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# Raman lasers at 1.171 and 1.517 $\mu$ m with self-frequency conversion in SrWO<sub>4</sub>:Nd<sup>3+</sup> crystal

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#### Abstract

We have grown and characterized a SrWO<sub>4</sub>:Nd<sup>3+</sup> crystal. Photoluminescence and Raman spectra are exhibited. We have obtained end-pumped self-stimulated Raman scattering in the two  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  and  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{13/2}$  laser channels leading to 1.171 and 1.517  $\mu$ m coherent radiations. The latter is close to the eye-safe range of wavelengths.

(Some figures in this article are in colour only in the electronic version)

### 1. Introduction

Nonlinear optical phenomena in crystals are intensively used to extend the wavelength range of commercially available lasers such as the Nd:YAG. In this context, stimulated Raman scattering (SRS) is a promising phenomenon with the advantages of solid-state technology: compactness of the devices, low maintenance. A general feature is that totally symmetric vibrations (breathing modes) of molecules or groups of atoms inside a crystal produce the most intense Raman lines [1]. Accordingly, nitrates, tungstates, molybdates, carbonates, vanadates are the most promising crystals for efficient SRS.

In the steady state in the limit of no pump depletion, the Stokes 1 intensity  $I_S$  grows as

$$I_{\rm S}(l) = I_{\rm S}(0) \exp(GI_{\rm P}l) \tag{1}$$

where *l* is the length of the Raman medium,  $I_P$  is the pump intensity and *G* is the Raman gain coefficient. *G* was determined for the main crystals [2]—for example  $G = 11 \text{ cm GW}^{-1}$  for Ba(NO<sub>3</sub>)<sub>2</sub>—as well as other relevant Raman parameters [3].

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A few Raman crystals accept luminescent doping such as Nd<sup>3+</sup> ion doping and therefore they offer both stimulated emission and the frequency shifting. These materials are bifunctional and have much interest. The most well known is KGd(WO<sub>4</sub>)<sub>2</sub>:Nd<sup>3+</sup>. It was operated in the two  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  and  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{13/2}$  laser channels and was able to generate wavelengths up to 1538 nm (the eye-safe wavelength) [4–9]. Let us quote also La<sub>2</sub>(WO<sub>4</sub>)<sub>3</sub>:Nd [10], PbWO<sub>4</sub>:Nd [11], YVO<sub>4</sub>:Nd, GdVO<sub>4</sub>:Nd [12]. More recently, SrWO<sub>4</sub>:Nd<sup>3+</sup> crystal was obtained with optimized growth conditions [13]. The Raman gain coefficient was measured to be 5 cm GW<sup>-1</sup> and the SRS-active mode frequency 921 cm<sup>-1</sup>. The self-frequency conversion of the  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  laser channel was investigated under flash-lamp pumping and efficient Stokes 1 at 1.18  $\mu$ m was produced.

In this paper, we give the spectroscopic properties of the SrWO<sub>4</sub>:Nd crystal relevant for self-stimulated Raman scattering. Then we describe end-pumped self-stimulated Raman scattering in the two  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  and  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{13/2}$  laser channels for generating 1.171 and 1.517  $\mu$ m radiations, the latter being close to the eye-safe domain: 1.5–1.57  $\mu$ m.

## 2. Crystal growth

The initial chemicals, analytical grade  $SrCO_3$ ,  $WO_3$  and spectral grade  $Nd_2O_3$ , were mixed separately in the molar ratio in an agate motor and then charged into a platinum crucible. They were heated to 1100 °C and kept at this temperature for seven days. After that, the chemicals were deposited in an iridium crucible and were placed in the DJL-400 furnace with a nitrogen atmosphere. The compounds were heated to a temperature 50 °C higher than the crystallization temperature for about 2 h to let the melt mix completely and homogeneously. A SrWO<sub>4</sub> seed was selected from the small crystals obtained by the spontaneous crystallization and it was used to grow bulky SrWO<sub>4</sub> crystal. The seeds, used in the subsequent experiments, were oriented parallel to the *c* axis. The rotation and pulling rates were 12–15 rpm and 1–1.2 mm h<sup>-1</sup>, respectively. When these procedures were over, the crystals were drawn out of the melt and cooled down to the room temperature at a rate of 12–30 °C h<sup>-1</sup>. The Nd<sup>3+</sup> concentration in the SrWO<sub>4</sub> sample was measured to be 0.33 wt% by the inductively coupled plasma optical emission spectroscopy method.

# 3. Characterization of the SrWO<sub>4</sub>:Nd<sup>3+</sup> crystal

The SrWO<sub>4</sub> crystal structure is tetragonal and then uniaxial (space group:  $I4_1/a$ ). The SrWO<sub>4</sub>:Nd<sup>3+</sup> sample was 4 mm in diameter and 4.1 cm in length. It was oriented along the *a* axis.

The sample was excited near 750 nm with a Laser Analytical Systems dye laser (styryl 8 dye) pumped by a pulsed frequency-doubled Nd:YAG laser from BM Industries (duration of pulses: 8 ns). The Nd<sup>3+</sup> emissions were detected through an HRS2 Jobin-Yvon monochromator equipped with a 1  $\mu$ m blazed grating (2.4 nm mm<sup>-1</sup> slit resolution). The detectors were a R1767 Hamamatsu photomultiplier and a nitrogen cooled Northcoast germanium cell. The decay time was recorded with a 9410 Lecroy oscilloscope.

The Raman spectra were excited with an  $Ar^+$  ion laser (514 nm wavelength) focused onto the crystal. The Raman backscattered spectra in *c* and *b* axis polarizations were recorded at room temperature through an Olympus BH2 microscope coupled to a Dilor XY monochromator with 1.1 cm<sup>-1</sup> for a 100  $\mu$ m slit and a nitrogen cooled CCD detector.

The emission spectra corresponding to the  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  and  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{13/2}$  transitions are given in figures 1 and 2 respectively in  $\sigma$  (electric field  $\perp c$ ) and  $\pi$  polarizations (electric



Figure 1. Polarized emission spectra of SrWO4:Nd<sup>3+</sup> crystal corresponding to the  $^4F_{3/2} \rightarrow \,^4I_{11/2}$  transition.



Figure 2. Polarized emission spectra of SrWO<sub>4</sub>:Nd<sup>3+</sup> crystal corresponding to the  ${}^4F_{3/2} \rightarrow {}^4I_{13/2}$  transition.

field  $\parallel c$ ). Their structure reflects the Stark splitting of the <sup>2S+1</sup>L<sub>J</sub> manifolds in the S<sub>4</sub> point group which is the Sr<sup>2+</sup> symmetry site. The peaks are rather broad and this is a sign of the disordered environments of the Nd<sup>3+</sup> ions due to electric charge compensation. In figure 1 the highest peak is at 1057.0 nm wavelength in  $\pi$  polarization. In figure 2 the highest peak has 1331.4 nm wavelength.

The lifetime of the emissions originating from the  ${}^{4}F_{3/2}$  level was measured to be 186  $\mu$ s. This value can be compared to those of PbWO<sub>4</sub>:Nd: 165/175  $\mu$ s [11], of KY(WO<sub>4</sub>)<sub>2</sub>: 125 and 70  $\mu$ s respectively for 1% and 5% Nd [14] and of KGd(WO<sub>4</sub>)<sub>2</sub>: 120  $\mu$ s for 1% Nd [15].

In figure 3 we give the Raman spectra of our *a*-oriented sample relevant for the selfstimulating Raman scattering experiment described in the two next sections. These are a[cc]a, a[bc]a and a[cb]a spectra (let us recall that in the  $\alpha[\beta\chi]\delta$  labelling  $\alpha$ ,  $\beta$  are respectively the direction and the polarization of the exciting beam,  $\chi$ ,  $\delta$  have the same meaning for the Raman scattered beam). Figure 3(a) shows that the *c* polarization is the more efficient for obtaining *c*-polarized Raman frequency conversion. This is very favourable because the emission spectrum in figure 1 has its highest peak at 1057.0 nm in *c* polarization, and so we expect lasing in this polarization. Figure 3(b) compared with 3(a) shows that *c* polarization



Figure 3. Raman spectra of SrWO<sub>4</sub>:Nd<sup>3+</sup> crystal relevant for self-stimulated Raman scattering.

excitation leads to lower Raman conversion in *b* polarization than in *c* polarization. So we expect (see the following sections) to get *c*-polarized self-stimulating Raman scattering.

# 4. Self-stimulated Raman scattering of the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ laser channel

The laser properties of the SrWO<sub>4</sub>:Nd<sup>3+</sup> crystal were first tested at the 1057 nm line corresponding to the  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  transition. The crystal was longitudinally excited in *c* polarization along its *a* axis at 747 nm in the  ${}^{4}F_{7/2} - {}^{4}S_{3/2}$  levels. The laser cavity was constituted of a plane input mirror with high transmission in the visible range and high reflectivity (>99.5%) near 1057 nm and an output concave mirror (radius of curvature: 15 cm, 99% reflectivity at 1057 nm and 60% at 1170 nm), the distance between the two mirrors being 7.5 cm. The pump was focused behind the crystal with a 17.5 cm focal length lens; the pump waist was measured by the knife method to be 230  $\mu$ m at the entrance face of the crystal.

Laser pulses were generated at 1057 nm and SRS self-frequency conversion pulses were observed at 1171 nm (Stokes 1) in *c* polarization. Both the laser and Raman pulses were observed in *c* polarization in agreement with the spectroscopic characterization of section 3. 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 4 versus the pump power. The threshold of the Stokes 1 SRS is close to that of the laser and the maximum Stokes 1 SRS conversion was 1.8%. The time evolutions of the pump, laser and Stokes 1 SRS pulses were measured with the 9410 Lecroy oscilloscope and are represented in figure 5. The



Figure 4. Laser and Stokes 1 output power from self-stimulated Raman scattering of the  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  laser transition.



Figure 5. The pump, laser and Stokes 1 time evolutions for the  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  transition.

150 ns delay between the pump and the laser is interpreted as the build-up time of the laser pulse. The Raman pulse is contained inside the laser one.

# 5. Self-stimulated Raman scattering of the ${}^4F_{3/2} \rightarrow {}^4I_{13/2}$ laser channel

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 with high transmission at 1060 and 750 nm and high reflection at 1331 nm, and an output concave mirror having 15 cm radius of curvature, high reflection at 1060 nm and 5% transmission at 1520 nm. Laser pulses were observed at 1331 nm and SRS Stokes 1 at 1517 nm, both in *c* polarization. The Stokes 1 at 1517 nm energy was measured with a Molectron pyrometer through an adequate interference filter and is represented in figure 6 versus the pump power. The maximum conversion efficiency of the pump towards the Stokes 1 SRS was 0.4%.

#### 6. Conclusion

We have grown and characterized a SrWO<sub>4</sub>:Nd<sup>3+</sup> crystal. Photoluminescence and Raman spectra are exhibited. We have obtained end-pumped self-stimulated Raman scattering in the



Figure 6. Stokes 1 output power from self-stimulated Raman scattering of the  $^4F_{3/2} \to \,^4I_{13/2}$  laser transition.

two  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$  and  ${}^{4}F_{3/2} \rightarrow {}^{4}I_{13/2}$  laser channels leading to 1.171 and 1.517  $\mu$ m coherent radiations. The latter is close to the eye-safe range of wavelengths.

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