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Self-frequency-sum mixing in Nd doped nonlinear crystals for laser generation in the three fundamental colours The NYAB case

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Abstract

When combined with a suitable nonlinear crystal host, several coincidences occurring in Nd^{3+} ions allow for simultaneous generation of laser radiation in the three fundamental colours; red, green and blue (RGB) in the same solid state laser crystal. We show experimentally this possibility in neodymium doped yttrium aluminium borate (NYAB) Nd^{3+}:$YAl_3(BO_3)_4$, although the same achievement is very likely to be possible in other nonlinear crystals containing Nd^{3+} laser active ions.

Keywords: Nd doped nonlinear crystals; Laser generation; Colour

1. Introduction

Since the discovery of the first laser, the ruby solid state laser [1], the advantageous properties of laser radiation — high brightness, spatial and time coherence — have been exploited in science and technology. During the last few years the number of laser hosts has increased drastically, most of them based on Nd^{3+} doped crystals [2,3].

At present, much research is performed towards obtaining colour displays which take advantage of the high brightness and spectral purity achievable from laser radiation. It has been long known (Young–Helmhotz theory of trichromacy) that different colours to human eye can be additively generated from weighted combinations of only three fundamental colours; red, green and blue (RGB) [4,5]. Therefore, it is advantageous to generate laser radiation in these colours in a single compact laser system in order to reduce cost, size and to obtain potential integration.

Up to now semiconductor diode lasers have not been able to provide full coverage of the requirements in every field of science, technology and industry, specially those requiring a combination of long operation lifetimes at room temperature, continuous wave operation, narrow spectral linewidth and good beam quality at moderate to high powers.

Discovery of the excellent characteristics of rare earth ions to produce infrared laser action — in particular Nd^{3+} — and nonlinear optics, has led to extensive research to combine both potentials. Nd^{3+} doped crystals are of great interest since they show strong optical absorption around 750 and 800 nm [2,3] so that they can be easily pumped by commercial laser diodes. Laser oscillation around 900, 1060 and 1300 nm has been demonstrated in a variety of Nd^{3+} doped crystals, corresponding to transitions from the metaestable state ($^4F_{3/2}$) to the lower Stark states $^4I_{9/2}$, $^4I_{11/2}$ and $^4I_{13/2}$. When pump and laser radiation are propagating in a nonlinear host, they could interact each other providing the possibility of generating visible laser radiation [6].

In our search for new visible lasers based on Nd^{3+} ions introduced in a nonlinear host, we have found a unique set of coincidences occurring in optical transitions leading to RGB laser radiation in the same solid state crystal. We here report on this achievement, and show how laser-diode-pumped compact, stable and efficient lasing pixels emitting RGB radiation can be obtained in the same crystal and can serve as the basis of future high brightness displays technology. In our first experiments we have used a particularly suitable nonlinear borate crystal $YAl_3(BO_3)_4$ (YAB) activated with Nd^{3+} ions (NYAB). Nevertheless, the wavelengths associated to optical transitions among the different quantum energy states of Nd^{3+} ions are not very sensitive to the crystal field of the host environment [2,3], and the processes described in this report leading to generation of RGB are very likely to be possible in other

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Nd^{3+} doped nonlinear crystals as for example, $LiNbO_3$:MgO [7], $LaBGeO_5$ [7], $GdCa_4O(BO_3)_3$ [8] and $BaCaBO_3F$ [9].

NYAB is a nonlinear crystal characterised by a space group *R*32. Its nonlinear properties are mainly due to the two BO_3 groups, being one in the 001 plane and the another one slightly tilted with respect to this plane. This is a noncongruent melting crystal and flux method has to be used for crystal growth usually producing crystals no longer than 1 cm. Nevertheless this crystal is of very high technological potential since it presents excellent transmission for the expected visible wavelengths, high damage resistance, high segregation coefficient for Nd^{3+} ions, absence of photorefractive damage and low laser thresholds for laser oscillation at 1 and 1.3 μm [10–12]. All these properties ensures that NYAB is an efficient infrared laser system under both low and high pump power densities. In addition, NYAB has relatively high nonlinear coefficient [7] and its dispersion relations for the refractive indexes allows for simultaneous generation of second harmonic generation and frequency sum mixing processes [13].

Laser oscillation at 1062 nm (corresponding to the $^4F_{3/2}\rightarrow{}^4I_{11/2}$ channel) was firstly used to generate visible laser radiation in the green and blue [14,15] and even in the UV spectral domain [16]. In addition a theoretical model has been developed in order to estimate the NYAB efficiency as a blue emitting device based on this infrared channel [17]. From this model the optimum conditions required in a self-sum-frequency mixing process have been determined. Unfortunately, red radiation can not be generated based on laser oscillation at 1062 nm. Recently, it has been demonstrated simultaneous RGB laser radiation from a NYAB crystal operating 1338 nm corresponding to the $^4F_{3/2}\rightarrow{}^4I_{13/2}$ laser channel [13]. As a consequence, in this work we are exclusively dealing to this infrared laser channel.

Table 1 summarises all the nonlinear processes obtained from an end pumped NYAB laser crystal. In this table the oscillating laser channel, interacting (fundamental) and resulting wavelengths, nonlinear process used in each case as well as the reference works are listed.

Table 1
Nonlinear processes obtained from an end pumped NYAB laser crystal[a]

Laser channel	Interacting wavelengths (nm)	Resulting wavelength (nm)	Process	Ref.
$^4F_{3/2}\rightarrow{}^4I_{11/2}$	590+1062	379	SSFM	[17]
$^4F_{3/2}\rightarrow{}^4I_{11/2}$	740+1062	436	SSFM	[16]
$^4F_{3/2}\rightarrow{}^4I_{11/2}$	808+1062	459	SSFM	[16]
$^4F_{3/2}\rightarrow{}^4I_{11/2}$	890+1062	480	SSFM	[16]
$^4F_{3/2}\rightarrow{}^4I_{13/2}$	1062+1062	531	SFD	[7]
$^4F_{3/2}\rightarrow{}^4I_{13/2}$	755+1338	481	SSFM	[12]
$^4F_{3/2}\rightarrow{}^4I_{13/2}$	807+1338	505	SSFM	[12]
$^4F_{3/2}\rightarrow{}^4I_{13/2}$	1338+1338	669	SFD	[13]

[a] The oscillating laser channel, interacting (fundamental) and resulting wavelengths, nonlinear process used in each case as well as the reference works are listed.

In this paper we experimentally show that red laser light can be obtained by self-frequency-doubling (SFD) of infrared laser radiation at 1338 nm. In addition, we also show how the coincidence that absorption bands best suited for pumping require radiation around 750 or 800 nm can lead to generation of tunable laser radiation around 490–500 nm (green) and around 475–488 nm (blue) via self-sum-frequency mixing (SSFM) of pump laser radiation with the fundamental laser radiation at 1338 nm. In this way, obtaining the three colours simultaneously requires the simultaneous use of two pump wavelengths.

The main properties of NYAB as an infrared laser gain medium as well as a nonlinear crystal are analysed. From the analysis of the dispersion relations for the refractive indices the optimum propagation directions (phase matching directions) for each nonlinear process have been determined. In addition the temperature dependence of these phase matching directions is reported. This data being of great interest since when NYAB is diode pumping relative high crystal temperatures are achieved.

2. Results and discussion

2.1. Characterisation of NYAB as a laser gain medium at 1338 nm

Fig. 1 shows the infrared laser power as a function of the absorbed pump power for different output coupler transmittances. Laser experiments were performed in a quasihemispherical cavity consisting of a flat dichroic input mirror (high reflectance at 1338 nm) and output coupler of 10 cm radius of curvature. Pump beam from the diode laser was focused into the crystal by a 5 cm focal lens. Several output mirrors of different transmittances

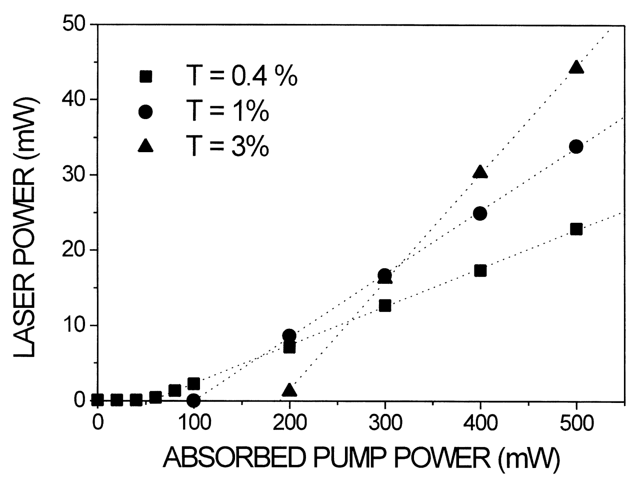

Fig. 1. Infrared laser power as a function of the absorbed pump power for different output coupler transmittances.

were available ($T = 0.4$, 1 and 3%). End pumping was performed by a CW laser diode emitting at 807 nm. Crystal used in laser gain experiments was a 5 mm long prism cut $\theta \cong 31°$ with respect to optical axis, being this the optimum direction for simultaneous RGB laser light generation. Pump power at threshold as low as 60 mW has been obtained when the high reflectance output coupler was used. Commercial diode lasers are in the hundred of milliwatts level so that they can efficiently be used to NYAB pumping.

It is well known that some physical parameters of great relevance on laser dynamics, such as internal optical loss factor and effective emission cross section can be determined from an appropriate analysis (Mode Overlap Formalism) of the laser gain experiments [11]. By this way the effective emission cross section has been estimated from data displayed in Fig. 1 to be $3.8 \cdot 10^{-20}$ cm^2 being this comparable to the values obtained in other Nd^{3+} doped laser crystals [7]. In addition, the low internal optical loss factor ($0.85\% \cdot cm^{-1}$) ensures laser oscillation with low pump thresholds even for long NYAB crystals.

Two beam laser spectroscopy has been previously used to estimate the presence of excited state absorption (ESA) of laser radiation [18]. This process consists in the absorption of one laser photon by one Nd^{3+} ion populating the metaestable state (unique populated excited state during laser oscillation). The influence of ESA in laser dynamics is an increase in the pump power at threshold. In the NYAB case, results showed that at the laser wavelength (1338 nm) no ESA is taking place.

2.2. Characterisation of NYAB as a self-frequency-doubling (SFD) and self-sum-frequency-mixing (SSFM) material

In what is known as sum-frequency mixing two photons of different energy add together to produce a single photon of total energy. Three waves coexist and are allowed to exchange energy [6]. The degenerate case in which two photons of same energy are added to produce a single photon of twice energy corresponds to second harmonic generation. These processes are realised involving electric charges contained in the nonlinear host crystal. Nonlinear frequency conversion processes are more efficient when in addition to energy, momentum is conserved:

$$\lambda_3^{-1} = \lambda_1^{-1} + \lambda_2^{-1} \quad \text{Energy} \tag{1}$$

$$\vec{k}_1 + \vec{k}_2 = \vec{k}_3 \quad \text{Momentum} \tag{2}$$

where λ_1 refers to infrared laser wavelength (1338 nm); λ_2 is the infrared laser wavelength in a SHG process and the pump wavelength in a SSFM process; and λ_3 is the resultant visible wavelength. Fig. 2 shows the visible laser wavelength as a function of the pump wavelength as resulting from a SSFM process. It should be noted here that the visible laser beam changes from blue to green when the pump wavelength is changed from 750 to 810 nm (Nd^{3+} absorption peaks). On the other hand the visible radiation generated by SHG is located at 669 nm resulting in a red beam.

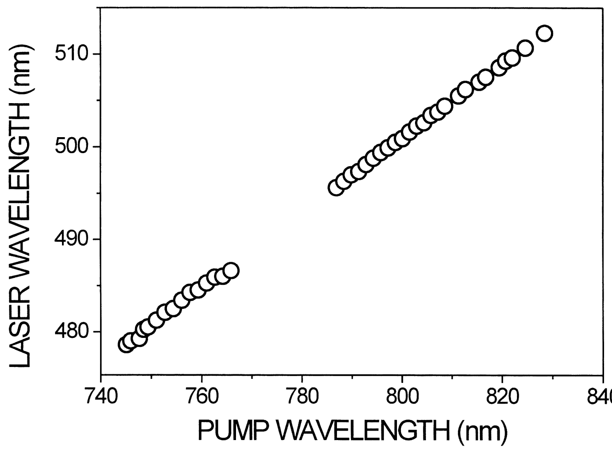

Fig. 2. Visible laser wavelength as a function of the pump wavelength as resulting from a SSFM process.

The rate and direction of energy transfer among the waves depend on their relative phases. To satisfy momentum conservation it its necessary to produce phase matching among the three waves participating in the process. In principle, the standard technique of efficient phase matching for self frequency sum mixing and second harmonic generation is the use of birefringence in nonlinear crystals, in which certain directions of propagation, θ_{PM}, favour the efficiency of conversion. In case of an uniaxial crystal of negative birefringence like NYAB, these directions are set by the conditions [6]:

$$n^e(\lambda_3, \theta_{PM})\lambda_3^{-1} = n^o(\lambda_1)\lambda_1 + n^o(\lambda_2)\lambda_2 \quad \text{In a Type I process} \tag{3}$$

$$n^e(\lambda_3, \theta_{PM})\lambda_3^{-1} = n^e(\lambda_1, \theta)\lambda_1 + n^o(\lambda_2)\lambda_2 \quad \text{In a Type II process} \tag{4}$$

In a Type I processes both radiations should be ordinary polarised. On the other hand, in Type II processes one laser radiation is ordinary polarised being the other extraordinary polarised. In the NYAB case pump efficiency is optimum when this is achieved by an ordinary polarised beam [11]. In addition previous works demonstrated that when laser cavity is aligned for low threshold, laser radiation is ordinary polarised. As consequence, for the NYAB case, visible laser light generation is only efficient by means of a Type I process.

Using the dispersion relations for the refractive indices reported previously for NYAB [19], we have solved θ_{PM} from Eq. (3). In a first step, we have considered the SFD process by assuming $\lambda_1 = \lambda_2 = \lambda_3/2$. We have then calculated the SFD phase matching direction a function of the fundamental laser wavelength (results are shown in Fig. 2 as a solid line). The θ_{PM} angle for SFD of the fundamental laser line at 1338 nm has been found to be 27°.

The phase matching direction for SFSM process can be obtained by similar way. In this case the laser wavelength is fixed (1338 nm) whereas the pump and visible wavelengths are related by energy conservation (see expression 1). The phase matching direction is now solved as a function of pump wavelength. Results are displayed in Fig. 2 (dotted line). By inspection of Fig. 3 we can determine that for a pump wavelength of 760 and 807 nm (where the absorption coefficient peaks) a value of $\theta_{PM} = 31.5$ and 30.8° are respectively obtained. As the θ_{PM} angle to generate red radiation by SFD ($\theta = 30.8°$) is close to these angles, both SFD and SSFM processes can occur in the same laser element. Although the angle acceptances full width at half maximum (FWHM) estimated for these processes in a 5 mm long crystal are around 0.5° [17], the use of focused beams (pump and laser) increases the FWHM remarkably. As we will see, these angular acceptances allow for simultaneous achievement of SFD and SSFM processes.

Recently it has been observed that when NYAB crystal is working as a laser system (and therefore optically pumped) it heats substantially [20]. Temperatures up to 80°C for pump powers around 1 W have been observed. The mechanism responsible of this crystal heating is the low ($\cong 0.2$) quantum efficiency associated to the $^4F_{3/2}$ state (defined as the number of radiative decays divided by the number of total decays from the metastable state) [21]. As a consequence, when $^4F_{3/2}$ state is populated (as it is during laser oscillation) an intense multiphonon decay is produced increasing crystal temperature.

The NYAB working temperature depends, mainly, on the pump power [20]. This dependence is shown as an inset in Fig. 4. When a laser device is to be designed the

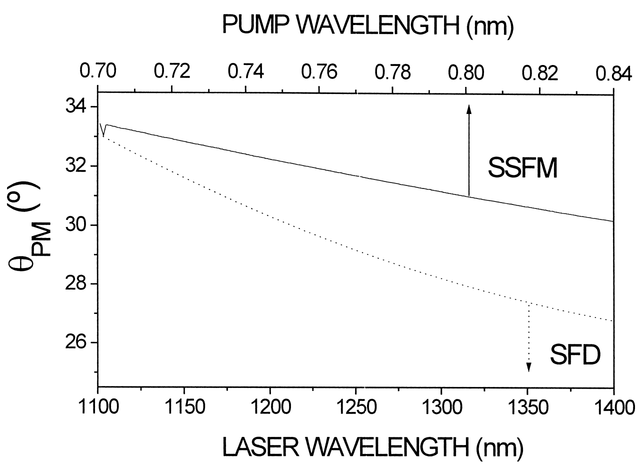

Fig. 3. Phase matching angle for SFD (solid line) and SSFM (dotted line) processes as a function of fundamental and pump wavelength, respectively.

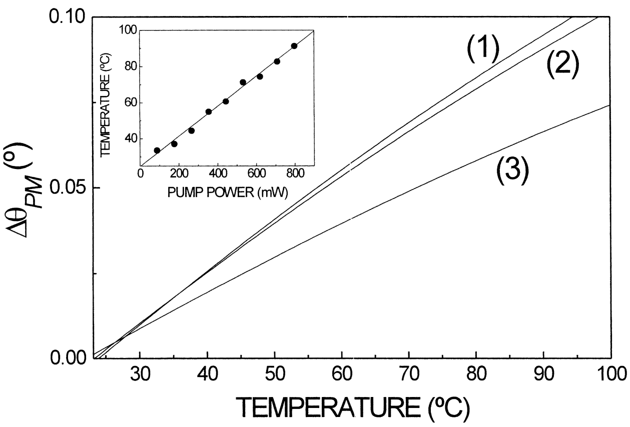

Fig. 4. Increment in the phase matching angle as defined in text for a SFD process of the fundamental laser line at 1338 nm (3) and also for a SSFM process between fundamental laser line (1338 nm) and pump radiation at 760 nm (2) and 807 nm (1). The crystal temperature as a function of pump power is shown as an inset.

first step is to choose the pump source, so that, the pump power (and therefore the crystal working temperature) is known. Since refractive indices depend on temperature [20], when solving the phase matching angle from Eq. (3), the final solution obtained is a function of crystal temperature ($\theta_{PM} \equiv \theta_{PM}(T)$). Fig. 4 shows the increment in the phase matching angle ($\Delta\theta_{PM}(T) = \theta_{PM}(T) - \theta_{PM}(T = 25°C)$) for a SFD process of the fundamental laser line at 1338 nm (red generation) and also for a SSFM process between fundamental laser line (1338 nm) and pump radiation at 760 nm (blue generation) and 807 nm (green generation). By combination of data displayed in Fig. 4, the optimum crystal orientation for each pump power can be determined.

2.3. Simultaneous RGB laser light generation

As was mentioned before simultaneous RGB radiation can be generated from a NYAB crystal operating at 1338 nm. For this, SFD and SSFM processes are generated in the laser gain medium. The wavelength of the SSFM radiation depends on the pump wavelength (see Fig. 2). Since green and blue radiations have to be generated, two pump wavelengths are required. This can be overcome by simultaneous pump from two laser sources emitting at the desired wavelengths. In our RGB experiments we have used a laser diode (807 nm) and a Ti:Sapphire laser (750 nm) to simultaneous pumping the NYAB crystal. An schematic representation of the experimental set-up used for these experiments is shown in Fig. 5. The two pump beams were made collinear by using a beam-splitter. The laser cavity and NYAB crystal were the same as that used for IR laser oscillation and described in Section 2.1.

Fig. 6 shows the spectrum of visible radiation obtained for a total pump power of 1.2 W (600 mW from both Ti:Sapphire and laser diode). The powers of green and blue

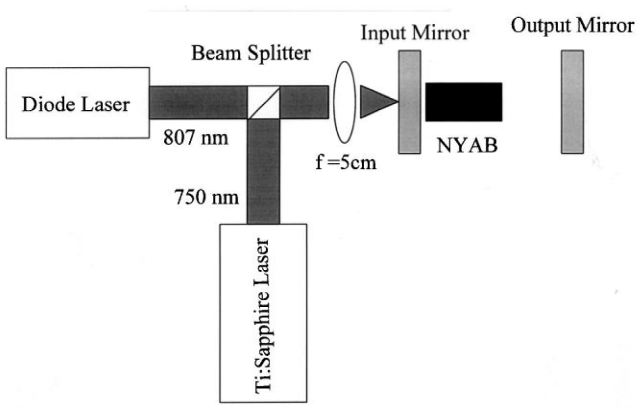

Fig. 5. Schematic diagram of the experimental set-up used for simultaneous RGB generation.

laser obtained are of the order of microwatts, due to poor phase matching, whilst the power of red laser generated is of order milliwatts.

Thus far, there is experimental evidence of the possibility of simultaneous generation of RGB. The natural question now is how efficient and compact can these systems be.

A first link on the chain of efficiency is infrared laser efficiency. Nd^{3+} ion based lasers are very efficiently pumped by means of laser radiation from GaAlAs emitting around 800 nm and 750 nm, allowing for compact diode-pumped all-solid-state lasers based on resonant pumping. On one hand GaAlAs are commercially available in high powers, and with electric-to-optical power conversion efficiencies in excess of 30%. Nd^{3+} has an intense absorption band around 800 nm and the emission from GaAlAs laser diodes can be used to pump Nd^{3+} ions resonantly. It is typical to find among Nd^{3+} doped materials, absorption coefficients of order 10 cm^{-1}, in which the pumping efficiency is almost unity. Theoretically, around 75% power of pump radiation can be converted to infrared laser power. In practice, slope efficiencies of order 50% are routinely obtained. This means that if diode lasers efficient convert electric-to-optical power, the efficiency of a diode pumped infrared laser from electric to optical power is still very high. Because minimum heat is transferred to the crystal by resonant pumping, air cooled systems can be developed. Pumping efficiency is therefore very high, and also results in high plug-in efficiency, potentially leading to battery operated devices.

The next question is how efficient nonlinear intracavity frequency conversion can be. Theoretical [22] and experimental work [23], demonstrate that a 100% of the infrared output power from a laser, can be intracavity converted to other frequency. In case of a self-frequency-doubled laser, it means that if one develops a system supplying say 100 mW of infrared output laser power, a suitable design of the system can lead to 100 mW of output power at the second harmonic. The key is that because the intracavity circulating power density is very high, only a few percent of it need to be converted to second harmonic to obtain 100 mW. The output coupling of the infrared laser is substituted by leaking of circulating infrared power in form of second harmonic. To realise this limit intracavity linear losses at the fundamental need to be very small, and the nonlinear material used must have a certain grade of nonlinearity. In case of a NYAB SFD laser in the green, 60 of those 100 mW have been successfully converted to green power [14]. It is expected that skilled laser cavity engineering can lead to 100% in the case of SFD and SSFM lasers. In this way, the overall efficiency of electric to second harmonic power is of order 10% (already demonstrated) although values around 20% are possible in principle. A 1 W GaAs diode laser could generate around 500 mW of green power, out of an electric power around 5 W (A real good efficiency if one keeps in mind that several energy conversions are realised in the process). This, in principle, is extensive to other intracavity frequency conversion processes.

This nonlinear frequency conversion efficiency relies on the possibility of simultaneous phase matching. The difficulty in obtaining good phase matching by use of birefringence can be overcome by techniques provided by nonlinear optics like quasiphase matching and skilled engineering.

The variation of the pump wavelength changes the spectral position of the laser radiation produced by SSFM (see Fig. 2). We now focus our attention in the green laser radiation generated by the interaction between pump (807 nm) and infrared (1338 nm) photons. It is well known that in a diode laser the emission wavelength depends on the working temperature. We have changed this diode temperature then looking at the green radiation wavelength. A total tuning range of 3 nm is obtained when the diode temperature is varied from 10° up to 30°C. This data ensures an output wavelength control by means of a thermal tuning of

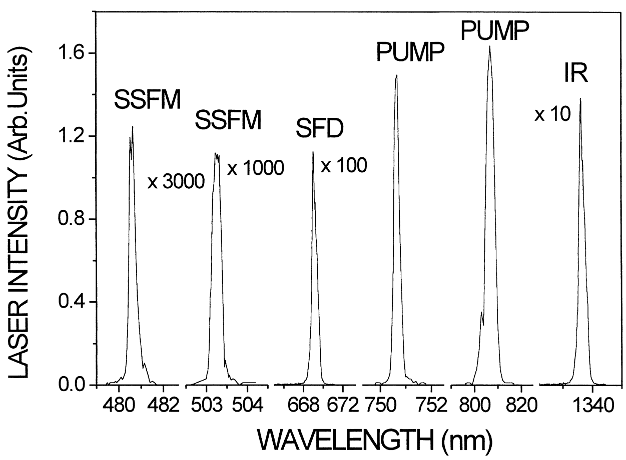

Fig. 6. Spectral distribution of the output laser beam obtained in RGB experiments. Note the different magnifications used for each line. The processes causing each line are also indicated.

diode laser. Finally, this small tunability range within the absorption band allows for some freedom in choosing pump wavelengths to optimise the gamma of colours.

3. Conclusions

Red, green and blue laser light has been generated at the same time by a single NYAB crystal operating at 1338 nm under simultaneous Ti:Sapphire and diode pumping. Red was generated by SFD the fundamental laser line at 1338 nm. Green and blue were generated by SSFM of the two pump beams with the infrared laser radiation. The optimum conditions for these two nonlinear processes have been determined as a function of wavelength and crystal temperature. Diode temperature induced tunable (3 nm) green laser radiation has also been demonstrated. Although the obtained pump to visible efficiencies are low, SFD together with SSFM emerge as a promising and efficient mechanism for visible laser light generation from diode pumping Nd^{3+} doped crystals.

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