Ten deep blue to cyan emission lines from an intracavity frequency converted Raman laser

Dimitri Geskus^a, Jonas Jakutis-Neto^b, Helen. M. Pask^c, Niklaus. U. Wetter^a* ^aCentro de Lasers e Aplicações, IPEN/SP, Av. Prof. Lineu Prestes, 2242 São Paulo, SP, Brazil; ^bInstituto de Estudos Avançados, IEAv-DCTA, Trevo Cel. Av. José A. A. do Amarante, 1, Putim 12228-001 São José dos Campos, SP, Brazil; ^cMQ Photonics, Department of Physics and Astronomy, Macquarie University, Sydney, NSW 2109,

Australia

ABSTRACT

Here we report on the generation of ten deep blue to cyan laser emission lines using an intracavity frequency converted Raman laser. The fundamental laser field of the intracavity Raman laser is based on the 3 level transition of a Nd:YLF laser crystal, providing a short wavelength at 903 or 908 nm. When combined with generation of a Stokes shifted field via intracavity stimulated Raman scattering (SRS) by a KGW Raman crystal, enables generation of laser emission in the deep blue to cyan wavelength regime via additional nonlinear frequency conversion. Output at several blue-green wavelengths was achieved, with quasi continuous wave (qcw) output powers of up to 1W. A detailed study of the spectral behavior of the underlying Raman laser processes revealed strong spectral broadening of the fundamental laser line at 908 nm to a width of up to 4 nm. The effect of the spectral broadening on the overall laser efficiency is analyzed.

Keywords: (140.3530) Lasers, neodymium, (140.3550) Lasers, Raman, (190.2620) Harmonic generation and mixing

1. INTRODUCTION

Solid-state intracavity frequency converted Raman lasers are able to efficiently deliver many "hard to reach" wavelengths in the near-infrared and visible spectral regions [1, 2]. The small stimulated Raman Scattering cross-section requires high intensity laser fields and extremely low loss cavities to enable resonant build-up of intracavity Stokes shifted laser fields. The currently commercially available high quality optical components such as crystals and dielectric coatings with complex spectral characteristics and durability enable the development of such intracavity Raman lasers, allowing continuous wave (cw) SRS laser operation [3]. In addition, intracavity sum frequency generation (SFG) and second harmonic generation (SHG) has been reported, resulting in highly efficient laser emission in the yellow-orange-red spectral region [4-6]. Here we exploit the ${}^{4}F_{3/2}$ - ${}^{4}I_{9/2}$ three level laser transition of Nd:YLiF₄ (Nd:YLF) and demonstrate intracavity SRS conversion using a KGW Raman crystal, providing cw and quasi continuous wave (qcw) laser oscillation in the 9XX nm wavelength interval. Via SHG and SFG using a LBO crystal inside the cavity we convert the intracavity fields to the visible wavelength range, demonstrating emission of ten spectral lines ranging from deepblue to cyan (451-495 nm), reaching Watt level qcw output power. These results highlight the wavelength agile character of the new generation intracavity frequency converted Raman laser sources, providing a suitable replacement for many other lasers in this field such as the Argon ion lasers and frequency doubled optically pumped semiconductor lasers (OPSLs).

Our recent achievements in intracavity Raman laser technology using the ${}^{4}F_{3/2} {}^{4}I_{9/2}$ three level laser transition of Nd:YLiF4 (Nd:YLF) resulted in laser operation at 3 different lines in the wavelength region between 908 nm and 990 nm [7], opening the way for Raman lasers in the blue spectral range. By applying SFG of the fundamental and or Stokes fields, directly allows for switching between a set of 3 blue wavelengths, e.g. 454 nm, 474 nm and 495 nm by simply adjusting the angle of the LBO crystal [8]. Utilizing an additional Stokes shift, and using two fundamental laser lines lead to the demonstration of ten emission lines in the deep-blue to cyan (452-495 nm) spectral range [9]. The demonstration of these ten blue emission lines, highlight the wavelength agile character of Raman lasers.

*nuwetter@ipen.br; phone +55 11 3133 9359; ipen.br

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We predict a further extension of the emission wavelength range by changing the Raman shift by an appropriate selection of other Raman transitions and/or selection of other Raman crystals. In addition higher order Stokes shifts allows for generation of an equally spaced set of wavelengths. The spacing and span of this set of wavelengths depends on the selected Stokes shift, and number of exploited orders of the Stokes shift. In this work we demonstrate a physically simple and cost-effective multi-color, blue laser source which could complement established laser sources such as Argon ion lasers, optically-pumped semiconductor lasers (OPSLs) [10-12] and optical parametric oscillators (OPOs) [13]. The spectral broadening of both fundamental as the Stokes shifted laser fields are analyzed and the effect on laser efficiency is studied using a mathematical model.

2. LASER EMISSION IN THE 9XX NM REGIME AND SPECTRAL BROADENING

We used a Nd(0.7%):YLF crystal to generate the fundamental laser field. The birefringence of Nd:YLF provides orthogonally polarized outputs at one of the two wavelengths of 903 nm (π) or 908 nm (σ). The relatively weak thermal lens and the large spacing of ground state energy levels of Nd:YLF lead to moderately-low reabsorption loss, making it a good candidate for three level laser operation. KGW was selected as the Raman-active crystal due to its moderately high Raman gain and weak thermal lens. Combining the two fundamental laser lines with the two main Raman shifts of KGW at 768 cm⁻¹ and 901 cm⁻¹ allows generation of four possible first Stokes wavelengths; 970 nm, 976 nm, 984 nm and 990 nm.

In this work we used a near-concentric cavity formed by two concave (ROC = 50 mm) mirrors with high reflectivity, R > 99.9 at 903-908 nm and R > 99.99 at 970-990 nm to efficiently resonate both the laser fundamental and Ramanshifted fields inside the cavity. The length and doping concentration of the 3 mm long Nd(0.7%):YLF crystal enables the pump transition to be nearly saturated, absorbing only 46% of the pump light under non-lasing conditions. The nearly saturated condition minimizes the reabsorption losses for the fundamental laser transition, which increases the laser efficiency. A fiber coupled diode laser operating at 797 nm was used as pump source. The laser diode was electronically pulsed, resulting in a qcw output power with a duty cycle of 1.7% at 25 Hz repetition rate. The output of the fiber was focused to a spot radius of 100 µm into the Nd(0.7%):YLF crystal. The Nd(0.7%):YLF crystal was not positioned in the center of the cavity, but a bit closer towards the pumped mirror, to allow for optimal overlap between pump mode and laser mode. The 10 mm long KGW crystal was placed in the center of the near concentric cavity where generation of SRS benefits from the highest intracavity intensities thanks to the small mode size of approximately 62 µm radius. The 10 mm long KGW crystal was cut for propagation along the Np optical axis; its facets had broadband AR-coatings providing low loss propagation of the fundamental and Stokes fields. The orientation of the KGW was adjusted by rotating it around its optical axis (Np), such that the E//Nm and E//Ng polarizations could be addressed providing SRS based on the 901 cm⁻¹ and 768 cm⁻¹ Stokes shift, respectively.

2.1 Spectral broadening

Depletion of the oscillating fundamental laser line by the intracavity generated SRS, causes a significant spectral broadening of the emission linewidth. It is known that this effect reduces the laser efficiency and that introduction of etalons can improve the laser efficiency significantly [14].



Figure 1. Emission spectra of the fundamental (left figure) and Stokes shifted laser emission (right figure) for various pump levels. The labels indicate the absorbed pump power (range: 2 - 32 Watts).

In figure 1 the emission spectra of the laser are presented for various pump levels. Here one can see that upon Stokes generation the spectrum of the fundamental laser emission starts to broaden. Stronger broadening is observed at higher intensities. The observed broadening of the fundamental laser line is extremely large, reaching a width of up to \sim 4 nm, which indicates that the fundamental laser is operating in the far wings of the natural emission line width of Nd:YLF, having a FWHM of only 3 nm [15]. Obviously this effect drastically reduces the intracavity gain of the fundamental laser line.

To calculate the impact of the spectral broadening on the laser performance we measured the broadening of the fundamental and the Stokes signal as a function of pump power for the set-up whose results are shown in figure 3. Based on the results of ref. [14], that show a clear saturation of the decrease of the effective gain factor caused by the broadening we chose to fit the broadening data with a saturation curve given by (1):

$$y = A \cdot \left(1 - \frac{1}{1 + B \cdot (P - P_{th})^2}\right) + y_0 \tag{1}$$

where P is the absorbed pump power, P_{th} the threshold pump power, A the maximum broadening (FWHM in nm) and y_0 the fundamental emission linewidth before Raman lasing occurs. B is a scaling factor that describes how fast the saturation of the fundamental and the Raman signal occur as a function of pump power.



Figure 2. Measured spectral broadening of fundamental and Raman signal.

It can be seen in figure 2, that the fundamental laser linewidth starts to broaden as soon as the Raman laser starts. We then assume that the effective gain is reduced linearly with increasing bandwidth. Based on this decrease of gain we calculated the expected laser performance using a rate equation model similar to the one in ref. [15] and compared the performance with the obtained laser performance, shown in figure 3. For the simulation we fixed all spectroscopic data except the cavity round trip losses [16, 17]. As a result we obtained good fit using cavity round-trip losses of 0.9% for the fundamental wave and 0.4% for the Stokes wavelength.



Figure 3. Measured laser performance (left) and simulated performance (right) when taking into account the limited available gain by the spectral broadened line width (full symbols) and without broadening (half symbols).

In figure 3 we see a good agreement between measured and simulated data for the case of spectral line broadening. In the absence of spectral broadening, the simulated Raman signal is approximately three times stronger. Also, the fundamental

laser signal is clamped to a lower value as soon as Raman lasing starts, when compared to the emission measured under line broadening. We anticipate that the introduction of etalons to restrict the emission line widths will significantly improve the laser performance of this laser system.

2.2 Cw laser operation compared to qcw laser operation at 990 nm

Cw Raman laser operation at 990 nm has been attempted using a 6 mm long Nd(0.7%):YLF crystal and a 10 mm long KGW crystal, inside the cavity. For comparison, the cavity parameters were almost identical to the previously reported qcw laser experiments [7], having a cavity length close to the stability limit of 11 cm and outcoupling efficiency of 0.28% at 990 nm. While increasing the pump pulse duration, to ultimately achieve cw laser operation, the physical cavity length was changed to 10.8 cm to accommodate a stronger thermal lens. At 18 W of absorbed pump power the Nd(0.7%):YLF crystal fractured, providing a maximum extracted cw output power of 27 mW at 990 nm. The measured cw output power of only 27 mW is much lower than the obtained qcw output power of ~400 mW at the same pump level. Cw laser operation appears to be more than a factor of 10 weaker compared to qcw laser operation, as depicted in figure 4.



Figure 4. Comparison between qcw output power taken from [7] and cw output power from similar laser configuration

The small cw output power can be explained to a large extend by the increase of the crystal temperature in combination with the three-level nature of the laser, affecting the laser performance significantly. In addition an elliptical output beam shape was observed, indicating a cylindrical thermal lens. More advanced strategies to compensate the thermal lens by a cylindrical lens [18], or adaptive optics [19] can improve the cw Raman laser performance.

3. LASER EMISSION IN THE BLUE

Introduction of SHG and SFG into the cavity, by inserting a nonlinear optical crystal such as LBO into the cavity, allowed us to generate 10 visible emission lines with wavelengths ranging from 452 nm to 495 nm. The related optical processes and the measured spectra are presented in figure 5.



Figure 5. Schematic of the intracavity laser processes (top part) and recorded emission spectra of each blue emission line. The inset shows a magnification of the 470-471 nm spectral region, revealing the two emission lines in this region.

For this experiment we employed a 3 mm long Nd(0.7%):YLF crystal and a 10 mm long LBO crystal , cut to $\theta = 90^{\circ}$, $\Phi = 16^{\circ}$ for critical phase matching. Both crystals were equipped with broadband AR-coatings for the fundamental and first Stokes wavelengths ranging from 903 till 990 nm. Both cavity mirrors, having a ROC of 50 mm, had broadband high reflection coatings for the same wavelength range. The extraordinary-polarized, with respect to the LBO crystal, blue laser emission was coupled out of the cavity through the output coupler, which had low reflectivity (R ~ 5%) for the deep blue to cyan wavelengths. A photograph of the operating laser is presents in the upper half of figure 6.



Figure 6. The top picture shows a photograph of the laser setup, while operating. The lower schematic shows the optical processes to generate 5 emission lines in the blue, based on a fundamental wavelength of 908 nm. Changing the fundamental laser line to 903 nm creates an additional set of 5 deep-blue to cyan emission lines, providing a total sum of 10 emission lines in the deep- blue to cyan spectrum generated by a single set of crystals.

By changing the angle of the LBO crystal, three blue emission lines could be generated; SHG of the fundamental field, SFG of fundamental and Stokes laser fields and SHG of the Stokes field. In the same way, two additional blue emission lines could be generated by rotating the KGW crystal, changing the wavelength of the Stokes field. Based on the fundamental field at 908 nm, 5 blue spectral lines were easily obtained as shown in the schematic of the lower part of figure 6.

The strong birefringent walk-off of the optical field inside the LBO crystal was used to select the polarization of the fundamental laser field. A simple tilt or translation of the concave outcoupling mirror made the cavity selective for the ordinary or the extraordinary polarization hence forcing the polarization of the laser and the wavelength of the fundamental field to 903 nm (π) or 908 nm (σ). Note that the orientation of the LBO crystal had to be adapted to maintain extraordinary polarization of the fundamental and/or Stokes field with respect to the LBO crystal to maintain the frequency conversion to the visible. For each fundamental wavelength five emission lines in the blue are obtained, resulting in a total of ten deep-blue to cyan spectral emission lines.

3.1 Experimental Results

Each individual spectral emission line was recorded using an Ocean Optics (HR4000) spectrophotometer with a resolution of 0.24 nm. The measured emission spectra are presented in the lower part of figure 10. The strongest blue output power of 0.94 W was extracted for the 474 nm emission line in qcw operation, having a M2 value of 1.08 and 1.17 for the X and Y direction respectively. A slight walk off of the laser beam in the LBO crystal resulted in a slight elliptical beam profile. Other strong blue emission lines at 495 nm and 470 nm were observed. The laser performance of the strongest blue laser lines are presented figure 7. Note that we only collected and reported the forward propagating light, we assume that the total extracted visible laser power is the double value, based on the assumption that the LBO crystal converts the intracavity infrared laser fields into equal portions in both the forward as backward propagating direction.



Figure 7. SFG and SHG generated blue laser power based on the 908 nm fundamental line and its Stokes fields at 976 nm and 990 nm.

Despite the larger Raman cross-section of the 768 cm⁻¹ Stokes shift [17], the corresponding laser emission lines at 470 nm and 488 nm provided lower output power with respect to the blue emission based on the 990 cm⁻¹ Stokes shift. The output power of the blue emission lines originating from the 903 nm fundamental laser line was very low and the oscillation tended to be unstable. The weak performance of blue laser emission based on the fundamental field at 903 nm, is attributed to laser operation at the short-wavelength edge of the mirror reflectivity band leading to increased intracavity losses for the fundamental field. Higher powers are expected with optimized coatings.

In addition cw laser operation with intracavity SFG was investigated. This experiment was based on a high-Q cavity using a 3 mm long (and therefore less absorbing) Nd(0.7%):YLF crystal, a 10 mm long KGW crystal and a 10 mm long LBO crystal inside the cavity. The obtained output power was 45 μ W at 474 nm, however the emission was 100% modulated resulting in an irregular train of pulses with durations of the order of 100 ns. Similar pulsating behavior of the Stokes emission was observed in our earlier report [7], this behavior suggests near or below threshold laser oscillation.

Blue laser emission of frequency doubled fundamental laser lines was easily observed using the previously described setup with the LBO crystal cut at a $\Phi = 16^{\circ}$ angle, however, the output power was clearly compromised by additional intracavity losses. The large angular tilt (~10°) of the LBO crystal to achieve an internal phase-matching angle of ~22.4° for SHG of the fundamental laser fields, strongly reduced the effectiveness of the AR-coatings of the LBO crystal. The use of temperature-tuned phase-matching would to be advantageous in avoiding this problem. Here we used a 3 mm long Nd(0.7%):YLF crystal and a 10 mm long LBO-crystal cut for $\theta = 90^{\circ}$, $\Phi = 21.9^{\circ}$ for the generation of the SHG of the fundamental laser fields to avoid the excessive losses. In this separate experiment the KGW crystal was removed from the cavity since the experiment does not require generation of the Stokes field. A qcw output power of 1.08 W at 454 nm having a threshold of 4 W absorbed pump power and a qcw output power of 0.51 W at 452 nm with a threshold of 6 W absorbed pump power was obtained.

4. DISCUSSION AND CONCLUSIONS

Here we report on the emission of ten deep-blue to cyan laser lines from a Nd(0.7%):YLF-KGW-LBO intracavity frequency converted Raman laser. A maximum extracted qcw output power of 0.94 W at 474 nm is achieved. Analysis of the fundamental processes involving the Stokes conversion, reveal a strong spectral broadening of the fundamental laser line, strongly suppressing the theoretical achievable laser performance. Based on our calculations the Raman laser performance can be improved by a factor three by use of an appropriate etalon to prevent broadening of the fundamental laser. Such an improvement would directly translate to a similar improvement of the achievable blue output power.

Watt level cw laser emission in the visible yellow to lime-green spectral region is easily achieved due to the absence of reabsorption of the fundamental laser field, which is typical for a four level laser transition [6]. In this work we see that the three level transition we employ in this work, heavily suppresses the output power, especially for cw laser operation. Moreover we see that the spectrum of the fundamental laser field is strongly broadened by depletion of its central line by the intracavity SRS conversion. We predict significant improvements for both the qcw Raman laser performance as well as the qcw visible laser output when restricting the broadening of the fundamental laser field by use of an intracavity etalon.

The ten blue laser emission lines demonstrated here highlight the wavelength agile character of intracavity frequency converted Raman lasers. At present the watt level laser power in the blue spectral region is primarily covered by economic frequency doubled optically pumped semiconductor lasers (OPSLs), or complex optical parametric oscillators (OPOs) providing continuous tuning ranges of up to 15 nm [20] and more than 50 nm [13], respectively. The intracavity frequency converted Raman laser demonstrated here complements these established laser sources. Intracavity frequency converted Raman laser sexcel in simplicity and robustness. Although the emission wavelength of the Raman laser cannot be continuously tuned, it can be configured in a manner that an operator can easily select between several discrete wavelengths that span the blue spectral region. Further, the use of different Raman transitions in KGW or other Raman crystals will enable spectral coverage to be designed. For example, multi order Stokes laser oscillations lection of the short 89 cm⁻¹ Stokes shift of KGW, would create a more closely spaced set of wavelengths, allowing for tuning over a small wavelength range [21] whereas the large stokes shift of 1332 cm⁻¹ in diamond [22] would create a more widely spaced wavelength set, ranging from blue to yellow.

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