\[ r(f) = \frac{1}{2}(f + 1)(f + 2). \]  

where \( r = 1 \) at single mode threshold. The significance of these equations is explained as follows. Consider a Nd:YAG laser with a transition linewidth (\( \Delta \lambda \)) of 0.5 nm \([16]\), \( n = 1.8 \), emission wavelength \( \lambda_0 = 1064 \text{nm} \). For the purpose of calculation, assume that \( d = 8 \text{mm} \) and \( l = 2 \text{mm} \). Putting these parameters in Eq. 1.1 the condition for SLM operation is obtained as, \( f(r) < 0.03 \). The inverse of this function is obtained by applying the value of \( f = 0.03 \) in Eq. 1.2 as, \( r(f) < 1.05 \). As \( r = 1 \) at the single mode threshold, it is inferred from Eq. 1.2 that the second mode will oscillate for a pump power 5% above threshold.

1.1.3 Review on SLM techniques

SLM inversion lasers employ a range of techniques for avoiding or mitigating the effects of spatial hole burning.

Unidirectional ring lasers

Ring lasers are configured to ensure that the beam travels unidirectionally, thus circumventing the standing-wave pattern problem and eliminating spatial hole burning \([12, 17]\). The conventional way of ensuring the unidirectional operation is by using an optical isolator \([17]\). An extension of this technique is the monolithic unidirectional ring laser or Miser, a type of ring laser resonator formed by a coated crystal \([18, 19]\). The optical isolator is incorporated into the resonator structure itself through its nonplanar light path and the applied magnetic field. The performance is also improved by reducing the size of the resonator \([19]\), which reduces the number of modes within the gain medium bandwidth. Using such an approach, an SLM laser with an excellent frequency stability of about 40 kHz was demonstrated.

Twisted-mode lasers

In this technique, birefringent elements are introduced at the two ends of the gain medium so that the linear polarisation is split into two orthogonal polarisations that transit through the gain medium. Each linear polarisation component forms a standing-wave pattern in the gain
medium which is $\pi/2$ out of phase with each other, resulting in a constant optical intensity in the laser, thereby suppressing the spatial modulation of gain [20]. For example, SLM operation of a Nd:YAG laser was obtained by placing two quarter-wave plates at each end of the laser rod and Brewster’s plate in front of one of the end mirror [21].

**Microchip lasers**

In some cases, it is possible to achieve SLM in a standing-wave cavity by shortening the cavity length so that the FSR of the cavity becomes larger than the gain bandwidth. Hence, only a single mode exists within the laser bandwidth, as illustrated in Fig. 1.2(a). This is easily accomplished in high gain microchip lasers which have a cavity length of a few mm and hence an FSR of hundreds of GHz [22]. A 730 $\mu$m long Nd:YAG laser demonstrated an SLM operation up to pump powers 40 times the threshold with a linewidth of about 5 kHz [22] but the output power was low.

**Frequency/wavelength selective elements**

Introducing frequency selective elements such as Bragg gratings or Fabry-Perot etalons or diffraction gratings in the cavity [23][24] represents a method for directly discriminating against neighbouring modes. Etalons may act as a mode selector [25], as shown in Fig. 1.2(b). Bragg gratings consisting of the periodic modulation of refractive index in a transparent material, allow a narrow range of wavelengths which satisfy the Bragg’s condition, $\lambda = 2nd$, where $d$ is the grating period. Usually, a combination of frequency selective elements is used in the cavity to achieve SLM [23][24].

---

**Figure 1.2:** Enforcing SLM by (a) reducing laser cavity length (gain bandwidth < FSR) (b) using Fabry-Perot etalon (gain bandwidth > FSR).
Gain medium positioning

Figure 1.3: Reducing spatial hole burning by placing a short length gain medium near one of the cavity mirrors.

Spatial hole burning is reduced if a gain medium of short length is placed near one of the end mirrors \cite{26,27}. This is because all the modes have a node at the end mirror as shown in Fig. 1.3 and are in phase at that location. Since the modes access similar volume and hence compete strongly, the second mode threshold is therefore raised considerably. In contrast, far away from the mirror, modes dephase as given by Eq. ?? and the probability for oscillation is high. Gain media such as heavily doped Nd:YVO$_4$ are well suited to this scheme. For example, SLM outputs has been observed using this technique for pump powers up to about 15 times above threshold \cite{13}. 
1.2 Increasing the power and wavelength range of SLM inversion lasers

The above outlines the methods for achieving SLM that have been demonstrated for specific cases (gain material, wavelength, power, CW or pulsed). However, no particular method is applicable in all cases. Furthermore, the amplitude and frequency stability depends on the precise length of the cavity which becomes a greater challenge to control as laser power increase. As a result, a range of strategies are used to diversify the performance range.

1.2.1 Power

Achieving high power in SLM inversion lasers is challenging, as the laser properties (such as beam quality, linewidth, wavelength tuning range) have to be preserved. It is because these properties are lost during the increase in the output power owing to the thermal lensing or stress effects in the gain medium. Some techniques such as injection locking or seeding overcome thermal and mechanical instabilities in high power SLM CW or pulsed lasers [28–31]. Another approach for power scaling is the use of master oscillator power amplifier (MOPA) systems, typically used for fibre lasers. By implementing large mode area fibres and several SBS suppression techniques, the output power of a single frequency laser using this architecture was scaled to about 0.9 kW [32].

1.2.2 Wavelength range

The spectral range of conventional inversion lasers is extended by wavelength conversion through nonlinear optical processes. These processes involve wave mixing such as second harmonic generation, sum and difference frequency generation, optical parametric generation and amplification or inelastic scattering such as stimulated Raman scattering (SRS) and stimulated Brillouin scattering (SBS) [33–37].

Optical parametric oscillators (OPOs) are a convenient method that uses second-order non-linear interactions to generate two coherent outputs at longer wavelengths (compared to the pump wavelength), thus accessing wide wavelength ranges (from visible, near-infrared to far-infrared, terahertz). The main attractions of SLM OPOs are narrow-linewidth, wide
wavelength tuning range (e.g., from 550 nm to 2830 nm \[38\]) and high output power (30 W of combined signal and idler output power \[39\]). These advantages enabled the wide utilization of SLM OPOs for spectroscopic and LIDAR applications. However, these lasers suffer from a few problems. Firstly, wavelength tuning in OPOs is achieved by altering the temperature or the crystal orientation to change the phase matching properties (a requirement for this nonlinear process) \[34\]. Although wide tuning range is its unique feature, phase-matching requirement complicates the cavity design \[41\]. Secondly, increase in thermal loading of the crystal at high pump powers due to the linear absorption results in beam quality degradation \[42\].

Raman lasers are also an efficient and convenient method to expand the spectral range of conventional lasers. SRS enables pump laser frequency down-conversion, with the energy difference between the pump and the output (Stokes) given by the vibrational mode (associated with the optical phonon) of the medium. Unlike other nonlinear processes, the conversion of a pump photon to a Stokes photon is automatically phase-matched. As adjusting the crystal temperature or angular orientation of the crystal to satisfy phase-matching condition can be avoided in Raman lasers, the cavity design is generally much simpler than in OPOs. The inherent Raman beam cleanup by the SRS process is also advantageous for the output beam quality \[43\]. However, the thermal loading in the crystal due to the inelastic nature of SRS process leads to thermo-optic effects such as thermal lensing, thermally induced birefringence which restricts the power scaling of these lasers \[35\].

### 1.3 Review of SLM Raman lasers

It is clear from Secs. 1.1.3 and 1.2.1 that there is a great diversity of SLM inversion lasers, in terms of both design and performance. However, to date, only a few SLM Raman lasers have been demonstrated and all these lasers \[44-49\], except perhaps in one case \[50\], have been "forced" to run SLM using standard techniques adapted from inversion lasers. Most of these cases are operating in CW regime.

The first reported SLM Raman laser was demonstrated in hydrogen with 8.2 mW maximum output power at 683 nm and 35% maximum conversion efficiency \[44\]. This demonstration
was significant since it is challenging to realize CW operation of linear Raman resonators where optical intensities are low. The difficulty is because SRS is a nonlinear process and high pump intensities are required to achieve the sufficient gain to initiate the process. Therefore, to develop CW Raman lasers, a high finesse cavity that was doubly resonant for the pump and the Stokes field enabled the build up the high intracavity intensities and reduce threshold to milliwatts range. SLM was achieved by the same concept that is used to realize SLM operation in microchip lasers. That is, the mode spacing of the cavity was about 2.9 GHz which was much larger than Raman linewidth of 510 MHz (owing to the pressure of gas cell), thus ensuring only one mode would attain threshold.

With progress in on-chip guided wave structures in the last 10-20 years, the development of microresonators offered high probability to realise Raman lasing due to the small mode volume and high quality factors. A CW Raman laser was demonstrated in a standing-wave silicon waveguide resonator pumped by a single-mode source at 1550 nm [50]. SLM output was observed at the Stokes wavelength of 1686 nm for a few mW of output power. The FSR of the 4.8 cm long waveguide is 0.9 GHz which is much smaller than the Raman linewidth of silicon (105 GHz [51]). This observation is notable because it is the first evidence that a standing-wave Raman laser operated SLM without being forced by any of the standard techniques used in inversion lasers. However, the authors did not specify as to how single-mode was achieved in the Raman laser and therefore, the reason for the observation is still not clear. Diamond has emerged as a promising Raman material for integrated-optics with some important advantages over silicon, owing to its wide spectral transparency range and reduced susceptibility for multi-photon effects. A diamond racetrack microresonator on a silica chip was demonstrated around 2 μm. SLM was achieved in this resonator as the FSR of this cavity (180 GHz) is larger than diamond Raman linewidth (45 GHz) [46]. However, the output power was in μW range.

Recently, the first bulk crystal CW Raman laser operating in SLM regime was demonstrated [48]. A Nd:YVO₄ monolithic self-Raman laser exhibited SLM during operation at temperatures lower than 125 K. The authors explained that monolithic structure and low temperature led to a large cavity mode spacing and narrower Raman linewidth respectively, which resulted in SLM operation. The laser generated 1.36 W maximum output power at
1.4 Thesis motivation and outline

1176 nm from 17.2 W pump power.

In the pulsed regime, intracavity elements have been shown to reduce linewidth \[52\] but SLM has been only achieved to date using injection seeding \[45, 49\]. For example, a single-mode pulsed external cavity methane Raman laser pumped by a narrowband laser was optimized for LIDAR at 1.5 microns by injection seeding \[49\]. A conversion efficiency of 43\% was achieved and the Stokes linewidth was measured to be 210 ± 34 MHz. The suitability of the laser as LIDAR transmitter was verified by detecting hydrogen cyanide. Injection seeding of both the pump at 1064 nm and a third-order Stokes of a pulsed external cavity barium nitrate Raman laser at 1559 nm achieved the required linewidth narrowing and mode stability for detecting carbon dioxide \[45\].

From the above, it is apparent that SLM development in these systems is still very immature, despite the fact that it has been 50 years since the first Raman lasers. A major reason for this slow development could be related to the more complex gain properties of Raman media and that this laser class has received generally much less development than inversion lasers. Although theory for SRS \[53, 54\] and laser dynamics have been well studied \[55–58\], to the author’s knowledge there have been no detailed studies on Raman gain characteristics with respect to SLM operation. Therefore, the aim of the thesis is to investigate the characteristic gain properties of the Raman medium, to study mechanisms which could reduce the mode competition in a standing-wave Raman cavities and to understand principles for SLM Raman cavity design.

1.4 Thesis motivation and outline

Recently, conceptually simple standing-wave Raman lasers operating in the CW regime have been demonstrated using diamond as the Raman medium. These systems are end-pumped lasers comprising of only the gain medium and two mirrors, and as detailed in the following Chapter, are capable of high power and efficiency. It is proposed that this system comprises an ideal system for investigating SLM operation. Indeed there has already been an indication that SLM is possible in such a system without the use of the methods outlined in Section 1.1.3.
Longitudinal modes
Δω = 45 GHz
Raman gain profile
FSR (Δω_0) = 1.68 GHz

Figure 1.4: The diamond gain profile and cavity longitudinal modes for a 78 mm long standing-wave cavity (Gain bandwidth (Δω_R) > FSR (Δω_l)).

An external cavity Raman laser based on a bulk single crystal diamond was pumped by a narrow-linewidth pump at 1064 nm and was found to provide SLM operation under certain conditions at output powers closer to the threshold [59]. However, it was reported to be stable only for about 100 ms [60]. Although the SLM was observed near threshold, the result is significant because the diamond Raman gain linewidth is 45 GHz and the mode spacing of 78 mm long cavity is 1.68 GHz. It implies that the diamond gain bandwidth supports about 26 longitudinal modes to oscillate in the cavity (as shown in Fig. 1.4). Since this thesis is aimed at developing SLM Raman lasers, the work on single-mode pumped diamond Raman laser demonstrated in [59] is relevant as it is the first evidence of SLM in a free-running standing-wave Raman laser and thus forms important background for this thesis.

The overall aim of the thesis is to demonstrate SLM operation in a standing-wave crystalline Raman laser. Chapter 2 proposes a novel conceptual approach to SLM Raman lasers that takes advantage of the unique properties of Raman gain. It introduces the selected cavity architecture and Raman materials (potassium yttrium tungstate (KYW) and diamond) used in this thesis. Chapter 3 details the investigation of KYW pumped by a broadband and narrow-linewidth laser sources. This chapter includes the observation of new spectral lines corresponding to lesser known Raman modes in the laser spectrum and simulations to understand the impact of these Raman modes on the laser performance. Chapter 4 presents the experimental results of a CW diamond Raman laser operating in SLM. It studies the reasons for the transition to multimode operation at high powers of a Raman laser and provides a
solution for achieving SLM operation. Chapter 5 explores the feasibility of expanding the spectral range of a CW diamond Raman lasers and the adaptability of the chosen cavity architecture in generating higher-order Stokes components and assesses its suitability for remote sensing applications. Chapter 6 describes the active stabilization of the cavity length of a CW diamond Raman laser using the Hänsch-Couillaud locking technique with the aim to increase mode stability and SLM power range. It details the implementation of the locking method by exploiting the stress-induced birefringence and polarisation dependent Raman gain properties of diamond. Lastly, Chapter 7 presents the conclusions of this thesis and outlines the future works.
This chapter explores the basic theory of stimulated Raman gain in the context of understanding the factors that determine mode competition and hence SLM stability. The theory is used to develop key concepts and the experimental approach to achieve high-power SLM operation in a standing-wave crystalline Raman laser cavity. Furthermore, the reasons for selecting the type of the Raman cavity configuration and the crystal are explained.

2.1 Theoretical considerations

In order to understand mode competition and hence SLM Raman laser design, the nature of Raman gain is compared and contrasted with inversion lasers. This includes a study of the line broadening mechanism for Raman gain and consideration of spatial hole burning in Raman lasers.
2.1.1 Gain saturation and mode competition in Raman lasers

The gain mechanism in Raman lasers relies on the coherent coupling of the pump and the Stokes field (amplitudes \( E_p, E_s \)) via the phonon field \((Q')\), as given by the following optical and phonon fields (propagating along \( x \) direction) involved in the laser [61].

\[
\begin{align*}
\frac{\partial E_s}{\partial x} - \frac{1}{v_s} \frac{\partial E_s}{\partial t} &= \frac{ig_s}{n_s} \omega_s E_p Q'^* , \\
\frac{\partial E_p}{\partial x} - \frac{1}{v_p} \frac{\partial E_p}{\partial t} &= \frac{ig_s}{n_p} \omega_p E_s Q' , \\
\frac{\partial Q'}{\partial t} + \frac{1}{T_2} Q' &= \frac{icn_s n_p \epsilon_0}{4\omega_s T_2} E_p E_s^* ,
\end{align*}
\]  

(2.1)  
(2.2)  
(2.3)

where subscripts \( s \) and \( p \) indicate Stokes and pump field respectively, \( \omega \) is the angular frequency, \( v \) is the group velocity, \( n \) is the refractive index, \( \epsilon_0 \) is the permittivity of free space, \( c \) is the speed of light, \( g_s \) is the steady-state Raman gain coefficient and \( T_2 \) is the dephasing time of the phonon field. Here, the SRS process is considered to be in the steady-state regime as the pump period is greater than the dephasing time \((T_2)\) of the optical phonons (of the order of tens of picoseconds for most crystals [36]). In the steady-state regime, the phonon term is eliminated from the Eqs. 2.1 and 2.2 of the Stokes and the pump fields to obtain their field intensities as a function of space [61],

\[
\begin{align*}
\frac{\partial I_s}{\partial x} &= g_s I_p I_s - \alpha_s I_s , \\
\frac{\partial I_p}{\partial x} &= - g_s \frac{\omega_p}{\omega_s} I_p I_s - \alpha_p I_p ,
\end{align*}
\]  

(2.4)  
(2.5)

where, \( \alpha_{s,p} \) are the absorption loss coefficients for the Stokes and pump fields. The principal differences in the gain mechanism between Raman and inversion lasers are as follows. Although some power is deposited in the medium in the form of phonon excitation, the amplification of Stokes modes originates from the power transfer from the pump via inelastic scattering by the phonon field. It is clear from Eq. 2.4 that the optical gain in the Raman gain medium is proportional to the product of the pump intensity and medium’s Raman gain coefficient. By contrast, in inversion lasers, the optical amplification is achieved by stimulated emission from energy stored in the medium as an inverted population.

In a standing-wave Raman cavity, the amplification of longitudinal modes of the cavity depends on how the gain saturates and hence the resulting mode competition. Eqs. 2.4 and 2.5 show that the Stokes field experiences exponential gain at the expense of the incident
pump above the Raman lasing threshold. The depletion of the pump by a particular Stokes longitudinal mode leads to a reduction of gain available for other modes, implying that the pump depletion is the mechanism for gain saturation.

The gain profile saturates differently depending on the pump linewidth. A standing-wave Raman laser pumped by a source having a narrower linewidth than the Raman gain bandwidth of the crystal is considered first. In this case, the pump availability is restricted to fewer modes than that supported by the Raman gain medium. The mode at the gain centre having the lowest lasing threshold would start depleting the pump, thereby suppressing the gain for neighbouring longitudinal modes and preventing them from lasing. Thus, the magnitude of the gain profile is reduced by the mode at the gain centre but the shape of the gain profile remains same (see Fig. 2.1(a)), implying that the gain saturation is homogeneous. This situation corresponds to the mode competition characteristics of homogeneously broadened inversion lasers. A similar conclusion was made for the Raman fiber laser reported by Babinet et al. [62]. They investigated the Raman gain profile and gain saturation mechanism extensively in a phosphosilicate fibre at high pump and Stokes powers. In that case, the pump linewidth was narrower than the Raman gain bandwidth and the authors found that the saturation of the Raman gain was homogeneous.

For the case of a broadband pump source (having a larger linewidth than the Raman gain bandwidth), the pump is depleted by the lowest threshold Stokes mode (at the gain centre). However, as pump frequencies separated from the Stokes by Raman frequency (within the full width of the Raman transition) are coupled, gain saturation leads to a hole in the centre of the pump spectrum of the order of full-width at half maximum (FWHM) of the Raman gain spectrum, as shown in Fig. 2.1(c). Mode competition is therefore only between adjacent longitudinal modes with approximately $\Delta \omega_R$. Outside this range, modes may attain threshold as pump power is increased leading to multi-longitudinal mode operation. Hence, mode competition is weakened away from the gain centre. The spectral hole burning in the pump profile is analogous to spectral hole burning in the populations of an inhomogeneously broadened inversion medium [63].
Figure 2.1: Gain saturation depending on the pump linewidth ($\Delta \omega_p$) and Raman linewidth ($\Delta \omega_R$). (a) Narrowband pumping, (c) broadband pumping, (b) and (d) unsaturated gain profile for narrowband and broadband pumping, respectively.

It is evident from the above discussion that the nature of gain saturation and mode competition of Raman lasers is dependent on the ratio of the pump and Raman linewidths. In principle, it is possible for media to have an inhomogeneously broadened Raman frequency spectrum or medium. For example, a medium may have inhomogeneities present where the local Raman frequency is shifted due to stress or defects. Note that the gain saturation for such a medium does not follow the same rules as for inversion lasers. Since the gain is proportional to pump intensity, gain saturates homogeneously (for a narrowband pump) whether the Raman frequency spectrum is homogeneously or inhomogeneously broadened.

### 2.1.2 Spatial hole burning free nature of Raman gain

In standing-wave inversion lasers, mode competition provided by the homogeneous broadening is reduced by spatial hole burning, which acts to reduce the threshold for multimode
operation. Therefore, it is important to examine the possibility of an analogous mechanism to “spatial hole burning” in standing-wave Raman lasers. The fact that there is no energy storage in the Raman medium makes the concept of spatial hole burning not readily applicable to Raman lasers.

In the case of an inversion laser, the spatial variation of gain is $\pi/2$ out of phase with the cavity standing-wave. Due to the availability of gain in the form of inverted population at the nodes of the standing-wave, an adjacent mode can utilize this gain to oscillate in the cavity. The pump field is depleted exponentially according to the relation for absorption, $I = I_0 e^{-\sigma_{ou} N_0 x}$, where $\sigma_{ou}$ and $N_0$ are the absorption cross-section and ground state population, respectively [12]. Since the pump field provides optical gain in a Raman laser, it is necessary to investigate whether undepleted pump is available at the nodes of the standing-wave pattern of the first longitudinal mode.

Consider a single-pass end-pumped Raman cavity where the Stokes field forms a standing-wave mode pattern between the cavity mirrors. Based on the Eqs. 2.4 and 2.5, the pump is exponentially depleted by the intense Stokes field at the antinodes, which implies that there is pump field at the nodes. However, in this case, the pump transits the gain medium along the axis of the cavity and is a monotonically decreasing function. Hence, there is no enhancement of gain at the nodes of the standing-wave cavity that would preferentially excite a second mode. This lack of spatial modulation of the gain signifies the absence of a mechanism to reduce mode competition, unlike the case of spatial hole burning in inversion lasers. Fig. 2.2 illustrates the principal difference between the spatial modulation of the gain and pump depletion in inversion and Raman lasers. By the same arguments, there is no spatial modulation of gain for a double-pass pumped Raman laser.

It is interesting to consider whether it would be more challenging to obtain SLM operation in side-pumped Raman lasers [64]. In this case, there is expected to be an undepleted pump field at the nodes of the Stokes mode which is available to excite a neighbouring mode of the cavity. In other words, side-pumping is anticipated to provide a spatial depletion of gain similar to that of spatial hole burning in inversion lasers.
Figure 2.2: Difference between the gain mechanisms for inversion and Raman lasers. In the inversion laser, the pump exponentially depletes in the cavity and the gain is available at the nodes for other modes. In Raman lasers, there is no energy storage and pump and optical gain is moving in the cavity such that there is no spatial depletion.

2.2 Experimental approach

The above arguments provide a prescription for obtaining SLM operation even in the case $\Delta \omega_R \approx c/2l$. A pump source having a narrower linewidth than the Raman gain profile is essential to produce homogeneous gain saturation. Furthermore, the Raman cavity should be end-pumped to utilize the spatial hole burning free nature of the Raman gain medium. In addition to these conditions, it is important to maintain the Stokes transverse mode to the fundamental transverse mode TEM$_{00}$ so that higher-order transverse modes cannot access the gain.
2.2 Experimental approach

2.2.1 Cavity design

There are two main classifications of standing-wave cavity configurations for realizing Raman lasers – intracavity configuration and external cavity configuration. In an intracavity configuration, the pump source and Raman gain medium are placed in the same resonator with the mirror reflectivities chosen to resonate the fundamental as well as the first Stokes. This enables high intracavity fundamental intensity, which helps in achieving sufficient Raman gain for Stokes lasing and is thus, favored for low threshold operations [65–67]. In an external cavity configuration, the Raman cavity is separated from the pump resonator [68, 69]. This architecture makes the CW operation challenging because small mode sizes and high pump intensities are required to reach the SRS threshold.

![Diagram of cavity configurations](image)

Figure 2.3: Doubly resonant and singly resonant external cavity configurations. Green solid line and dashed line indicate single-pass pumping and double-pass pumping, respectively.

The external cavity Raman laser provides several important practical advantages over an intracavity configuration. The mutual separation of the pump and Raman cavities avoids interactive issues that would otherwise complicate experiments. Thus, pump laser characteristics such as power, frequency behaviour and spatial properties, can be controlled independently of the Raman laser response. This configuration also relaxes cavity mirror specifications and the associated increase in the laser-induced damage threshold. The beam quality management is easier at higher powers because the complexity of managing thermal...
lensing and the stability of the laser mode, is reduced. Therefore, the external cavity configuration is selected for the Raman lasers presented in this thesis.

An external cavity may be chosen to be resonant at either or both the Stokes and pump wavelengths. The former is referred to as singly resonant cavity and the latter is a doubly resonant cavity. Owing to the resonance of both the optical fields, the Raman lasing threshold is considerably reduced for a doubly resonant configuration. However, doubly resonant cavities are restricted in practice to single frequency pumping. Thus, a singly resonant external cavity configuration is more adaptable for investigations in this thesis as it allows investigation with a range of pump bandwidths.

### 2.2.2 Choice of Raman gain crystals

<table>
<thead>
<tr>
<th>Properties</th>
<th>Diamond</th>
<th>KGW</th>
<th>Ba(NO$_3$)$_2$</th>
<th>KYW [70, 71]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crystal structure</td>
<td>cubic</td>
<td>monoclinic</td>
<td>cubic</td>
<td>monoclinic</td>
</tr>
<tr>
<td>Raman shift (cm$^{-1}$) at room temperature</td>
<td>1332.3</td>
<td>901</td>
<td>1047.3</td>
<td>905</td>
</tr>
<tr>
<td></td>
<td></td>
<td>768</td>
<td></td>
<td>765</td>
</tr>
<tr>
<td>Raman gain coefficient</td>
<td>10 [72]</td>
<td>3.3 (901)$^a$</td>
<td>11</td>
<td>3.6 (905)</td>
</tr>
<tr>
<td>(cm/GW)@1064 nm pump wavelength</td>
<td></td>
<td>4.4 (768)</td>
<td></td>
<td>3.6 (765)</td>
</tr>
<tr>
<td>Raman linewidth at room temperature</td>
<td>1.5</td>
<td>7.8</td>
<td>0.4</td>
<td>6-8</td>
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<td>FWHM (cm$^{-1}$)</td>
<td>5.9</td>
<td></td>
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<td>Thermal conductivity at room temperature</td>
<td>2000</td>
<td>$k_a=2.6$</td>
<td>1.2</td>
<td>3.3$^b$</td>
</tr>
<tr>
<td>(W/m.K)</td>
<td></td>
<td>$k_b=3.8$</td>
<td></td>
<td></td>
</tr>
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<td></td>
<td></td>
<td>$k_c=3.4$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Thermal expansion coefficient</td>
<td>1.1</td>
<td>1.6-8.5 [73]</td>
<td>13 [74]</td>
<td>2-8.5 [73]</td>
</tr>
<tr>
<td>(10$^{-6}$/ K)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$dn/dT$ at room temperature (10$^{-6}$ K$^{-1}$)</td>
<td>15</td>
<td>0.4</td>
<td>20</td>
<td>0.4</td>
</tr>
<tr>
<td>Optical transparency (µm)</td>
<td>0.23 - 100</td>
<td>0.3 - 5</td>
<td>0.3 - 1.8</td>
<td>0.34 - 5.5</td>
</tr>
</tbody>
</table>

$^a$Raman Gain coefficient for different Raman modes. $^b$averaged over the three principal directions.
2.2 Experimental approach

Selecting the optimal crystal for CW operation requires consideration of several parameters. Table 2.1 lists several crystals and their properties which have been commonly used to realize Raman lasers either in intracavity or external cavity configurations. Two parameters that are particularly relevant for CW operation are the Raman gain and the thermal conductivity. It is desirable to have crystals with high Raman gain to achieve low Raman lasing threshold and high thermal conductivity to mitigate any thermal effects in the media. This heating is due to the quantum defect of the SRS process and impurity absorption of the resonated Stokes. From the table, it is clear that diamond and barium nitrate have the highest Raman gain, which may be one indicator as to why these crystals are the only ones so far to demonstrate CW output in an external cavity configuration [68, 75]. However, thermal lensing in the barium nitrate crystal, arising from the poor thermal conductivity, limited the conversion efficiency to 5% and maximum achievable Stokes output power to 164 mW [68]. A CW external cavity Raman laser based on diamond was able to scale output power up to 381 W, without major thermal problems in the crystal, due to its superior thermal conductivity [75]. Diamond, therefore, was chosen as the primary material candidate for investigating SLM Raman lasers.

Despite the suitable parameters of diamond, a preliminary demonstration of an external cavity diamond Raman laser, pumped by a single-mode laser source did not show SLM operation at Stokes powers above threshold [59]. As the first step in realizing SLM in Raman lasers, it is helpful to determine whether the material properties of diamond contribute to the multimode behaviour. Therefore, a crystal with contrasting material properties is investigated, to compare with diamond. Potassium yttrium tungstate (KY(WO$_4$)$_2$, KYW), was chosen due to its moderate Raman gain and better thermal conductivity compared to barium nitrate. It has been successfully used for expanding the spectral coverage of inversion lasers in intracavity configurations [76-78].

**Potassium yttrium tungstate (KYW)**

Potassium yttrium tungstate (KY(WO$_4$)$_2$, KYW) is a prominent member of the class of double metal tungstates that has been studied extensively in CW and pulsed intracavity [77, 79] and pulsed external cavity configurations [80, 81]. It has a moderate Raman gain coefficient (3.6 cm/GW) and thermal conductivity, as shown by comparison in Table 2.1, as well as a
high optical damage threshold and robust mechanical properties. It is comparatively easy to dope with rare-earth ions such as Nd$^{3+}$, Er$^{3+}$, which enabled their use as gain medium for self-Raman lasers [67, 80]. This class of Raman crystal also has the practical advantage that multiple phonon modes have high Raman gain coefficients and are selectable by laser polarization (see also below) [70, 82].

Figure 2.4: Crystal structure of KYW; K = blue, Y = green, W = brown and O = red [71, 83].

Double metal tungstates have the general formula, $A^{+1}B^{+3}(WO_4)_2$, where A = alkali metal cation and B = trivalent metal or rare-earth cation. It has a monoclinic crystal structure (C2/c and I2/c space groups) and is, therefore, a biaxial crystal. In this structure, oxygen atoms form three classes of polyhedra in KYW — it bonds with tungsten (W), potassium (K) and yttrium (Y) atoms to form WO$_6$, KO$_{12}$ and YO$_8$ respectively [83]. Due to the various vibration modes, the KYW Raman spectra contain multiple Raman lines, as shown in Fig. 2.5. The two strong phonon modes at 765 cm$^{-1}$ and 905 cm$^{-1}$ correspond to the symmetric stretching vibrations of the WOOW and WO molecular groups, respectively [84–86]. These modes can be accessed separately by changing the polarisation state of the pump radiation with respect to the crystallographic axes of the crystal.

The directions of the three orthogonal refractive index axes $N_m$, $N_g$ and $N_p$ with respect to the crystallographic axis in the I2/c space group are shown in Fig. 2.6. Each of these crystallo-optic axes have been labelled based on the refractive index axes which are $n_m$, $n_g$ and $n_p$, respectively, with $n_g > n_m > n_p$. In Raman laser experiments, the crystal is usually cut
2.2 Experimental approach

Figure 2.5: Raman spectrum of KYW for two excitation geometries [86].

for propagations along the $N_p$ axis to provide high gain for the two primary modes. If the pump polarisation is oriented along $N_g$ axis which is about 17.5° from the crystallographic $c$ axis, then 765 cm$^{-1}$ phonon mode is accessed. When the pump polarisation is aligned along $N_m$ axis which makes an angle of 58° with the crystallographic $a$ axis, the 905 cm$^{-1}$ phonon mode has the highest gain. $N_p$ axis is parallel to the crystallographic $b$ axis.

Figure 2.6: The crystallo-optic axes with respect to the crystallographic axes [70].

Diamond

SRS in diamond was observed as early in 1963 [87], but the cost and poor availability of good optical quality diamond crystals limited its use in optical applications only until recently. The advances in chemical vapour deposition (CVD) technology led to the development of cheaper, and low defect, low birefringence, high purity and high optical quality diamond, invoking interest in its potential use in optics and lasers [88].
Diamond has a broad transmission range, high Raman gain, high damage threshold, excellent thermal conductivity and low thermal expansion coefficient (see Table 2.1). Each of these properties makes diamond an attractive material for Raman frequency conversion. The high gain of diamond lowers the Raman lasing threshold and the required crystal length. Furthermore, the superior thermal conductivity and low thermal expansion reduce thermal effects in the crystal [61]. Combining the thermal properties with high damage threshold, diamond is an outstanding candidate for power scaling of crystalline Raman lasers [75]. The broad transmission window offers a large spectral coverage. Moreover, the large Raman shift of 1332 cm\(^{-1}\) allows for generating wavelengths further from the fundamental in a single Stokes shift.

![Figure 2.7: The two interpenetrating FCC lattices (red and blue) and the direction of the Raman vibrational mode along [111] is indicated by blue arrows [60].](image)

Diamond belongs to the face-centered cubic (FCC) structure (\(O_h^7\) –Fd\(\bar{3}\)m space group), where each carbon atom is linked to four other carbon atoms through a covalent bond in regular tetrahedrons. The lattice consists of two interpenetrating FCC lattices displaced of the cubic cell by one-quarter of the length of the diagonal, as shown in Fig. 2.7. The crystallographic axis and dimension of the single crystal diamond used in this thesis are illustrated in Fig. 2.8. The diamond is grown along [100] by the CVD process. The [110] axis is selected
as the propagation direction [69, 75, 89].

Figure 2.8: The dimensions and crystallographic orientation of diamond. (a) Side view and (b) end view [90].

![Diagram](image)

Figure 2.9: Raman spectrum of diamond.

The first-order Raman scattering corresponds to the vibrational modes that involve the movement of two FCC lattices along the direction of the linking carbon-carbon bond. At the phonon dispersion zone-centre, the two transverse and longitudinal vibrational modes converge to result in a triply degenerate Raman mode. As shown in the Fig. 2.9, the Raman spectrum contains a single phonon mode at 1332.3 cm$^{-1}$ at room temperature. The Raman gain coefficient of diamond is highest when the pump polarisation is oriented along <111> direction, which is also the direction of the carbon-carbon bond and the induced vibrations [61]. This was also experimentally verified by measuring the lasing threshold and efficiencies of an
external cavity diamond Raman laser for different orientations of the pump polarisation [89]. This axis is oriented at an angle of 54.7° from the <001> axis.

In this thesis, optical grade single crystal CVD diamond (Raman-grade or "ultra-low birefringence") manufactured by Element Six Ltd. was used. It had a nitrogen impurity of approximately 20-40 ppb and an absorption coefficient in the range 0.001-0.004 cm\(^{-1}\) at 1064 nm [90]. The birefringence was typically < 10\(^{-5}\) (in the visible and near-infrared range) perpendicular to the growth direction [91]. The diamond crystals from the same batch were used to investigate the effect of stress-induced birefringence on the CW external cavity Raman laser performance [91]. It was found that linear birefringence (as well as small variations in coating and surface quality) determined the Raman lasing threshold. The output Stokes polarization was seen to be aligned to the birefringence axis which provides the maximum gain. An analytical modelling was developed to study the effect of resonator and crystal parameters on the efficiency of a quasi-CW external cavity diamond Raman laser [92]. It showed that the absorption loss affect the efficiency directly and increase the lasing threshold. However, a crystal from the same batch was used to scale the output power of a quasi-CW diamond Raman laser to 381 W with 61% conversion efficiency [75], implying that the absorption loss is low enough to not impact the laser efficiency severely.

2.3 Conclusion

This chapter outlined the key concepts and the experimental approach to achieve the aim of the thesis. The fundamental difference in gain dynamics of conventional inversion lasers and Raman lasers was examined. It was found that the gain saturation and mode competition is dependent on the pump and Raman linewidth. As there is no optical energy stored in the Raman cavity and the gain arises from coherent coupling between the pump and Stokes fields via a phonon field, there is no mechanism analogous to "spatial hole burning" in narrow-linewidth end-pumped Raman lasers.

The external cavity configuration was chosen to demonstrate the SLM Raman laser for its advantages in design simplicity, improved thermal lens management and avoided interactions with the pump gain medium. The crystals —KYW and diamond were selected as two
interesting candidates for investigating the SLM properties of Raman lasers.
This chapter describes the development of a CW external cavity Raman laser (ECRL) based on KYW with the aim to provide an initial study of factors influencing the output spectrum and to determine whether the material properties of diamond were responsible for the multi-mode behaviour observed previously for the diamond ECRL of ref. [60]. The chosen design configuration is a cavity that is only resonant at the Stokes wavelength to reduce complexity and allow flexibility to pump with a multi-longitudinal mode laser. Since this is the first reported attempt of a CW ECRL using a tungstate crystal, it was deemed prudent to first use a high power pump source to ensure the Raman lasing threshold is exceeded.

The basic Raman laser characteristics of lasing threshold, slope efficiency, spectrum are studied. The complex Raman spectrum of KYW, which is found to play a critical role in
determining the output spectrum and laser efficiency, is characterized and modelled to understand its role on the laser performance.

In the second part of the Chapter, an attempt to attain SLM is made by using a narrow bandwidth CW pump. The longitudinal mode spectrum was investigated.

### 3.1 Multi-longitudinal mode pumping

#### 3.1.1 ECRL cavity design

As discussed in chapter 2, the external cavity configuration was selected for realizing the Raman lasers described in this thesis. The main challenge to demonstrate the Raman laser in the CW regime using this configuration is to achieve a moderate Raman lasing threshold. The threshold for Raman lasing for single pass pumping is given by

\[ P_{th} = \frac{T + 2.\alpha.l_R}{2.g.l_R} \pi.w_p(0)^2 \]  

where \( \alpha, l_R, g, T \) and \( w_p(0) \) are loss coefficient (crystal absorption and scattering), crystal length, Raman gain coefficient, output coupler transmittance and pump spot radius respectively. According to this equation, Raman lasing is achieved when the round-trip gain \( (g.l_R) \) exceeds the cavity losses \( (T + 2.\alpha.l_R) \). It is evident from the above relation that the tight focusing of the pump significantly reduces the threshold. To provide simpler alignment and reduce cavity loss, it is an advantage to reduce the number of surfaces in the cavity. Due to these reasons, a concentric 2-mirror resonator is favoured. Although a folded cavity also provides a method for obtaining small mode sizes, the minimization of cavity losses is made more difficult due to the additional mirrors.

In a concentric cavity, the cavity length is equal to the sum of the radii of curvature (ROC) of the mirrors. The stability condition of a linear cavity configuration without a thermal lens in the resonator is given by \( 0 \leq g_1 g_2 \leq 1 \), where \( g_1 = (1 - L/R_1) \), \( g_2 = (1 - L/R_2) \), and \( L \) and \( R_1, R_2 \) are resonator length and ROC of the mirrors.

Since the cavity length is approximately twice the mirror ROC, both \( g_1 \) and \( g_2 \) are close to \(-1\), the configuration is therefore near the limit of the cavity stability condition. So, the
cavity length is reduced slightly to ensure the \( g_1 \) and \( g_2 \) parameters are in the stability region. This can be referred to as a "near-concentric" configuration. Being near to the stability limit, the cavity is very sensitive to alignment and changes in length.

![Diagram](image)

Figure 3.1: (a). Schematic of concentric cavity configuration. (b). Stability diagram for two mirrors.

Due to the quantum defect of the SRS process, heat is unavoidably deposited into the Raman crystal and thus thermal lensing is an important factor to consider in the design of the resonator. With a thermal lens of focal length \( f \) placed in the centre of a linear resonator with flat mirrors, the stability parameters \( (g_1 \text{ and } g_2) \) are modified as \( g = g_1 = g_2 = 1 - \frac{L}{2f} \) [93]. The equation implies that the cavity is destabilized if the focal length of thermal lens reaches \( f = L/4 \). To alleviate this thermal effect, use of shorter radius of curvature (ROC) mirrors (and hence shorter cavity) is recommended [60].

For a cavity non-resonant at the pump frequency, the cavity is made highly reflective for the Stokes to provide a modest threshold. The Raman crystal facets are also anti-reflection (AR) coated at the pump and the first Stokes wavelengths to minimize reflection losses. To avoid second-order Stokes lasing in the cavity from the high intracavity intensity of the first Stokes, it is desirable that the reflectivity of the input and output couplers for the second Stokes wavelength are as low as possible.
Another important design consideration is the matching of the pump waist size to the Stokes mode in the cavity [92]. The desired cavity mode is the fundamental Gaussian mode (TEM$_{00}$) and therefore, to suppress higher-order modes, the pump spot size in the Raman medium should not be larger than the radius of the fundamental Stokes mode. This is achieved by focusing optics placed before the input coupler (such as a plano-convex lens). Fig. 3.2 illustrates the near-concentric Raman cavity configuration. It has to be noted that there is a limit to the focusing of the pump [94] and the optimal ratio of Stokes waist radius to the pump waist radius should be about 1.5 to attain minimum threshold. This is the optimum point which provides a good spatial overlap between pump and the Stokes modes and high pump intensities to increase gain [92, 94].

![Figure 3.2: Schematic of near-concentric Raman cavity configuration](image)\[\Delta L = 2R_1 (= R_2) - L.\]

The mode size of the pump is smaller than the mode size of Stokes.

**KYW ECRL cavity design**

Input and output mirrors of 50 mm ROC were selected as it enabled a short resonator of length 120 mm. The calculated Stokes mode size was 63 µm. The lasing threshold ($P_{th}$) for the double-pass pumped Raman laser was estimated by modifying the Eq. 3.1 with a factor of two in the denominator to account for double pass pumping. Since the loss coefficient, $\alpha$ for the crystal was unknown, a loss value of $\alpha = 0.017\%$ cm$^{-1}$. The published Raman gain coefficients of the two primary Raman modes of KYW are both 3.6 cm/GW [71]. These two modes enable conversion from 1064 nm to 1158 nm and 1177 nm via the 765 cm$^{-1}$ and 905 cm$^{-1}$ Raman modes respectively. Using these values, the calculated threshold for the 765 cm$^{-1}$ mode and a 0.2% output coupler transmittance is 10 W for a pump spot radius of 30 µm in the crystal. As the Stokes mode size at 1177 nm in the same cavity length and the
output coupling used in the experiment are slightly greater than for 1158 nm, the threshold of
the KYW Raman laser emitting at 1177 nm corresponding the 905 cm$^{-1}$ mode is estimated
to be 17 W for the OC transmittance of 0.4%. Note that the reflection losses at the end faces
of the crystal were not accounted for in the above equation and therefore, the experimental
threshold is anticipated to be higher.

The pump focussing lens is selected to ensure a pump mode spot smaller than the Stokes at
the midpoint of the KYW crystal.

### 3.1.2 Experimental arrangement

**Raman laser configuration**

![Figure 3.3: Schematic diagram of KYW ECRL](image)

A polarized Nd:YAG laser operating at 1064 nm with 200 W peak power in 250 µs pulses,
operating at 40 Hz is used as the pump. The beam profile was near Gaussian with an $M^2 < 1.5$. The output linewidth was measured to be 0.09 nm (ie., much smaller than the Raman linewidht of 0.8 nm). A 50 mm long KYW crystal was placed at the midpoint of a 120 mm long cavity formed by two 50 mm radius-of-curvature mirrors as shown in Fig. 3.3. The end faces of the KYW crystal were coated with broadband AR coatings from 1000 to 1200 nm to reduce reflection losses. A half-wave plate was used to align the pump polarisation along either the $N_g$ or $N_m$ crystallo-optic axis, as illustrated in Fig. 3.4.

The pump focusing lens was selected to achieve the aforementioned overlap with the Stokes mode at the midpoint of the KYW crystal. The collimated pump beam with 800 µm radius ($1/e^2$) was tightly focussed into the crystal using a singlet lens of 50 mm focal length to produce a 23 µm radius in the KYW crystal. The input coupler was selected for high
transmittance ($T = 96.8\%$) at 1064 nm and high reflectance ($R > 99.9\%$) at the first Stokes (1158 nm). Two output couplers OC1 and OC2 were used having 99.89\% and 98.6\% reflectance at 1158 nm, respectively. A thermal power sensor was used to measure the Stokes output power. The laser spectrum was recorded using a spectrometer (Ocean Optics NIR 512), and a CCD camera (WinCamD, DataRay Inc.) was employed to measure the spatial beam profile.

Figure 3.5: Reflectance spectra of the input coupler (IC) and the two output couplers (OC1 and OC2) used in the KYW Raman laser. The measurement uncertainty is 0.2\% and 0.5\% for measurements of OC1 and OC2, respectively. The vertical dashed lines indicate the pump and Stokes wavelengths observed experimentally (described in Sec 3.2.4).
3.1.3 Thermal considerations

Since the phonon dephasing time of KYW ($T_2 = 1.5 \text{ ps}$ [95]) is much shorter than the pulse duration of the pump (250 $\mu$s), the Raman process falls under the steady-state Raman regime. In order to understand the evolution of the thermal effects in the crystal and its effect on the laser performance, the time constant for the establishment of steady-state thermal gradients in the crystal is calculated using $\tau = \frac{\omega_p^2 \rho C_p}{\kappa}$ which is the relation for a circular laser beam in a rectangular slab. Here $\omega_p$, $\rho$, $C_p$ and $\kappa$ are the pump spot size, material density, specific heat and thermal conductivity respectively [96]. Using values given in Table 3.1, $\tau \approx 500 \mu$s for a pump spot radius of 23 $\mu$m [96, 97]. Therefore, for 250 $\mu$s pump pulses the KYW Raman laser is operating in a quasi-CW regime and some time resolved behaviour may be expected during within each pulse due to the evolution of a thermal lens.

Gaussian beam calculations (e.g., using LASCAD) show that the thermal lens strength needed to destabilize the cavity is 143 diopters and above (corresponding to a focal length of less than 7 mm). Even though a slight reduction in the cavity length compensates for the thermal lens focal length, the Stokes mode size in the center of the crystal decreases which will ultimately affect the efficiency of pump conversion and output beam quality.

The thermally-induced lens strength $f^{-1}$ for an isotropic crystal is given by the following equation,

$$\frac{1}{f} = \frac{P_{\text{dep}}}{2\pi K_c \omega_p^2} \left[ \frac{dn}{dt} + (n_0 - 1)(\nu + 1)\alpha_T + 2n_0^3 \alpha_T C'_{x,y} \right],$$  \hspace{1cm} (3.2)

where $P_{\text{dep}}$, $\omega_p$, $\frac{dn}{dt}$, $\nu$, $\alpha_T$, $n_0$, $K_c$ and $C'_{x,y}$ are the heat deposited in the crystal, pump spot size, thermo-optic coefficient, Poisson ratio, thermal expansion coefficient, principal refractive index, thermal conductivity and photoelastic coefficient, respectively [98]. The first term in the equation indicates the temperature dependence of the refractive index through the thermo-optic coefficient. The second term quantifies the end-bulging effect of the thermal lens [99]. Since KYW is anisotropic, the Poisson ratio used in the equation (0.3; see Table 3.1) is an estimate based on a direction-averaged value [99]. The third term corresponds to the photo-elastic effect or the thermal strain induced variation in refractive index [99]. The contribution from this third term is negligible compared to the thermo-optic and end-face bulging terms [100]. Hence, the required power deposited in the KYW to establish a thermal
Continuous-wave external cavity KYW Raman laser

A lens of $f = 7 \text{ mm}$ is found to be 0.3 W. Given the quantum defect ($\lambda_s/\lambda_p - 1$) is 0.088 for $\lambda_p = 1064 \text{ nm}$ and $\lambda_s = 1158 \text{ nm}$, and assuming negligible impurity absorption, this corresponds to 3 W of Stokes power.

<table>
<thead>
<tr>
<th>Properties</th>
<th>KYW [71,101]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density ($\text{g/cm}^3$)</td>
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</tr>
<tr>
<td>Specific heat capacity ($\text{J/gm.K}$)</td>
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<tr>
<td>Thermal conductivity ($\text{W/cm.K}$)</td>
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<td>Refractive index at 1064 nm</td>
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<tr>
<td>Poisson ratio</td>
<td>0.3</td>
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<tr>
<td>Thermo-optic coefficient ($\frac{dn}{dt}$) (K$^{-1}$)</td>
<td>$-12.4 \times 10^{-6}$</td>
</tr>
<tr>
<td>Thermal expansion coefficient ($\alpha_T$) (K$^{-1}$)</td>
<td>$15.9 \times 10^{-6}$</td>
</tr>
</tbody>
</table>

3.1.4 Experimental results

**Laser performance using output coupler OC1**

The pump polarisation was aligned along the $N_g$ crystallo-optic axis to provide maximum gain on the 765 cm$^{-1}$ mode. Lasing at the first Stokes wavelength (1158 nm) was observed above 18 W pump power, which is reasonably close to the predicted threshold of 13 W. From Fig. 3.6(a), it can be seen that the slope efficiency increased from 0.03% to 0.1% at pump powers above 60 W. The maximum conversion efficiency reached only 0.04%, which is fifty times lower than a previously reported CW ECRL based on barium nitrate [68] and a thousand times lower than for diamond [75]. There was no evidence of thermal roll-off at higher pump powers and the output beam remained TEM$_{00}$ across the measured power range (shown in the inset of Fig. 3.6(a)). The residual pump power was found to increase with pump power. Fig. 3.7(a) shows the shape of the pump (blue) pulse. The initial spikes in the pump waveform seen in the first 150 ns from turn-on are related to the relaxation oscillations and have peaks values about three to five times higher than the average intensity. (Note that the step seen on the trailing edge of the pump waveform is due to an artifact of the system brought about from a small delay (30 $\mu$s) between in the turn-off time of the two
3.1 Multi-longitudinal mode pumping

Figure 3.6: (a). On-time output power versus pump power for pump polarisation aligned parallel to the N\textsubscript{g} axis. The inset shows the Stokes beam profile at 92 W pump power. (b) Laser output spectrum at 50 W pump power, including the pump line at 1064 nm and various Stokes components. The brackets indicate the Raman phonon modes responsible for the various Stokes wavelengths (765 cm\textsuperscript{-1}, 905 cm\textsuperscript{-1} and 87 cm\textsuperscript{-1}, respectively).

Pump modules in the pump laser.) It can be seen from the Fig. 3.7(a) and (b) that the Stokes waveform also replicates many of the features from the pump, although it is noted that the period of oscillations is extended compared to that observed in the pump. The time-averaged behaviour is qualitatively similar to the pump and no decay that might be expected from a thermal effect in the Stokes intensity was observed. The Stokes waveform showed some fast fluctuations and spiking behaviour in Fig. 3.7(a) and (b) respectively.

Figure 3.7: (a). Trace of pump (blue) and Stokes (red) intensity near threshold (b). Trace of Stokes intensity at 60 W pump power.
Measurement of the Raman laser output spectrum (see Fig. 3.6(b)) revealed, apart from the desired first Stokes emission at 1158 nm, radiation at 1177 nm related to the 905 cm\(^{-1}\) phonon mode, as well as several emission lines with a small spacing of about 10 nm, corresponding to the 87 cm\(^{-1}\) phonon mode as indicated by the brackets in the spectrum. The following trend in the output spectrum with increasing pump power was seen: The threshold for first Stokes at 1158 nm was attained first. With an increase in pump power of few watts from the threshold, output at a Stokes wavelength of 1168 nm was observed which we show below is consistent with "cross-cascading" from 1158 nm via a low frequency Raman mode of KYW at 87 cm\(^{-1}\). Cross-cascading refers to the process of cascaded stimulated Raman scattering (SRS) via two or more different phonons [102]. As the pump power is further increased, the Stokes wavelength at 1177 nm corresponding to the 905 cm\(^{-1}\) from 1064 nm attains threshold, which also is subsequently cross-cascaded via the 87 cm\(^{-1}\) mode to 1188 nm with only a small further increase in pump power. This is followed by further cascaded Stokes shifts from 1168 nm to 1182 nm and from 1182 nm to 1194 nm via 87 cm\(^{-1}\). In all, the 765 cm\(^{-1}\) shift followed by 3 cross-cascaded shifts via the 87 cm\(^{-1}\) phonon mode, as well as the 905 cm\(^{-1}\) shift followed by 1 cross-cascaded shift via the 87 cm\(^{-1}\) phonon mode was observed.

The fast fluctuations and spikes (see Fig. 3.7(a) and (b)) were initially believed to be an effect of the cross-cascading process. However, the first Stokes and cross-cascaded Stokes waveforms were studied individually and the spiking behaviour was observed in each of these waveforms and therefore, ruling out such an effect. These observations are tentatively attributed to the dynamics associated with competition between spatial, longitudinal or polarization modes. During these experiments, blue emission was seen emerging from the beam path within the crystal. Such emission is routinely seen in similar tungstate Raman lasers and is attributed to an up-conversion process involving Tm\(^{3+}\) ion impurities [103].

The minimum Raman lasing at the first Stokes wavelength (1177 nm) for pump polarisation aligned to the \(N_m\) crystallo-optic axis was attained above 22 W pump power. The measured threshold is close to the predicted threshold value. Similar cascading behaviour was observed as shown in the Fig. 3.8. As the pump power was increased by 10%, a cross-cascaded Stokes shift to 1209 nm was observed involving a different low frequency mode at
Figure 3.8: Output spectrum showing the residual pump (1064 nm) and three Stokes-shifted wavelengths generated from the 905 cm\(^{-1}\) and cross-cascaded 225 cm\(^{-1}\) phonon modes. The brackets specify the Raman modes responsible for the first and cascaded Stokes wavelengths (905 cm\(^{-1}\) and 225 cm\(^{-1}\), respectively).

225 cm\(^{-1}\). A further 225 cm\(^{-1}\) Stokes shift to 1241 nm occurred with a threshold of 90 W. The observed higher output power at 1209 nm than 1177 nm is attributed to the higher output coupling at the longer wavelength for OC1 as shown by the dashed lines in the Fig. 3.5.

**Laser performance using output coupler OC2**

Figure 3.9: On-time output power versus pump power. The laser output spectrum in the inset was measured at 110 W pump power and the pump intensity is attenuated using a long pass filter. The brackets indicate the Raman modes responsible for the Stokes wavelengths (765 cm\(^{-1}\) and 87 cm\(^{-1}\), respectively).
With the pump polarisation oriented along the $N_g$ crystallo-optic axis, the laser exhibited a higher threshold pump power of 90 W which was expected due to the higher transmittance at the Stokes wavelengths. The output power increased linearly with the pump power achieving a slope of 4% as shown in Fig. 3.9 and a maximum conversion efficiency of 2%. This conversion efficiency is comparable to the aforementioned CW barium nitrate laser [68]. The output spectrum contained the first Stokes wavelength at 1158 nm and the cross-cascaded Stokes wavelength at 1168 nm corresponding to the 765 cm$^{-1}$ and 87 cm$^{-1}$ Raman modes respectively.

For the pump polarisation aligned to $N_m$, the higher output coupling provided by OC2 led to an increase in threshold (to 120 W). Cross-cascading by the 225 cm$^{-1}$ mode was observed at a pump power of 130 W.

### 3.2 Analysis

The above results show that the laser behaviour is characterized by low efficiency and a complex output spectrum involving multiple cascaded Raman shifts. This is unusual compared to previously reported ECRLs (pulsed and CW). The reasons are examined in this section.

#### 3.2.1 Thermal analysis

The power deposited in the crystal is given by $P_{dep} = \left(\frac{\lambda_s}{\lambda_p}\right)P_{gen}^s + P_{int}^s \alpha l_R$ where the two terms correspond to contributions from the SRS process and the impurity absorption respectively. The power coupled into the Stokes and the phonon field is obtained from the pump depletion. Since pump depletion in the crystal was small (< 0.02 W), the heat deposited in the crystal is therefore much lower and well below the calculated value for destabilizing the cavity (0.3 W). This is consistent with absence of thermal effects noted in the Stokes waveform (Fig. 3.7(a) and (b)) and slope efficiency (Fig. 3.6(a)). Thus thermal effects in the laser are deduced to be weak for the investigated output powers and the origin of the laser inefficiency must be attributed to other factors. It should also be noted that the contribution by impurity absorption is neglected here. However, it is not negligible and can lead to the thermal effects in the crystal.
3.2.2 Raman gain coefficients for the low frequency modes

Although most reports on double metal tungstate Raman lasers utilize the two primary phonon modes, the low-frequency modes in KYW and corresponding modes in similar materials have been observed previously. Kaminskii et al. [71] and Hanuza and Macalik [86] reported strong Raman modes of frequency 87 cm\(^{-1}\) and 225 cm\(^{-1}\) in the spontaneous Raman spectrum of KYW that have been attributed to a librational mode of WO\(_n\) and a translation mode of Y\(^{3+}\) ions, respectively. In the similar crystal KGW, laser operation on the analogous mode at 84 cm\(^{-1}\) was observed in an intra-cavity Raman laser designed for lasing on the primary 768 cm\(^{-1}\) mode [104].

Even though they have been identified previously in the literature, the gain coefficients of these low-frequency modes have not been reported. Spectrometer characterization requires a system with high resolution and with filtering capable of resolving features close to the probe wavelength.

Figure 3.10: Spontaneous Raman spectra of KYW for the pump polarisation (E) parallel to (a) the N\(_g\) and (b) the N\(_m\) crystallo-optic axis. The insets to (a) and (b) show fitted line shapes (after baseline correction) of the 87 cm\(^{-1}\) and the 225 cm\(^{-1}\) modes, respectively.

In order to address this deficiency, the linewidth and gain coefficients for the low frequency modes were determined using a high-resolution (< 1 cm\(^{-1}\)) Raman spectrometer (LabRAM HR Evolution, HORIBA Ltd.) which enables investigation of spectral features as low as 3.5 cm\(^{-1}\). The probe beam from the 532 nm laser incorporated in the spectrometer was
propagated along the crystallographic b-axis (the lasing direction) with its polarisation vector varied using a half-wave plate. Fig. 3.10(a) shows the three main lines at 87 cm\(^{-1}\), 765 cm\(^{-1}\) and 905 cm\(^{-1}\) when the pump polarisation is oriented along the N\(_g\) axis. It can be deduced from the figure that the stationary gain coefficient of the 87 cm\(^{-1}\) line is higher than the main 765 cm\(^{-1}\) line since its peak value is higher than the main mode. Moreover, the width of the 87 cm\(^{-1}\) line (1.1 cm\(^{-1}\)) is much narrower than the 765 cm\(^{-1}\) line (8.7 cm\(^{-1}\)). For the pump polarisation aligned to N\(_m\) (see Fig. 3.10(b)), the 225 cm\(^{-1}\) line is observed but in this case the peak is smaller than the main 905 cm\(^{-1}\) line.

Table 3.2: Linewidths, relative peak intensities and Raman gain coefficients (g) of the KYW Raman modes for pump polarisations parallel to the N\(_g\) and N\(_m\) axes.

<table>
<thead>
<tr>
<th>Raman mode (cm(^{-1}))</th>
<th>Linewidth (cm(^{-1}))(a)</th>
<th>Relative peak intensities(a)</th>
<th>g (cm/GW)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>N(_g)</td>
<td>N(_m)</td>
<td>N(_g)</td>
</tr>
<tr>
<td>87</td>
<td>1.11 ± 0.01</td>
<td>0.57 ± 0.02</td>
<td>2.564 ± 0.013</td>
</tr>
<tr>
<td>765</td>
<td>8.69 ± 0.02</td>
<td>8.82 ± 0.02</td>
<td>1.21 ± 0.05</td>
</tr>
<tr>
<td>905</td>
<td>6.88 ± 0.003</td>
<td>6.89 ± 0.003</td>
<td>1.0 ± 0.01</td>
</tr>
<tr>
<td>225</td>
<td>–</td>
<td>5.89 ± 0.013</td>
<td>–</td>
</tr>
</tbody>
</table>

\(a\) Error values are derived from the variance of fit parameters to Voigt profiles.

Fitting to the profiles reveals that the linewidth of the 87 cm\(^{-1}\) mode is 1.1 cm\(^{-1}\), which corresponds to a dephasing time \(T_2 \approx 9.6\) ps (from \(T_2 = 1/(\pi \nu \Delta \nu)\)). The instrument width is determined from the Voigt fit of the barium nitrate spontaneous Raman spectrum which has a reported Lorentzian linewidth of 0.4 cm\(^{-1}\) for 1047 cm\(^{-1}\) phonon mode \([61]\). The \(T_2\) value is slightly longer than the first-order mode in diamond (\(T_2 \approx 7.1\) ps), but shorter than for the primary (A\(_g\)) mode in barium nitrate (\(T_2 \approx 26\) ps) \([61]\). The linewidths of the primary Raman modes at 765 cm\(^{-1}\) and 905 cm\(^{-1}\) agree with previously published values to within measurement uncertainty \([73]\). By comparing the relative peak intensities of the respective modes and referencing to a reported value of 3.6 cm/GW in the literature for the 905 cm\(^{-1}\) gain coefficient \([71]\), the gain coefficients for the 87 and 225 cm\(^{-1}\) are 9.2 and 2.5 cm/GW respectively. Note that no uncertainty was reported for the 905 cm\(^{-1}\) reference value, hence the uncertainty for the low-frequency Raman modes have not been quoted here.

The low frequency Raman shifts are seen in cross-cascading since the output coupler provides
strong feedback at the associated wavelengths (see Fig. 3.5). The Stokes shifts from the pump wavelength are not seen as those wavelengths are non-resonant in the ECRL. For example, the first Stokes emission for the 87 cm\(^{-1}\) mode, which would occur at 1074 nm, does not reach threshold.

### 3.2.3 Analytical model for a single cascaded Stokes shift

Since the laser spectrum showed multiple cross-cascading involving three phonon modes, it is relevant to first understand the effect of cross-cascading on the pump and the Stokes field in the cavity. Therefore, a simple analytical model was developed to show the impact of a second Stokes field via a second phonon mode on the flow of power from the pump field to the first Stokes field in a Raman laser.

Firstly, the case for an ECRL with only a single shift operating in steady-state regime is reviewed. Since they are time-independent, the following coupled differential equations describe the evolution of the pump and the Stokes beam intensities in the Raman laser,

\[
\frac{dI_s(z)}{dz} = g I_s(z) I_p(z),
\]

(3.3)

\[
\frac{dI_p(z)}{dz} = -\frac{g I_p(z) I_s(z)}{\eta},
\]

(3.4)

where, \(g\) is the Raman gain coefficient at the Stokes wavelength, \(I_p\) and \(I_s\) are the pump and Stokes intensities and \(\eta = \lambda_p/\lambda_s\) is the quantum defect with \(\lambda_p\) and \(\lambda_s\) are the pump and the Stokes wavelength. It is assumed that the beams are collimated in the cavity, the fields are in the steady-state regime and the absorptive losses are ignored for simplicity. Therefore, the primary losses in the cavity are due to the output coupling loss, \(-\ln R_s\), where \(R_s\) is the reflectivity of the output coupler at the first Stokes wavelength. The pump intensity \(I_{p_{th}}\) required to reach lasing threshold is obtained when the steady-state gain and cavity losses are equal, therefore,

\[
\int_0^{2l} I_p(z)dz = -\frac{\ln R_s}{g} = I_{p_{th}} 2l.
\]

(3.5)

The round trip growth of the Stokes intensity (through two passes in the Raman crystal of length \(l\)) is obtained by integrating Eq. 3.3

\[
I_s(2l) = I_s(0) e^{2g \int_0^{2l} I_p(z)dz},
\]

(3.6)
The depletion of the pump beam after the double pass in the cavity is given by,

\[ I_p(2l) = I_p(0)e^{-4glT_s}. \]  

(3.7)

Here, \( T_s \) is the average Stokes intensity defined as \( \frac{\int_0^{2l} I_s(z)dz}{2l} \). It is evident from the above equation that the increase in the Stokes intensity is accompanied by an exponential decrease in the residual pump power. The result of the calculation of the above equations is illustrated in Fig. 3.11 for a 99.5% output reflectance at the first Stokes wavelength. The figure depicts the pump depletion by the Stokes field in a Raman laser without cascading.

Figure 3.11: Incident pump intensity versus Stokes and residual pump intensity for a Raman laser without cascading.

In order to see how a cross-cascaded Stokes field interacts with the pump and the Stokes field intensity, the following rate equation is introduced for the cross-cascaded Stokes intensity \( I_{cs} \) for which the first Stokes acts as the pump,

\[ \frac{dI_{cs}(z)}{dz} = \eta_{cs}g_{cs}I_s(z)I_{cs}(z). \]  

(3.8)

where \( g_{cs} \) is the Raman gain coefficient for the low-frequency mode. By integrating Eq. 3.8, the round trip growth of cross-cascaded Stokes intensity is obtained as

\[ I_{cs}(2l) = I_{cs}(0)e^{\eta_{cs}g_{cs}I_s2l}. \]  

(3.9)

Since at steady state, \( R_{cs} = I_{cs}(0)/I_{cs}(2l) \), the threshold condition for cross-cascaded Stokes intensity is

\[ T_s = \frac{-\ln R_{cs}}{g_{cs}\eta_{cs}4l}. \]  

(3.10)
Eq. [3.10] shows that the intracavity Stokes is constant above the threshold. This clamping of Stokes intensity at the onset of cross-cascading, as indicated by Eq. [3.7], adversely affects the power transfer from the pump. Substituting Eq. [3.10] into Eq. [3.7] yields

\[ I_p(2l) = I_p(0)(R_{cs})^\frac{g}{g_{cs}g_{cs}}. \] (3.11)

The above equation shows that the residual pump power as a fraction of the input \((I(2l)/I(0))\) is constant in the regime of cross-cascaded lasing and is a function of output reflectivity at cross-cascaded Stokes line. Fig. 3.12 shows the dynamics of the pump and the Stokes intensity in the presence of a cross-cascaded Stokes calculated from the above equations for \(R_s = 92\%\) and \(R_{cs} = 50\%\), and \(g = 4.5\) cm/GW and \(g_{cs} = 5\) cm/GW. The saturation of pump depletion is represented by the increase in the residual pump power accompanied by the clamping of the first Stokes intensity at the threshold for cross-cascading.

Figure 3.12: Laser behaviour with cascading. The Stokes and residual pump power as a function of input power. \(I_{\text{threshold}1}\) and \(I_{\text{threshold}2}\) indicate 1st Stokes threshold and cross-cascaded Stokes thresholds respectively.

For the Raman laser presented in this chapter, the calculated threshold for cascading is only marginally higher than the first Stokes threshold as \(R\) for the first and the cross-cascaded Stokes is similarly high (> 98% for OC1 and 2) and \(g_{cs}\) is larger than \(g\). Substituting the experimental values for output coupler reflectivity and Raman gain coefficient in Eq. [3.11] yields a pump depletion rate of < 1%. Therefore, in this laser, cross-cascading clamps the first Stokes intensity and saturates the pump depletion leading to a low conversion efficiency, and at the same time preventing thermal effects in the crystal.
3.2.4 Numerical modelling for multiple cascading

The above analytical model describes the contrast in the dynamics of the pump and first Stokes field intensities in the Raman laser with and without the presence of a single cross-cascaded Stokes field. However, in the present experiments, there is a multitude of cross-cascaded Stokes components generated through the interactions of up to three phonon modes and therefore, more equations with additional terms for each cascaded Stokes components are required. The coupled differential equations used in the analytical model become more complicated and an analytical solution seems unlikely. So, a numerical model was adapted to include the effect of the low-frequency Raman modes to gain insights into the dynamics underpinning the experimental observations. It is based on coupled time-dependent rate equations describing the amplification and depletion of the pump and different Stokes fields via SRS \[105,106\].

In this model, the pump and Stokes intensities are considered uniform as a function of position inside the cavity, and anti-Stokes generation, Raman four-wave mixing and dispersion effects are neglected. The generalized equations for the pump, the two first Stokes and the two cross-cascaded Stokes fields from each of the first Stokes fields are:

\[
\frac{dI_p}{dt} = \frac{I_{pump}}{t_r} - \frac{2l_{RGa}}{t_r} I_p I_a - \frac{2l_{RGb}}{t_r} I_p I_b - \frac{L_s - \log(R_{1p} R_{2p})}{t_r} I_p
\]  \hspace{1cm} (3.12)

\[
\frac{dI_a}{dt} = \frac{2l_{RGa} \eta_a}{t_r} I_p I_a - \frac{2l_{RGc} \eta_c}{t_r} I_a I_a - \frac{L_s - \log(R_{1a} R_{2a})}{t_r} I_a + \frac{2K I_R}{t_r} I_p
\]  \hspace{1cm} (3.13)

\[
\frac{dI_{a1}}{dt} = \frac{2l_{RGc} \eta_c}{t_r} I_a I_{a1} - \frac{2l_{RGc} \eta_c}{t_r} I_a I_{a2} - \frac{L_s - \log(R_{1a} R_{2a1})}{t_r} I_{a1} + \frac{2K I_R}{t_r} I_a
\]  \hspace{1cm} (3.14)

\[
\frac{dI_{a2}}{dt} = \frac{2l_{RGc} \eta_c}{t_r} I_a I_{a2} - \frac{2l_{RGc} \eta_c}{t_r} I_a I_{a3} - \frac{L_s - \log(R_{1a} R_{2a2})}{t_r} I_{a2} + \frac{2K I_R}{t_r} I_a
\]  \hspace{1cm} (3.15)

\[
\frac{dI_b}{dt} = \frac{2l_{RGb} \eta_b}{t_r} I_p I_b - \frac{2l_{RGc} \eta_c}{t_r} I_b I_{bi} - \frac{L_s - \log(R_{1b} R_{2b})}{t_r} I_b + \frac{2K I_R}{t_r} I_p
\]  \hspace{1cm} (3.16)

\[
\frac{dI_{b1}}{dt} = \frac{2l_{RGc} \eta_c}{t_r} I_b I_{b1} - \frac{2l_{RGc} \eta_c}{t_r} I_b I_{b2} - \frac{L_s - \log(R_{1b} R_{2b1})}{t_r} I_{b1} + \frac{2K I_R}{t_r} I_b
\]  \hspace{1cm} (3.17)

\[
\frac{dI_{b2}}{dt} = \frac{2l_{RGc} \eta_c}{t_r} I_b I_{b2} - \frac{2l_{RGc} \eta_c}{t_r} I_b I_{b3} - \frac{L_s - \log(R_{1b} R_{2b2})}{t_r} I_{b2} + \frac{2K I_R}{t_r} I_{b1}.
\]  \hspace{1cm} (3.18)
where, $I_{\text{pump}}$ is the initial pump intensity, $g_a$ and $g_b$ denotes the gain-coefficients at 765 cm$^{-1}$ and 905 cm$^{-1}$ modes respectively, $g_c$ is the gain coefficient for 87 cm$^{-1}$ mode, at the pumping wavelength. $\eta_i$ is quantum defect for the $i^{\text{th}}$ Stokes shift from the pump wavelength. $I_R$ is the Raman crystal length and $L_s$ is the dissipative loss excluding out-coupling loss and $K$ is the spontaneous scattering loss. $t_{\text{rt}}$ is the round trip time of the cavity and $R_{1p}$ and $R_{2p}$, $R_{1a,b}$ and $R_{2a,b}$, and $R_{1ai,bi}$ and $R_{2ai,bi}$ are the input and output coupler reflectivities at the pump, first Stokes and $i^{\text{th}}$ cross-cascaded wavelengths, respectively. The OC1 and OC2 reflectivities for each of the Stokes wavelength were taken from Fig. 3.5. The cavity length was 120 mm and the pump spot size for OC1 and OC2 were 23 µm and 39 µm, respectively. Since the absorption coefficient and reflection losses of the crystal were unknown, the model agreed well with the experimental observations when the round trip intracavity dissipative losses (scatter and absorption) were taken to be in the range 0.8% to 1.0%. The intracavity pump intensity $I_p$ and the first and the $i^{\text{th}}$ cross-cascaded Stokes intensities $I_{a,b}$ and $I_{ai,bi}$ were solved numerically using standard differential equation solver (NDSolve function) in Mathematica and the corresponding output powers were calculated and compared with the experimental data.

Since the gain coefficients of the 765 cm$^{-1}$ and 87 cm$^{-1}$ modes were not precisely known for each crystallo-optic axis, the best results were obtained when the gain coefficients for the 765 cm$^{-1}$ and 87 cm$^{-1}$ modes were set to 4.5 cm/GW and 9.0 cm/GW respectively. The gain coefficients are within the uncertainty range of the gain coefficients derived from the spontaneous Raman spectra (see Table 3.2 and 3.3). With these values, the calculated threshold at 1158 nm for OC1 was 17.8 W (which is in close agreement with the measured value of 18 W).

Table 3.3: Raman gain coefficients obtained by the numerical model (for pump polarisation parallel to $N_g$, pump wavelength: 1064 nm).

<table>
<thead>
<tr>
<th>Raman mode (cm$^{-1}$)</th>
<th>Gain coefficients (cm/GW )</th>
</tr>
</thead>
<tbody>
<tr>
<td>87</td>
<td>9.0</td>
</tr>
<tr>
<td>765</td>
<td>4.5</td>
</tr>
<tr>
<td>905</td>
<td>3.6</td>
</tr>
</tbody>
</table>

The intensities for each Stokes wavelength as a function of pump power are generated by
the model. For a visual comparison with the experimental laser spectrum along $N_g$, the model results were plotted as Lorentzian lines at each Stokes wavelength with a linewidth of 0.7 nm. The spectra were modelled by using a Lorentzian lineshape function (probability density function and Cauchy distribution in Mathematica). The strength of each Stokes spectral line was calculated by multiplying the output power of each line with this function. The modelled spectra (dashed lines) for OC1 and 2 agree well with the experimental spectra (solid lines) as shown in Fig. 3.13(a) and (b), respectively. The model qualitatively and quantitatively reproduced the experimental observations: the onset of the cross-cascaded Stokes shifts at low thresholds accompanied by poor pump depletion and poor conversion efficiency.

![Figure 3.13: Measured spectra (solid lines) and corresponding modelled spectra (dashed lines) for (a) OC1 at different pump powers and (b) OC2 at 110 W.](image)

It can be seen from the model that the threshold for the cross-cascaded Stokes at 1168 nm is achieved with only a slight increase in pump power of about 2 W. It also demonstrates the clamping of the intracavity first Stokes intensity with the onset of 1168 nm lasing, which restricts the transfer of power from pump to first Stokes. This is confirmed by the stepwise increase in the residual pump in the model. When the pump power is further increased by 5 W, the first Stokes at 1177 nm corresponding to the 905 cm$^{-1}$ is seen, which is soon followed by cross-cascading to 1187 nm via the 87 cm$^{-1}$ mode.

In the case of OC2, the model shows the threshold for the first Stokes at 1158 nm at 95 W which is close to the experimental value of 90 W. The threshold for the first Stokes at 1177 nm
related to the 905 cm\(^{-1}\) is achieved at 120 W pump powers which is same as the experimental threshold. The number of the cross-cascaded Stokes wavelengths for OC2 is smaller than for OC1 because of the former’s lower reflectivity (98% compared to 99.7% for OC1) at all Stokes wavelengths.

Both the analytical and the numerical model shows that the cross-cascading leads to the clamping of the first Stokes intensity, which inhibits the pump depletion and limits the overall conversion efficiency. However, the numerical model also shows that a further cascade unclamps the first Stokes intensity and thus restores effective power transfer from the pump. This could eventually lead to a large recovery in the slope efficiency. It is evident from the numerical model that the value of slope efficiency changes from low to high in alternating fashion with odd and even orders of cascading. The reason that this laser has an overall poor efficiency is because the threshold for odd-order cascade is very near to the even-order which causes the slope efficiency to remain low for most of the power range. The calculated output power and thus, slope efficiency for OC1 were significantly higher than the measured values. It may be because as the threshold for the cross-cascading depends on the intracavity losses, a slight change in the loss value changes the slope efficiency. However, the model reproduced the much higher output power observed for OC2 compared to OC1 which is attributed to the increase in threshold for cascaded Stokes components leading to improved pump depletion.

### 3.2.5 Discussion

The efficiency and spectrum achieved from the multimode pumping, and the modelling show that the performance of the Raman laser is significantly impacted by the low-frequency Raman modes, due to their high gain and the high mirror reflectivity at wavelengths within 250 cm\(^{-1}\) of the first Stokes lines. Therefore the impact of these modes has to be taken into account when designing a Raman laser based on KYW or crystals with high-gain, low-frequency Raman modes, and a high cavity reflectivity as is typical for CW systems.

The negative impact of cross cascading on laser efficiency appears too extreme in the present case. There have been numerous reports of efficient Raman lasers using double-metal-tungstate crystals such as KYW and KGW (which has a very similar Raman spectrum to KYW) demonstrated in CW/pulsed intra/external cavity configurations [77, 79, 80, 82, 104].
Continuous-wave external cavity KYW Raman laser

[107][111], and only one reports cross-cascading as a power-limiting factor at high power [81]. Most of these devices operated with the pump polarisation aligned along the Nm axis where the 905 cm\(^{-1}\) mode is dominant, and did not report cross-cascading [77][79][80][107][110]. One possible reason is that the low-frequency mode (225 cm\(^{-1}\)) has lower gain than the main mode. Also the resulting Stokes shift is about 20 nm from the first Stokes so the output coupler reflectivity at the cascaded wavelength may be more differentiated from the first Stokes wavelength. Nevertheless, a nanosecond pulsed ECRL [82] has reported cross-cascading via the analogous 204 ± 3 cm\(^{-1}\) in KGW from the first Stokes mode (901 cm\(^{-1}\)). In that case, the authors attributed the low slope efficiency (in comparison to that obtained in other ECRLs) mainly to thermal effects in the crystal and the poor pump beam quality, but did not discuss the effect of cross-cascading.

A few papers have reported cross-cascading in KGW lasers with pump polarisation aligned parallel to Ng. Cross-cascading via the 84 cm\(^{-1}\) mode is observed at elevated powers, which coincides with an observed sudden decrease in slope efficiency in case of a ns-pulse pumped KGW ECRL with a low-reflectivity output coupler [81]. However, the mechanisms by which the cross-cascading resulted in reduced efficiency were not discussed. Cross-cascading on the low-frequency Raman mode has been observed in some CW intracavity and self-Raman lasers with highly reflective output couplers, although any adverse effect of cross-cascading on the laser efficiency was not discussed [104][111]. Since the boundary conditions for power flow in intracavity Raman lasers are different to ECRLs, a direct comparison of their results with observations from this laser is not appropriate. In general, most other reports of efficient KGW and KYW lasers do not refer to cross-cascading [77][79][80][107]. This could be due to the specific loss properties of the resonators used (e.g. mirror coatings) or insufficient resolution in the characterization of the output spectrum.

Typically cross-cascading would be considered a parasitic process that should be avoided to improve laser efficiency. To achieve good efficiency, the cavity losses at the first and the cross-cascaded Stokes should be increased to extend the pump power range between the first and the cross-cascaded thresholds. However, this requires sophisticated mirror coatings with significant changes in reflectivity (tens of percent) over a spectral range of a few to a few tens
of nanometers, which is likely to be expensive and impractical. Introducing wavelength selective elements such as etalons, gratings etc. into the cavity to suppress the cross-cascading may be a more viable alternative.

The presence of the high gain low-frequency modes may, on the other hand, be a source of benefit. Cross-cascading could be used to simultaneously generate multiple closely spaced wavelengths which may be of use in spectroscopy or in the generation of terahertz radiation at integer multiples of the mode frequency (2.6 THz in the case of 87 cm\(^{-1}\)) through difference frequency mixing of first and/or cross-cascaded Stokes waves. Furthermore, Raman beam conversion for the purposes of either brightness enhancement \[112\] through the Raman beam cleanup effect, or beam combination, could be achieved with much lower quantum defect than with conventional Raman modes in crystals. This may pave the way for a novel approach to high-power Raman lasers with possible advantages over diamond, for example, due to its relative ease of growth of large crystals with good damage threshold and moderate thermal properties.

3.3 Narrow linewidth pumping

The original aim of the KYW study was to use narrow-linewidth pumping as a test bed to investigate the SLM concepts developed in Chapter 2 and compare the results with a diamond Raman laser. However, the preceding Sections have proved that the laser spectrum has multiple spectral lines owing to the cross-cascading process via the low-frequency phonon modes. Therefore, the focus of the section is shifted to the investigation of the longitudinal mode spectrum of individual Stokes wavelength components.

3.3.1 Experimental arrangement

**KYW Raman laser**

In this setup, the pump is a continuous-wave, narrowband and tunable distributed feedback (DFB) laser (TOPTICA Photonics, model DL DFB BFY) which is amplified to a maximum power of 50 W at 1064 nm with \(M^2 = 1.05\) by a Yb fibre amplifier (IPG Photonics, model YAR-LP-SF), as shown in Fig. 3.14. It has a linewidth of 5 MHz which is significantly
narrower than the KYW Raman gain bandwidth (179 GHz). Hence, the first requirement of homogeneous gain saturation is satisfied for testing the concept developed in Chapter 2. The stability of the pump center wavelength was measured to be about 40 MHz as shown in Fig. 3.15.

Although the performance of the KYW Raman lasers operating with OC1 and OC2 was poor, the lowest Raman lasing threshold was provided by OC1. Since the power available at the input of the Raman cavity is up to 40 W (some power is lost in the optical isolator and the optics placed in the beam path), OC1 was chosen as the output coupler to minimize the threshold. The pump was chopped at 33% duty cycle with a pulse duration of 10 ms. Since the pump pulse duration is longer than the time to establish steady-state thermal gradients, the Raman laser reaches a CW operating regime during each pulse. The half-wave plate was used to align the pump polarisation to $N_g$. The Raman cavity setup is the same as that described in Section 3.2.2.
3.3 Narrow linewidth pumping

3.3.2 Output power and efficiency

The Raman lasing threshold for Stokes wavelength at 1158 nm and laser efficiency obtained with single-mode pumping were same to that obtained for multimode pumping over the same pump power range as shown in Fig. 3.16(a). Above the Raman lasing threshold, a slight decrease in the residual pump power was observed (see the encircled part in the Fig. 3.16(a)). However, as expected, it increased and was comparable to the incident pump power at higher pump powers. Consequently, the pump depletion in the crystal is negligible and hence, the overall conversion efficiency is minimal.

![Output Stokes power and residual pump power vs pump power](image)

Figure 3.16: Output Stokes power (red) and the residual pump power (black) versus pump power with the pump polarisation oriented along the N_g axis using OC1.(b) Laser output spectrum at different pump powers.

Fig. 3.16(b) shows the laser spectrum at different pump powers. As expected, with only a slight increase in pump power, cross-cascaded Stokes wavelength at 1168 nm via the 87 cm\(^{-1}\) phonon mode from first Stokes emission at 1158 nm corresponding to 765 cm\(^{-1}\) was observed. Also, the first Stokes wavelength at 1177 nm and its cross-cascaded line at 1188 nm corresponding to the 905 cm\(^{-1}\) mode and the 87 cm\(^{-1}\) mode respectively were seen as the pump power was increased.

3.3.3 Spectral diagnostics

The laser output comprising first Stokes (1158 nm), cross-cascaded Stokes (1168 nm) and the first Stokes corresponding to 905 cm\(^{-1}\) (1177 nm) was separated into its separate components.
using a 300 lines/mm reflective diffraction grating, as shown in Fig. 3.17. The separation between the reflected first-order 1168 nm and 1177 nm Stokes wavelengths was small and therefore, two right-angled prisms were employed to further separate the two Stokes beams. The longitudinal mode structure of 1158 nm and 1168 nm were investigated by a Fabry-Perot interferometer (FPI) and an optical spectrum analyzer (OSA).

Figure 3.18: (a). The output from the single-mode fiber amplifier measured by the Fabry-Perot interferometer. The linewidth of the pump mode is limited by the resolution of the FPI. (b). The output from the single-mode fiber amplifier measured by the optical spectrum analyzer. The linewidth of the pump mode is 0.06 nm which is close to the resolution limit of the spectrum analyzer.
The confocalscanning FPI (Thorlabs, SA210) provides a free spectral range (FSR) of 10 GHz with a resolution of 67 MHz. A convex lens of focal length 100 mm was used to couple the Stokes beam into the FPI. The FPI detector was monitored using an oscilloscope. The etalon scan rate of 10 GHz in 4 ms was calibrated using the single-mode fiber amplifier output (Fig. 3.18(a)).

The laser spectrum was also monitored with an OSA (Anritsu, MS9710C) in order to calculate the overall width of each spectral line. The resolution of the instrument is 0.05 nm. The Stokes beam was coupled into a multimode fiber and then fed into the OSA. To monitor the Stokes center wavelength, another laser spectrum analyzer (Bristol instruments, model 771A-NIR) was employed. The laser spectrum analyzer has an operating wavelength range from 520 to 1700 nm providing a wavelength accuracy of ±0.2 ppm and a spectral resolution as high as 2 GHz. The pump wavelength stability (see Fig. 3.15) was recorded using this instrument.

### 3.3.4 Longitudinal mode structure

The mode structures of the first Stokes and cross-cascaded Stokes wavelengths at 1158 nm and 1168 nm respectively, at pump powers close to threshold were investigated for the laser operating CW. The FPI scans revealed that each Stokes line was operating on multiple longitudinal modes. Fig. 3.19 shows the multiple modes within an FSR of the FPI for Stokes
wavelengths at 1158 nm and 1168 nm at the maximum output power. The FSR of the cavity is 869 MHz (optical cavity length = 172.5 mm, including 50 mm long KYW crystal with refractive index of $N_g$ axis at 1158 nm is 2.05 [71]). Some of the lines in the scan have this spacing. The interferogram is expected to be complex due to long finite time and the overlapping nature of the scan for spectra of width larger than 10 GHz. Also, as the KYW faces were plane and parallel, etalon effects may further complicate the spectrum.

![Figure 3.20: First Stokes spectrum at 1158 nm measured by the OSA.](image)

An OSA was employed to measure the overall spectrum width. Fig. 3.20 shows the 1158 nm Stokes width was approximately 0.232 nm which corresponds to 52 GHz or approximately 58 longitudinal modes.

### 3.3.5 Discussion

The Raman laser pumped by a single-mode source exhibited multiple longitudinal modes at various Stokes powers, even those close to the threshold. This observation, which is similar to that obtained for a CW diamond Raman laser [60], suggests that the multimode behaviour of the diamond Raman laser is not unique to diamond material characteristics. To understand the causes of mode instability in Raman lasers is challenging in the present system as the spectral dynamics are too complex.

It is clear from this chapter that cross-cascading not only limits the conversion efficiency but it also complicates the mode spectrum. Therefore, it is judicious to proceed with the
investigation of the longitudinal mode structure of CW diamond ECRL on account of its much simpler Raman spectrum.

3.4 Conclusion

This chapter presented a detailed investigation of spectral features and cross-cascading and multi-Stokes dynamics owing to low-frequency phonon modes in a KYW ECRL. Surprisingly, unlike CW barium nitrate ECRL, thermal effects did not play a significant role in reducing the conversion efficiency. Instead, cross-cascading via low-frequency modes was found to inhibit the conversion of the first Stokes orders, limiting conversion efficiency to « 1%. A high-resolution spontaneous Raman spectra were recorded along the N_g and N_m crystallo-optic axes which revealed the high gain coefficients of these low-frequency modes, particularly 87 cm\(^{-1}\) mode. Analytical and numerical models were developed to explain the effect of cross-cascading on the laser efficiency and the modelled results agreed well with the experimental observations. It can be concluded that, for a Raman crystal with one or more strong, low-frequency Raman modes, cross-cascading may be a major factor for consideration in Raman cavity setup, particularly for CW configurations that rely on highly resonant cavities.

The low-frequency Raman mode at 87 cm\(^{-1}\) in KYW (84 cm\(^{-1}\) in KGW) nevertheless indicates potential for future applications as multi-wavelength laser sources of high beam quality and as a low-quantum-defect Raman material for brightness enhancement and/or beam combination.

Since cross-cascading complicated the study for causes of the multimode operation of ECRL, it was decided to continue the investigation in CW diamond Raman ECRL where the problem of cross-cascading can be readily avoided owing to the pure single Raman mode of the diamond.

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The results are published in Soumya Sarang et al. "High-gain 87 cm\(^{-1}\) Raman line of KYW and its impact on continuous-wave Raman laser operation." Optics Express 24.19 (2016): 21463-21473.

Chapter Contributions: All experimental work was conducted by the author. An existing numerical code written by Dr. Oliver Lux was modified to enable analysis of cross-cascading results.
The previous Chapter showed that KYW is not as well suited to efficient CW Raman laser operation in the external configuration as diamond in terms of efficiency and simplicity of the output spectrum. As a result, diamond is seen as a more promising system for investigating SLM operation. In this chapter, an external cavity diamond Raman laser pumped by a single-mode laser source at 1064 nm is used as the test bed to investigate the spatial hole burning free nature of Raman gain concept developed in Chapter 2. The laser performance is characterized and conditions for achieving SLM are investigated.

4.1 Narrow-line width pumped DRL

The basic cavity design is similar to that used for KYW Raman laser except that it is adapted to take into account diamond’s Raman frequency shift, length and gain/loss properties. The
Single longitudinal mode diamond Raman laser

diamond phonon mode at 1332.3 cm\(^{-1}\) enabled Raman conversion from 1064 nm to 1240 nm. To estimate the threshold for the double-pass pumped DRL, the same equation is used as for KYW Raman laser (Eq. 3.1 with a factor of 2 in the denominator). The crystal absorption loss (\(\alpha\)) is taken to be 0.17\% cm\(^{-1}\) from the ref. [92] as the diamond crystal used in this experiment belonged to the same manufacturing batch. The steady-state Raman gain coefficient (\(g\)) is taken as 10 cm/GW from Table 2.1. Using these values, the calculated threshold is 16 W for a 0.5\% output coupler transmittance and a pump spot radius of 30 \(\mu\)m in the centre of the 8 mm long diamond crystal.

### 4.1.1 Experimental arrangement

![Figure 4.1: Schematic diagram of diamond ECRL](image)

The experimental arrangement is shown in Fig. 4.1. The DRL consisted of an 8 mm \(\times\) 4 mm \(\times\) 1.2 mm low nitrogen, low-birefringence, single-crystal CVD diamond (Element Six Ltd.) placed in the middle of the 102 mm long resonator between 50 mm ROC input coupler and output coupler. The diamond end-faces were AR coated at the pump and the Stokes wavelength to minimize the intracavity losses (\(R < 1\%\) at 1064 nm, \(R < 0.25\%\) at 1240 nm). The resonator input coupler was highly transmitting (\(T=97.2\%\)) at the pump wavelength and highly reflective (\(R=99.9\%\)) at the Stokes wavelength, whereas the output coupler was highly reflective (\(R=99.9\%\)) at the pump wavelength and partially transmitting (\(T=0.43\%\)) the Stokes radiation. Fine control of the cavity length was necessary for SLM operation and was achieved using a piezoelectric translation stage (PZT) on the input coupler.

The pump source was the same CW narrowband Yb-fiber amplifier as used in Sec 3.4.1 for pumping the KYW laser. The pump linewidth of 5 MHz satisfies the first requirement for
generating a homogeneous gain spectrum for testing the SLM concept developed in Chapter 2. The pump beam was guided into the DRL in the same way as described for KYW Raman laser. A convex lens of focal length 1 m was placed in the beam path to collimate the slightly diverging pump beam. A convex lens of focal length 50 mm was used to focus the pump beam into the diamond, resulting in a focal spot size of 40 µm. The Stokes mode size was calculated to be 57 µm. A half-wave plate was used to orient the pump polarisation along the <111> axis which provides the maximum Raman gain [69, 91].

The output Stokes was collimated by a convex lens of focal length 75 mm, and a long-pass filter (Thorlabs, FEL1150) was used to filter any residual light. The longitudinal mode structure of the Stokes emission was investigated using a scanning Fabry-Perot interferometer and the Stokes centre wavelength was simultaneously monitored by a laser spectrum analyzer (as described in Sec. 3.4.3).

### 4.1.2 Output power and efficiency

The first Stokes at 1240 nm was achieved at a threshold of 13.4 W pump power and increased linearly with a slope of 37%. The threshold is in good agreement with the value predicted in Section 4.2 (16 W). The back reflected residual pump power from the DRL cavity was also measured from the reflected beam by the input polarizer of the optical isolator to investigate the pump depletion. Initially, it was seen that the residual pump power increased with incident pump power even above the Raman laser threshold (apart from a small power range just above threshold). This is in contrast to the expected exponential decrease predicted in the Sec.3.3.3 (see Fig. 3.11). Such increasing residual pump power was often found to be a symptom of poor cavity alignment and coincident with low efficiency.

To improve pump depletion and therefore laser efficiency, a new procedure was adopted for cavity alignment. The concentric circular fringes of the pump radiation resulting from the formation of a Fabry-Perot interferometer by the DRL cavity mirrors were observed using a laser beam profiler placed behind the output coupler and provided a good indication of alignment between the cavity and pump beam axes. The residual pump power was examined in this case and found to better follow the expected decrease, as shown in Fig. 4.2(a). Though the threshold in this case rose slightly to 15.6 W, the slope efficiency and overall conversion
efficiency increased to 60% and 37% respectively (as illustrated in Fig. 4.2(b)). The maximum output Stokes power obtained was 14 W from 39 W pump power.

The beam quality of the Stokes output at different Stokes powers was measured by focusing the Stokes beam by a convex lens of focal length 250 mm. The beam radius ($\omega$) was recorded at different positions ($z$) by a beam profiler which was translated (across this focussed beam). The $M^2$ was calculated using

$$\omega^2 = \omega_0^2 \left(1 + \frac{M^4 \lambda^2}{\pi^2 \omega_0^4} (z - z_0)^2\right)$$  \hspace{1cm} (4.1)$$

where $\omega_0$ is the beam radius at the focus position at $z_0$. Beam quality factor ranging from 1.0 at low Stokes power to 1.2 ± 0.04 at maximum Stokes power were determined, thus confirming that the DRL operated almost completely in fundamental transverse mode (as shown in the inset of Fig. 4.3(a)). Fig. 4.3(a) shows the output beam quality measurement at 14 W Stokes power. Stokes wavelength tuning was achieved by varying the DFB setpoint.
4.1 Narrow-line width pumped DRL

Figure 4.3: (a). Measurement of beam quality of the Stokes beam at maximum Stokes power. The inset shows the Stokes beam profile at maximum Stokes power. (b). Output Stokes wavelength tuning versus DFB seed laser temperature.

temperature, which tuned the pump wavelength from 1062.85 to 1065.62 nm at a tuning rate of 80 pm/K. The tuning range was limited by the safe operating temperature range of the DFB laser. The Stokes output wavelength was thus, varied from 1238.12 to 1241.86 nm at a tuning rate of 117 pm/K as shown in Fig. 4.3(b).

4.1.3 Longitudinal mode structure

The FSR of the DRL cavity is calculated to be 1.3 GHz (optical cavity length = 113 mm, including 8 mm long diamond with refractive index at 1240 nm is 2.388). Therefore, approximately 35 longitudinal modes fit within the FWHM of the Raman gain spectrum of diamond (45 GHz). Using the scanning Fabry Perot interferometer, SLM was successfully observed for up to 4 W of Stokes power depending on the alignment and the cavity length. In general, it was found that cavity alignments that yielded high slope efficiency and low residual pump power (due to good pump-Stokes mode overlap) were also likely to lead to the highest SLM output power. For Stokes powers higher than 4 W, mode hopping was observed and subsequently the appearance of multiple lasing cavity modes as shown by the FPI spectrum in Fig. 4.4(b). For Stokes powers above 10 W, the mode spectrum was highly multimode (see Figs. 4.4(c) and (d)). Since the modes lying outside the FPI FSR overlapped on the modes in the same measured time interval, it was difficult to determine the number of lasing longitudinal modes in Figs. 4.4(c) and (d).
Figure 4.4: Fabry Perot scans for (a) 1 W, (b) 6 W, (c) 10 W, and (d) 16 W of Stokes power.

The stability of the Stokes centre wavelength was also monitored using the laser spectrum analyzer. During the SLM operation, the Stokes wavelength fluctuated in the range of 80 MHz (1 standard deviation) over several tens of seconds, as illustrated in Fig. 4.5(a). However, when the laser was operating in multimode regime, the fluctuations were in the range of tens of GHz, as shown in Fig. 4.5(b), which is of the order of the diamond Raman linewidth. The reasons for the multimode behaviour are discussed in Sec. 4.2.

4.1.4 Analysis and discussion

The demonstration of the 4 W SLM DRL, operating 80% over threshold, is consistent with the concept of spatial hole burning free Raman gain described in Chapter 2. SLM operation was achieved in a long standing-wave resonator, with the gain medium placed at the midpoint. To the authors knowledge, there have been no reports of similar SLM lasers based on inversion media operating at that pumping level and output power.

Fig. 4.6 shows the longitudinal modes under the diamond Raman bandwidth. Considering the close spacing of the longitudinal modes (1.3 GHz), the gain difference between the
mode at the Raman gain maximum and the second mode is about 0.3%, the DRL is expected to show multiple longitudinal modes much closer to the threshold than observed experimentally if spatial hole burning is present. In order to more quantitatively contrast the Raman laser SLM performance with an inversion laser, the threshold prediction for a second longitudinal mode is calculated using the theory introduced in Sec. 1.1.2. The critical parameters are $d$, $n$, $l$, $\Delta \lambda$ and $\lambda_0$ which denote the gain medium length, refractive index, absorption depth, FWHM of the laser transition and first longitudinal mode wavelength respectively. Since the pump is double-passed in the cavity, twice the length of diamond is taken as the absorption depth $l$, which is 16 mm. The inversion laser transition linewidth $\Delta \lambda$ is substituted with the diamond gain bandwidth, which is 0.23 nm at 1239 nm ($\lambda_0$). The refractive index at 1239 nm is 2.39. By putting these values in Eq. 1.1 and Eq. 1.2, the condition for single-mode laser operation is obtained as $r(f) < 1.06$. It implies that the threshold of the second longitudinal

Figure 4.5: Time dependence of Stokes centre frequency during the (a) single-mode operation (b) multimode operation.
mode induced by spatial hole burning is attained for a pump power 6% above the single-mode threshold. Thus the pumping level for the SLM DRL (80% over threshold) is significantly higher than the predicted multimode threshold for an equivalent inversion laser. Based on this discussion, it can be deduced that the observation of SLM in the free-running DRL is attributed to the spatial hole burning free nature of the Raman gain.

### 4.2 Thermal effects

During the DRL experiments, several phenomena were observed that are pertinent to stable and controlled SLM operation and are described in the following Sections.

#### 4.2.1 Variation of Raman frequency on diamond temperature

Monitoring of the Stokes output spectrum using the OSA (described in Sec. 3.4.3) showed that the Stokes wavelength decreased with increasing pump power. At the same time, it was also noted that the copper mount on which the diamond was placed was heating up significantly by more than 100 K. These observations motivated the simultaneous measurement of the temperature of the diamond and the variation of the Stokes wavelength. The Raman frequency shift was calculated for each pump power by obtaining the difference between the pump and Stokes wavelengths at each of these pump powers. It was also noted that the pump
4.2 Thermal effects

centre wavelength also decreased slightly with pump power (1.56 GHz over the full range), hence this was also factored into the calculation. The Raman shift was found to vary by about 35 GHz for an increase in diamond temperature of approximately 100 K.

Using the relation \[ \omega_R(T) = \omega_R(0) - \frac{C}{(e^{D(h\omega_R(0)/kT)} - 1)}, \]
which calculates the shift in Raman frequency as a function of temperature \( T \), a change in diamond temperature of 1 K leads to a change in Raman shift of 0.3 GHz. The values of the parameters \( \omega_R(0) \) denoting the Raman shift at 0 K, \( C \) and \( D \) which are the free parameters, are taken from the reference as 1333 cm\(^{-1}\), 61.14 cm\(^{-1}\) and 0.787 respectively. \( h \) and \( k \) are Planck’s and Boltzmann’s constants respectively. Since the measured variation in Raman shift is 35 GHz for a temperature change of 100 K, the observed change in Raman shift is consistent with the calculated value.

![Graph](image)

Figure 4.7: Raman frequency (in wavenumbers) calculated from the pump-Stokes frequency difference for operation without and with the chopper (black and red), and with the chopper and temperature controller (TC) (blue).

This effect could be minimized by better controlling the diamond temperature. Chopping of the pump beam and the use of feedback-controlled Peltier element (placed between the plates of the copper mount) was used to stabilize the mount and diamond temperature to within 0.5 K. This reduced the variation in the Stokes wavelength, and hence the Raman shift, to within 0.08 cm\(^{-1}\) (2.4 GHz) over the investigated pump power range. Fig. 4.7 illustrates the variation in the Raman shift with increasing pump powers for three cases discussed here.
4.2.2 Variation of Stokes power on cavity length

![Graph showing behavior in Stokes power during cavity shortening and lengthening](image)

Figure 4.8: Behaviour in the Stokes power during (a) during cavity shortening and lengthening; (b) increasing and decreasing of the pump wavelength (pump power = 17.2 W, Stokes power = 1.8 W).

When the cavity length was scanned gradually by several microns using the PZT stage, the output Stokes power was seen to follow periodic fluctuations with contrasting behaviour observed depending on whether the cavity was shortened or lengthened (as shown in Fig. 4.8(a)). During shortening, periodic sharp dips in Stokes power were observed between gradual rises in Stokes power whereas for lengthening sharp peaks were seen between intervals of zero Stokes power. The oscillations were typically irregularly spaced with an average period of approximately 0.5 μm. Similar behaviour was reproduced when the pump laser wavelength was tuned up or down as illustrated in Fig. 4.8(b). The behaviour for an increasing wavelength was similar to cavity shortening, whereas decreasing wavelength was similar to cavity lengthening.

Fig. 4.9 shows the periodic behaviour for the pump and Stokes powers for several ramp rates applied to the PZT. The scan range for each ramp rate is 6 microns and the same number of peaks and dips were observed for a given scan distance for slow scan rates (< 6 microns/sec). When the cavity length was increased, the spacing between the sharp peaks was about (550 ± 30) nm corresponds to approximately half the pump wavelength as the laser acts as a scanning Fabry Perot interferometer. Owing to the low-finesse at the pump wavelength and high-finesse at the Stokes wavelength, the pump peaks and dips were less
4.2 Thermal effects

Figure 4.9: Periodic peaks and dips in pump (blue) and Stokes (wine) intensity at different ramp frequencies (red) applied externally to the PZT stage (pump power = 20 W, Stokes power = 2.5 W). The scan range is 6 microns in each case.

pronounced than for the Stokes. Moreover, the peaks and dips of both the pump and the Stokes powers were less pronounced as the scan rate was increased. For slow rates, the pump and the Stokes powers slowly increase over a larger time duration at the turning points of the scan. To check whether this behaviour is due to hysteresis in the PZT, the experiment was repeated for pump powers below the Raman lasing threshold. This waveform showed pump transmission fringes, thereby confirming that the distinct oscillatory behaviour (sharp peaks and dips) is a cavity resonance effect associated with Raman lasing.

4.2.3 Analysis and discussion

The heating of the diamond through SRS and impurity absorption leads to changes in the optical cavity length $\Delta L$ through the thermo-optic effect and thermal expansion, and is given by

$$\frac{\Delta L}{\Delta T} = d\left(\frac{dn}{dT} + n\alpha\right)$$ (4.2)
The values of the thermo-optic coefficient \( (dn/dT = 15 \times 10^{-6} \text{ K}^{-1}) \) and thermal expansion coefficient \( (\alpha = 1.1 \times 10^{-6}) \) are taken from Table 2. Therefore, \( \Delta L/\Delta T = 140 \text{ nm/K} \) for the diamond of length \( d = 8 \text{ mm} \). Given that laser action alters \( \Delta L \) through heating, it is reasonable to expect that there is coupling between the Stokes power and cavity resonance. Specifically, it is proposed that a primary effect is the pump wavelength comes into and out of resonance as the cavity is tuned.

In order to verify this, a numerical model was developed to solve the cavity enhancement of the pump and the rate equations of the pump and Stokes powers as a function of the scanned mirror separation. The mirror scan is modelled as a triangular ramp function. The optical cavity length due to temperature is calculated using Eq. 4.2. The temperature rise \( (\Delta T) \) is calculated from the relation \( Q = mC\Delta T \), where \( Q, m \) and \( C \) denote the heat deposited, mass and specific heat of diamond respectively. The heat deposited \( (Q) \) is calculated from the relation given in Sec. 3.2.1 (contribution of impurity absorption is ignored). The Stokes power is calculated by the coupled rate equations of the pump and the Stokes. The resonant enhancement of the intracavity pump power \( (P_{\text{int}}) \) is calculated from the following equation [114]:

\[
P_{\text{int}} = \frac{P_{\text{inc}}}{P_{\text{inc}}} \left[ (1 - \sqrt{R_iR_o})^2 + 4\sqrt{R_iR_o} \sin^2(2\pi L/\lambda_p) \right]^{-1}.
\] (4.3)

where, \( P_{\text{inc}}, R_i, R_o, L \) and \( \lambda_p \) are the incident pump power, input coupler reflectivity, output coupler reflectivity, cavity length and pump wavelength respectively. Based on Eq. 4.3 with \( R_i = 2.8\% \) and \( R_o = 99.9\% \), the intracavity pump power varies between 73\% and 144\% of the incident power as the cavity length was scanned.

Fig. 4.10 shows the modelled transmitted pump behaviour when the cavity length is ramped at the scan rate 0.14 microns/sec. Owing to the absence of thermally induced cavity length below the threshold (Fig. 4.10(a)), the effective cavity length follows the linear progress of the external cavity length introduced by the ramp and the pump follows the Airy function, as given by Eq. 4.3 which agrees well with the experimental observation. It can be seen from the Fig. 4.10(b) that the change in effective cavity length above Raman lasing threshold is not a linear progression. Instead, the effective cavity length which is a combination of thermally induced cavity length due to heat deposition and physical cavity length has a step-wise progression. This effective cavity length modifies the dynamics of the intracavity pump power.
4.2 Thermal effects

Figure 4.10: Modeled intracavity pump and Stokes power for (a) pump power below Raman lasing threshold, (b) pump power and Stokes power above Raman lasing threshold, at a scan rate 0.14 microns/sec.

which in turn affects the Stokes power. Due to this coupling, the pump and the Stokes power show distinct oscillatory behaviour similar to experimental observation.

Figure 4.11: Simulated resonance enhancement of pump power and Stokes power above Raman lasing threshold at a scan rate of 14 microns/sec.
The simulation also agrees well with the experimental results observed for the faster scan rate as illustrated in Fig. 4.11. In this case, as the external cavity length changes rapidly, the stepwise progression of the effective cavity length is less pronounced as the heat deposition in the crystal is reduced with the increase in scan rate and hence, the pattern of the pump and the Stokes powers are not as well-defined as in the case of low scan rates. Based on the good qualitative agreement between this model of thermal effects and experimental observations, these are believed to be the primary cause of the peculiar periodic behaviour of the pump and Stokes powers.

The distinct oscillatory trend of the pump and the Stokes power observed during shortening and lengthening of the cavity as shown in Fig. 4.9 can be explained as follows. As the pump wavelength comes into resonance, the intracavity pump power increases leading to an increase in the intracavity Stokes power. The higher Stokes generation gives rise to increased heat deposition in the diamond, leading to an increase in the optical cavity length via the thermo-optic effect and thermal expansion. This increase in optical cavity length combined with the increase in external cavity length reinforces the increase in effective cavity length, due to which the pump wavelength rapidly shifts out of the cavity resonance. Thus, the Stokes power falls off sharply to zero and remains in the same state until the pump reaches the next cavity resonance, at which point the process repeats. This results in the sharp peaks of Stokes power followed by longer periods of zero Stokes power seen in Fig. 4.9. The observed period of oscillation is consistent (approx. 0.5 micron) with half pump wavelength borne out by the model.

In contrast, during shortening of the cavity, the pump and the Stokes powers increase as the pump comes into resonance, leading to increased thermal loading in the diamond. The increase in optical cavity length is counterbalanced by the active cavity length shortening which retains the pump wavelength in the resonance for a longer duration. However, further shortening of the external cavity length overcomes this balance, pushing the pump wavelength out of cavity resonance and leading to the laser falling below threshold, which is revealed as sharp dips in power in the Fig. 4.9. The absence of Stokes power results in zero thermal loading in diamond, reducing the optical cavity length and bringing the pump wavelength
rapidly back into resonance, at which point the cycle repeats.

Thus, in the present system, output power instability is attributed to the coupling between the Stokes power and optical cavity length via the thermo-optic effect and thermal expansion of diamond. This coupling mechanism is also responsible for the observation of mode hopping as a small change in the cavity length causes the modes to sweep through the gain. As the output power increased, the modal stability also decreased which resulted in the multimode operation. A similar thermally induced optical cavity length effect has also been observed in singly and doubly resonant OPOs \[115, 116\] with producing analogous asymmetric power patterns as a function of cavity length. Although these lasers used a thermal self-locking technique to stabilize the output frequency, they suggested active stabilization of the cavity length using feedback techniques for long-term stability. Thus, it is also critical for active cavity length stabilization in the DRL to improve the frequency stability and to achieve higher SLM output powers.

4.3 Spectral effects in a high Q resonator

![Stokes power versus incident pump power for the output coupler (T = 0.0096% at Stokes wavelength).](image)

Figure 4.12: Stokes power versus incident pump power for the output coupler (\(T = 0.0096\%\) at Stokes wavelength).

As part of the SLM laser characterization experiments, an output coupler with reduced transmission (\(T = 0.0096\%\) at Stokes wavelength) was used. This DRL had a Raman lasing threshold at 17 W and the Stokes power increased linearly with a 2.2% slope efficiency as shown in Fig 4.12. As expected, the low output efficiency resulted from the low output
coupling relative to input coupler losses.

Upon measuring the residual pump power dependence on pump power it was found that, apart from a small decrease just above the threshold, it increased at a high rate as shown in Fig. 4.13(a). Interestingly, when the output spectrum was simultaneously monitored with this measurement, another spectral line separated from the SRS line by 0.36 nm (70 GHz) was observed, which coincided with the observation of increasing residual pump powers as illustrated in Fig. 4.13(b). A peak at a similar spacing was observed in a previous DRL [75] (72 GHz) which was identified as the backward stimulated Brillouin scattering (SBS) frequency shift of diamond for a longitudinal acoustic mode.

![Figure 4.13](image)

Figure 4.13: (a) Residual pump power versus incident pump power and the threshold is indicated by the vertical dashed line. (b) OSA spectrum showing the second output line attributed to SBS.

In order to confirm that the observed additional spectral line is due to SBS, the Brillouin frequency shift from 1239 nm was calculated. The Brillouin frequency shift ($\Delta \omega_B$) from an excitation frequency ($\omega_p$) is given by

$$\Delta \omega_B = 2.\omega_p.n.(v_s/c)\sin(\theta/2)$$  \hspace{1cm} (4.4)$$

where $\theta$, $\omega_p$, $v_s$, $c$ and $n$ are the scattering angle, Stokes frequency, velocity of the acoustic wave, speed of light in vacuum and the refractive index respectively [117]. For the relevant case here of backscattered SBS, $\theta = \pi$. The velocity of sound in CVD diamond is related to the combination of the elastic moduli ($S$) and density of diamond ($\rho$) as, $v_s = \sqrt{S/\rho}$. 
Following Ref. [75], the velocity for the longitudinal acoustic mode along \(<110>\) axis is taken as \(\sqrt{c_{11} + c_{12} + 2c_{44}}/2\rho \text{ cm/s} \) [117]. By putting the values of \(c_{11}, c_{12}, c_{44}\) and \(\rho\) as \(10.764 \times 10^{12}, 1.252 \times 10^{12}, 5.774 \times 10^{12}\) and \(3.515 \text{ g/cm}^3\) respectively, in the above relation, the velocity of sound is calculated to be 18,308 m/s respectively. Using this value in the Eq. 4.4, the Brillouin frequency shift is calculated to be 2.35 cm\(^{-1}\). As the calculated shift is consistent with the experimentally observed separation of 2.35 cm\(^{-1}\) (70 GHz), it is confirmed that the observed spectral line is SBS.

The intracavity Stokes intensity in the present case of the higher finesse cavity was approximately doubled compared to the setup with the other output coupler, thus enhancing the probability for SBS generation. The poor slope efficiency (along with the increase in the residual pump power) is attributed to the cross-cascaded SBS shift by analogy with the cross-cascading in KYW reported in Chapter 3. Although SBS was occasionally observed, the threshold for SBS appeared to be highly sensitive to cavity alignment and thus could be suppressed with a minor alignment adjustment. Nevertheless, SBS may pose a challenge for achieving an SLM DRL, particularly when the intracavity Stokes intensity is very high.

4.4 Conclusion

The spatial hole burning free nature of the Raman gain enabled SLM operation in a standing-wave DRL despite the cavity mode spacing being 35 times smaller than the diamond Raman gain-width. SLM operation up to 4 W of Stokes power was achieved. At higher output powers, the DRL operated in a multimode state owing to the coupling between the Stokes and the optical cavity length via the thermo-optic effect and thermal expansion. Thus, precise control of the diamond temperature as well as active stabilization of the cavity length using conventional feedback mechanisms appears to be necessary for power scaling and stabilization of the SLM laser. SBS was also observed in the DRL with low output coupling and may present a challenge for the SLM operation.

The results are published in O. Lux, S.Sarang et al. "Intrinsically stable high-power single longitudinal mode laser using spatial hole burning free gain.” Optica 3.8 (2016): 876-881. Chapter Contributions: The experimental work was jointly conducted and analysed by the author and Dr. Oliver Lux. I acknowledge the assistance of Dr. Oliver Lux in modelling and analysing the thermal effect results.
Single longitudinal mode diamond Raman laser
Single-longitudinal-mode continuous-wave lasers operating in the wavelength range from 1400 nm to 1600 nm are important for remote sensing applications where the laser beams are propagated through the atmosphere and there is risk of indirect exposure to the laser beam, since wavelengths longer than 1400 nm (commonly, but somewhat misleadingly, referred to as "eye-safe") are strongly absorbed by the front of the eye, reducing exposure to the retina. Thus, maximum permissible exposures to this laser radiation are much higher than at other wavelengths in near-IR. These wavelengths also coincide with the absorption transition of key molecular gas species in the atmosphere. Erbium-doped fiber lasers, Er:YAG lasers and OPOs are important current technologies operating in this wavelength regime, which is used for LIDAR applications [118, 119]. However, these sources have limitations and disadvantages with respect to the pumping technology, output power and beam quality. CW diamond Raman lasers have already proved to be an efficient method for frequency conversion [74]. Diamond is an interesting alternative for generating eye-safe wavelength at 1485 nm via cascaded Raman shift from first Stokes wavelength at 1240 nm.
Thus far there have only been two reports of cascaded CW crystalline ECRLs \[120, 121\]. A second-Stokes CW barium nitrate ECRL operating at 576.7 nm was reported in 2009, but achieved a maximum conversion efficiency of 0.2\% \[120\]. Very recently, building on new modelling of cascaded-Stokes lasing in CW ECRLs, a second-Stokes CW DRL at 1485 nm was demonstrated with a conversion efficiency of 44\% \[121\]. This advance provides a new opportunity to explore the potential for SLM operation of an efficient second Stokes DRL.

The aim of this chapter is to investigate and achieve SLM operation of a 1485 nm CW external cavity DRL through cascaded Stokes lasing from 1064 nm via two Raman shifts. In order to investigate the effect of spectrally-selective cavity elements on the stability of the SLM Stokes output, a volume Bragg grating is employed as an output-coupler mirror for the second Stokes. The suitability of this laser for remote sensing applications is further investigated by performing water vapour detection measurements.

### 5.1 Second Stokes diamond Raman laser

#### 5.1.1 Cavity design

The theory of cascading used for the description of the second-order Stokes DRL in this section is adapted from the ref. \[121\], which is similar to the theory outlined in Chapter 3 for cross-cascading, except that the same phonon mode is participating in the two-step Raman conversion. For the laser operating in the CW regime, it is necessary to minimize the incident pump power required to attain second Stokes threshold. To reduce the second Stokes threshold, very high output mirror reflectivity at the first Stokes wavelength \((R=99.9\%)\) is used to enhance the intracavity first Stokes power which is the pump for the second Stokes. The Raman lasing threshold of this laser is determined by using the equation using \[121\]

\[
\frac{\bar{P}_s}{A} = \frac{-\ln R_{2s} + 2\alpha L}{g_{2s} A l} \quad (5.1)
\]

where, \(\bar{P}_s, R_{2s}, \alpha, l, g_{2s}, A\) are average intracavity first Stokes power, output mirror reflectivity at second Stokes, absorption loss of the crystal, length of diamond, gain coefficient of the phonon mode and the effective area of the interaction between the pump and Stokes beams, respectively. It can be seen from the Eq. \[5.1\] that the first Stokes power is constant with pump
power above the second Stokes lasing threshold which is consistent with the cross-cascading theory developed in Chapter 3.

Following the equations from the reference, the residual pump power $P_p(2l)$ after the onset of second Stokes is determined

$$P_p(2l) = P_p(0)(R_{2s})\frac{1}{\eta_{2s}} e^{\frac{2\alpha l}{\eta_{2s}}}$$  \hspace{1cm} (5.2)

where $\eta, \eta_{2s}$ are the quantum defect of the first Stokes and second Stokes processes respectively. The residual pump power increases with incident pump power ($P_p(0)$). The generated second Stokes power $P_{2sout}$ is obtained [121] by

$$P_{2sout} = \eta\eta_{2s}(P_p(0) - P_{pth}(0))(1 - R_{2s}\frac{1}{\eta_{2s}} e^{\frac{2\alpha l}{\eta_{2s}}})$$  \hspace{1cm} (5.3)

where $P_{pth}$ is the intracavity pump power at the threshold of the second Stokes generation.

If the output mirror reflectivity at the second Stokes wavelength is very high, the threshold for the second Stokes will be close to the first Stokes threshold which results in a poor pump conversion to the first Stokes and consequently, a low second Stokes conversion as shown in Eq. 5.3. Therefore, to achieve appreciable second Stokes power, the output mirror reflectivity at the second Stokes wavelength must be low [121].

![Figure 5.1: Simulated plot of residual pump power, first Stokes power and second Stokes power versus incident pump power.](image)
The values of the $\alpha$ and $g_{2s}$ are taken to be 0.375% and 10 cm/GW 121, respectively. Since $\eta = 1064/1240$ and $\eta_{2s} = 1240/1485$, the quantum-limited efficiency $\eta \eta_{2s} = 72\%$. The reflectivity of the output mirror at 1240 nm and 1485 nm is taken to be 99.99% and 70% respectively. The spot radius at the centre of diamond for the pump, first Stokes and second Stokes is taken to be as 33 $\mu$m, 38 $\mu$m and 40 $\mu$m, respectively. By putting these parameters into the Eqs. 5.1 and 5.2 the threshold for the double-pass pumped second Stokes DRL is calculated to be 6.7 W and the laser slope efficiency 16%. Fig. 5.1 shows the simulated trend of the residual pump power, first Stokes and second Stokes powers with incident pump power, which shows the characteristic cascading behaviour seen previously in Chapter 3 as the input power is sequentially increased above the first and second Stokes thresholds.

### 5.1.2 Experimental arrangement

![Figure 5.2: Schematic diagram of a second Stokes DRL; LPF – low pass filter.](image)

The experimental setup (see Fig. 5.2) is essentially the same as the narrow-linewidth pumped DRL presented in Chapter 4. The cavity mirrors were exchanged to those suitable for efficient second-Stokes generation using the reflections specifications shown in Table 5.1. The new output coupler had a 75 mm radius of curvature and the cavity length was altered to 125 mm long cavity to produce the suitable Stokes mode size. The diamond was anti-reflection coated for the pump as well as first and second Stokes wavelengths ($R = 1.1\%$ at 1485 nm) to reduce reflection losses. It was placed at a distance of 46 mm and 71 mm from the input and output couplers, respectively.

A convex lens of focal length 75 mm with anti-reflection coatings at the second Stokes wavelength positioned at 25 mm from the output coupler was used to collimate the output.
5.1 Second Stokes diamond Raman laser

Table 5.1: Input and output coupler reflectivities.

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Input coupler (R%)</th>
<th>Output coupler (R%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1064</td>
<td>0.65</td>
<td>99.997</td>
</tr>
<tr>
<td>1240</td>
<td>99.95</td>
<td>99.99</td>
</tr>
<tr>
<td>1485</td>
<td>99</td>
<td>70</td>
</tr>
</tbody>
</table>

beam. A long-pass filter (Thorlabs, FEL1300) was used to separate the second Stokes wavelength ($T = 84\%$) from the pump and the first Stokes wavelength. Since the second Stokes wavelength was outside the useful wavelength range of the scanning Fabry-Perot interferometer employed in the earlier experiments, the longitudinal mode spectrum was investigated using the laser spectrum analyzer (Bristol instruments, model 771A-NIR, see instrument details on Sec.3.3.3, Chapter 3).

5.1.3 Experimental results

Output power and efficiency

![Graph](image)

Figure 5.3: The first Stokes power (red), second Stokes power (blue) and the residual pump power (black) versus incident pump power.

The DRL had a threshold of 4 W for the first Stokes at 1240 nm and 6 W for second Stokes at 1485 nm, which are good agreement with the predicted values. The second Stokes output power increased linearly with a slope efficiency of 25\% and the maximum power was 7 W at 34 W incident pump power, as shown in Fig. 5.3. The measured value of slope
efficiency was close to the predicted value (16%). The trend of the residual pump, first Stokes and second Stokes powers are in good agreement with the model presented in Section 5.2.1 (see Fig. 5.1). By varying the DFB laser temperature, the pump wavelength and thus first Stokes wavelength were tuned which consequently tuned the second Stokes wavelength from 1483 nm to 1488 nm corresponding to about 750 GHz frequency range, as illustrated in Fig. 5.4.

Figure 5.4: Wavelength tuning of the second Stokes DRL by varying the DFB laser temperature.

**Longitudinal mode structure**

Figure 5.5: The second Stokes DRL operated in single-mode regime at second Stokes power of 0.1 W at DFB laser temperature of 28°C.

Up to second Stokes power of 0.1 W, a single spectral peak was observed on the laser spectrum analyzer which implies that the laser was operating in SLM regime (see Fig. 5.5). As
the Stokes power increased, multiple spectral lines were seen (see Fig. 5.6(a)) indicating that the operational regime shifted to multimode. The central wavelength stability was also investigated and it showed fluctuations of about 8 GHz, as shown in Fig 5.6(b), consistent with the diamond Raman gain bandwidth.

![Image](image_url)

Figure 5.6: The second Stokes DRL operated in multimode regime above 0.1 W. (a) Wavelength fluctuations of the order of 8 GHz. (b) Multiple peaks indicating multimode state.

It was explained earlier in Chapter 4 that thermally-induced optical cavity length changes destabilize the mode spectrum and hence, the multiple longitudinal modes were observed. In comparison to the first Stokes DRL, the thermal load in diamond is exacerbated in a second Stokes DRL. This is for two reasons. Firstly, the intracavity first Stokes intensity is higher (almost doubled due to the higher reflectivity of the output coupler at the first Stokes than the one presented in Chapter 4) which causes an increased impurity absorption. Secondly, the quantum defect of the second Stokes generation process also contributes to the heating of the diamond. This increases the coupling between the Stokes power and the optical cavity length via diamond thermal expansion and the thermo-optic effect leading to an increased rate of cavity destabilization than in first Stokes DRL and hence resulting in a lower maximum SLM power. The maximum SLM power in the second-Stokes laser may also be limited by the saturation in pump depletion as noted for the first Stokes laser in Chapter 4. The increase in the residual pump power at pump powers above the second Stokes threshold suggests that there will be increasing gain available to other longitudinal first-Stokes modes as pump power is increased. Thus, multiple longitudinal modes in the first Stokes is also likely to lead to
earlier onset of multimode operation of the second Stokes.

5.2 Second Stokes laser incorporating a VBG

Since the laser was operating in multimode regime, a wavelength selective element was selected to assist in stabilizing SLM. For this purpose, a reflecting volume Bragg grating (VBG) was chosen. A VBG is an optically transparent bulk material consisting of a periodic variation of the refractive index. It reflects only the wavelength ($\lambda_c$) which satisfies the Bragg condition which is dependent on the refractive index of the VBG material ($n_{eff}$) and grating period ($\Lambda$),

$$\lambda_c = 2n_{eff}\Lambda.$$  \hspace{1cm} (5.4)

VBGs offer many advantages over other intracavity optical filters such as etalons, as they have good optical and mechanical stability and can withstand high laser power which is useful for power scaling of the lasers. Although their wavelength bandwidth is larger than the etalons, the power in the higher diffraction orders in VBG is low and fewer orders are easy to be filtered out in comparison to etalons [122, 123].

However, in the present DRL, a VBG is not desirable as an intracavity filter since its incorporation increases the losses which are crucial for CW operation. Therefore, it was decided to use VBG as an output mirror, thereby modifying the cavity configuration into coupled cavity configuration. Coupled cavity lasers employing VBGs have been a popular technique to achieve narrow-linewidth and even SLM operation in conventional inversion lasers and OPOs [124, 125]. In this configuration, the VBG provides highly wavelength-selective feedback to improve discrimination against secondary laser modes.

5.2.1 VBG cavity design

As discussed previously in Section 5.2.1, for a better pump conversion to the first Stokes and to the second Stokes wavelength, a high output coupling at the second Stokes wavelength is used. Therefore, the VBG is specified for low reflectivity (56%) at the second Stokes wavelength. A VBG (OptiGrate Corp.) was 5 mm $\times$ 5 mm $\times$ 10 mm in dimension, made of photosensitive silicate glass with a diffraction efficiency (reflection) of 55.5% at 1486 nm at
normal incidence with a 100 pm (13.6 GHz) reflection bandwidth (FWHM) was obtained.

The reflectivity spectrum of the grating (as shown in Fig. 5.7) was obtained using

\[
 r = \frac{\sinh^2(l \sqrt{k^2 - \sigma^2})}{\cosh^2(l \sqrt{k^2 - \sigma^2}) - \sigma^2/k^2}
\]  

(5.5)

where \( l \) and, \( k \) and \( \sigma \) indicates the length of the VBG and coupling coefficients respectively. The coupling coefficients are defined as follows: 

\[
 k = \frac{\pi \delta n}{\lambda}, \quad \zeta = \frac{2 \pi \delta n}{\lambda}, \quad \sigma = \zeta + \delta, \quad \text{where,} \quad \delta n \quad \text{is the modulation of the refractive index.}
\]

The detuning \( \delta \) is the difference between the incident wavelengths \( \lambda \), and the central wavelength \( \lambda_c \) is

\[
 \delta = 2 \pi n_{eff} (1/\lambda - 1/\lambda_c).
\]  

(5.6)

Figure 5.7: Simulated plot for VBG reflectivity versus incident second Stokes wavelength with a maximum diffraction efficiency of 55.5% at 1486 nm (using \( \delta n = 4.55 \times 10^{-5} \)) with the mode spacing of the near-concentric Raman (shorter) cavity inside the spectrum (shown for comparison).

The values of parameters \( n_{eff}, \lambda_c, l \) are taken as 1.4448, 1486 nm and 10 mm respectively. It is clear from the figure that the bandwidth of the VBG is broad considering the number of cavity longitudinal modes lying within the bandwidth (approximately 11 modes). The reflectivity for wavelengths outside of the bandwidth is very low with the first side-lobes having only 4% diffraction efficiency, thereby providing higher discrimination to wavelengths.
outside the VBG bandwidth.

![Schematic diagram of the coupled cavity second Stokes DRL](image)

Figure 5.8: Schematic diagram of the coupled cavity second Stokes DRL; LPF –low pass filter, VBG –volume Bragg grating.

The near-concentric Raman cavity (described in Sec.5.1.2) was converted to a coupled cavity configuration by placing a VBG behind the output coupler at a distance of 105 mm, as shown in Fig. 5.8. Thus, the VBG acted as the output mirror ($T = 44.5\%$ at 1486 nm) for a coupled cavity of overall length 230 mm. The second Stokes beam was collimated by a $f = 75\text{ mm}$ convex lens before being incident on the VBG.

### 5.2.2 Longitudinal mode structure

![Normalized intensity vs. Wavelength](image)

Figure 5.9: At the second Stokes power of 0.5 W, a single peak is seen in the spectrum of the second Stokes Raman laser when the VBG is employed in the cavity.
The DFB laser temperature was adjusted to about 35\(^0\)C to tune the pump wavelength thereby altering the second Stokes wavelength to match the VBG resonance wavelength at 1486 ±0.1 nm (see Fig. 5.4). It can be clearly seen from the second Stokes spectrum that a single peak is visible (see Fig. 5.9) when the VBG was incorporated into the cavity as opposed to the multiple peaks shown in Fig. 5.6(a) when the VBG was not employed. Since the VBG provided a discrimination in the diffraction efficiency of about 0.5% to the longitudinal modes adjacent to the central mode, it acted as a spectral filter and a single mode was achieved for second Stokes powers up to 0.5 W, despite its broad bandwidth. It has to be noted that the DFB laser temperature was slightly adjusted within ±1\(^\circ\)C (but coincided with VBG resonant wavelength) to obtain the spectrum shown in the figure.

![Figure 5.10: (a). The centre wavelength of second Stokes DRL was monitored for over 5 mins when the VBG was employed (b). The wavelength fluctuations were within 43 MHz in the highlighted region of (a).](image)

The stability of second Stokes centre wavelength was also monitored. Fig. 5.10(a) shows a time series of the laser wavelength over 5 mins. As shown in the figure, the laser remained in the SLM regime for most of the time except for some larger fluctuations (multimode transition). During the last 1.5 mins, the fluctuations were within 0.32 pm (43 MHz) as illustrated in Fig. 5.10(b). However, in some measurements, after a few transitions between SLM to multimode state, the SLM regime was completely lost and cavity length adjustments (i.e. the position of output coupler and VBG) had to be performed to bring the laser back to SLM. The reasons for this switching between SLM and multimode regime were the same as described in the Section 5.2.3. So, active stabilization of the cavity length through feedback techniques
was found to be necessary in order to achieve high-power SLM operation for longer time durations.

Figure 5.11: Mode-hopping of the second Stokes DRL corresponding to twice the mode spacing.

The FSR of the inner cavity comprising of the input and output mirrors is 1.1 GHz for a cavity length of 125 mm. In the course of the analysis of the wavelength stability, the second Stokes DRL exhibited mode-hopping as shown in Fig. 5.11. As opposed to the first Stokes DRL, the spacing between these longitudinal modes was 14.1 pm (1.9 ±0.1 GHz) which corresponds to about twice the FSR of the inner cavity.

Figure 5.12: Histogram representing the mode-hopping by about 14 pm (a) when VBG was off-resonance, (b) when VBG was on resonance in the laser setup.
Following the observation of the "double mode-hop", a histogram representing the counts of the measured wavelengths in bins of 0.2 pm was produced from all the wavelength stability measurements carried out when the VBG was off-resonance and on-resonance, as shown in Fig. 5.12. The histograms show that the separation between the longitudinal modes is about 14 pm (1.9 GHz) confirming that the laser preferentially skips one successive longitudinal mode of the inner cavity.

### 5.2.3 Second Stokes mode spacing

This observation of double mode-hops led to a study into its cause. In the present case, the coupled cavity length comprising the input coupler, output coupler and the VBG acting as the output mirror is 230 mm and thus, the mode spacing of the coupled cavity is calculated to be 0.6 GHz. Therefore, the expected mode-hop spacing is approximately an integer multiple of the coupled cavity mode spacing as well as the inner cavity mode spacing (1.9 GHz ≈ 2 * \(\Delta\nu_{\text{inner}}\) ≈ 3 * \(\Delta\nu_{\text{coupled}}\)). However, since the double mode spacing was also observed when the VBG was off-resonance, this phenomenon could not be traced back to the coupling of the two cavities, although the peaks in the histograms are more pronounced in the resonance case. But due to the low reflectivity of VBG and the high intracavity losses introduced by the lens and the filter, the finesse of the coupled cavity is rather low (about 0.52 per round trip, excluding the reflection losses at the collimating lens) so that its influence on the mode spacing is only minor. Moreover, this explains the fact that the exact length of the coupled cavity was not crucial for the stable SLM operation. It is clear from the discussion here that the coupled cavity configuration is not a contributing factor for the observation.

Another possibility for the double mode-hop is through the seeding mechanism for the second Stokes generation which promotes certain longitudinal modes to oscillate in the cavity. Seeding can be either through spontaneous Raman scattering or the parametric process of Raman assisted four-wave mixing (FWM). As in the case of first Stokes laser, it is expected that the stimulated second Stokes scattering is initiated by the quantum noise associated with the spontaneous second Stokes scattering as in the case for the stimulated first Stokes scattering. However, if the initiating noise was provided by spontaneous scattering then it would seed all the cavity modes and the filtering of the longitudinal modes would not occur.
The other seeding process is through FWM which is considered a dominant seeding mechanism for the higher Stokes order according to Ref. [127]. FWM is a parametric third-order nonlinear process which involves three waves to generate a fourth wave at a different frequency. FWM generation near the second-Stokes consists of mixing two first Stokes fields (of frequency $\nu_1$) with one pump field (frequency $\nu_0$). Thus, the second Stokes component is generated by, $\nu_2 = 2\nu_1 - \nu_0 = \nu_1 - \nu_r$, where $\nu_r$ is the Raman frequency [128].

Since it is a parametric process, the efficiency of the process is strongly dependent on the phase matching or momentum conservation condition. However, in the case of second Stokes DRL, the three optical fields –pump and first Stokes field and the second Stokes field are collinear. So, in this case, the seeding mechanism can only be non-phase-matched FWM.

The pump and the first Stokes fields are in resonance with the inner cavity and therefore, the first Stokes frequency is an integer multiple of the mode spacing of the inner cavity $\Delta \nu$, given by $\nu_1 = \nu_0 - n\Delta \nu$. Since second Stokes frequency is $\nu_2 = 2\nu_1 - \nu_0$, which can be written as $\nu_0 - 2n\Delta \nu$ due to energy conservation. Clearly, the first Stokes seeds only the second Stokes frequency spaced by the double the inner cavity mode spacing. So, the spacing between two successive second Stokes modes will be, $\nu_2' - \nu_2 = (2\nu_1' - \nu_0) - (2\nu_1 - \nu_0) = (\nu_0 - 2(n+1)\Delta \nu) - (\nu_0 - 2n\Delta \nu) = 2\Delta \nu$, which is twice the mode spacing of the inner cavity. Therefore, it can be concluded that the FWM seeding mechanism is responsible for the observation of the double mode jumps in the Raman laser.

### 5.2.4 Discussion

The above demonstration has shown that external cavity configuration is suited for higher order Raman generation and thus extends the achievable wavelength range. Further extension to visible and UV wavelengths may also be obtained using the intracavity frequency doubling and tripling [90]. The observation of double mode spacing may be beneficial in realizing SLM in higher-order Raman laser which is explained by the following mode spacing concept. As the spacing between the second Stokes modes is increased by twice the mode spacing when seeded by FWM, it is presumed that the spacing between the consecutive modes of $n^{th}$ order Stokes Raman lasers will be $n$ times the cavity mode spacing. This signifies that
5.3 Remote sensing application–water vapour detection

the number of longitudinal modes of higher-order modes within the Raman gain profile will be reduced by the Stokes order when initiated by FWM. Furthermore, better discrimination of modes (either by Raman gain or the intracavity frequency selective elements) can be accomplished due to a lesser number of modes in a higher-order Raman laser.

5.3 Remote sensing application–water vapour detection

Figure 5.13: Transmission spectrum of ammonia (red) and water vapour (black) from HITRAN database (temperature=293 K, pressure=14 mbar, path length=3.8 m). The dashed line indicates the strongest absorption line for ammonia and water vapour at 1486.134 nm and 1486.713 nm, respectively.

As mentioned in Chapter 1, LIDAR applications require high-power SLM laser sources with good beam quality and long-term stability. In order to determine the feasibility of second-Stokes laser as a reliable LIDAR transmitter, an absorbing gas species in the tuning range of the laser was investigated under laboratory conditions. In this tuning range, there are absorption lines of the two gas species — water vapour and ammonia, as shown in Fig. 5.13. As the detection of water vapour was simple to be conducted in the lab environment, it was chosen for the absorption measurements in this experiment. The strongest absorption line of water vapour in the laser wavelength range is at 1486.713 nm (6726.2488 cm$^{-1}$) [129].

Water vapour is a significant greenhouse gas in the atmosphere which traps more heat than carbon dioxide accounting for about 60% of the warming effect [130]. It is also an important
component of Earth’s climate systems. So, monitoring of water vapour concentration in the atmosphere is crucial for monitoring climate changes.

### 5.3.1 Experimental arrangement

![Experiment Setup Diagram]

Figure 5.14: Setup for detecting water vapour; LPF –low pass filter, VBG –volume Bragg grating, PD –InGaAs photodiode, M1,M2 –reflecting mirrors, WM –wedge mirror.

Fig.5.14 illustrates the setup for water vapour detection based on absorption spectroscopy. For this purpose, the light beam was divided into two parts using a wedge –one was sent to examine the absorbing gas and another was the unattenuated part referred to as reference signal. The weak part was sent to the laser spectrum analyzer to record the output second Stokes wavelength variation. A photodiode placed near the coupling lens of the spectrum analyzer provided the reference signal. The major portion of the second Stokes beam was propagated through the lab over a path length of a few meters. This transmitted beam was then detected by a second InGaAs photodiode. The ratio between the transmitted and reference signal was measured over a time-scale simultaneously with the wavelength variations to obtain the absorption spectrum. A mechanical chopper and a lock-in amplifier was used to improve signal to noise ratio.

The resonance wavelength of the VBG was shifted by heating VBG using a temperature...
controlled oven (Ekspla TK1 with KK1). It enabled tuning of the resonance wavelength of VBG from 1486.0 nm to 1486.6 nm (with 1 pm accuracy) in order to achieve a wavelength stabilized Raman laser coinciding with the absorption lines of water vapour.

5.3.2 Experimental results

![Graph](image)

Figure 5.15: The measured transmission of the second Stokes DRL through the air along with a Voigt profile fit and a calculated transmission based on spectroscopic data from the HITRAN database [131].

The laser wavelength was tuned to 1486.713 nm. Since the laser wavelength lies outside the VBG resonance wavelength, it was difficult to maintain a stable SLM operation over longer time scales and hence the measurement error was large. The temperature and relative humidity in the lab were 20°C and 60% respectively. The partial pressure of water vapour was calculated using Tetens equation to be 14 mbar [132]. The measured transmission was averaged in wavenumber bins of width 0.02 cm\(^{-1}\). Comparing experimental data with the simulated spectra from HITRAN and Voigt fitting from calculations described in [133], the simulated values were within the error range of the measured values as shown in Fig. 5.15. The self-broadening and air-broadening caused the broadening of the absorption line with an FWHM of 0.15 cm\(^{-1}\) (4.5 GHz) and the minimum transmission of 75%. The good agreement
obtained between measured and calculated spectra shows the feasibility of this laser to be employed for water vapour detection.

5.3.3 Discussion

The results obtained for water vapour detection reveal a promising alternative to OPOs in satisfying the requirements as a LIDAR transmitter. The spectral range of the inversion lasers can be extended using Raman lasers with automatic phase matching, unlike in OPOs [134]. Another major drawback in OPOs is the thermal degradation of the beam quality as mentioned in Chapter 1. In contrast, Raman lasers can provide an output with diffraction-limited beam quality owing to the Raman beam cleanup [43, 45].

However, the stability of the VBG stabilized Raman laser over time was poor and the signal to noise ratio was also low. One of the reasons for poor frequency stability was that the laser operating wavelength was lying outside the VBG resonance wavelength and thus, retaining the SLM operation was difficult. Another limitation to maintain the SLM stability is the thermally-induced changes in the optical length of the diamond as described in Sec.5.1.3. Therefore, an active cavity length stabilization technique is recommended to compensate for these changes and enhance frequency stability for a longer duration.

5.4 Conclusion

A CW tunable SLM external-cavity second Stokes DRL emitting in the "eye-safe" wavelengths from 1483 to 1488 nm was demonstrated at output power up to 0.1 W when using a simple 2-mirror standing-wave cavity. The cavity was modified into a coupled-cavity configuration by incorporating a volume Bragg grating as the feedback element. The VBG enabled the laser to operate in SLM state at higher power (0.5 W) with frequency stability better than 50 MHz over 1 to 2 minutes. This frequency stabilized laser was successfully employed to detect water vapour and therefore, proving the competitiveness of Raman laser technology for LIDAR applications.

A double mode-hop was observed during the investigation of the second Stokes longitudinal mode spectrum. The study of this increased effective mode spacing showed that the
non-phase matched four-wave mixing which is the dominant seeding mechanism for second Stokes is responsible for the behaviour. This feature may be advantageous for realizing stable SLM in higher-order Raman lasers.

The main issue highlighted in this Chapter and in Chapter 4 for achieving stable SLM operation is the need of active cavity length stabilization to compensate for the optical cavity lengths induced by the thermal effects in diamond. Thus the next step is to select a locking technique to stabilize the laser cavity length to achieve high SLM output power over longer periods.


Chapter Contributions: The experimental work was jointly conducted and analysed by the author and Dr. Oliver Lux. I acknowledge the assistance of Dr. Oliver Lux in analysing the water vapour detection results.
Second Stokes SLM laser
Active cavity length stabilization

The DRLs presented in Chapters 4 and 5 demonstrated SLM operation but suffered from instability when attempting to scale to higher powers. The frequency stability during the SLM operation was not high (e.g., 80 MHz for tens of seconds reported in Chapter 4). To improve the frequency stability and increase the SLM output power, an active stabilization of the cavity length through a feedback technique is investigated in this Chapter. The work presented is aimed at providing active cavity length stabilization without adding additional components to the cavity that could deleteriously impact the efficiency of the high-Q resonator.

A variant of Hänsch-Couillaud method was chosen to stabilize the cavity by exploiting the crystal birefringence demonstrated in ref. [135]. The initial idea was to implement this method by using the (significant) in-grown stress birefringence of the diamond. However, it is shown that the locking may be achieved by another mechanism that is tentatively explained through a mechanism based on polarisation dependent pump depletion by SRS.
6.1 Cavity length stabilization technique

Wavelength stabilization of a SLM laser is achieved by feedback control of its cavity length. For this purpose, the laser wavelength $\lambda_L$ is continuously monitored with respect to a reference standard to determine the drift of the laser wavelength. This reference wavelength $\lambda_R$ is provided either by the transmission fringe of a stabilized Fabry-Perot interferometer [136], an atomic or molecular transition or a stabilized laser [137]. The light beam from the laser and the reference are sent to a wavelength comparator. The wavelength comparator produces an output signal proportional to the deviation of the laser wavelength from the reference wavelength. This output error signal is then sent to an electronic feedback controller which alters the laser cavity length via a piezo translator of the cavity mirror to minimize this error signal to zero and thus, ensures $\lambda_L = \lambda_R$. Depending on whether the reference wavelength corresponds to the maximum or side of the transmission peak of a stabilized Fabry-Perot interferometer, the locking is referred to as the top-of-fringe locking and side-of-fringe locking respectively.

![Figure 6.1: Schematic of laser wavelength stabilization [138].](image)

In the top-of-fringe technique, the laser wavelength is compared with the transmission peak of the reference FP interferometer. If the laser is simply locked to the maximum of the resonance peak, it is difficult to predict the direction of drift of the cavity length from the intensity measurement since a drift in both sides introduces the same change in the error signal. A common solution to the problem is to modulate the laser frequency or cavity length to create a modulation of the transmission fringe which is proportional to the derivative of the original signal. This determines the drift as the zero position of the derivative corresponds
to the maximum of the peak, while the sign (positive or negative) of the slope indicates the
direction of the drift as shown in Fig. 6.2(a). As a result, this locking technique requires
complex electronics to modulate the transmission peak [139].

Figure 6.2: (a). The locking signal of the top-of-fringe method calculated for a cavity having
an input mirror reflectance of 70%. Here, the laser is locked at the zero-crossing of the signal
corresponding to the maximum of the fringe. (b). The red crosses indicate the capture range.

On the other hand, the side-of-fringe method is simpler as the locking signal is obtained
without modulation. The signal-to-noise ratio of this locking technique is low as the method
is sensitive not only to fluctuations in laser frequency but also its intensity. The capture range
of a locking technique is defined as the highest frequency deviation the locking system can
tolerate and still be brought back to the lock-point. In the case of the side-of-fringe method,
the capture range is limited to one side of the peak, and is small, particularly for the narrow
cavity fringe of the high finesse cavities. The top-of-fringe method has generally a larger
capture range, extending to adjacent cavity modes on each side of the fringe as illustrated in
Fig. 6.2(b). Owing to these disadvantages of the side-fringe locking, the top-of-fringe is the
preferred method for stabilizing lasers.
6.1.1 Hänsch-Couillaud technique

The earlier section described locking techniques to stabilize lasers using stabilized FP cavities. These techniques can also be applied to stabilize enhancement cavities to single-mode pump lasers [140, 141]. A commonly used technique for stabilizing enhancement cavity is Hänsch-Couillaud (HC) method. It is a straightforward method top-of-fringe type locking without the need of a modulation. In this method, an element in the cavity produces different losses to orthogonal polarisations of the light beam in the cavity [139]. This change in the polarisation is then analyzed to generate a dispersion-shaped locking signal directly. Fig. 6.3 shows the schematic diagram of the HC method which is described as follows.

![Hänsch-Couillaud locking setup diagram]

Figure 6.3: Setup for Hänsch-Couillaud locking; IC–input coupler, OC–output coupler, PZT–piezoelectric translation stage, QWP–quarter-wave plate, BS–beam splitter, PBS–polarizing beam splitter, PD1 and PD2–photodetectors.

A polarizer is inserted into the cavity such that the polarisation axis of the incident light is at an angle with the transmission axis of the polarizer so that the pump beam can be decomposed into two orthogonal linearly polarized components. When a change in the cavity length pushes the pump beam ($I_i$) out of resonance with the cavity, it causes a relative phase shift between the orthogonal polarisation components because the component parallel to the transmission direction of the polarizer ($I_a$) experiences a frequency dependent phase shift,
whereas the perpendicular component \((I_b)\) is simply reflected by the input mirror without undergoing any phase change. This relative phase shift results in an elliptical polarisation which is analyzed by a detection system comprising a quarter-wave plate, polarizing beam splitter and two photodetectors (as shown in Fig. [6.3]). An electronic control system with a feedback loop then regulates the cavity length. The difference between the two signals detected by the photodetectors \((I_a\) and \(I_b)\) provides the locking signal for the HC scheme [139]

\[
I_a - I_b = I_i.2.\cos\theta.\sin\theta \frac{T_1.F.\sin\delta}{(1 - F)^2.4.F.\sin^2\delta/2},
\]

(6.1)

where \(\theta\) is angle between the transmission axis of the polarizer and the polarisation axis of the incident beam, \(\delta\) represents the phase mismatch in the two components due to the cavity length change, \(T_1\) is the transmittance of input coupler, \(F\) is amplitude ratio per roundtrip accounting for the intracavity losses and the two reflections at the mirrors which is given by the relation [142]

\[
F = \sqrt{R_1R_2(1-T)^2},
\]

(6.2)

where \(R_1\), \(R_2\) and \(T\) represents the input and output mirror reflectivities, and intracavity losses due to both absorption and reflection from the diamond respectively. As shown in Fig. [6.4], an asymmetrical locking signal similar to that in Fig. [6.2] is generated by this method but without the need for signal modulation and lock-in electronics.

![Figure 6.4: Calculated pump fringe and locking signal by the HC method for an input mirror reflectance of 70% for \(\theta = 45^\circ\). The lock point and the zero-crossing is depicted by the vertical and horizontal dashed line, respectively.](image)

Birefringence of the cavity elements is ignored in the HC scheme. However, a variant of this
scheme has been successfully adapted to stabilize cavities which manifest variations in the polarisation of the resonant light due to the birefringence in the crystal [135] or dielectric cavity mirrors [143] without needing a polarizer inside the cavity. Here, the birefringence induces the necessary polarisation dependent phase mismatch to generate an HC-type locking signal.

In this Chapter, it is aimed to use this variant of the HC method that uses birefringence to achieve stabilization. Even though diamond is an isotropic crystal, it often shows stress-induced birefringence due to grown-in defects during production [144]. CVD single crystal diamond used in the laser experiments typically have birefringences of the order of $10^{-5}$-$10^{-6}$ at near-infrared wavelengths. As illustrated in Fig. 6.5, the basic setup of the HC variant method is similar to the original (see Fig. 6.3) with a birefringent crystal replacing the polarizer. The difference in refractive indices of the crystal for e-ray and o-ray results in a phase difference, providing the locking signal which is detected as the difference between the intensities of the detector signals, $I_a - I_b = |E_a|^2 - |E_b|^2$ [135]. The amplitude of the detector signals ($E_a$ and $E_b$) is obtained by solving the Jones matrix for a rotated quarter-wave plate.

Figure 6.5: Setup for HC variant locking using crystal birefringence; IC–input coupler, OC–output coupler, PZT–piezoelectric translation stage, QWP–quarter-wave plate, BS–beam splitter, PBS–polarizing beam splitter, PD1 and PD2–photodetectors.
by an angle $\theta$

\[
\begin{pmatrix}
E_a \\
E_b
\end{pmatrix} =
\begin{pmatrix}
\cos(\theta) & \sin(\theta) \\
\sin(\theta) & \cos(\theta)
\end{pmatrix} \cdot
\begin{pmatrix}
1 & 0 \\
0 & i
\end{pmatrix} \cdot
\begin{pmatrix}
\cos(-\theta) & \sin(-\theta) \\
\sin(-\theta) & \cos(-\theta)
\end{pmatrix} \cdot
\begin{pmatrix}
E_r^x \\
E_r^y
\end{pmatrix}
\]

(6.3)

where $E_{r,x,y}$ is the reflected beam represented as

\[
E_{r,x,y} = E_{i,x,y} \left\{ \sqrt{R_1} - \frac{T_1.F.(\cos\delta_{e,o} - F + i\sin\delta_{e,o})}{\sqrt{R_1((1 - F)^2 + 4.F.\sin^2(\delta_{e,o}/2))}} \right\},
\]

(6.4)

where, $R_1$ is the input mirror reflectance, $\delta_e$ and $\delta_o$ represents the phase change of the e-ray and o-ray respectively.

In this case, there are two pump resonance fringes for e-ray and o-ray reflected by the cavity, therefore, each of the resonances produces a dispersion-shaped locking signal. Thus, the locking signal has two sharp edges representing the e-ray and o-ray, as shown in Fig. 6.6. The cavity is locked on either of the steep zero-crossings.

### 6.2 Experimental arrangement

This section describes the Raman laser cavity arrangement, stabilization control and diagnostic setup for investigating the longitudinal mode spectrum for the Stokes output used in
the experiment.

### 6.2.1 Raman laser configuration

The schematic diagram of the Raman cavity and the cavity stabilization setup is shown in Fig. 6.7. The pump source was the narrow-linewidth laser described in earlier chapters. The Raman cavity configuration was same as described in Chapter 4, except for the cavity mirror specifications. In this experiment, the reflectivity of the input coupler (IC) was 0.74% and 99.94% at the pump and Stokes wavelengths, respectively, and the output coupler (OC) had a reflectivity of 90% and 99.5% at the pump and Stokes wavelengths, respectively. Therefore, there is no resonant enhancement of the pump in the cavity. The effect of the low Q cavity on the laser stabilization experiments is discussed in Section 6.4. The single-crystal diamond used in this experiment had a stress-induced birefringence (measured by Metripol polarimetry without thermal load [91]) of approximately $5 \times 10^{-6}$ at near-infrared wavelengths. The birefringence was uniform over the whole cross section of the diamond crystal. The pump beam experienced a retardance by the diamond birefringence which provided the source of a locking signal for cavity stabilization [135]. There are alternative stabilization methods where the Stokes beam is monitored for maintaining SLM operation and is discussed in the
Sec. 7.1 of next Chapter.

The detection system is as described for the original HC method [139]. The beam sampler (BS) back-reflected the pump beam from the Raman cavity and into the detection system. A dichroic mirror (DM; HT @ pump, HR @ Stokes) filtered out any residual Stokes in the beam. The resultant elliptically polarized pump beam was converted by a quarter-wave plate (QWP) such that the power incident on the silicon photodetectors (PD1, PD2), after passing through the polarizing beam splitter (PBS), were approximately equal. The photodetector signals were sent to the stabilization controller (Laselock digital, TEM Messtechnik). The difference between the photodetector signals provided the necessary locking signal for the stabilization controller to regulate the DRL cavity length via a control signal fed to the piezoelectric translator (PZT) on which the output coupler was mounted. The laser and the detection system were covered to minimize the disturbances due to air flows and acoustical vibrations.

6.2.2 Stabilization control

The stabilization controller consists of an input section (differential amplifier), a scan generator and a PID (proportional, integral, derivative) servo-loop to generate a control signal to achieve top-of-fringe stabilization and an output amplifier to drive the mirror piezo. The scan generator produced a triangular waveform to sweep through the pump fringes of the Raman cavity. A set-point is chosen for the PID loop which continuously compares the actual input signal difference to this set-point to generate a control signal to counterbalance the deviation from the set-point. The deviation from the set-point serves as the error signal for this loop. The controller also contains a logic module that relocks the laser when the control signal was driven out of the operating range of the error signal. In experiment, adjustment of the gains of P, I and D enabled the circuit to rapidly find the lock point and cavity stabilization.

6.2.3 Spectral diagnostics

Fig. 6.8 illustrates the diagnostic setup for the investigation of the DRL output. A convex lens (CL) of $f = 75$ mm collimated the output beam which was split into a transmitted and reflected beam by a beam splitter (BS1). The transmitted output beam was passed through
a long-pass filter (LPF1; Thorlabs FEL1100) to separate the pump from the Stokes output. A wedged beamsampler (WB) reflected this Stokes beam into two beams. One part was fiber-coupled into an OSA (Anritsu, MS9710C) to monitor the laser output spectrum with a resolution of 0.05 nm. The other part was sent into a 10 GHz scanning Fabry-Perot interferometer (Thorlabs, SA210) to enable investigation of the longitudinal modes. A power meter measured the beam power of transmitted part by the wedge sampler. The total Stokes output power was calculated by calibrating the transmission of the optical train from the laser to the power meter.

The reflected portion from BS1 was split by another beam splitter (BS2). The transmitted beam was passed through another LPF2 at normal incidence to separate the Stokes from the pump. The Stokes output was focussed by a convex lens of $f = 25$ mm to the laser spectrum analyzer (Bristol Instruments, model 771A-NIR) to record the stability of the Stokes centre wavelength. A part of the Stokes output reflected from the BS2 was passed through a tilted LPF3 to monitor the transmitted pump resonances on the oscilloscope.

Figure 6.8: Experimental setup for diagnosing the mode structure of the DRL; FL–focussing lens, CL–collimating lens, IC–input coupler, OC–output coupler, PZT–piezoelectric translation stage, WB–wedge shaped beamsampler, BS–beamsplitter, LPF–low pass filter, GM–gold mirror, PD–photodetector.
6.3 Laser characterization without cavity length stabilization

Since the cavity mirror reflectivities were different from that used for the first Stokes demonstration described in Chapter 4, it was necessary to characterize the performance of the Raman laser before implementing the stabilization technique.

6.3.1 Output efficiency and power

![Graph showing Stokes power and residual pump power as a function of input pump power.]

Figure 6.9: Stokes power (squares) and residual pump power (circles) as a function of input pump power.

The first Stokes output at 1240 nm was observed above a pump power of 23 W. The Stokes power increased linearly from threshold with increase in pump power with a slope of 70% (see Fig. 6.9). The overall conversion efficiency was 21%. The residual pump power was also measured and it decreased monotonically with higher pump powers after the onset of the Stokes, as shown in the figure. This implies that there was good depletion of the pump above the threshold which was found to be an important requirement for SLM as discussed in Chapter 4.

6.3.2 Longitudinal mode spectrum

Fig. 6.10(a) shows a single mode within the FSR of the interferometer at Stokes output power of 2.1 W, implying that the laser operated in the SLM regime up to this power level. As
expected from the findings in Chapter 4, multimode operation was observed for powers above 2.1 W output power. The overlapping of longitudinal modes (see Fig. 6.10(b) and (c)) was also observed during the multimode operation (more than 8 modes > 10 GHz above 3 W Stokes power). These observations are similar to those reported in Chapter 4.

### 6.4 Locking signals: below and above laser threshold

Most of the locking experiments reported in the literature were performed with highly resonant cavities \cite{135, 140, 145, 146}. It was necessary to test whether the diamond birefringence and present cavity finesse are sufficient to provide the necessary steep zero-crossing locking signal to stabilize the cavity. This section presents a theoretical and experimental analysis of the HC locking signals for low finesse DRL below and above the Raman lasing threshold. In the following sections, the term "cavity finesse" is used with respect to the pump wavelength unless explicitly stated otherwise.
6.4 Locking signals: below and above laser threshold

6.4.1 Calculated locking signal below threshold

To estimate whether this mirror reflectivity and the diamond birefringence provide the required locking signal for stabilization, locking signals for the present experimental conditions were calculated using Eqs. 6.3 and 6.4. The dephasing ($\Delta$) per round-trip induced by the birefringence ($\Delta n$), defined as $\Delta = (2\pi/\lambda)\Delta n 2l$, is approximately 0.5 rad. Values for absorption and reflection loss were obtained from the laser threshold and slope efficiency using the model described in ref. [92] and were found to be 0.017% per mm and 0.1% per face, respectively. Using the input and output reflectance values in Eq. 6.2, $F$ is calculated to be 0.08.

![](image)

Figure 6.11: Calculated locking signal plot for 0.5 rad round-trip dephasing for 0.74% input mirror reflectivity. The lock point and the zero-crossing is depicted by the vertical and horizontal dashed line, respectively.

The result is plotted against the reflected pump resonance as shown in Fig. 6.11. It can be clearly seen from the plot that the calculated locking signal is not same as the expected locking signal as shown in Fig. 6.6. Although the locking signal was sinusoidal instead of the typically utilized HC dispersive locking signal, the stabilization of cavity lengths of the master and slave resonators of an injection-seeded Nd:YAG laser was successfully demonstrated in ref. [147]. According to the authors, locking is achieved for a sinusoidal locking signal if the peak of the resonance coincides with a steeply varying part of the locking signal. But in the present case, the pump resonance is in phase with the modelled signal and thus, there is no lock point indicating that the current cavity cannot be stabilized. The absence of a lock point is because the cavity is low-finesse which results in sinusoidal signals for the e-ray, o-ray and hence also the locking signal.
From the above discussion, it is evident that either diamond birefringence or the cavity finesse should be increased to achieve a usable locking signal for the stabilization controller. Therefore, locking signals were also calculated for birefringence values 3.2 and 6.4 times higher (i.e., $1.6 \times 10^{-5}$ and $3.2 \times 10^{-5}$) which induced round-trip dephasings of 1.5 and 3 rad, respectively.

![Figure 6.12: Calculated locking signal plots for round-trip dephasing of 1.5 and 3.02 rad respectively, for a low finesse DRL. The lock point and the zero-crossing is depicted by the vertical and horizontal dashed line, respectively.](image)

As shown in Fig. 6.12, as the birefringence increases, the peak of the reflected pump resonance coincides with the slopes of the locking signals and a lock point is achieved for approximately 3 rad dephasing. As shown in calculated plots, a zero-crossing steep locking signal can only be achieved if the diamond birefringence is 10 times higher. Although selecting diamond with higher birefringence is a possible solution, there is also an alternative approach through increasing cavity finesse. Locking signals for $R_1$ of 9%, 50%, 70% and 90% were calculated and plotted with the reflected pump resonance to compare the usability of each locking signal. As shown in the Fig. 6.13, the reflected resonance and locking signal for 9% reflectivity are in phase and thus, there is no lock point at this reflectivity. As the mirror reflectivity increases, round-trip loss decreases, the dephasing becomes prominent which is evident by the appearance of the edges representing both the orthogonal polarisations. Although the dephasing is noticeable for 50% reflectivity; the lock point is still absent as the dip of the reflected pump...
6.4 Locking signals: below and above laser threshold

resonance corresponds to the dip of the locking signal. As the reflectivity increases to 70%, the edges representing the e-ray and o-ray are more pronounced and a predicted lock point occurs at the slope of the locking signal, near to the zero-crossing. The calculated signal reaches the zero-crossing for 90% reflectivity providing the best lock position out of all these reflectivities. Moreover, this locking signal is similar to that shown in Fig. 6.6 (calculated for the same cavity finesse), although the two edges are not as sharp owing to the reduction in birefringence. A significant inference obtained from these theoretical plots is that even though the birefringence induced dephasing is low, the high finesse of the cavity assists in creating a signal sufficient for locking [148]. It should be noted that the above calculations only illustrate the lock-point and does not take into consideration of the noise on the locking signal. Since signal-to-noise ratio determines the duration and frequency stability, these calculations do not provide quantitative information on locking stability.

![Figure 6.13: Calculated locking signal plots for 0.5 rad round-trip dephasing for 9%, 50%, 70% and 90% input mirror reflectivities. The lock point and the zero-crossing is depicted by the vertical and horizontal dashed line, respectively.](image-url)
6.4.2 Measured locking signal below threshold

![Diagram](image)

Figure 6.14: Time traces of pump resonances (black) and generated locking signal (red) for low finesse cavity without Raman lasing. The locking signal is out of phase with the pump resonance.

The generated locking signal obtained for a low finesse cavity at pump power below Raman lasing threshold is shown in Fig. 6.14. As can be seen from the figure, the locking signal was simply a mirror image of the transmitted pump resonances. Thus, the experimental locking signal is in broad agreement with the theoretical prediction (see Fig. 6.11). Owing to the absence of a lock point, the stabilization controller was not able to lock the cavity.

In order to verify the theoretical prediction for the locking signal obtained for input mirror reflectance of 90%, an experiment was conducted using input and output couplers of 90% reflectance at the pump wavelength. For this experiment, the DFB seed laser was used as the pump. The focussing lens of \( f=50 \) mm was replaced by 75 mm to have a better spatial matching to the cavity mode in the diamond due to a change in the pump spot size incident on the focussing lens before the Raman cavity.

A dispersion-shaped locking signal was produced as shown in the Fig. 6.15 which assisted in the successful stabilization of the cavity, thus verifying the theoretical prediction. The figure also shows higher transverse modes while scanning the cavity which may be owing to the jittering on the output coupler due to the motion of the PZT stage. The inset of the figure
6.4 Locking signals: below and above laser threshold

Figure 6.15: Locking signal (red) for corresponding pump fringes (black) obtained from a high-finesse cavity by exploiting the diamond birefringence. The inset shows two edges (representing o and e-rays) above the zero-crossing.

shows two edges above the zero-crossing which represent the o and e-ray resonances similar to the modelled plot obtained for $R_1 = 90\%$ (see Fig. 6.13), although the feature is not as sharp and equal. The reason for the decrease in sharpness is already discussed in Sec. 6.5.1. The reason for unequal edges in the experimental plot is investigated using simulation.

The symmetry between the sharp edges representing the o-ray and e-ray is determined by the balance between the two photodetector signals ($I_a$ and $I_b$) which is varied by altering the angle of the quarter-wave plate ($\theta$ given in Eq. 6.3). Therefore, the locking signals were calculated for various $\theta$ values as shown in Fig. 6.16. The locking signals for $\theta = 0^\circ$ and $90^\circ$ are identical to the original HC locking signal. The two photodetector signals are balanced at $\theta = 45^\circ$, thus the edges corresponding to the o- and e-rays are symmetrical although the edges are not as sharp as in Fig. 6.6. As the balance between the two photodetector signals is made unequal, the edges become less symmetrical as illustrated for $\theta = 30^\circ$ and $\theta = 60^\circ$. This asymmetry agrees well with the experimental locking signal shown in the inset of Fig. 6.15.
In conclusion, these experiments showed that the locking technique based on crystal birefringence was unsuccessful in stabilizing the low-finesse cavity for the present DRL configuration. The calculations reveal that the locking of a passive cavity is likely to be effective only if either the diamond birefringence or the cavity finesse are higher.

### 6.4.3 Measured locking signal above threshold

Figure 6.17: Locking signal (red) generated for the pump fringes (black) in the low finesse cavity above Raman lasing threshold. The steep portion of the locking signal coincides with the pump peak resonance indicated by the vertical line.

Despite the predicted absence of a suitable locking signal for the passive cavity, the behaviour of the system when lasing was anticipated to be much more complex due the role
6.4 Locking signals: below and above laser threshold

of Raman conversion on the back-reflected pump field. As a result, locking of the low finesse cavity was attempted for pump powers above laser threshold. As shown in Fig. 6.17 a locking signal different from that obtained for below threshold (see Fig. 6.14) was produced with the steep portion corresponding to the peak of the transmitted pump resonance when the pump power was increased above Raman lasing threshold. Using this locking signal, the cavity was successfully stabilized (see Sec. 6.5.4 below for further detail) and provided a promising path forward for further investigation.

6.4.4 Calculated locking signal above threshold

Since the cavity and crystal parameters are same for both below and above threshold, it is deduced that the locking mechanism is attributed to the effect of Stokes generation. In this section, an explanation for the generation of the locking signal above Raman lasing threshold is proposed.

According to the Raman scattering tensor for diamond, the Stokes polarisation is parallel to the pump polarisation when it is oriented along the <111> axis [89]. However, the present laser is high-Q at the Stokes wavelength (intracavity Stokes power of approximately 2 kW at the maximum output Stokes power) with a stress-induced birefringent diamond inside the cavity. According to [91], the multiple roundtrips of the Stokes wavelength increases the influence of diamond birefringence on the Stokes polarisation such that it is oriented along the birefringence axis which provides maximum gain depending upon pump polarisation as shown conceptually by the diagram in Fig. 6.18. It can be seen from the Fig. 6.19 that the scattering efficiency (gain) values are different for the two orthogonal pump polarisations. These pump polarisations would experience different levels of depletion by the Stokes field. This is expected to induce a polarisation dependent loss in the pump beam which may form the basis for a locking signal.

In order to obtain further evidence for this proposed mechanism, the locking signal for above threshold operation was calculated by including an additional loss factor in the amplitude ratio ($F$) term for the o-ray and e-ray in Eq. 6.4. This term is modified to

$$F_{x,y} = \sqrt{R_1 R_2 (1 - T)^2 (1 - F_{loss,x,y})^2}$$

where $F_{loss,x,y}$ is the loss factor to represent the
differential pump depletion. Thus Eq. 6.4 is modified to

\[
  E_{x,y}^r = E_{x,y}^i \left\{ \frac{\sqrt{R_1} - \frac{T_{1,F_{x,y}}(\cos \delta_{e,o} - F_{x,y} + i \sin \delta_{e,o})}{\sqrt{R_1}((1 - F_{x,y})^2 + 4 F_{x,y} \sin^2(\delta_{e,o}/2))}}{\sqrt{R_1}} \right\}
\]

(6.5)

Figure 6.19: Scattering efficiency plot as a function of pump and Stokes polarisation [60]. The black lines represent maximum scattering efficiency. The red crosses indicate the loss terms for two orthogonal polarisations.

To determine values for \( F_{\text{lossx}} \) and \( F_{\text{lossy}} \), a basis is chosen so that two orthogonal polarisations are amplified independently of each other [149]. For the purpose of calculation, it is assumed that the birefringent axis (Stokes polarisation) is approximately at 60°, and the relative gain values corresponding to this angle and the orthogonal direction (60° + 90° = 150°), see red crosses in Fig. 6.19, provide loss terms for the orthogonal pump polarisations. Assuming that the pump depletion is about 50%, both the scattering efficiency values are
multiplied by 0.5 to estimate the loss for each pump component $F_{lossx}$ and $F_{lossy}$. By substituting these values in $F_{lossx,lossy}$ in Eq. 6.5 the locking signal was calculated as illustrated in Fig. 6.20. It is evident from the figure that the dip of the pump resonance coincides with the steep portion of the locking signal curve. So, although the calculated locking signal is not dispersive as obtained for the HC method, this signal can be used by the stabilization controller.

Figure 6.20: Calculated locking signal plot along with the pump resonance for round-trip dephasing of 0.5 rad in diamond for the low-finesse, above Raman lasing threshold case. The lock point and the zero-crossing is depicted by the vertical and horizontal dashed line.

### 6.5 Laser output with cavity length stabilization

With a locking signal achieved during Raman laser operation, the longitudinal mode spectrum and frequency stability were investigated. Moreover, the stability of the locking technique is reviewed.

#### 6.5.1 Performance of the stabilized DRL

The stabilization of the laser enabled SLM operation at higher output powers. Up to 7.2 W SLM power was observed as confirmed by the FP plot illustrated in Fig. 6.21 in contrast to the maximum of 2.1 W for the unstabilized laser. Continuous SLM operation was observed several seconds at this power level. The stability improved to several minutes when the Stokes power was lowered to about 4.5 W.
Figure 6.21: FP interferometer plot shows SLM at 7.2 W Stokes power.

Figure 6.22: (a) The Stokes wavelength stability at 4.5 W. The inset shows the wavelength fluctuations of about 125 MHz. (b) FPI plot confirming SLM during the initial period. (c) FPI plot showing that the large wavelength fluctuations in (a) correspond to multi-longitudinal mode operation. The black line above the longitudinal modes in (b) and (c) represents the measured Stokes output power.
The centre wavelength was monitored for about 3 mins using the laser spectrum analyzer. The Stokes wavelength fluctuated within 125 MHz and the FPI showed SLM for about 2 mins as shown in Fig. 6.22(a) and Fig. 6.22(b) respectively. The output power was also stable during this duration (illustrated by the black line above the longitudinal modes in the figure). After about 2 minutes, the laser reverted to multimode operation for several seconds (see FPI plot in Fig. 6.22(c)), after which the laser returned to SLM (not shown in the figure). During multi-longitudinal mode operation, the output power was highly unstable (see also Fig. 6.22(c)). As the laser wavelength drifted, the lock lasted only over several seconds after which a new set-point and PID setting was required to maintain SLM.

6.5.2 DRL instability

Figure 6.23: Output centre wavelength as function of time showing transitions between single-mode to the multimode regime at a Stokes power of 3.2 W.

In order to investigate the stability, the centre wavelength was monitored using the laser spectrum analyzer at various Stokes powers. As shown in Fig. 6.23, the laser operated in the SLM regime at 3.2 W of power for about 40 seconds after which the laser switched to the multimode state but the controller was able to recover the laser back to single mode. The transition from single mode to multimode and back continued for some time before the locking was completely lost leading to the multimode operation of the laser.

There were instances at higher Stokes powers (>4 W) where the laser wavelength was less stable. In these cases, the laser wavelength was constant for tens of seconds after which
the locking was lost resulting in wavelength fluctuations for several seconds. Eventually, the controller was able to retain the lock which brought the operation back to the SLM state but at different Stokes output wavelength corresponding to a different longitudinal mode. In Fig. 6.24(a), the separation between the two wavelengths is 6.6 pm (1.3 GHz, FSR of the laser) implying that the cavity was stabilized to the adjacent longitudinal mode while in Fig. 6.24(b), the separation was double the FSR implying the neighbouring mode was skipped. The methods for improving the stability of the locked cavity are discussed in Sec. 6.7.

### 6.6 Observation of SBS

When the output coupler of the Raman laser was adjusted to achieve more output power, at a specific alignment, another spectral line separated by about 0.36 ± 0.01 nm (70 GHz) from the Stokes wavelength was observed in the OSA which is attributed to SBS (as discussed in Sec 4.4). Fig. 6.25 shows the less intense SBS line along with the SRS for several Stokes output powers above 2 W. The conditions under which SBS is obtained were not found to be easily reproduced and are not fully understood. Since the study of SBS in the Raman cavity is not the focus of the current work, it was not investigated in detail.

During attempts to stabilize the cavity when SBS was present, it was found that the locking signal was chaotic and lacking a usable lock-point. As discussed in Sec. 4.4, SBS is cross-cascaded from Raman line. Therefore, using the knowledge of cross-cascading from Chapter 3 and experimental observation of residual pump power (see Fig. 4.13(a)), it is...
6.7 Discussion

Many papers have reported the successful adaptation of HC variant locking scheme by exploiting birefringence to stabilize laser cavities \cite{135, 140, 143, 145, 146, 150}. A majority of expected that the onset of SBS clamps the Raman field and leads to an increase in residual pump field. As the stabilization technique is dependent on the differential pump depletion (as described in Sec. 6.5.4), the saturation of pump depletion at the onset of SBS is likely to be responsible for the instability and inability to lock the DRL. This argument is further validated by the fact that the locking was achieved when SBS was eliminated from the cavity by adjusting the tilt of the output mirror.

Although SBS is a parasitic effect in the realisation of SLM Raman lasers, it also provides an opportunity for investigating a nonlinear effect only recently observed for the first time in diamond \cite{75}.

Figure 6.25: OSA spectra at Stokes powers of 2.6 W, 4.7 W and 7.3 W showing another spectral line attributed to SBS in the unstabilized laser.
the aforementioned demonstrations \cite{135, 140, 145, 146, 150} used a second harmonic crystal having a birefringence \cite{151} about $10^5$ times higher than diamond. The only exception is the stabilization of a cavity using the weak birefringence of the dielectric cavity mirrors which introduced a very low phase difference of about $10^{-5}$ rad \cite{143}, but the cavity finesse was $10^4$ larger than the DRL presented in this Chapter. These demonstrations corroborate the experimental results and simulations that diamond birefringence is too low to be exploited with the existing cavity parameters to achieve HC variant stabilization. Attempts were made to enhance the locking stability such as reducing air fluctuations, improving the mechanical stability of the optical elements, and adjusting the PID loop parameters. However, the locking did not improve. Another problem in all these experiments was the quality of the piezo translation stage, which limited the bandwidth response of the stabilization controller. An improved piezo actuator placed directly on the output mirror is expected to improve the integrity of the cavity control system.

The results show that "Raman" locking is possible, but the slope of the locking signal at the lock-point is low, hence inherently unstable. It is not clear how this could be improved without a better understanding of the Raman locking mechanism. In order to develop a more robust system, a path forward could be provided either by increasing the diamond birefringence or the cavity finesse. Increasing birefringence may be readily achieved by introducing an additional birefringent material into the cavity, such as waveplate or harmonic crystal and relying on the original variant of HC locking method. The latter may be a convenient arrangement for future SLM DRLs operating in the visible. Increasing the cavity finesse may be readily achieved by increasing the input mirror reflectivity. An alternative approach is to introduce a Brewster-cut diamond to enable the differential pump loss thus, generate an HC locking signal with an added advantage of low reflection losses for Stokes \cite{152}.

The Pound-Drever-Hall (PDH) technique is also a method that may be considered in the future studies. In this method, the pump laser frequency is modulated by an external electro-optic or acousto-optic phase modulator to produce sidebands at the modulation frequency. The reflected pump from the cavity is detected and compared with the modulation source frequency which generates a locking signal with wider capture range (approximately twice the cavity linewidth) than obtained with HC method. This method is insensitive to power
and spatial drifts; however, it is limited by the modulation frequency and bandwidth of the photodetector. As the detection is based on pump reflection, the problem of the low signal-to-noise ratio owing to the low finesse at the pump wavelength may limit the stability of this locking technique. In addition to PDH locking method, injection locking of the low-finesse DRL cavity with a low-power narrowband seed laser (eg. diode laser) at the Stokes wavelength is another possible option for realizing a stable high-power SLM Raman laser.

## 6.8 Conclusion

A variant of Hänsch-Couillaud technique was investigated for the first Stokes diamond Raman laser by exploiting the diamond birefringence. Stabilization of the cavity length of the DRL was achieved only above the Raman lasing threshold. Although the birefringence was not sufficient to lock the low-finesse cavity, the diamond Raman gain properties and birefringence effect on Stokes polarisation generated the HC locking signal above lasing threshold. SLM operation was achieved for 7.2 W Stokes power using this "Raman" locking mechanism. The cavity was stable only for short time duration due to the low signal-to-noise ratio of the locking signal contributed by the low finesse at the pump wavelength. The HC locking signal calculations provide a useful tool for understanding locking configurations and stability. In future designs, locking through birefringence requires that either the cavity finesse or the birefringence of diamond must be higher in order to improve SLM and frequency stability at higher output powers.

Chapter Contributions: All experimental work and analysis of the results were conducted by the author.
Conclusion

Single longitudinal mode lasers are a core technology for many applications in industry, defence and quantum information science such as atom cooling [2], gravitational wave sensing [1], remote sensing applications [3], optical metrology and interferometry [4]. Some of these applications impose stringent requirements on operating wavelength, spectral tunability, and output power. The demand for robust, efficient and reliable sources mean that solid-state laser technology is almost universally a preference, if not a requirement. However, inversion-based solid-state lasers provide limited wavelengths, output powers. Therefore, alternative solid-state laser technologies were explored. Even though crystalline Raman lasers provide an efficient method to extend the available spectral range of conventional lasers, the development of SLM Raman lasers to date, is relatively immature. Therefore, the aim of this thesis was to investigate the fundamental characteristics of Raman gain in crystals that pertain to SLM operation, and to exploit this knowledge to advance SLM diamond laser technology.

The concepts of gain saturation and mode competition in Raman lasers which were not
well-understood were developed in Chapter 2. These studies revealed that gain saturation in Raman lasers is dependent on the ratio of the pump linewidth and Raman gain bandwidth. Moreover, there is no spatial hole burning for longitudinally pumped Raman lasers. These concepts laid the foundation for the approach pursued in the thesis. The externally-pumped configuration avoided thermal effects and spectral broadening effects present in the intracavity Raman lasers [56, 57]. In order to compare the effects of the material properties on the longitudinal mode structure of the Raman laser, two Raman crystals were chosen for study.

Chapter 3 provided new insights into the operation dynamics and efficiency of CW Raman lasers based on crystals with multiple phonon modes by elucidating the effect of low-threshold cross-cascading on the laser efficiency. Although the low-frequency phonon mode at 87 cm$^{-1}$ of KYW was previously identified in the literature, this Chapter provided the first measurements of its Raman gain coefficient and dephasing time. The complexity of the KYW Raman spectrum was found to be seriously detrimental to laser efficiency and thus was deduced to be a poor candidate for the SLM study. The impact of cross-cascading on the laser output spectrum revealed that a crystal with a more simple and "pure" Raman spectrum is highly desirable for SLM operation. Diamond’s purely single phonon mode in its Raman spectrum, in addition to its high Raman gain and excellent thermal conductivity, distinguish it as a much better candidate for SLM operation. Although the low-frequency phonon mode was observed as a parasitic effect for the KYW laser, it is useful for demonstrating multi-wavelength laser sources, terahertz generation and as low quantum defect mode for beam brightness and/or beam combination.

Chapter 4 reported the first SLM operation of a first Stokes DRL up to 4 W output power. The SLM output power and operational level above the threshold were much higher than those reported in the literature for inversion lasers without incorporating spatial hole burning mitigation techniques [13, 153]. The experimental results in this Chapter corroborated the concepts of spatial hole burning free Raman gain developed in Chapter 2. This Chapter also identified further challenges for SLM operation. The unstable and multimode operation of the laser at higher output powers was found to be caused by coupling between the intracavity Stokes power and the optical cavity length via thermal expansion and thermo-optic effect of diamond. This result demonstrated additional passive or active stabilization techniques are
required for achieving SLM operation at higher Stokes powers. This Chapter also reported observations of SBS in diamond under certain operating conditions.

In Chapter 5, an efficient CW second Stokes DRL at 1485 nm was demonstrated for the first time through cascaded Stokes generation from 1064 nm via 1240 nm \([121]\). It was found that the SLM power range was much more limited in this case as the gain saturation is stalled once second Stokes attained threshold. An approach incorporating a volume Bragg grating as a frequency selective cavity mirror, enabled SLM operation up to 0.5 W second Stokes output power. As the laser output wavelength at 1485 nm coincides with strong absorption lines of some atmospheric gases such as ammonia and water vapour, there was an opportunity to investigate the feasibility of this laser as a LIDAR transmitter. Successful detection of water vapour was achieved in laboratory conditions. The cavity length was continuously adjusted to maintain SLM, as a result of the same diamond thermal effects observed in the previous Chapter. Thus, these results further emphasized the need for active stabilization of the cavity length.

Chapter 6 reported an SLM operation up to 7.2 W Stokes power by employing active stabilization of the laser. It was initially aimed to achieve locking below Raman lasing threshold by exploiting stress-induced diamond birefringence using a variant of Hänsch-Couillaud method. However, investigation of the locking signals by simulation and experiment below threshold revealed that the diamond birefringence was not sufficient to generate a usable locking signal for a low cavity finesse at the pump wavelength. However, locking above Raman lasing threshold was found to be successful. This was explained by a polarization dependent pump depletion through the combined effects of birefringence on the Stokes polarization and the diamond Raman gain. The results presented in this Chapter are noteworthy since the polarization dependent depletion due to the Raman gain of diamond provides a new variant of the Hänsch-Couillaud technique potentially suitable for locking CW diamond Raman lasers. However, the low signal-to-noise ratio of the locking signal and the observation of SBS under certain cavity mirror adjustments, limited the duration of stable SLM operation and frequency stability of the laser.
7.1 Ongoing and future projects

This Section outlines the directions for the future research arising from the results obtained in this thesis.

7.1.1 Knowledge in the current locking mechanism

A hypothesis was proposed in Chapter 6 to explain the observation of the locking signal above Raman lasing threshold. In order to more closely examine whether Stokes generation causes a polarisation dependent pump depletion, more theoretical and experimental development is suggested. A model for rotation of the pump polarisation based on the coupling of the Stokes field with pump field components could be rigorously calculated using the Raman tensors. Such a model may be supported by an experimental investigation of pump polarization rotation due to Stokes generation for both the CW and pulsed DRLs. These studies could provide a better understanding of the "Raman" locking mechanism observed above threshold and thus, aid in future design of stable high-power SLM DRLs.

7.1.2 Linewidth and frequency stability

The locking mechanism described in this thesis suffered from a low signal-to-noise ratio (SNR) of the locking signal, leading to short-term locking and frequency instability. In order to improve these characteristics at higher output powers, the cavity finesse at the pump wavelength has to increase. For single-mode pumping, an enhancement cavity designed to be resonant for both the pump and Stokes is a good option as it would also provide a low Raman lasing threshold for CW operation.

Alternative stabilization techniques such as Pound-Drever-Hall (PDH) may also provide advantages for achieving SLM. The PDH method has a wider capture range than HC method (approximately twice the cavity linewidth) and insensitive to power and spatial drifts, thus providing better tolerance against cavity perturbations (acoustic vibrations, air flows). Since this method also detects the pump reflectance, the finesse at the pump wavelength is likely to be an important design parameter for this technique. Injection locking of the DRL cavity by a narrow-linewidth low-power tunable CW laser (diode laser) at 1240 nm is another viable option to enforce SLM operation. Rings are conventionally preferred over linear cavities...
and are a good option for realizing SLM DRLs. Indeed progress has already been made in this area [154]; recently an SLM ring DRL was demonstrated using HC locking method to generate 883 nm at 1 W. The unidirectionality was achieved by two methods: introducing a parametric loss for one direction due to sum frequency generation using a nonlinear crystal or introducing gain for one direction by a reinjecting mirror.

It is interesting to consider what determines the ultimate linewidth limits for an SLM Raman laser as the spontaneous emission into the laser mode, which is mostly responsible for the Schawlow-Townes linewidth limit for inversion lasers [155], is small in comparison. One paper predicts that the spectral linewidth of the laser may be reduced below Schawlow-Townes linewidth by more than one order of magnitude [156]. According to the authors, highly coherent, narrow-bandwidth source with sub-Poissonian intensity noise may be achieved by Raman lasers. Therefore, it will be interesting to investigate the linewidth of SLM Raman laser once highly stable SLM operation is achieved.

7.1.3 Frequency stability for broadband pumping

The stabilization techniques discussed in Sec. 7.1.2 rely on narrow-linewidth (< 5 MHz) pumping; however, the ability to pump with broader pump sources is of interest for developing SLM DRL employing a greater range of pump laser technologies. SLM operation is likely to be achieved by injection seeding of the DRL cavity by an SLM diode laser at the Stokes wavelength. However, locking the cavity length is not possible by detecting pump reflectance. In this case, active cavity length stabilization has to be achieved by PDH or HC methods based on the detection of Stokes reflectance with respect to the reference wavelength provided by the SLM diode laser wavelength. Since the cavity finesse at the Stokes wavelength is large, the SNR of the locking signals is anticipated to be high enough to achieve long-term locking.

7.1.4 SBS

SBS was recently observed in diamond for the first time [75] and this thesis reports the first follow-up confirmation of that result, and the first case of SLM-pumped hybrid SBS/SRS. It was observed as a parasitic effect as it was found to interfere with the cavity locking on the occasions that it was seen. More study is required to better understand the operating
conditions that bring or suppress SBS. However, this newly observed nonlinearity in an extreme optical material presents a new opportunity to develop diamond Brillouin lasers. Brillouin fiber lasers [157, 158] and microring resonators [159, 160] are known to benefit from significant linewidth narrowing owing to acoustic damping and cavity feedback. The combination of diamond thermal properties and SBS linewidth-narrowing offers potential for obtaining ultra-narrow bandwidth diamond Brillouin lasers required for applications such as coherent optical communication, gyroscopy, atom cooling, spectroscopy and microwave photonics.

7.1.5 Generating other wavelengths

It has been recently demonstrated that the high intracavity power provided by the external cavity configuration enables generation of visible wavelengths through intracavity frequency doubling [90]. Moreover, the large birefringence of the nonlinear crystal ($10^5$ times larger than the diamond) is predicted to be beneficial for the stabilization of the cavity by HC type locking. Frequency doubling of the current system would produce tunable wavelength sources ranging from 619 to 621 nm. Such sources could replace diode lasers for applications such as in spectroscopy, atom cooling and holography.

Applications demanding other wavelengths may be addressed by pumping with the appropriate pump wavelengths. For example, the wavelength at 589 nm is significant for laser guide stars used in adaptive optics systems for astronomy and aerospace applications (eg. tracking of space debris). A narrow-linewidth, high-power (20 W), diffraction-limited laser source is required for exciting sodium ions in the mesosphere (90 to 110 km above the Earth’s surface). This wavelength may be generated by frequency doubling a DRL pumped at 1018 nm which produces a Stokes wavelength at 1178 nm. High-power pump source at 1018 nm is provided by conventional Yb fibre laser technology [161].

7.1.6 Applicability to other Raman materials

One significant insight from this thesis is that it is challenging to demonstrate an efficient CW external cavity Raman laser based on a crystal with low-frequency phonon modes. To ensure high conversion efficiency, it is better to utilize Raman crystals with single phonon mode or
fewer phonon modes (without high gain low-frequency modes) to avoid cross-cascading. This condition restricts the use of many Raman crystals in the CW external cavity configuration such as crystals belonging to tungstates [162], vanadates such as Nd:GdVO₄ [163]. Although barium nitrate has fewer phonon modes, it is not suitable for use in CW regime owing to the poor thermal conductivity of the crystal [68]. Therefore, prospects for practical Raman SLM lasers using materials other than diamond appear to be limited.

7.2 Closing words

The results presented in this thesis have enabled a better understanding of the gain mechanisms of crystalline Raman lasers with respect to the mode competition and SLM operation. This thesis also includes the first results of SLM operation of first Stokes and second Stokes CW external cavity Raman lasers based on diamond. The insights into cascading dynamics identified Raman spectrum as another key parameter that determines the selection of Raman crystals for CW external cavity configurations. The work presented in this thesis opens up new methods for achieving SLM operation and provided a potential variant of the Hänsch-Couillaud technique suitable for locking diamond Raman lasers. The realisation of SBS using SLM pumped diamond Raman lasers provides an opportunity to develop Brillouin lasers based on diamond to be pursued in the future.
Related Publications

A.1 Peer-reviewed journal articles

This appendix includes the following published journal articles which I have authored/co-authored and are relevant to this thesis:


A.2 Conference presentations


High-gain 87 cm$^{-1}$ Raman line of KYW and its impact on continuous-wave Raman laser operation

SOUMYA SARANG,* ROBERT J. WILLIAMS, OLIVER LUX, ONDREJ KITZLER, AARON MCKAY, HADIYA JASBEER, AND RICHARD P. MILDREN

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Abstract: We report a quasi-continuous-wave external cavity Raman laser based on potassium yttrium tungstate (KYW). Laser output efficiency and spectrum are severely affected by the presence of high gain Raman modes of low frequency (< 250 cm$^{-1}$) that are characteristic of this crystal class. Output spectra contained frequency combs spaced by the low frequency modes but with the overall pump-to-Stokes conversion efficiency at least an order of magnitude lower than that typically obtained in other crystal Raman lasers. We elucidate the primary factors affecting laser performance by measuring the Raman gain coefficients of the low energy modes and numerically modeling the cascading dynamics. For a pump polarization aligned to the $N_g$ crystallo-optic axis, the 87 cm$^{-1}$ Raman mode has a gain coefficient of 9.2 cm/GW at 1064 nm and a dephasing time $T_2 = 9.6$ ps, which are both notably higher than for the 765 cm$^{-1}$ mode usually considered to be the prominent Raman mode of KYW. The implications for continuous-wave Raman laser design and the possible advantages for applications are discussed.

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OCIS codes: (140.3550) Lasers, Raman; (140.3580) Lasers, solid-state.

References and links

Solid state Raman lasers are convenient and efficient devices for extending the wavelength range of inversion based lasers [1]. External cavity Raman lasers (ECRLs) offer simplicity and flexibility over other configurations for designing the resonator in terms of thermal-lens management and mirror reflectivities by placing the Raman crystal in a separate cavity singly and flexibility over other configurations for designing the resonator in terms of thermal-lens management and mirror reflectivities by placing the Raman crystal in a separate cavity singly resonant to the Stokes wavelength [2]. However, achieving continuous-wave (CW) operation is challenging as high-intensity pumping, small mode sizes and low-loss resonators are required to achieve moderate lasing thresholds [3–5]. The first CW ECRL used barium nitrate and was limited to 5% conversion efficiency and 164 mW output power due to strong thermal lensing in the crystal [3]; whereas more recent work using diamond has enabled conversion...
efficiency exceeding 60% and output powers up to 380 W due to diamond’s excellent thermal properties and high Raman gain [6]. However, since the Raman shift in diamond is relatively large (1332 cm\(^{-1}\)), there is still demand for crystalline Raman lasers with greater flexibility in terms of output wavelength, despite having less output power potential.

Table 1. Comparison of KYW Properties with Other Raman Crystals Used in CW-ECRLs

<table>
<thead>
<tr>
<th></th>
<th>Barium nitrate (cubic) [7]</th>
<th>Diamond (cubic) [7]</th>
<th>KYW (monoclinic) [8,9]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Raman shift (cm(^{-1}))</td>
<td>1047.3</td>
<td>1332.3</td>
<td>765 (E (\parallel) (N_0))</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>905 (E (\parallel) (N_a))</td>
</tr>
<tr>
<td>Raman gain for 1064 nm pumping (cm/GW)</td>
<td>11</td>
<td>10(^a)</td>
<td>3.6 (765 cm(^{-1}))</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>3.6 (905 cm(^{-1}))</td>
</tr>
<tr>
<td>Thermal Conductivity (W/m.K)</td>
<td>1.2</td>
<td>2000</td>
<td>3.3(^b)</td>
</tr>
</tbody>
</table>

\(^a\)For polarizations parallel to a [111] direction [10].
\(^b\)Average value over the three principal directions.

The class of double metal tungstates—of which potassium gadolinium tungstate (K\(\text{Gd}(WO_4)_2\), KGW) and potassium yttrium tungstate (K\(\text{Y}(WO_4)_2\), KYW) are prominent examples—offer the distinct advantage of two strong phonon modes around 765 cm\(^{-1}\) and 905 cm\(^{-1}\) that have similar Raman gain coefficients and can be accessed separately by changing the polarization state of the pump radiation with respect to the crystallographic axes of the crystal [8, 9]. As shown in Table 1, they also have thermal conductivity approximately three times that of barium nitrate [11], a moderate Raman gain coefficient, a high optical damage threshold and robust mechanical properties [12, 13]. The two aforementioned modes correspond to the symmetric stretching vibrations of the WO\(_\text{OO}\) and WO molecular groups, respectively [14–16]. Although most reports on double metal tungstate Raman lasers utilize these two phonon modes, others have been observed. Kaminskii et al. [9] and Hanuza and Macalik [16] reported strong Raman modes of frequency 87 cm\(^{-1}\) and 225 cm\(^{-1}\) in the spontaneous Raman spectrum of KYW that have been attributed to a librational mode of W\(_\text{O}_n\) and a translation mode of Y\(^{3+}\) ions, respectively. Laser operation on a low frequency mode (84 cm\(^{-1}\)) was observed in an intra-cavity KGW Raman laser which was designed for lasing on the prominent 768 cm\(^{-1}\) mode [17]. In this work, the output wavelengths resulted successive Stokes shifts from the 768 cm\(^{-1}\) and 84 cm\(^{-1}\) modes. Following [18], we refer to this process of cascaded stimulated Raman scattering (SRS) via two or more different phonons as cross-cascading. Cross-cascading has also been observed in single-pass excitation of other crystals. On the other hand, lasing on the 225 cm\(^{-1}\) phonon mode has not been observed to our knowledge. Similar cross-cascading processes have been utilized for multi-wavelength Raman lasers in other double metal tungstate devices [19]. Therefore double metal tungstate crystals offer a greater choice of Stokes wavelengths than diamond or barium nitrate, in addition to reduced susceptibility to thermal effects compared with barium nitrate.

In this paper, we demonstrate a quasi-CW ECRL employing KYW pumped at 1064 nm to generate laser radiation at 1158 nm and 1177 nm, corresponding to the first Stokes components of the two prominent high-energy phonon modes. However, we find that cross-cascading involving the 87 cm\(^{-1}\) and 225 cm\(^{-1}\) Raman modes plays a critical role in determining the output spectrum and laser efficiency. Polarization-dependent spontaneous Raman spectra of the crystal have been recorded to understand the manifestation of the observed phonon modes for different pumping conditions, and a numerical model was developed to analyze the role of cross-cascading and its impact on ECRLs. The 87 cm\(^{-1}\) mode has been found to have a gain coefficient, significantly higher than the two modes often regarded as the primary Raman modes.
2. Experimental setup

2.1 Raman laser setup

The pump source was a polarized Nd:YAG laser, multimode at 1064 nm, delivering 200 W of power in 250 µs pulses at 40 Hz repetition rate and having $M^2$ less than 1.5 [20]. A half-wave plate was utilized to align the pump polarization along either the $N_g$ or $N_m$ crystallo-optic axis. The pump beam was focused into a 50 mm long KYW crystal which was placed in a near concentric 120 mm long cavity, as shown in Fig. 1. The end faces of the KYW crystal were coated with broadband AR coatings from 1000 to 1200 nm to reduce reflection losses.

![Fig. 1. Schematic diagram of the experimental setup. IC = input coupler; OC = output coupler.](image)

Fig. 1. Schematic diagram of the experimental setup. IC = input coupler; OC = output coupler.

The Stokes output power was measured using a thermal power sensor. The output spectrum was recorded using a spectrometer (Ocean Optics NIR 512), and the spatial beam profile was measured employing a CCD camera (WinCamD, DataRay Inc.).

2.2 Raman microscopy

A high-resolution (< 1 cm$^{-1}$) Raman spectrometer (LabRAM HR Evolution, HORIBA Ltd.) incorporating a 532 nm laser was employed to record the spontaneous Raman scattering spectra. The probe beam was propagated along the crystallographic $b$-axis (the lasing direction) with its polarization vector varied using a half-wave plate.
3. Results

3.1 Laser performance

For OC1, the first Stokes at 1158 nm had a threshold of 18 W. Above 60 W of pump power, the Stokes output increases with a slope efficiency of 0.1%, reaching a maximum conversion efficiency of 0.04% (see Fig. 3(a)). The efficiency was fifty times lower than that obtained with a CW barium nitrate ECRL [3] and a thousand times lower than in a CW diamond ECRL [6]. There was no evidence of output saturation and the spatial beam profile of the Stokes output remained TEM$_{00}$ across the investigated power range (see inset of Fig. 3(a)). Therefore, the cause of the inefficiency was deduced to be primarily non-thermal in nature. The time constant for establishing steady-state thermal gradients in KYW for a pump spot radius of 23 µm is approximately 500 $\mu$s [20, 21], thus the laser pulses are in a regime of thermal non-equilibrium and the temperature gradients are even smaller than expected for the stated output power. Measurements of the residual pump power show that the amount of pump depletion in the Raman crystal was negligible, confirming that a non-thermal mechanism was inhibiting power transfer in the Raman crystal.

![Fig. 3. (a). Output Stokes peak power versus pump peak power when using OC1. The pump polarization direction was parallel to the $N_g$ axis. The inset shows the Stokes beam profile at 92 W pump power. (b). Laser output spectrum at 50 W pump power, containing the pump line at 1064 nm and various Stokes components. The brackets indicate the Raman modes responsible for the various output wavelengths (765 cm$^{-1}$, 905 cm$^{-1}$ and 87 cm$^{-1}$, respectively).](image)

The Raman laser output spectrum shown in Fig. 3(b) reveals, in addition to the first Stokes lines at 1158 nm and 1177 nm corresponding to 765 cm$^{-1}$ and 905 cm$^{-1}$ shifts, several emission lines spaced by approximately 10 nm which is consistent with the 87 cm$^{-1}$ phonon mode in KYW. At pump powers near the Raman laser threshold, only the first Stokes line at 1158 nm was attained. However, when the pump power was slightly increased (less than 1.2 times threshold), cross-cascaded Stokes generation involving the 87 cm$^{-1}$ mode occurred, generating radiation at 1168 nm. With further increase in pump power to about 26 W, the Stokes line at 1177 nm corresponding to the 905 cm$^{-1}$ Raman mode was also observed, followed by a Stokes shift from 1177 nm to 1188 nm via the 87 cm$^{-1}$ Raman mode. This is followed by further cascaded Stokes shifts from 1168 nm to 1182 nm and from 1182 nm to 1194 nm via 87 cm$^{-1}$ Raman mode.
For OC2, which showed higher transmission at the Stokes wavelengths, the laser exhibited a higher threshold pump power of 90 W, as expected. The Stokes power increased linearly with a slope efficiency of 4%, as shown in Fig. 4. The maximum conversion efficiency was 2% which is comparable to the aforementioned CW ECRL based on barium nitrate [3]. As for OC1, thermal effects in the Raman laser were negligible and the output spectrum exhibited a cross-cascaded spectral line due to the 87 cm\(^{-1}\) mode. At 110 W pump power, the output spectrum contained the primary 765 cm\(^{-1}\) Stokes mode and a single cross-cascaded line at 1168 nm (see inset of Fig. 4).

In a further experiment, we oriented the pump laser polarization along the \(N_m\) crystallo-optic axis using OC1, which suppressed lasing on the 765 cm\(^{-1}\) shift and enabled first Stokes lasing via the 905 cm\(^{-1}\) Raman mode. The threshold was 42 W, while cross-cascading to 1209 nm involving the 225 cm\(^{-1}\) mode was observed for pump powers above 50 W (see Fig. 5). For 90 W of input power, the first cross-cascaded line was stronger than the 905 cm\(^{-1}\) shifted line which we attribute in part to a higher transmission for the longer wavelengths for this output coupler. The threshold pump power for the second 225 cm\(^{-1}\) Stokes shift to 1241 nm was 90 W.

**Fig. 4.** Output Stokes peak power versus pump peak power when using OC2. The laser output spectrum in the inset was measured at 110 W pump power and the pump intensity is attenuated using a long pass filter. (Note that the shoulders on the short-wavelength side of the more intense lines in the inset is an artifact of the spectrometer.) The brackets indicate the Raman modes responsible for the Stokes wavelengths (765 cm\(^{-1}\) and 87 cm\(^{-1}\), respectively).

**Fig. 5.** Output spectrum at 90 W pump peak power for the pump polarization aligned to the \(N_m\) axis and when using OC1. The spectrum shows the pump wavelength at 1064 nm and three Stokes-shifted wavelengths generated from the 905 cm\(^{-1}\) and cross-cascaded 225 cm\(^{-1}\) phonon modes. The brackets indicate the Raman modes responsible for the first and cascaded Stokes wavelengths (905 cm\(^{-1}\) and 225 cm\(^{-1}\), respectively).
Similar to the observation for a $N_g$ pump polarization, the higher output coupling provided by OC2 led to an increase in threshold (to 120 W). Cross-cascading by the 225 cm$^{-1}$ mode was observed at a pump power of 130 W.

3.2 Raman spectrum of KYW

Although the 87 cm$^{-1}$ and 225 cm$^{-1}$ Raman modes have been previously identified, their gain coefficients have not been previously reported. In order to determine the gain coefficients for these shifts, we recorded high-resolution spontaneous Raman spectra for the two principal crystallo-optic directions.

![Fig. 6. Spontaneous Raman spectra of KYW for the pump polarization (E) parallel to (a) the $N_g$ and (b) the $N_m$ crystallo-optic axis. The insets to (a) and (b) show fitted line shapes (after baseline correction) of the 87 cm$^{-1}$ and the 225 cm$^{-1}$ modes, respectively.](image)

For the pump polarization oriented parallel to $N_g$, as shown in Fig. 6(a), there are three main lines at 87, 765 and 905 cm$^{-1}$. The peak value of the 87 cm$^{-1}$ mode is substantially higher than for the 765 cm$^{-1}$, indicating a higher stationary Raman gain coefficient. As is clearly apparent in the figure, the linewidth of the 87 cm$^{-1}$ mode is much narrower than that of the 765 cm$^{-1}$ mode, which suggests that the higher gain coefficient is in part attributable to a much longer dephasing time. Voigt profiles were fitted to the individual peaks of the instrument-broadened spontaneous Raman spectra after baseline correction in order to determine the Raman linewidths as well as the ratio of the gain coefficients. The results, summarized in Table 2, show that the stationary Raman gain coefficient of the 87 cm$^{-1}$ mode is higher than that of the primary modes. It has a Raman linewidth of 1.1 cm$^{-1}$, corresponding to a dephasing time $T_2 \approx 9.6$ ps which, for comparison, is slightly longer than the first-order mode in diamond ($T_2 \approx 7.1$ ps), but shorter than for the primary ($A_g$) mode in barium nitrate ($T_2 \approx 26$ ps) [7]. For the pump polarization aligned parallel to $N_m$, shown in Fig. 6(b), the 905 cm$^{-1}$ line has the highest peak value with the 225 cm$^{-1}$ line slightly lower. The linewidths of the primary Raman modes at 765 cm$^{-1}$ and 905 cm$^{-1}$ are in agreement with previously published values to within measurement uncertainty [22]. By referencing to a reported value of 3.6 cm/GW in the literature for the 905 cm$^{-1}$ gain coefficient [9] and comparing the relative peak intensities of the respective Raman modes, we determined the gain coefficients at 1064 nm for the 87 cm$^{-1}$ and 225 cm$^{-1}$ to be 9.2 cm/GW and 2.5 cm/GW, respectively.
Table 2. Linewidths and Relative Peak Intensities of the Raman Modes for \(N_g\) and \(N_m\) Axes

<table>
<thead>
<tr>
<th>Raman mode (cm(^{-1}))</th>
<th>Linewidth (cm(^{-1}))(^{a})</th>
<th>Relative peak intensities(^{a})</th>
</tr>
</thead>
<tbody>
<tr>
<td>87</td>
<td>1.11 ± 0.01 E</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.57 ± 0.02 E</td>
<td></td>
</tr>
<tr>
<td>765</td>
<td>8.69 ± 0.02 E</td>
<td></td>
</tr>
<tr>
<td></td>
<td>8.82 ± 0.02 E</td>
<td></td>
</tr>
<tr>
<td>905</td>
<td>6.88 ± 0.003 E</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6.89 ± 0.003 E</td>
<td></td>
</tr>
<tr>
<td>225</td>
<td>5.89 ± 0.013 E</td>
<td></td>
</tr>
</tbody>
</table>

\(^{a}\)Error values are derived from fits to Voigt profiles.

4. Model analysis

We have observed that the presence of the low-energy Raman modes has a profound effect on the quasi-CW performance of a KYW ECRL. However, according to previous reports of KYW and KGW Raman lasers in other configurations (i.e. either pulse-pumped, intra-cavity, or both) [12, 23, 24], highly efficient operation (many tens of percent) can, in principle, be obtained without noticeable impact from low-energy modes. In order to elucidate the disparity between our observations and previous work, we have developed a numerical model to describe the power performance and spectral properties of the KYW ECRL. It is based on coupled rate equations describing the amplification and depletion of the pump and different Stokes fields via SRS [25, 26]. In this model, the pump and Stokes intensities are considered uniform as a function of position inside the cavity, and anti-Stokes generation, Raman four-wave mixing and dispersion effects are neglected. The generalized equations for the pump and first Stokes fields are:

\[
\frac{dI_p}{dt} = \frac{I_{pump}}{t_{rt}} - \frac{2I_{p}\gamma_{p} I_{p}}{t_{rt}} I_{p} - \frac{2I_{p}\gamma_{s} I_{s}}{t_{rt}} I_{s} - \frac{L_a - \log(R_{p,1p})}{t_{rt}} I_{p},
\]

(1)

\[
\frac{dI_{a}}{dt} = \frac{2I_{p}\gamma_{a} \eta_{a}}{t_{rt}} I_{p} I_{a} - \frac{2I_{p}\gamma_{s} \eta_{s} I_{s}}{t_{rt}} I_{s} I_{a} - \frac{L_a - \log(R_{s,1a})}{t_{rt}} I_{a} + \frac{2K I_{a}}{t_{rt}} I_{a},
\]

(2)

\[
\frac{dI_{b}}{dt} = \frac{2I_{p}\gamma_{b} \eta_{b}}{t_{rt}} I_{p} I_{b} - \frac{2I_{p}\gamma_{s} \eta_{s} I_{s}}{t_{rt}} I_{s} I_{b} - \frac{L_a - \log(R_{s,2b})}{t_{rt}} I_{b} + \frac{2K I_{b}}{t_{rt}} I_{b},
\]

(3)

where \(I_{pump}\) is the initial pump intensity, \(\gamma_{p}\) and \(\gamma_{s}\) are the gain coefficients of the primary Raman modes at 765 cm\(^{-1}\) and 905 cm\(^{-1}\), respectively, \(\gamma_{s}\) denotes the gain coefficient of the 87 cm\(^{-1}\) mode, at the pumping wavelength, and \(\eta_{i}\) is the quantum defect for the \(i^{th}\) Stokes shift from the pump. \(K\) is the spontaneous Raman scattering factor, \(\gamma_{p}\) is the Raman crystal length, \(t_{rt}\) is the round-trip time of the cavity, \(L_a\) is the dissipative loss excluding the out-coupling loss, and \(R_{p,1p}\) and \(R_{s,1a}\) and \(R_{s,2b}\) are the input and output coupler reflectivities at the pump and Stokes wavelengths, respectively. The mirror reflectivities at each of the Stokes wavelengths were obtained from transmission measurements of Fig. 2. The cavity length was 120 mm and the pump spot sizes were 23 \(\mu\)m and 39 \(\mu\)m for OC1 and OC2, respectively. The model agreed well with the experimental observations when the round trip intracavity dissipative losses (scatter and absorption) were taken to be in the range 0.8% to 1.0%. \(I_p\) is the intracavity intensity for the pump, \(I_{a,b}\) and \(I_{a,b,1}\) are the intracavity intensities for the first Stokes and cascaded Stokes components respectively. Equations for cascaded Stokes fields were also used in the model, where the pump field is replaced by the first or next-lower order Stokes field. These intracavity intensities were solved numerically using Mathematica, and
the corresponding output powers were calculated and compared to the experimental data. The best agreement was achieved when the ratio of the gain coefficients for the 87 cm$^{-1}$ and 765 cm$^{-1}$ modes with respect to the 905 cm$^{-1}$ mode was set to 2.5 and 1.25, respectively, which are within the uncertainty range of the gain coefficients derived from the spontaneous Raman spectra (compare Tables 2 and 3).

Table 3. Raman Gain Coefficients Obtained by the Numerical Model (for Pump Polarization Parallel to $N_g$, Pump Wavelength: 1064 nm) by Comparison with Experiment

<table>
<thead>
<tr>
<th>Raman modes (cm$^{-1}$)</th>
<th>Gain coefficients (cm/GW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>87</td>
<td>9.0</td>
</tr>
<tr>
<td>765</td>
<td>4.5</td>
</tr>
<tr>
<td>905</td>
<td>3.6</td>
</tr>
</tbody>
</table>

The model yields the intensities for each Stokes wavelength as a function of pump power. For a visual comparison with the experimental laser spectrum along $N_g$, we plotted the model results as Lorentzian lines at each Stokes wavelength with a linewidth of 0.7 nm. The modelled spectra are shown as dashed lines in Fig. 7(a) and (b) for OC1 and 2, respectively, along with the measured spectra (solid lines). Since we used an ECRL configuration which is non-resonant at wavelengths near the pump, first Stokes laser operation at 1074 nm generated directly from the pump via the 87 cm$^{-1}$ mode does not reach threshold.

In all cases the model qualitatively reproduces what was observed experimentally: the onset of cascaded Stokes shifts at low thresholds accompanied by poor pump depletion and poor conversion efficiency. Using the coefficients in Table 3, the calculated laser threshold at 1158 nm for OC1 was 17.8 W in close agreement with the experimental value of 18 W. The model also shows that, with only a slight increase in pump power of about 2 W, the threshold for cross-cascaded Stokes at 1168 nm is achieved. This leads to clamping of the intracavity first Stokes intensity which subsequently stalls the transfer of pump power to the first Stokes. This is confirmed by a stepwise increase in residual pump power (not shown here). For a further rise in pump power by 5 W, the first Stokes related to the 905 cm$^{-1}$ mode (at 1177 nm) is also seen, followed by a cross-cascaded Stokes line at 1187 nm owing to the interaction with the 87 cm$^{-1}$ mode. The OC1 reflectivities for all the Stokes wavelengths are high and approximately equal ($99.7 \pm 0.1\%$), and thus promote the cross-cascading process.

In the case of OC2, the calculated threshold for lasing at 1158 nm (765 cm$^{-1}$ mode) is 95 W, which is close to the measured threshold of 90 W. The first Stokes component corresponding to the 905 cm$^{-1}$ mode (at 1177 nm) is obtained after an increase in pump power.
of approximately 30 W. These values agree well with those measured, confirming the accuracy of the model and the deduced Raman gain coefficients. It is evident from the model that the number of cascaded Stokes wavelengths is larger for OC1 compared to OC2 owing to the former’s higher reflectivity.

Although cross-cascading leads to clamping of a first Stokes line accompanied by stalling of the Stokes conversion efficiency, the model also shows that for pump powers higher than the threshold for a further cascade leads to a large recovery in the slope efficiency. This is because the cross-cascaded Stokes intensity becomes clamped which in turn unclamps the first Stokes intensity and power transfer from the pump. Indeed, using the model it was found generally that the slope efficiency changes from low to high values in an alternating fashion for odd and even number of cascades. The reason the laser efficiency remains poor overall is because the threshold for the odd-order cascade is very close to the previous one causing the slope efficiency to remain low for most of the power range. Although model derived thresholds are highly sensitive to losses, it reproduces well the higher efficiency observed for OC2 compared to OC1 and confirms that this is facilitated by the offsetting of cross-cascading to higher powers.

5. Discussion

The experimental results and the model show that the low frequency Raman modes have a major impact on the performance of the KYW laser, due to their high gain and the high output coupler reflectivity at wavelengths within 250 cm$^{-1}$ of the first Stokes lines. The resultant low-threshold cross-cascaded Stokes shifting obstructs the efficient conversion of power from the pump to the first Stokes line, thus limiting the overall conversion efficiency. Therefore the impact of these modes must be taken into account when designing Raman lasers based on KYW or crystals with similar Raman spectral features. However, there have been numerous reports of efficient Raman lasers using double-metal-tungstate crystals such as KYW and KGW (which has a very similar structure and Raman spectrum to KYW) [12, 17, 19, 23, 24, 27–31], and only one that considers cross-cascading as a power-limiting factor [32].

One possible reason that these devices exhibited higher efficiency and were not apparently severely impacted by cross-cascading is that they operate with pump polarization along the $N_m$ axis where the gain coefficient of the 225 cm$^{-1}$ mode is lower than the 87 cm$^{-1}$ and 905 cm$^{-1}$ Raman modes [12, 19, 23, 24, 27–29]. In addition to the lower gain for this mode, the cross-cascaded wavelength is about 20 nm apart from the first Stokes so the output coupler reflectivity for the cascaded shift may be more differentiated from that of the first Stokes. Nevertheless, cross-cascading from the first Stokes mode (901 cm$^{-1}$) via the 204 ± 3 cm$^{-1}$ mode in KGW (equivalent to the 225 cm$^{-1}$ mode in KYW) has been reported in a nanosecond-pulsed ECRL [30]. In that case they could not achieve slope efficiency similar to that obtained in other ECRLs and attributed it mainly to thermal effects in the crystal and the poor pump beam quality (cross-cascading was not discussed).

There are several papers which have reported cross-cascading in KGW lasers with pump polarization along $N_g$. In the case of a ns-pulse pumped KGW ECRL with a low-reflectivity output coupler [32], cross-cascading via the 84 cm$^{-1}$ mode was observed at elevated powers, coinciding with a sudden decrease in slope efficiency. This may be evidence that cross-cascading is playing a similar role in limiting efficiency as observed in our work. In some CW intracavity and self-Raman lasers operating with low output coupling, cross-cascading on the low frequency Raman mode has been observed, although any adverse effects of cross-cascading on the laser efficiency was not discussed [17, 31]. Since the boundary conditions for power flow in intracavity Raman lasers are different to ECRLs, we cannot directly compare their results with our observations. Finally, there are other reports of efficient KGW and KYW lasers operating in the same orientation in which cross-cascading was not observed [12, 19, 23, 24]. This could be due to the specific loss properties of the resonators used (e.g. mirror coatings) or insufficient resolution in the characterization of the output spectrum.
Typically cross-cascading would be considered a parasitic process that should be avoided to improve laser efficiency. This can be achieved by tailoring the OC reflectivity at the first and cross-cascaded Stokes wavelengths in order to extend the range of pump power between the first and cascaded-Stokes thresholds. However, this would require sophisticated mirror coatings with significant changes in reflectivity (tens of percent) over a spectral range of only a few to a few tens of nanometers. This is likely to be costly and impractical. Introducing wavelength selective elements such as etalons or gratings into the cavity is an alternative approach for suppressing cross-cascading.

Even though the low frequency Raman mode at $87 \text{ cm}^{-1}$ reduced the efficiency of this laser, it may be advantageous for other applications. This high gain mode could be used to simultaneously generate multiple closely spaced wavelengths which may be of use in spectroscopy or in terahertz radiation generation at integer multiples of 2.6 THz ($87 \text{ cm}^{-1}$) through difference frequency mixing of first and/or cross-cascaded Stokes waves. Furthermore, Raman beam conversion for the purposes of either brightness enhancement [33] through the Raman beam cleanup effect, or beam combination [34], can be enabled with much lower quantum defect than with conventional Raman modes in crystals. This may pave the way for an alternative approach to diamond in terms of high-power Raman lasers with possible advantages arising from the ease of growth of large tungstate crystals with good damage threshold and moderate thermal properties.

6. Conclusion

We have demonstrated an external cavity quasi-CW KYW Raman laser operating at multiple Stokes wavelengths. Cross-cascading involving the $765 \text{ cm}^{-1}$ mode and the $87 \text{ cm}^{-1}$ mode was observed when the pump polarization was aligned along the $N_g$ axis, while a similar effect was present via $905 \text{ cm}^{-1}$ and $225 \text{ cm}^{-1}$ modes when the pump polarization was parallel to the $N_m$ axis. Numerical modelling and experimental results confirmed that the early onset of cascaded Stokes components severely limits the pump depletion and thus, lowers the overall conversion to the output Stokes beam. The gain coefficients and dephasing times of the low frequency modes were calculated from high-resolution spontaneous Raman spectra. It is concluded that for a Raman crystal with one or more strong, low-frequency Raman modes, cross-cascading can be a major factor for consideration in Raman cavity design, particularly for CW systems that rely on highly-resonant cavities. The high gain coefficient of the $87 \text{ cm}^{-1}$ Raman mode in KYW ($9.2 \text{ cm/GW}$ at 1064 nm) indicates potential for future applications as multi-wavelength laser sources of high beam quality and as a low-quantum-defect Raman material for brightness enhancement and/or beam combination.

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A fundamental advantage of lasers is their ability to produce a large number of photons in a single optical mode, yet this is achieved in only a minor fraction of devices due to the instability mechanism called spatial hole burning. Here, we exploit the spatial hole burning free nature of a stimulated scattering gain medium to demonstrate single longitudinal mode (SLM) operation in a generic standing wave cavity. A continuous wave diamond Raman oscillator with multi-Watt-level output power and a frequency stability of 80 MHz is demonstrated without use of additional mode-selective elements. Mode stability is addressed by considering the coupling of the Stokes power with thermally induced optical path length changes in the gain medium. The results foreshadow a novel approach for greatly extending the power and wavelength range of SLM laser sources, and with potential advantages for achieving sub-Poissonian intensity noise and sub-Schawlow–Townes linewidths.

OCIS codes: (140.3550) Lasers, Raman; (140.3570) Lasers, single-mode; (140.3600) Lasers, tunable; (140.6810) Thermal effects.

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1. INTRODUCTION

High-power lasers operating on a single longitudinal mode (SLM) form the basis of the highest precision measurements in nonlinear optics and spectroscopy as well as for applications in remote sensing, gravitational wave detection [1] and laser cooling [2]. Stable SLM in standing wave inversion lasers is impeded by spatial hole burning [3], the periodic modulation of gain induced along the laser axis by saturation of the inversion in the antinodes of the standing wave, which causes instability of the mode due to competition from nearby longitudinal modes that occupy a volume that is less saturated. Approaches for overcoming or circumventing spatial hole burning can be classified into schemes involving intracavity frequency-selective elements [4], the twisted-mode technique [5], injection seeding [6], unidirectional ring laser designs [7], and short-cavity lasers [8]. Spatial hole burning is also often reduced by placing a gain medium with a short absorption depth at one end of the resonator [9]. However, simultaneously satisfying requirements of robustness and high power at wavelengths demanded by many single-mode applications remains an ongoing challenge.

For laser gain media that rely on stimulated scattering, such as Raman and Brillouin scattering, gain saturation occurs via a distinctly different process than inversion lasers. There is no energy storage in the Raman medium so that the gain is not necessarily spatially modulated in the same manner as inversion lasers [10]. Hence, it is expected that mode competition effects are dictated by other factors. To date, SLM stimulated scattering lasers have been realized only in systems that employ the same techniques to enforce SLM employed in inversion lasers [11–15]. One possible exception is in the first demonstration of a cw silicon Raman laser [13]. However, unfortunately, there were few details reported about how this was achieved.

In this article, we show that the nature of end-pumped stimulated scattering gain media provides a novel method for realizing SLM laser operation in standing wave cavities without the use of any additional cavity elements. A SLM Raman laser is demonstrated for the gain medium placed at the midpoint of a standing wave cavity having a mode spacing approximately 35 times smaller than the gain linewidth. It is shown that when using diamond as the Raman medium there is potential for the scheme to generate high power (many Watts and beyond) and at wavelengths that are not easily accessible by conventional laser sources.

The principle underpinning SLM generation is qualitatively described as follows. In stimulated scattering the gain arises from the coherent coupling between the pump field $E_p$ and the Stokes field $E_S$ via a phonon field $Q = Q_e \cdot \exp[i(k_r - \omega_0 t)]$ with amplitude [16]

$$Q_e = \frac{N(\partial \sigma / \partial Q)_{\rho} E_p E_S^*}{\omega_0^2 - \omega_0^2 - i2\omega_0 \Gamma}, \quad (1)$$

where $N$ is the density of scattering units, $\partial \sigma / \partial Q$ is the spatial derivative of the polarizability tensor, $\omega_0$ is the phonon frequency at $k_r = 0$, and $\Gamma$ is a damping constant. From Eq. (1), the phonon field is driven in intense regions of the Stokes standing wave. This provides a positive feedback mechanism that further drives the cavity mode and reinforces its stability (see Fig. 1). Neighboring modes are suppressed by gain saturation, which
occurs in Raman lasers due to depletion of the pump field. Thus, provided the node and antinode regions are uniformly pumped, which is generally the case for end-pumped systems, the positive feedback provides an intrinsic mechanism for mode stability. This is in contrast to inversion lasers in which the gain is depressed in antinodal regions, providing negative feedback and causing longitudinal mode instability.

Although the principle may be applicable to Brillouin and Raman gain, a Raman system is more suitable for demonstrating the concept as its bandwidth (10–1000 GHz) is typically much wider and many times the typical mode spacing of lab-scale resonators [3].

2. STANDING WAVE DIAMOND RAMAN OSCILLATOR

We have investigated the possibility for SLM operation in a standing wave Raman cavity using diamond as the active medium. Diamond is a high-gain stimulated Raman scattering (SRS) medium (~10 cm/GW at 1 μm [17]) with beneficial thermal properties. Its exceptionally high thermal conductivity [2000 W/(m·K) at 300 K] [18], moderate thermo-optic coefficient (dn/dT ≈ 15 × 10⁻⁶ K⁻¹ at 300 K) [19], and low thermal expansion coefficient (α = 1.1 × 10⁻⁶ K⁻¹ at 300 K) [20] allow the material to operate continuously as an end-pumped Raman laser with negligible thermally induced lensing or birefringence [21–24]. As a result, it is well suited for investigating the longitudinal mode properties as separately as possible from thermal effects induced in the medium.

A low-nitrogen, low-birefringence, CVD-grown single-crystal diamond (ElementSix, Ltd.) with dimensions of 8 mm × 4 mm × 1.2 mm was placed on a copper block in the center of a near-concentric optical cavity. In order to generate a Lorentzian gain profile, the diamond was end-pumped using a narrowband and tunable distributed feedback (DFB) laser (TOPTICA Photonics, model DL DFB BFY), amplified by a Yb fiber amplifier (IPG Photonics, model YAR-LP-SF) [Fig. 2(a)]. Up to 40 W of diffraction-limited beam quality (M² = 1.05) was available with high frequency stability (40 MHz over one h).

The wavelength was tunable in the range from 1062.8 to 1065.6 nm by varying the operating temperature of the DFB laser with a thermal tuning rate of 80 pm/K. The tuning range was limited by the bandwidth of the DFB laser while the Yb fiber amplifier, in principle, offered an operating wavelength range from 1030 to 1070 nm. An optical isolator was used to prevent optical feedback between the pump and the Raman laser. After passing through a half-wave plate that was used to align the pump polarization to the diamond axis providing the highest Raman gain [25,26], the pump beam was focused into the diamond using a plano-convex lens with a f₁ = 50 mm focal length, resulting in a focal spot radius of 40 μm.

The cavity was 102 mm long, consisting of two plano-concave mirrors of 50 mm radius of curvature. The input mirror (IM) was highly transmitting (T = 97.2%) at the pump wavelength and highly reflective (R = 99.9%) at the Stokes wavelength, whereas the output coupler (OC) reflected the pump radiation (R = 99.9%) and partially transmitted the Stokes radiation (T = 0.43%). The end facets were AR-coated for the Stokes wavelength to minimize intracavity losses. Active temperature stabilization of the diamond mount with an accuracy of 0.5 K was achieved by a feedback-controlled Peltier element, while the cavity length was controlled with a resolution of 20 nm by means of a piezoelectric translation stage (PZT) on the input mirror. The emitted Stokes radiation was collimated using a second plano-convex lens with f₂ = 50 mm focal length and finally separated from the weak pump radiation that leaked through the cavity utilizing a long-pass filter.

3. LASER PERFORMANCE AND SINGLE LONGITUDINAL MODE OPERATION

Above the laser threshold of 12 to 15 W pump power, the Stokes power increased linearly with a slope efficiency of 62%. At the maximum injected pump power of 37 W the Raman laser provided up to 14 W of output power, which corresponds to a conversion efficiency of 38% (Fig. 3). This is close to the optimal values obtainable for the given pump power, beam waist radius, crystal length, and OC transmittance [23]. Measurement of the beam propagation factor yielded M² values in the range from 1.0 to 1.1, indicating near diffraction-limited beam quality. This was also confirmed by analysis of the transverse intensity distribution of the Stokes beam (see inset of Fig. 3).
The longitudinal mode structure of the Raman laser was characterized using a scanning Fabry–Pérot interferometer (FPI) (Thorlabs, model SA210) with a free spectral range (FSR) of 10 GHz and a spectral resolution of about 60 MHz. Simultaneously, the Stokes center wavelength was monitored by using a laser spectrum analyzer (Bristol Instruments, model 771A-NIR), which provided an accuracy of 50 MHz in the 1.24 μm spectral range. The FPI spectra showed that SLM output was obtained at Stokes power up to 4 W. The measured FWHM linewidth in this power range was limited by the resolution of the FPI (Fig. 4). At 1 W, the SLM output was stable apart from occasional mode hops caused by thermal drift (there was no active stabilization of the cavity length). At a higher power (1–4 W), the SLM output was less stable and observed for periods of several tens of seconds before either a mode hop or a short period of multimode behavior. At a higher power (>4 W), the output tended to be always multimode; indeed at a very high power (>8 W) the number of oscillating modes was sufficiently high to cause complex interferograms (see the bottom of Fig. 4) due to the overlap of successive interference orders of the scanning FPI.

SLM operation was observed for pump powers up to 80% above threshold, depending upon the precise alignment and cavity mirror spacing. In contrast, inversion lasers of a similar gain bandwidth and mode spacing are only stable for powers marginally above the threshold. The gain of a second mode spaced by several hundreds of MHz/K, as described in [29], while 

\[ \frac{\omega}{R} \cdot \frac{\hbar}{k_B} - 0.75 \exp \left( B \hbar \omega_R / k_B T \right) \]

denote empirical factors for inversion standing wave lasers predicts that spatial hole burning induces secondary modes at pump powers less than 5% above the threshold for the present laser parameters. Therefore, it is evident that spatial hole burning is absent in Raman lasers, as expected.

The SLM wavelength was tunable from 1238.1 to 1241.9 nm [Fig. 2(b)] by scanning the pump wavelength from 1062.8 to 1065.6 nm. The respective wavelength difference corresponds to the first Stokes Raman shift in diamond, which has a room temperature value of \( \omega_R / 2\pi = 39.941 \) THz [19]. As the pump power was increased well above threshold, the Stokes output frequency was observed to decrease by as much as 35 GHz. Based on temperature measurements of the diamond, the decrease in the Stokes wavelength is consistent with a shift in the center Raman frequency due to heating of the diamond crystal according to the relation 

\[ \omega_R(T) = \omega_R(0) - A \cdot \exp \left( B \hbar \omega_R / k_B T \right) \]

Here, \( A = 1.68 \) THz and \( B = 0.75 \) denote empirical factors [19] and \( \omega_R(0) = 39.953 \) THz is the Raman frequency at \( T = 0 \) K [29], while \( h \) and \( k_B \) are Planck’s and Boltzmann’s constant. According to this relation, a temperature variation of 1 K results in a variation in Stokes radiation frequency of about 0.3 GHz. Hence, precise control of the diamond temperature is crucial when aiming at high wavelength stability of the Raman laser output. Consequently, the Peltier element was employed in the subsequent experiments for active temperature control. This led to reduced temperature drifts of less than 0.5 K and allowed for fine tuning of the mean Stokes wavelength with a resolution of 200 MHz.

### 4. COUPLING BETWEEN STOKES POWER AND CAVITY LENGTH

In the regime of SLM operation, the stability of the Stokes wavelength was in the range of 80 MHz over periods of several tens of seconds, as shown in Fig. 3(a). On longer time scales, the fluctuation range is on the order of hundreds of MHz. This is several times larger than the variation in the pump frequency and was traced back to temperature fluctuations in the diamond that affect the spectral properties in two ways. First, it leads to a change in the Raman shift by several hundreds of MHz/K, as described above. Second, it gives rise to changes in the optical path length due to thermal expansion and thermo-optic changes in the refractive index. The combined effects lead to an effective cavity length

![Fig. 3](image-url)  
Output power (red circles) and conversion efficiency (green squares) of the external Raman oscillator. The error bars indicate the standard deviation. The inset shows the transverse intensity distribution of the Stokes beam at maximum output power.
Fig. 5. Wavelength stability of the Raman laser emission over three minutes for (a) 1.2 W and (b) 10 W Stokes power. The colored area represents the standard deviation from the mean value, which is indicated by the central dashed line.

change $\Delta L_{th} = d \cdot \left( \frac{dn}{dT} + n_0 \cdot \alpha \right) \approx 140 \text{ nm/K}$, which substantially influences the mode competition dynamics of the Raman laser and introduces power instabilities.

Since the amount of heat deposited in the crystal is proportional to the Stokes power due to the quantum defect of the inelastic scattering process, the thermally induced wavelength fluctuations are especially strong at elevated Stokes powers. As a result, mode hopping and ultimately multimode operation is observed as the Stokes power is increased. In this regime, the center frequency fluctuates by tens of GHz [Fig. 5(b)], which is on the order of the Raman linewidth of the interacting vibrational mode (45 GHz [19]).

Fig. 6. Resonant enhancement of the diamond Raman laser: (a) the measured variation of the intracavity pump (blue line) and Stokes output power (red line) upon scanning of the cavity length and (b) a theoretical simulation of the dynamics taking into account thermal effects in the diamond that affect the optical cavity length and lead to a complex feedback mechanism. The dashed lines show the behavior below the threshold.

for a much greater period of the cycle. It was also found that shapes of the pulses for the pump and Stokes were influenced by the scan rate.

The contrasting behavior observed as a function of the scan direction and rate are explained by thermal effects in the diamond. As the intracavity pump power rises by virtue of increasing resonant enhancement, the Stokes power increases accordingly, resulting in larger Stokes-induced heat deposition in the crystal. The thermal loading induces a further positive change (increase)
in optical path length. Thus, in case of active lengthening of the cavity, the compounding effect of the Stokes-induced heat deposition leads to an enhanced rate of increase in the effective cavity length toward the optimum for Stokes power. Once the cavity length is in resonance with the pump wavelength, the thermal effect immediately pushes the cavity beyond resonance. Hence, the Raman laser power falls sharply below threshold until the next resonance is approached. For further cavity lengthening, the cycle manifests as periodic peaks spaced by $\lambda_p/2$. In contrast, active shortening of the cavity counteracts the thermally induced change in optical cavity length thereby extending the length range over which high Stokes power is obtained.

A numerical model has been developed to simulate these effects. Figure 6(b) shows the calculated temporal behavior of the pump and Stokes power together with the external change in cavity length $\Delta L_{ext}$. Below the Raman laser threshold (dashed lines), the ratio of the cavity to incident power ($P_{cav}/P_{inc}$) follows the function

$$
\frac{P_{cav}}{P_{inc}} = \left[1 - \sqrt{R_{th}R_{OC}(1 - \gamma)}^2 + 4\sqrt{R_{th}R_{OC}(1 - \gamma)} \cdot \sin^2(2\pi L/\lambda_p)\right]^{-1},
$$

with $\gamma = 0.01$ denoting the intracavity losses and $L$ being the cavity length. Above the threshold, the deposited heat is assumed to increase linearly with Stokes power giving rise to a thermally induced length change $\Delta L_{th}$ which in turn alters the intracavity pump power.

This feedback leads to an effective length change $\Delta L_{cav} + \Delta L_{th}$, which shows a stepwise progression that substantially modifies the dynamics of $P_{cav}/P_{inc}$ from a below-threshold cavity (i.e., for a passive Fabry–Pérot cavity). The contrasting behavior for shortening and lengthening the cavity is reflected in the Stokes power, which depends explicitly on $P_{cav}$.

5. CONCLUSION

Apart from its impact on the power stability of the Raman laser, the coupling between the Stokes power and the cavity length is an important mechanism that impedes stable SLM operation at high power. The coupling is most strongly derived via thermal expansion and the thermo-optic effect, but is also weakly influenced through the temperature dependence of the Raman center frequency. This conclusion is supported by the fact that SLM operation has not previously been observed in Raman lasers, except in the single case of the cw silicon Raman laser described in [13]. Here, we propose that the silicon waveguide gain medium was in excellent thermal contact to the substrate, which in combination with its low average power reduced the thermal effects in the Raman medium. In bulk and waveguide devices, active stabilization of the cavity length will be vital for obtaining stable SLM operation at higher powers.

The results foreshadow an alternative approach to SLM lasers with major strengths in simplicity and oscillator power. The linewidth limits are dictated by the detailed properties of the pump source and the contrasting nature of the gain medium [30]. As a result, an interesting question is raised on whether the SLM Raman lasers may lead to practical methods for achieving sub-Poissonian intensity noise and sub-Schawlow–Townes linewidths with important benefits for applications demanding ultra-high coherence and spectral power density. Diamond is an excellent candidate for exploring such directions due to its wide transmission range, lack of multiphoton parasitic loss, and power handling capacity.

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Single longitudinal mode diamond Raman laser in the eye-safe spectral region for water vapor detection

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Abstract: We report a narrowband and tunable diamond Raman laser generating eye-safe radiation suitable for water vapor detection. Frequency conversion of a tunable pump laser operating from 1063 to 1066 nm to the second order Stokes component in an external standing-wave cavity yielded 7 W of multimode output power in the wavelength range from 1483 to 1488 nm at a conversion efficiency of 21%. Stable single longitudinal mode operation was achieved over the whole tuning range at low power (0.1 W), whereas incorporation of a volume Bragg grating as an output coupler enabled much higher stable power to be attained (0.5 W). A frequency stability of 40 MHz was obtained over a minute without active cavity stabilization. It was found that mode stability is aided via seeding of the second Stokes by four-wave mixing, which leads to a doubling of the mode-hopping interval. The laser was employed for the detection of water vapor in ambient air, demonstrating its potential for remote sensing applications.

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References and links


1. Introduction

Powerful eye-safe lasers with high spectral purity are required for active remote sensing of atmospheric trace gases. In particular, air- and space-borne light detection and ranging (lidar) systems aiming at accurate concentration measurements of the most important greenhouse gases (GHGs) carbon dioxide, methane and water vapor rely on high-power, frequency-stable laser sources emitting at specific absorption lines of the measured gas species. In order to meet the stringent demands in terms of power performance as well as of the spectral and spatial laser properties, injection-seeded optical parametric oscillators (OPOs) and amplifiers (OPAs) are currently employed as laser transmitters [1,2]. For instance, OPO/OPA systems operating at 1571 nm and 1645 nm have been developed for the air-borne lidar system CHARM-F which allows for simultaneous measurement of carbon dioxide and methane column concentrations [3]. A similar laser transmitter will be realized for the upcoming German-French satellite mission MERLIN (Methane Remote Sensing Lidar Mission) which strives for precise quantification of spatial and temporal gradients of atmospheric methane columns with low bias [4]. In recent years, resonantly pumped Er:YAG lasers have been investigated as an alternative for carbon dioxide and methane detection. Here, the laser directly operates at 1617 nm or 1645 nm which enables higher efficiency and smaller footprint of the overall system [5–8]. However, both concepts based on OPOs and Er:YAG lasers exhibit deficiencies regarding the beam quality, especially at high power levels due to thermally-induced beam distortions.

Another promising approach for realizing lidar transmitters is provided by Raman lasers which allow for efficient frequency conversion of mature laser systems to selected emission wavelengths suitable for trace gas detection [9]. Apart from their compactness, major
advantages of Raman lasers are derived from the automatic phase matched nature of stimulated Raman scattering, which diminishes thermal dephasing and enables Raman beam cleanup [10]. The latter describes the fact that the spatial gain profile experienced by the generated Stokes beam is a convolution of the pump and Stokes fields which converges to a Gaussian distribution, thus providing fundamental transverse mode (TEM$_{00}$) output and diffraction limited beam quality.

Furthermore, recent studies have shown that single longitudinal mode (SLM) operation, which is a prerequisite for narrowband laser emission, can be readily obtained in standing-wave Raman lasers without the use of line selective elements due to the lack of spatial hole burning [11]. From the multitude of Raman crystals, CVD diamond has been demonstrated to be an excellent material for high-power frequency conversion due to its high Raman gain coefficient and its beneficial thermo-mechanical properties, which in combination with the Raman beam cleanup effect, avoids detrimental thermal lensing and offers high-brightness output [12–16]. Following this approach, efficient eye-safe laser generation was obtained in a diamond Raman laser, providing 16 W power at 1485 nm with near-diffraction-limited beam quality [13]. Furthermore, intracavity frequency-doubling of a cw diamond Raman laser has been accomplished, yielding watt-level, tunable output with excellent beam quality in the visible spectral region [17].

In this study, we show that, aside from their excellent power and beam quality performance, diamond Raman lasers additionally allow for the generation of frequency-stable and narrowband output at selected GHG absorption lines in the near-infrared spectral region. For this purpose, an external cavity diamond Raman laser operating in SLM was developed which was tunable from 1483 to 1488 nm, while water vapor in the ambient air was chosen as absorbing gas species to demonstrate the laser’s potential for trace gas detection. Water vapor is the principal GHG due to its large atmospheric abundance and its role as a key amplifier of global warming [18]. Precise measurement of the atmospheric water vapor concentration is therefore essential to check and improve climate models and to provide more accurate climate change and weather predictions.

In the course of our investigations, we studied the influence of a volume Bragg grating (VBG) on the spectral properties of the Raman laser. VBGs are rapidly emerging as compact and robust optical elements for spectral narrowing and mode-selection in all types of lasers [19]; however, until now their applicability in crystalline Raman lasers has not been demonstrated. Finally, we show that the effective mode spacing of a SLM Raman laser scales with the Stokes order, thus benefiting the stability of single-mode operation in higher-order Stokes Raman lasers.

Fig. 1. Experimental setup of the second Stokes Raman lasers: (a) single-cavity configuration for tunable output from 1483 to 1488 nm, (b) coupled-cavity configuration employing a volume Bragg grating (VBG).
2. Experimental setup and Raman laser performance

The experimental setup of the tunable external cavity second Stokes Raman laser is depicted in Fig. 1(a). A single-frequency distributed feedback (DFB) laser (TOPTICA Photonics, model DL DFB BFY), amplified by an Yb fiber amplifier (IPG Photonics, model YAR-LP-SF), was employed as a pump source, delivering up to 40 W cw output power at diffraction-limited beam quality ($M^2 = 1.05$) and high frequency stability (40 MHz over one hour). The pump wavelength was tunable in the range from 1062.8 to 1065.6 nm by varying the operating temperature of the DFB laser with a thermal tuning rate of 80 pm/K. Optical feedback between the pump and the Raman laser was prevented by using an optical isolator, while a half-wave plate was utilized to ensure polarization of the pump radiation along the $[111]$ axis of the diamond, thus providing highest Raman gain [20].

A plano-convex lens with $f_{L1} = 50$ mm focal length was used to focus the pump beam into the low-nitrogen, low-birefringence, CVD-grown single-crystal diamond (ElementSix, Ltd.) which was placed on a copper block in the center of a near-concentric optical cavity, resulting in a pump spot diameter in the diamond of about 66 $\mu$m. The rectangular crystal with dimensions of 8 mm $\times$ 4 mm $\times$ 1.2 mm was anti-reflection-coated at the first and second Stokes wavelength showing transmission of 99.5% at 1.24 $\mu$m and 98.9% at 1.48 $\mu$m, respectively.

The Raman oscillator was formed by two concave mirrors with radii of curvature of 50 mm and 75 mm, respectively. The mirrors were spaced by 125 mm, leading to an optical cavity length of 136 mm considering the refractive index of the diamond. Both mirrors were highly reflective at the first Stokes wavelength ($R_{M1}(\lambda_{St1})$ = 99.95%, $R_{M2}(\lambda_{St1})$ = 99.99%), generating intracavity first Stokes field powers in the kW range. The input coupler (M1) also highly reflected the second-order Stokes radiation ($R_{M1}(\lambda_{St2})$ = 99.0%), while the output coupler (M2) partially transmitted this component ($T$ = 30%). The choice of mirror specifications, especially high output coupling at the second Stokes wavelength, were based on insights recently gained from analytical modeling of cw external-cavity Raman lasers [21].

Measurement of the laser performance showed low threshold (6 W) for both first and second Stokes generation, while the first Stokes power remained nearly constant once the second Stokes field arose, as displayed in Fig. 2(a). Above the second Stokes threshold, the first Stokes field merely acts as a mediator between the pump and the second Stokes fields, so that efficient conversion to the latter is achieved [21]. The maximum second Stokes power was measured to be 7 W at 34 W pump power, corresponding to a conversion efficiency of 21%.

The output wavelength was continuously tunable by varying the temperature of the DFB pump laser diode, realizing a tuning range from 1483 to 1488 nm. The spectral characteristics of the Raman laser emission were studied using a laser spectrum analyzer (Bristol Instruments, model 771) with a wavelength accuracy of 30 MHz at 1.48 $\mu$m. Here, the transition from multimode to single-mode operation was readily observed by the appearance of a smooth Lorentzian line shape as well as a significant reduction of the frequency fluctuations from several GHz to a few tens of MHz, as shown in Fig. 2(b). SLM operation of the Raman laser was obtained for output powers up to approximately 0.1 W. However, multimode operation was observed at higher powers. Thermally induced changes in Raman shift and optical path length are considered to be the major reason for limiting the SLM power [11]. Thermal loading of the diamond is aggravated compared to the first Stokes Raman laser due to the strong intracavity first Stokes field (hence larger impurity absorption) and the additional heat load due to the cascaded Stokes process. This stronger coupling between Stokes power and optical cavity length results in a reduced maximum SLM output power compared to that observed for a first Stokes laser (up to 4 W [11]).
Fig. 2. Performance of the second Stokes diamond Raman laser: (a) Output power of the first and second Stokes radiation as well as residual pump power versus pump power, (b) Raman laser spectrum depending on the temperature of the DFB pump laser diode.

3. Wavelength stabilization using a volume Bragg grating

To increase the SLM power and to improve the frequency stability on longer time scales, a volume Bragg grating (VBG) (OptiGrate Corp) was incorporated into the system, according to the scheme in Fig. 1(b), by placing it 100 mm behind mirror M2. The VBG was designed to have a peak diffraction efficiency (reflectivity) of 55% at 1486.0 nm wavelength at normal incidence with a reflection bandwidth of about 100 pm (FWHM). In this way, it acted as the second Stokes output coupler and formed a coupled cavity of about 250 mm optical length with the inner cavity formed by M1 and M2. A plano-convex lens \( f_{L2} = 75 \text{ mm}, \) AR-coated at \( \lambda_{St2} \), which was placed at a distance of 25 mm from M2, collimated the beam onto the VBG to facilitate a stable resonator mode, while a long-pass filter (LPF), which was highly transmitting at the second Stokes wavelength \( (T \approx 84\%) \), was utilized to suppress the pump and first Stokes radiation leaking through the inner cavity. Wavelength tuning of the VBG-stabilized Raman laser was accomplished by scanning the pump laser wavelength in combination with heating the grating in a temperature-controlled oven. The latter allowed shifting the VBG peak wavelength from 1486.0 to 1486.6 nm with an accuracy of about 1 pm (135 MHz).

The influence of the VBG on the spectral purity of the Raman laser was investigated by recording its spectrum for the second Stokes tuned on- and off-resonance with the grating peak. Figure 3(a) shows both cases, measured at 0.5 W output power. Multimode operation was evident when the Raman laser was tuned off-resonance so that the VBG was transparent for the second Stokes radiation, leading to side-bands in the Raman laser output spectrum and frequency fluctuations of about 4 GHz. In contrast, oscillation of a single longitudinal mode was observed when the pump laser wavelength was set such that the second Stokes wavelength matched the room temperature VBG peak wavelength at 1486.00 nm and optical feedback was provided. As depicted in Fig. 3(b), the stability of the center wavelength was about 40 MHz over periods of one to two minutes, which is in the order of the pump frequency fluctuations. Hence, the utilization of the VBG facilitates SLM operation as it improves the mode discrimination despite its broad bandwidth of about 100 pm.
As shown in Fig. 4, measurement of the temporal variation of the center wavelength over several minutes revealed the occurrence of mode-hops which is attributed to heating of the diamond and its mount. Owing to the aforementioned impurity absorption and Stokes generation, the diamond and mount increases in temperature by tens of Kelvin within a few minutes, which leads to an increase of the optical path length and alters the Raman shift frequency [11]. In principle, this problem can be readily overcome by active cooling of the diamond mount. The mode-hops were measured to be (1.8 ± 0.2) GHz which corresponds to twice the mode spacing of the inner first Stokes cavity. The reason for this is explained as follows.

In the case of the first Stokes mode, the frequency is an integer multiple of the inner cavity mode spacing $\Delta \nu$ and lies close to the peak of the Raman gain near 1240 nm. The second Stokes mode will experience gain due to the first Stokes field as its pump, and be seeded by spontaneous Raman scattering and the result of non-phase-matched four-wave mixing (FWM) of the fundamental frequency $\nu_0$ with the first Stokes frequency $\nu_{St1} = \nu_0 - n \cdot \Delta \nu$, where $n$ is a positive integer. While the former process potentially seeds all cavity modes, the latter only provides a seed at $2\nu_{St1} - \nu_0 = \nu_0 - 2n \cdot \Delta \nu$ due to energy conservation. Hence, we deduce from the observed mode-hop interval of $2\Delta \nu$ that four-wave mixing is the dominant seeding mechanism. It should be noted that the above explanation presumes that the optical lengths of the coupled cavities formed by M1 and M2, and M1 and the VBG are chosen such that the cavities are in resonance. However, due to the low finesse of the latter which is further diminished by intracavity losses introduced by lens L2 and the long-pass filter, the exact cavity lengths are not critical for stable SLM operation.

The concept of increased mode spacing may be transferrable to higher Stokes orders. As the frequency of the $m$th Stokes order is seeded by FWM, spaced from the fundamental by $m \cdot n \cdot \Delta \nu$, the number of available longitudinal modes within the Raman gain bandwidth is reduced by factor $m$. This may be a useful feature, as it enables secondary modes to be more easily discriminated, e.g. by the gain profile or inserted frequency selective cavity elements, and thus assists in SLM stability.
4. Water vapor absorption measurements

The suitability of the second Stokes SLM diamond Raman laser for LIDAR applications was verified by performing absorption measurements of water in air. For this purpose, the output beam of the Raman laser was first separated into two portions, as depicted in Fig. 5. While a weak part was guided to a laser spectrum analyzer which monitored the temporal variation of the radiation wavelength, the major part passed through a mechanical chopper and propagated through the lab over a path length of a few meters before being incident on an InGaAs photodiode. The chopper-modulated output signal was fed into a lock-in amplifier together with the chopper frequency to increase the signal-to-noise ratio of the transmitted signal. A second photodiode, placed close to the coupling lens of the spectrum analyzer, provided a reference signal. The path length difference between the “reference beam” and the “transmitted beam” was 3.8 m. The temporal evolution of the ratio between the transmitted and reference signal was measured simultaneously with the wavelength variations to determine the correlation between laser wavelength and water vapor absorption.
The laser was tuned to a H$_2$O absorption line at 1486.713 nm (6726.2488 cm$^{-1}$), which was the strongest line in the tuning range of the Raman laser. However, it was located outside of the VBG tuning range, making it difficult to sustain stable mode-hop free operation on longer time scales. The temperature and relative humidity in the laboratory were measured to be $T = 20.0^\circ$C and $r \approx 60\%$, corresponding to a water vapor partial pressure of about 14 mbar. The simulated transmission spectrum for the given experimental parameters and using spectroscopic data from the HITRAN database [22] is plotted in Fig. 6. Due to self- and air-broadening of the absorption line, the FWHM of the profile accounts for about 0.15 cm$^{-1}$ (4.5 GHz), while the minimum transmission is in the range of 75%. These values were within the error margins of the experimental data which was obtained by averaging the measured relative transmission in wavenumber bins of width 0.02 cm$^{-1}$. Thus, the developed Raman laser was demonstrated to be feasible for water vapor detection.

5. Conclusion

We have demonstrated SLM operation of a diamond Raman laser emitting in the eye-safe spectral region. Efficient frequency conversion of a tunable pump laser to the second order Stokes component produces 7 W multimode output power in the range from 1483 to 1488 nm. Implementation of a volume Bragg grating increased the single-mode output power from 0.1 W to 0.5 W, while improving the frequency stability over time scales of several minutes. Analysis of the long-term frequency stability revealed that the effective mode spacing of the Raman laser is twice the cavity mode spacing which can be explained by seeding of the second Stokes by four-wave mixing and represents a beneficial inherent property of higher-order Raman lasers when aiming at SLM operation. Finally, the Raman laser was successfully employed for water vapor detection. Here, significant reduction of the measurement error is expected by improving the laser frequency stability, e.g. by using a VBG whose room temperature peak wavelength matches the center wavelength of the selected absorption line.

Detection of other gas species can be accomplished by adapting the current system to use a greater fraction of the Yb fiber amplifier gain spectrum (e.g. from 1010 to 1120 nm), thus enabling access to major portions of the near-infrared via first (1165 – 1320 nm) and second Stokes (1380 – 1600 nm) generation. Therefore, it is expected that SLM Raman lasers based on the developed concept represent a promising alternative to existing OPO/OPA and erbium-based laser sources applied for remote sensing of atmospheric gases.
With a view to their applicability in lidar systems, Raman lasers offer higher spectral brightness than erbium lasers of comparable complexity which show either lower SLM output powers ranging from tens to a few hundred mW [5,6] or lower spectral purity [8]. Compared to the currently employed OPO/OPA systems, Raman lasers are superior in terms of compactness and beam quality. The latter is diminished in OPOs and OPAs especially at elevated power levels, owing to thermally induced aberrations in the nonlinear crystal which lead to $M^2$ values in the order of 2 [1]. In contrast, the Raman beam-cleanup effect enables diffraction-limited output in the eye-safe spectral region even at high output powers beyond 100 W [21].

In general, the potential for power scaling, especially of diamond Raman lasers, opens new opportunities for developing high-power SLM lasers which are of great interest not only for remote sensing applications, but also for other research areas such as gravitational wave detection and laser cooling. Furthermore, extension of the available emission wavelengths to the visible spectral range can be achieved by subsequent second harmonic generation, reaching, for instance, 698 nm which represents the wavelength of the $^1S_0 \rightarrow ^3P_0$ clock transition in Sr atomic clocks [23].

Future investigations will primarily aim to implement active cavity length control to enhance stabilization, e.g. by applying the Hänsch-Couillaud method. In particular, the further development of the proposed concept strongly depends on the need to compensate for thermally-induced changes in optical length of the Raman crystal so that stable SLM operation is achieved on longer time scales.

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