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Stimulated Raman scattering and second harmonic generation from a Nd : Y V O4 laser at 1064 nm to generate 587 nm.

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Stimulated Raman scattering and second harmonic generation from a $Nd:YVO_4$ laser at 1064 nm to generate 587 nm.

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Abstract. We have investigated a light generation at 587 nm based on a combination of two nonlinear processes, stimulated Raman scattering (SRS) in $Nd:GdVO_4$ and second harmonic generation (SHG) in BBO, which efficiently convert the 1064 nm fundamental wavelength of a $Nd:YVO_4$ laser to yellow light. An average power of $0.72\,W$ was measured at the output of 1174 nm before SHG with only $3.7\,W$ pumping.

1. Introduction

For high harmonic generation from Sodium vapor, the atoms have to be brought to a excited state before the driving field is applied. This excitation is performed by a 587 nm pulse in the ps time scale. The excitation by two successive pulses allows to reach close to 100% population in the excited state [1]. As we know, generation of yellow light directly by using solid state laser is not yet possible. However, it has been obtained by various approaches in the past years, including sum frequency generation with 1064 nm and 1319 nm obtained from mode locked $Nd:YVO_4$ lasers [2] and Raman shifting picosecond pulses in $LiIO_3$ followed by second harmonic generation and by OPO and SFG from a mode locked $Nd:YAG$ laser[3].

In this work, we report high efficiency conversion by using $Nd:GdVO_4$ Neodymium doped Gadolinium Vanadate for Raman shift which is followed by frequency doubling from a 1064 nm $Nd:YVO_4$, 10 ps pulse duration. It is one of the active Raman scattering material that measured a $885cm^{-1}$ frequency shift. Thus, it will give us 1174 nm in the first Stokes, with a high Raman gain coefficient of around $4.5\,cm/GW$ [4].

Solid state Raman shifting crystals are attractive because they offer a higher gain and better thermal properties than gaseous media. $Nd:GdVO_4$ was known in 1994 as a material for diode pump laser [5], as well as Raman crystal. In the following sections, some of the important aspects of SRS are given. The last section presents the results of our experiment in a simulation and experiment.

2. Theory

We expose the theory of our experiment in two parts, stimulated Raman scattering(SRS) theory first and second harmonic generation (SHG) next. As the second harmonic generation is a known theory we spend more time on the stimulated Raman scattering.

2.1. Stimulated Raman scattering(SRS)

It is known that the dipole moment per unit volume; or the polarization, relates to the strength of the incident electro- magnetic field:

$$P_i = \sum_j \chi_{ij}^{(1)} E_j + \sum_{j,k} \chi_{ijk}^{(2)} E_j E_k + \sum_{j,k,l} \chi_{ijkl}^{(3)} E_j E_k E_l + ... \tag{1}$$

The third term of the equation gives the Raman scattering. This term also results in the different kinds of nonlinear optical phenomena (e.g. two-photon absorption, Brillouin scattering and self-focusing).

Scattering is a redirection of incident light when is traveling through a medium. Light and matter interaction gives a periodically perturbation of electron orbits. This perturbation or oscillation occurs in the same frequency of incident light. Then, the molecules get a periodic separation of charge which is named induced dipole momentum. Since this oscillation is a source of electro-magnetic radiation, large amount of light is scattered at the identical frequency of the incident light. This process referred as elastic scattering. However, small amount of scattered light is at different frequencies, as inelastic one. Raman scattering is an example of such inelastic scattering.

Therefore, photons are inelastically scattered when they pass through the Raman media. Some of them are scattered in lower frequencies than incident light frequency (Stokes scattering) and other ones in higher frequencies (Anti-stokes scattering). We will summarize some important aspects of Raman scattering.

The value of the induced dipole moment $\bar{p}$ can be shown in the following form:

$$\bar{p} = \alpha \bar{E} \tag{2}$$

where α is the polarizability and E is the strength of the electric field of the incident EM wave. The polarizability is a property of the material which relates to the structure of the molecular and nature of the bands. As known, for incident EM wave, the electric field is expressed as:

$$\bar{E} = E_0 \cos(2\pi\nu_0 t) \tag{3}$$

where ν_0 is the frequency of the incident beam. If we combine equations 1 and 2 we obtain the time-dependent induced dipole moment in the form:

$$\bar{p} = E_0 \cos(2\pi\nu_0 t) \tag{4}$$

Since the ability to perturb the local electron cloud of a molecular structure relates to the relative position of individual atoms, the polarizability is a function of the instantaneous

position of each atoms. If we define $E_{vib} = (J + \frac{1}{2})h\nu_{vib}$ as the vibrational energy of a particular mode, where J is the vibrational quantum number ($J = 0, 1, 2, ..$), ν_{vib} is the frequency of the vibrational mode, h is the Planck constant, as we know, for any molecular band, the individual atoms have specific vibrational modes, in which the vibrational energy level are quantized in a manner similar to the electronic energy.

Based on the particular vibrational mode, the physical atoms displacement dQ around their equilibrium position is written:

$$dQ = Q_0 \cos(2\pi\nu_0 t) \tag{5}$$

where Q_0 is the maximum displacement around the equilibrium position. For such small displacements, the polarizability may be approximated by a Taylor expansion, namely,

$$\alpha = \alpha_0 + \frac{\partial\alpha}{\partial Q} dQ \tag{6}$$

where α_0 is the polarizability of the molecular mode at the equilibrium position. Due to the vibrational displacement of Equ. (5), the polarzability may be given as:

$$\alpha = \alpha_0 + \frac{\partial\alpha}{\partial Q} Q_0 \cos(2\pi\nu_0 t) \tag{7}$$

Finally, by using a trigonometric identity after substituting Equ. (7) into Equ. (3) we obtain:

$$p = \alpha_0 E_0 \cos(2\pi\nu_0 t) + \left(\frac{\partial\alpha}{\partial Q}\frac{Q_0 E_0}{2}\right)\{\cos[2\pi(\nu_0 - \nu_{vib})t] + \cos[2\pi(\nu_0 + \nu_{vib})t]\} \tag{8}$$

Final equation presents three terms for induced dipole moments. Each term results a different scattering in a different frequency (ν_0 , $\nu_0 - \nu_{vib}$, $\nu_0 + \nu_{vib}$). First one in same frequency of incident light is an elastic scattering (e.g. Mir or Rayleigh); Two last terms are inelastic one in lower and higher frequencies, which referred to as Stokes and Anti-Stokes scattering, respectively. [7]

Raman scattering like other light scattering can be spontaneous scattering or stimulated. Stimulated Raman scattering (SRS) differs from spontaneous Raman scattering in a number of ways; it appears at high intensity light, it occurs in light beams of small divergence and, finally, its spectrum of stimulated emission shows distinct line narrowing. From scattering theory, we obtain an exponential light amplification:

$$N_s^* = N_s^*(0)\exp(gI_L l) \tag{9}$$

where g denotes the gain factor of the individual scattering process, I_L the intensity of the incident light beam, l the length of interaction and $N_s^*(0)$ the photon flux at z=0. As is seen from Equ. (9), the amplification is determined by the gain factor g of the specific process, the intensity of incident beam I_L and the interaction length l.

The main part of stimulated scattering light are in the forward and backward direction within the light beam propagation of small angular divergence. This small beam angle is mainly caused by a geometrical effect. The interaction length is much larger than the beam diameter. Since the amplification grows exponentially with the interaction length Equ. (9) a narrow cone around the beam (both direction) is observed.

There are two temporal cases, a steady state regime and a transient one, which are important for our further investigation. When the pump pulse duration is much longer than the dephasing time which is intrinsic to the material used ,($\tau_p >> T_R$) we are in the steady state regime. Conversely, for the transient regime we have the pump pulse duration smaller than the dephasing time ($\tau_p << T_R$) and the spectral width of pump laser $\Delta\nu_p$ is much broader than the Raman line homogeneous broadening $\Delta\Omega_R = (\pi c T_R)^{-1}$ ($\Delta\nu_p >> \delta\Omega_R$) [8].

For quantitative description of SRS we require a set of differential equations which introduce light intensities in the medium in both direction (backward and forward direction)[6]. This set of equations is:

$$\frac{n_l}{c}\frac{\partial I_L}{\partial t} + \frac{\partial I_L}{\partial z} = -g_L(I_{s1}^F I_L + I_{s1}^B I_L) - \beta I_L \tag{10}$$

$$\frac{n_{s1}}{c}\frac{\partial I_{s1}^F}{\partial t} + \frac{\partial I_{s1}^F}{\partial z} = g_{s1} I_{s1}^F I_L - \beta I_{s1}^F \tag{11}$$

$$\frac{n_{s1}}{c}\frac{\partial I_{s1}^B}{\partial t} - \frac{\partial I_{s1}^B}{\partial z} = g_{s1} I_{s1}^B I_L - \beta I_{s1}^B \tag{12}$$

Where g_L and g_{s1} are gain factor and they are related by: $g_{s1} = g_L w_{s1}/w_L$. I^B and I^F are backward and forward intensity respectively. Furthermore, β is the absorption coefficient. A full study of stimulated Raman emission needs the solution of the coupled equations together with higher Stokes components. After solving the set of Equs. (10, 11 , 12) we consider only, the on-axis term. Since we neglect backward scattering, we omit the superscript F:

$$\frac{\partial I_L}{\partial t} = -g_L I_{s1} I_L - \beta I_L \tag{13}$$

$$\frac{\partial I_L}{\partial t} = -g_L(I_{si} I_{si-1} - I_{si+1} I_{si}) - \beta I_L, I = 1, 2, 3, ... \tag{14}$$

We will use the last set of equation to simulate SRS, for finding right crystal we need know how much shift is required to achieve $1174 - 1180$ nm Since we need around $885 - 912\ cm^{-1}$ shift to obtain $1174 - 1180$ nm and due to available options we studied several crystal which are presented in Tab. 1.

Barium Nitrate, $Ba(NO_3)_2$ is well known Raman crystal which has the highest Raman gain coefficient. The gain coefficient affects the threshold for Raman laser. $Ba(NO_3)_2$ for Raman application is more optimal in case of ns and longer pulses. Due to the pulse duration we are using, This is not applicable for our experiment.

The potassium gadolinium tungstate $KGd(WO_4)_2$ or KGW single crystal is considered as a new and capable material for optical and laser applications. Because

Table 1. Some Physical properties of Raman crystals. [8]

Name	Raman shift[1]	Raman gain[2]	Damage threshold[3]	Dephasing Time[4]
KGW	901	3.3	10	2
$Nd:GdVO_4$	885	4.5	20	3.5
$Ba(NO_3)_2$	1048	11	0.4	26

$1: cm^{-2}$, $2: cm/GW$, $3: GW/cm^2$, $4: ps$

of its low structure symmetry and high χ^3-nonlinearity, KGW offers an unusual wealth of properties and $901\,cm^{-1}$ Raman shift, It was our first choice. We use plenty time to get result from this crystal. Because of high Raman gain and low damage threshold, a damage in the crystal was observed during the experiment.

Then, we decided to use $Nd:GdVO_4$. Neodymium doped Gadolinium Vanadate ($Nd:GdVO_4$) is a well known material for diode pumped laser. Similar to $Nd:YVO_4$ crystal, $Nd:GdVO_4$ crystal also exhibits high Raman gain, low Raman threshold, high damage threshold and low absorption coefficients at pumping wavelengths, which result from the excellent fit of the neodymium dopant in the crystal lattice. Thus, it is an excellent choice for our work.

2.2. Second harmonic generation (SHG)

Second harmonic generation is the most common nonlinear process which can be easily dealt with. Comprehensive theories of this process is exposed in [9]. The main aspects of the theory of SHG generation are summarized in the following.

Second harmonic generation can be observed in crystals without inversion of symmetry. It can be risen by $\chi^{(2)}$ nonlinearity, where a pump beam generates another beam with twice its frequency in the media. In most cases, the pump wave is delivered as a laser beam, and the doubled frequency is generated. The output beams are propagating in the same direction with a small angle which is called the walk-off angle. The wave equation at 2ω is well known in the following form:

$$\frac{\partial E(2\omega)}{\partial z} = -\frac{i\omega}{n_{2\omega}c} d_{\text{eff}} E^2(\omega) e^{i\Delta k z} \tag{15}$$

where it is assumed that losses are negligible. The slowly varying envelope approximation is used to achieve the expression ($\Delta k = k(2\omega) - 2k(\omega)$).

For no pump depletion ($E(2\omega) << E(\omega)$) the amplitude $E(\omega)$ remains basically constant during the interaction. Then, if $E(2\omega, z = 0)$ is 0 the electrical field can be obtained in the following equation:

Table 2. Results for BBO and KDP from SLNO. [10]

Crystal	Type	Phase match angle[1]	Wave length range[2]	Damage Threshold[3]
BBO	$o+o=e(I)$	21.5	190-1780	50
KDP	$o+o=e(I)$	42.0	200-1500	2.5-5

$1:\theta[degree]$, $2:nm$, $3:GW/cm^2$

$$E(2\omega, z=l) = -\frac{i\omega d_{\text{eff}}}{n_{2\omega}c}E^2(\omega)\int_0^l e^{i\Delta kz}dz = -\frac{i\omega d_{\text{eff}}}{n_{2\omega}c}E^2(\omega)l\,\frac{\sin(\Delta kl/2)}{\Delta kl/2}e^{i\Delta kl/2} \tag{16}$$

Also the optical intensity can be achieved by using $I = n/2\sqrt{\epsilon_0/\mu_0}|E|^2$,:

$$I(2\omega, l) = \frac{2\omega^2 d_{\text{eff}}^2 l^2}{n_{2\omega}n_\omega^2 c^3\epsilon_0}\left(\frac{\sin(\Delta kl/2)}{\Delta kl/2}\right)^2 I^2(\omega) \tag{17}$$

The phase matched condition, $\Delta k = 0$, gives the high conversion efficiency. If the process is not phase matched, the generated waves go out of phase and the generation at $2w$ will be lost.

In this work, Second harmonic generation has been used to obtain 587 nm laser from a 1064 nm source. The process starts with pumping the source into a Raman crystal in order to generate the 1174 nm light which is fed through a SHG crystal to double its frequency. Due to the phase matched and the beam properties, the right SHG crystal was chosen by using SNLO software (Tab. 2). Beta Barium Borate (BBO) is a good choice because of availability, price and efficiency. BBO crystal combines very wide transparency, moderately high nonlinear coupling, high damage threshold and good chemical and mechanical properties. Phase matching in BBO crystal covers a wide range, yielding SHG from 190 nm to 1780 nm.

2.3. Simulation Results

In this section, simulation of stimulated Raman Scattering in $Nd:GdVO_4$ will be discussed. In previous sections we discussed about theory of the stimulated Raman scattering and we got a set of coupled equations. Also, we studied in the properties of $Nd:GdVO_4$. As mentioned before we used Equs (12, 13) to simulate SRS. To solve the equations we use the ODE45 Matlab differential solver package. Here, we set the initial values of the fundamental input intensity and the Stokes intensity. Our assumption for the fundamental input intensity was $5\,GW/cm^2$ and, it was $10^{-12}\,GW/cm^2$ for the Stokes. We made our calculation for two different lengths of matter interaction. We dealt with 10 cm length crystal to show first Stokes and higher order Stokes since there

are more Stokes in much longer interaction length (Fig 1). The plot shows the pumping intensity and the intensity of the Stokes. As presented, we have a slight slope in each Stokes in maximum since the media absorbs a part of incident beam energy. This amount depends on β, absorption coefficient. Another point that should be considered is width of Stokes intensities which varies related to the gain factors.

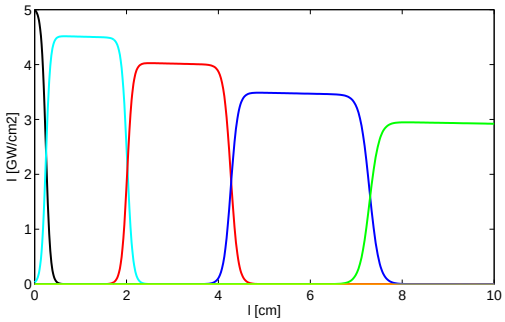

Figure 1. Calculated intensity versus length of interaction. Input intensity $5\,GW/cm^2$ for 10 cm interaction length

Since $Nd:GdVO_4$ was not commercially available in 10 cm, we conducted our work with two 1.2 cm $Nd:GdVO_4$ crystals in series. This leads to a more practical result. Thus we made our simulation in two stages. For doing this, we should consider the output of the first crystal as the input for the second crystal in both wavelengths. The results for the first crystal and second are shown in Fig. 2. We can see the amount of amplification for the 1st Stokes is not very pronounced. Based on our calculation the 1st Stokes intensity reached $0.031\,GW/cm^2$ in output which is enough to be used as a seed for the second crystal.

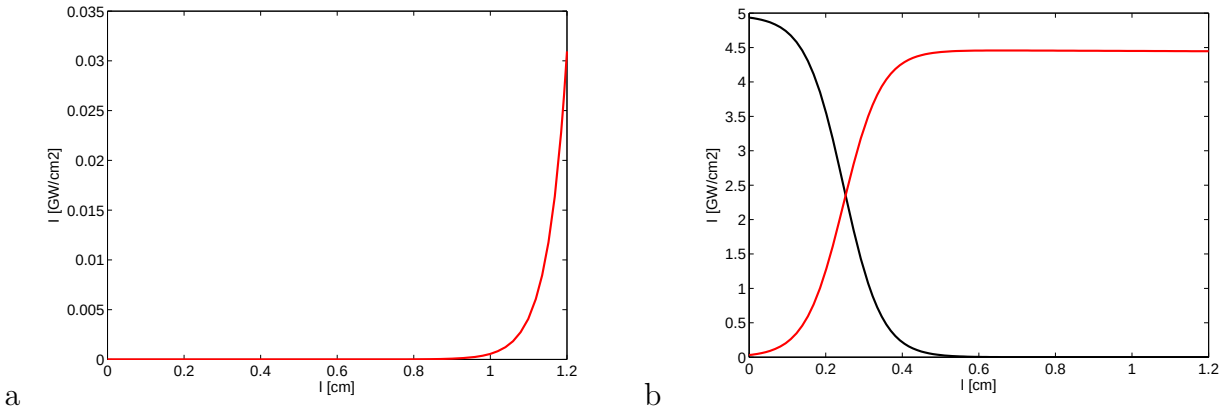

Figure 2. Calculated intensity versus length of interaction, a) First crystal with $10^{-12}\,GW/cm^2$ amount of initial value of first Stokes , b) Second crystal with $0.031\,GW/cm^2$ amount of initial value of first Stokes.

The next plot (Fig. 3) is the intensity versus Raman gain factor G. The relation between Raman threshold, length and input intensity is represented in this figure. As shown in the figure, Raman gain for maximum conversion is around 10. By using $G = I_p l g_0$, we calculated the best input power to run our experiment. Without considering in other losses (eg. backward scattering, high order Stokes, ...), the calculated value was $1.85\,GW/cm^2$. These losses may play an important role in the experiment result.

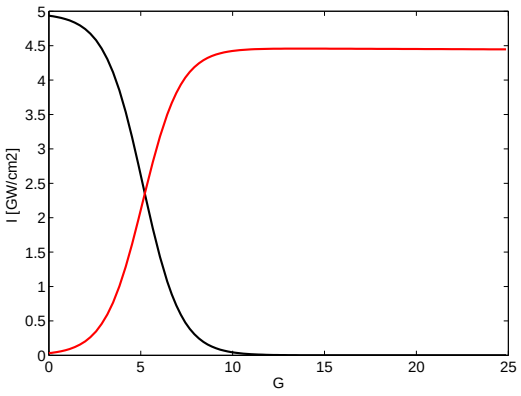

Figure 3. Calculated Intensity of input and first Stokes versus G.

2.4. Experimental Results

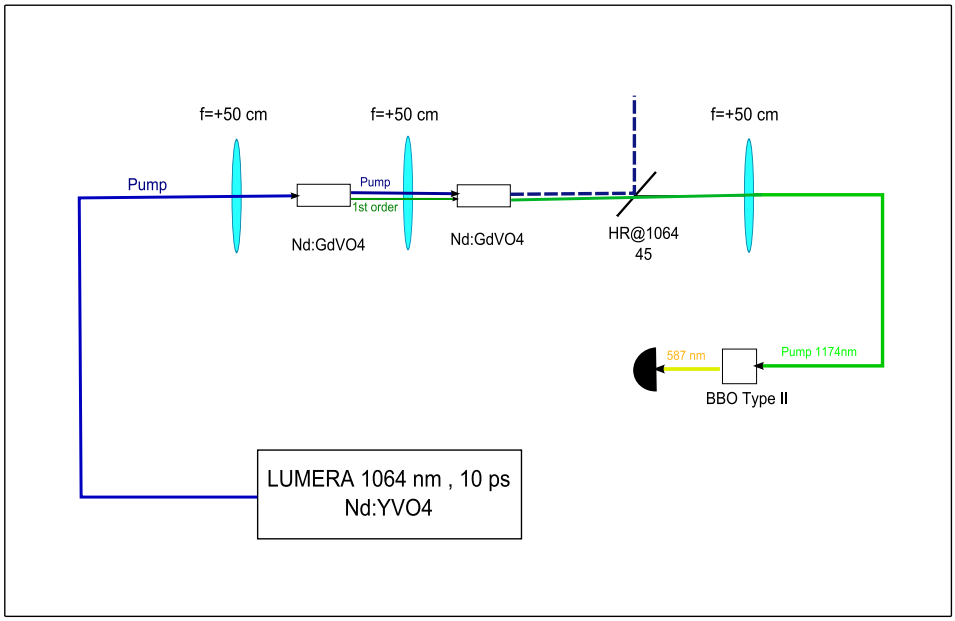

Figure 4. Experimental setup

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The experimental setup is shown in Fig.4. The pump laser used in the experiment is a HYPERAPID from LUMERA laser GmbH. The beam has a wavelength 1064 nm with 10 ps pulse duration, 100 kHz repetition rate and 3 mm beam diameter. The output can reach up to 40 W. The Raman generation will be achieved in a two-stage system. Using a 50 cm focal length lens we focused our beam down to 150 μm such that high intensity beam is reached to Raman scattering threshold generation in the first $Nd:GdVO_4$. In the second stage $Nd:GdVO_4$, we imaged the first crystal using a +10 cm focal length lens to the second crystal. For preventing the fundamental wavelength to reach the detector we used a dielectric mirror coated for high reflection at 1064 nm and 45 degree incidence after the $Nd:GdVO_4$ crystal. The whole light focused to the detector by the use of a 5 cm focal length lens.

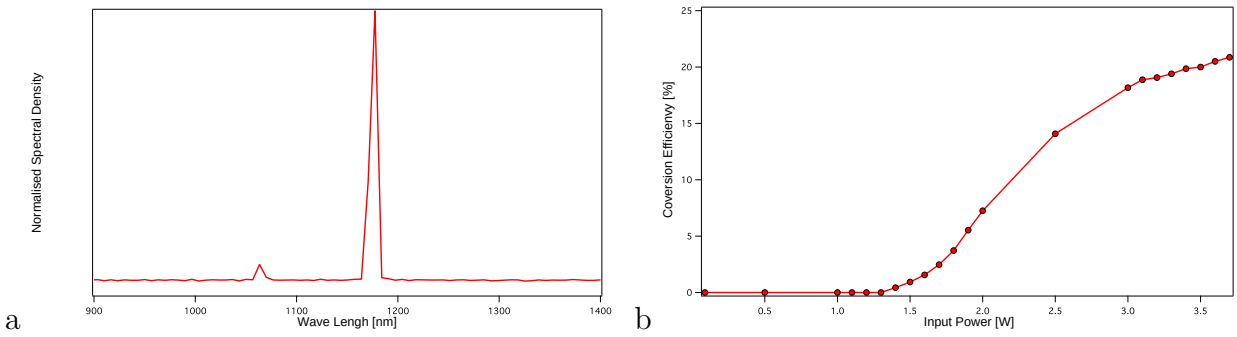

Figure 5. a) Spectrum of the output at 1174 nm, b) Measured conversion efficiency versus input power

Our measurement for Raman scattering has been plotted in Fig.5. As shown, the conversion efficiency exponentially grows until saturating around 20%. The Raman threshold is about $1.5\,GW/cm^2$. When we reached up to $7.3\,GW/cm^2$ we noticed a change in output power. This phenomenon has been observed three times. As no physical damage is observed, we thought that color centering in starting to occur. As we are working with high intensity beams, the issue of damage has to be considered for every optics.

The second step of experiment gives us the light at 587 nm by frequency doubling after getting 1174 nm from $Nd:GdVO_4$. A BBO crystal has been used to achieve SHG. The input power for SHG is 20% of the fundamental input before Raman shift. SHG has been observed as shown in Fig. 6.

3. Conclusion

We have demonstrated a practical approach with a new crystal for the development of a solid-state laser source in the 587 nm, based on $Nd:YVO_4$ laser employing efficient Raman shifting. Our results showed that we can achieve 20% conversion effeciency in

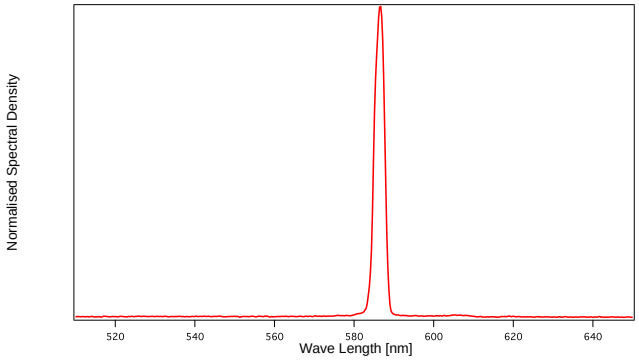

Figure 6. Spectrum of yellow light 587 nm

the first Stokes wavelength at 1174 nm and SHG in a BBO crystal has been achieved as a proof of principle.

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