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Eye-safe laser radiation from stimulated Raman scattering frequency self-conversion in $KGd(WO_4)_2$:Nd^{3+}

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Abstract
Under short-pulsed longitudinal pumping at 600 nm we obtained intracavity stimulated Raman scattering frequency self-conversion of the 1352.5 nm laser line in $KGd(WO_4)_2$:Nd^{3+}. The wavelength which was produced is eye-safe at 1539.5 nm and the conversion efficiency was 1.26% with uncoated faces of the 3 cm long crystal.

1. Introduction

Raman scattering is a third optical non-linear process which occurs in gases, liquids and solids. Stimulated Raman scattering (SRS) shifts the wavelength of a laser radiation leading to a new one with high coherence, brightness and peak power and low divergence. This process is of special importance in crystals because of the advantages of solid-state technology (compactness of the devices, low maintenance). So the Raman spectroscopic properties (frequency shift, Raman linewidth, peak intensity) of various crystals have been extensively investigated: nitrates, carbonates, tungstates, molybdates, phosphates, niobates [1, 2]. The Raman gain coefficients of the most efficient materials have been measured: 11 cm GW^{-1} for $Ba(NO_3)_2$, 6 cm GW^{-1} for $KGd(WO_4)_2$, 5.5 cm GW^{-1} for $CaCO_3$.

Such a crystal can be used as a single-pass Raman cell, inside an external cavity or inside the laser cavity (intracavity Raman laser). For example the radiation from a LiF:F_2^- colour centre laser (tunable from 1.1 to 1.25 μm) was converted with a single-pass SRS cell constituted of $Ba(NO_3)_2$ (or $KGd(WO_4)_2$) in the 1.23–1.37 μm (first Stokes) and in the 1.43–1.6 μm spectral ranges (second Stokes) [3] with conversion efficiencies of 60% and 20% respectively. In [4], an external cavity containing $Ba(NO_3)_2$ was pumped either by the 1064 nm or by the 1318 nm line of a flash-lamp-pumped YAG:Nd laser; it was able to produce efficiently several Stokes components and in particular radiation in the eye-safe region around 1.55 μm. In [5], an intracavity Raman laser using $Ba(NO_3)_2$ inside a flash-lamp-pumped YAG:Nd laser operating at 1.318 and 1.338 μm produces 1.535 and 1.556 μm in the eye-safe range with 48% efficiency.

Another attractive configuration is based on self-active materials in which the non-linear optical effect and the laser emission are produced by the same crystal, simplifying the device. In the area of second-order processes, the best-known device is the self-frequency-doubling laser and an overview is presented in [6]. In the field of SRS frequency self-conversion, the laser oscillation on the 1068 nm line of Nd^{3+}-doped $KGd(WO_4)_2$ was first used and Raman shifted by the host. Nanosecond ([7], efficiency not measured) and picosecond [8] regimes were exhibited under flash-lamp pumping and wavelengths were produced in the 0.97–1.5 μm range. Then the laser oscillation of the 1351 nm line of $KGd(WO_4)_2{:}Nd^{3+}$ was used to reach a wavelength safe for the human eye: 1538 nm [9]. In this case the efficiency of the Raman self-conversion was 0.15%. The crystal was side-pumped with Xe flash lamps and had its faces antireflection coated, leading to pulses with 12 ns duration at 10 Hz repetition rate.

In the present work, we have used a short-pulsed longitudinal pumping and obtained intracavity SRS frequency self-conversion of the 1352 nm laser line in $KGd(WO_4)_2{:}Nd^{3+}$. The wavelength which was produced is eye-safe at 1539 nm and the conversion efficiency was 1.26% with uncoated faces.

Our crystal was grown by a modified Czochralski technique, to a size of 30 mm. It is oriented along the *b*-axis and the Nd concentration is 2.54 at.%. Its main physical properties can be found in [10].

2. Stimulated Raman scattering self-conversion of the 1068 nm Nd^{3+} laser line

The laser properties of the $KGd(WO_4)_2{:}Nd^{3+}$ crystal were first tested on the 1068 nm line corresponding to the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition. The crystal was longitudinally excited along the *b*-axis at 600 nm in the $^4G_{5/2}$–$^2G_{7/2}$ levels with the beam of a Laser Analytical System dye laser (dye: rhodamine 6G), pumped by a frequency-doubled Nd:YAG laser from BM Industries. The polarization of the pump leading to the highest absorption was N_m and the corresponding absorption spectrum is shown in figure 1 (we recall [10] that for light propagation along

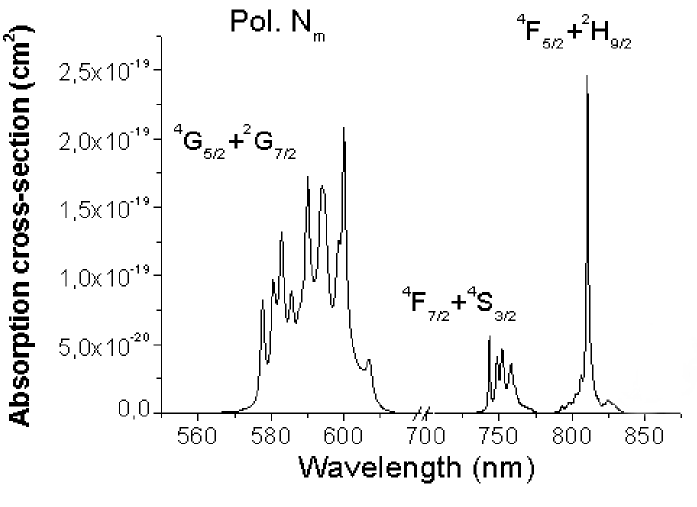


Figure 1. The absorption spectrum of the $KGd(WO_4)_2{:}Nd^{3+}$ crystal in the *b*-direction for the N_m-polarization.

the *b*-direction, the two orthogonal polarizations parallel to the principal axes of the optical indicatrix are called N_m and N_g, corresponding respectively to the values 1.986 and 2.033 of the refractive indices at 1.06 μm).

The laser cavity was constituted of a 16MLB183 plane input mirror with high transmission in the visible range and high reflectivity (>99.5%) near 1068 nm and an output concave mirror (radius of curvature: 15 cm; 99% reflectivity at 1068 nm and 60% at 1180 nm), the distance between the two mirrors being 7.5 cm. The pump was focused behind the crystal with a 19 cm focal length lens; the pump waist was measured, by the knife method, to be 420 μm at the entrance face of the crystal.

Laser pulses were generated at 1068.2 nm and SRS self-frequency conversion pulses were observed at 1181.7 nm (Stokes 1) and 1322.1 nm (Stokes 2) in N_m-polarization. The wavelengths have been measured with a Jobin-Yvon HRS2 monochromator carefully calibrated at the second order with the 546.07 and 690.716 nm lines of a Hg lamp and the 753.58 nm line of a Ne lamp. So the Stokes shift is found at 899 cm^{-1} in our Nd-doped $KGd(WO_4)_2$, although it is at 901.5 cm^{-1} in the undoped $KGd(WO_4)_2$ [10] for the A_g active vibrational mode. The time evolutions of the pump, laser and Stokes 1 SRS pulses were measured with a digital 9410 Lecroy oscilloscope and are represented in figure 2. The 60 ns delay between the pump and the laser are interpreted as the time for de-excitation from the $^4G_{5/2}$–$^2G_{7/2}$ levels to the $^4F_{3/2}$ initial laser level. This interpretation was confirmed by testing a new excitation at 750 nm in the $^4F_{7/2}$–$^4S_{3/2}$ levels. In this latter case, with a similar pump pulse duration, a similar pump waist and a similar cavity length, the delay between the pump and the laser pulses was reduced to 10 ns, corresponding to the reduced transition time of the $^4F_{7/2}$–$^4S_{3/2} \rightarrow {}^4F_{3/2}$ de-excitation.

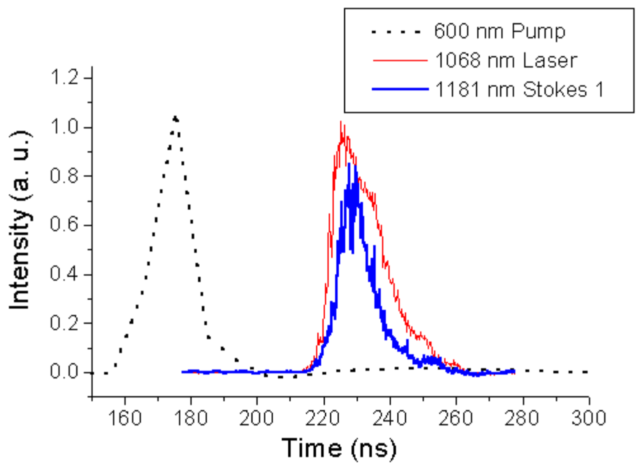


Figure 2. The time evolutions for the pump at 600 nm, laser at 1068.2 nm and Stokes 1 SRS pulses.

The energies of the laser and the Stokes 1 SRS pulses were measured with a Molectron pyrometer through adequate interference filters and are represented in figure 3 versus the pump power. The threshold of the Stokes 1 SRS is higher than that of the laser, and the maximum Stokes 1 SRS conversion was 4%.

3. Eye-safe laser radiation from SRS self-conversion of the 1352.5 nm Nd^{3+} laser line

The laser emission on the 1351 nm line corresponding to the Nd^{3+} $^4F_{3/2} \rightarrow {}^4I_{13/2}$ transition in $KGd(WO_4)_2$ has already been demonstrated under flash-lamp pumping [11] and under

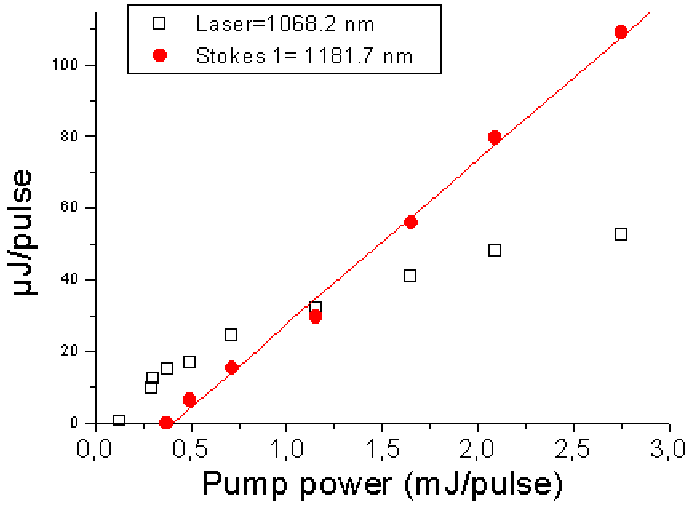


Figure 3. The energies of the laser at 1068 nm and the Stokes 1 SRS pulses versus the pump power.

longitudinal pumping [12]. It is possible because the effective emission cross-section has significant magnitude despite the excited-state absorption (ESA) cross-section occurring in the same range of wavelengths, corresponding to the $^4F_{3/2} \rightarrow {}^4G_{7/2} + {}^4G_{9/2} + {}^2K_{13/2}$ transition (figure 4). In figure 5 we show the emission, along the *b*-axis, of our sample and the ESA extracted from [13]. These spectra are calibrated in cm^2 with the Judd–Ofelt treatment extended to anisotropic crystals [14] using the following intensity parameters:

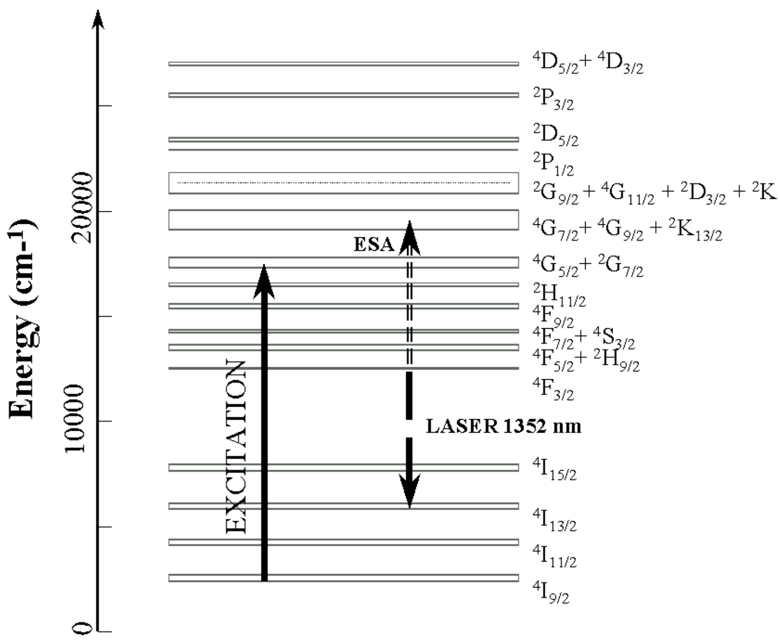
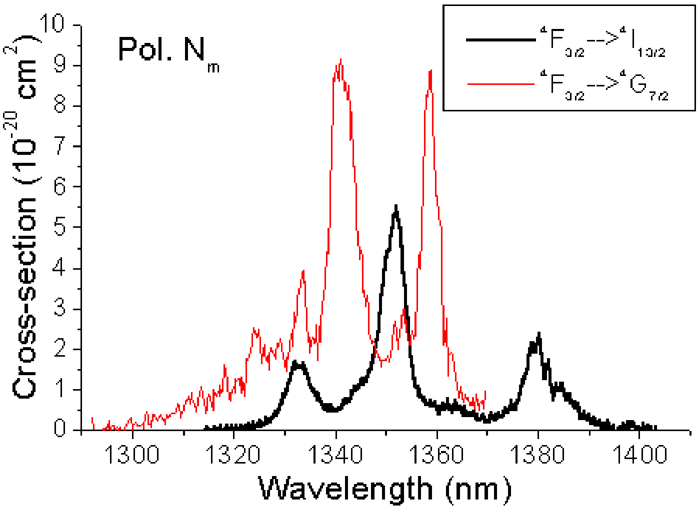
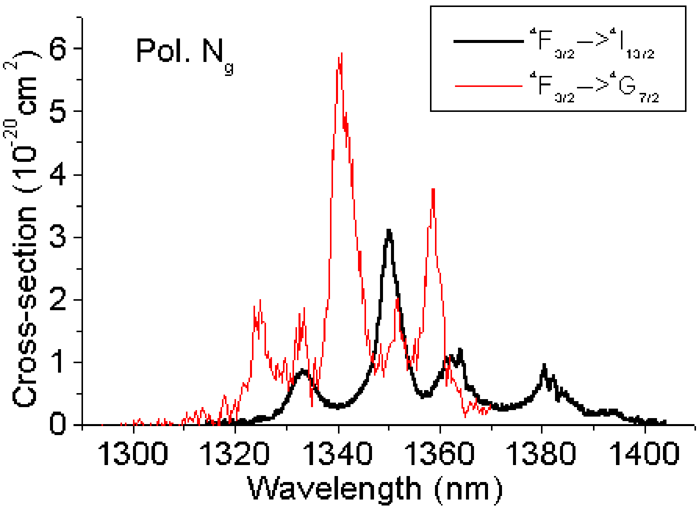


Figure 4. The energy level scheme of Nd^{3+} in the $KGd(WO_4)_2$ crystal.



(a)



(b)

Figure 5. Emission and ${}^4F_{3/2}$ excited-state absorption cross-sections along the *b*-axis in (a) N_m-polarization and (b) N_g-polarization in Nd^{3+}-doped $KGd(WO_4)_2$ crystal.

$\Omega_{2m} = 0.674 \times 10^{-19}$ cm^2, $\Omega_{4m} = 0.221 \times 10^{-19}$ cm^2, $\Omega_{6m} = 0.206 \times 10^{-19}$ cm^2 (N_m-polarization), $\Omega_{2g} = 0.265 \times 10^{-19}$ cm^2, $\Omega_{4g} = 0.152 \times 10^{-19}$ cm^2, $\Omega_{6g} = 0.145 \times 10^{-19}$ cm^2 (N_g-polarization).

The laser tests were done with the same conditions of pumping as in the previous section. The cavity was constituted of an entrance plane mirror and an output concave mirror having 15 cm radius of curvature. Their coatings were identical: 99.7% reflectivity at 1352 nm, 90% reflectivity at 1540 nm, 4% reflectivity at 1068 nm and high transmission in the visible range. Laser pulses were observed at 1352.5 nm and SRS Stokes 1 at 1539.5 nm in N_m-polarization. Their energies were measured with a Molectron pyrometer through adequate interference filters and are represented in figure 6 versus the pump power. The threshold of the Stokes 1 SRS is

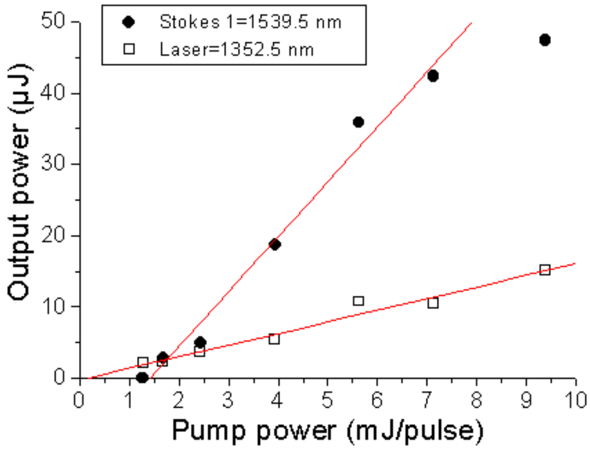


Figure 6. The energies of the laser at 1352.5 nm and the Stokes 1 SRS at pulses 1539.5 nm versus the pump power.

higher than that of the laser and the maximum conversion efficiency of the pump towards the Stokes 1 SRS was twice 0.63% (=1.26%), taking into account the fact that the SRS Stokes 1 escapes from the entrance mirror with the same energy as through the output mirror.

4. Conclusions

We have used a short-pulsed longitudinal pumping at 600 nm and obtained SRS frequency self-conversion of the 1352.5 nm laser line in $KGd(WO_4)_2:Nd^{3+}$. The wavelength which was produced is eye-safe at 1539.5 nm and the conversion efficiency was 1.26% with uncoated faces of the crystal. The yield can be improved by using antireflection-coated faces, an input mirror highly reflective at the Stokes 1 wavelength and an output mirror with a larger radius of curvature more adapted to our pump waist. Despite our non-optimal experimental conditions, we have obtained a higher conversion efficiency than in previous SRS self-conversion in the eye-safe region (0.15%) [9].

Acknowledgment

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