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Pump and probe measurements of optical amplification at 584 nm in dysprosium doped lithium niobate crystal

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ABSTRACT

Usually the Dy^{3+} ions have been used as laser sources associated with the $^6H_{13/2} \rightarrow {}^6H_{15/2}$ transition about 3 μm. However, these ions exhibit emissions in the blue (480 nm) and in the yellow (575 nm) that are promising for laser operations. In this work, positive optical gain about 1.9 cm^{-1} (~8.2 dB/cm) has been observed at 584 nm in trivalent dysprosium doped lithium niobate crystal, Dy:$LiNbO_3$, using a pump and probe experimental setup. High power laser pulses at 475 nm have been used as the pump source in order to strongly populate the $^4F_{9/2}$ level of the Dy^{3+} ions due to ground state absorption. Low signal beam cw radiation at 584 nm has been used as the probe beam to stimulate the emission associated with the $^4F_{9/2} \rightarrow {}^6H_{13/2}$ electronic transition of the Dy^{3+} ions. The process has been modelled as a four level system and the level populations have been analysed and simulated in order to study the gain dynamics. Optical amplification of the probe signal has been observed during the first 300 μs, which represents a good agreement between the measured lifetime of the $^4F_{9/2}$ level and the proposed simulation.

1. Introduction

Single crystal of lithium niobate ($LiNbO_3$) (denoted LN) has been studied intensively during several decades and its properties are well documented in numerous papers [1,2]. LN crystals are considered as promising versatile materials in optics and photonic science and technology. They are widely used in optical modulators, optical frequency doublers, optical parametric oscillators, holographic data storage, and even for acoustical memory as reported [3,4]. The LN crystals doped with rare earth ions become luminescent media able to generate and amplify light [5–9]. This ability, combined with inherent non-linear properties and the LN transparency, extending from 350 to 1800 nm, offers a possibility to design self Q-switching and self-frequency doubling laser source.

Dy^{3+}-doped crystals or glasses have been considered as promising laser active materials able to emit radiation associated with the $^6H_{13/2} \rightarrow {}^6H_{15/2}$ transition of Dy^{3+} around 3 μm [10]. Little attention was paid to the visible emission originating in the $^4F_{9/2}$ state situated at about 21,000 cm^{-1} [9,11]. The intense emission in yellow region near 575 nm offers the potential of laser operation in the four-level-scheme since the related transition terminates in the $^6H_{13/2}$ level which is situated well above the ground $^6H_{15/2}$ level. Feasibility of laser operation in Dy^{3+}-doped tungstate crystal in the $^4F_{9/2} \rightarrow {}^6H_{13/2}$ channel (yellow emission) and in the $^4F_{9/2} \rightarrow {}^6H_{11/2}$ channel (red emission) has been demonstrated upon flash-lamp pumping at liquid nitrogen temperature [12]. Commercialization of blue laser diodes opens new possibilities of optical pumping; therefore, potential laser transitions in the visible are to be reconsidered.

With this intention, we report here on the room temperature (RT) optical amplification properties of Dy^{3+} doped LN at around 580 nm. The gain dynamics of this process has been analysed by using the corresponding rate equations that describe the excited state population changes. A good agreement between the experimental data and the simulation has been found and supports the proposed excitation scheme.

2. Experimental

The samples of this study with a Dy^{3+} concentration of 1.94% have been reported in a previous work [11,13]. Measurements of optical amplification were carried out in a pump and probe experimental setup (see Fig. 1). The pump radiation was provided by an optical parametric oscillator (OPO) syntonized at 475 nm with high energy pulses between 25 and 75 mJ/cm^2 of about 5 ns of duration. The probe beam was obtained by a continuous 400 W lamp combined with a monochromator, resulting in a signal power density of 195 $\mu W/cm^2$ at 584 nm with a spectral width of 5 nm.

The incident of pump and probe beams (linearly polarized) were directed normal to the surface of the sample (parallel to the

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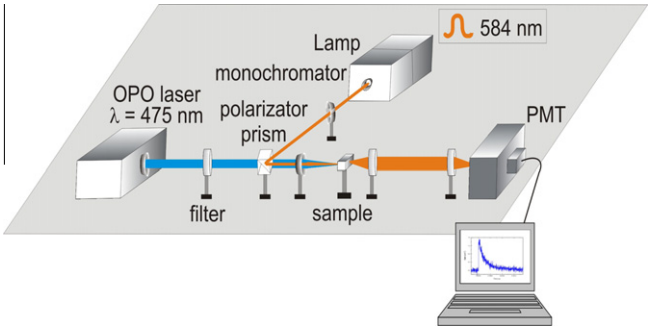

Fig. 1. Experimental setup used in the pump and probe experiments.

c-axis) which was situated after a 1 mm diameter pinhole. A dichroic mirror was employed to align both beams. In order to cover only the whole area of the pinhole, the pump beam was focused by a 20 cm focal length lens.

The detection chain was formed by a TRIAX-180 monochromator with 1 nm resolution and the output of the photomultiplier tube was registered by a digital oscilloscope TEKTRONIX-2430A for temporal analysis of the decay curves.

To determine the optical gain, two kinds of emission spectra were measured. In the first one, the pump and probe beams were present simultaneously, while in the second one the probe was blocked. Both spectra were compared after subtracting the continuous background due to the probe.

3. Results and discussion

The Dy^{3+}: $^6H_{15/2} \rightarrow {}^4F_{9/2}$ ground state absorption (GSA) is centred about 475 nm. In the pump and probe experiments, the high power pump pulses at 475 nm induce resonant GSA strongly populating the $^4F_{9/2}$ excited level. In these conditions, a probe beam tuned at 584 nm can induce a relaxation process involving the stimulated emission of a photon at the same frequency. The energy levels diagram shows the proposed mechanisms to find optical amplification in this setup (see Fig. 2).

As mentioned in the experimental section, two emission spectra are recorded: the first one with pump and probe present and the second one when the probe is blocked. These spectra are given in Fig. 3. An increase of the detected intensity at the signal wavelength, 584 nm (indicated by an arrow in the figure), can be clearly observed. This increment is due to the stimulated emission

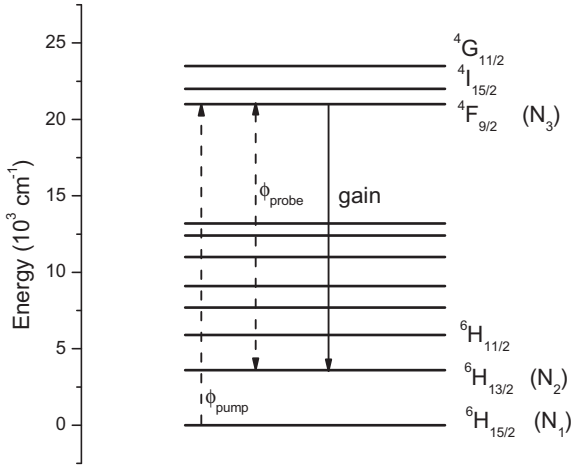

Fig. 2. Energy level diagram of Dy^{3+} ions.

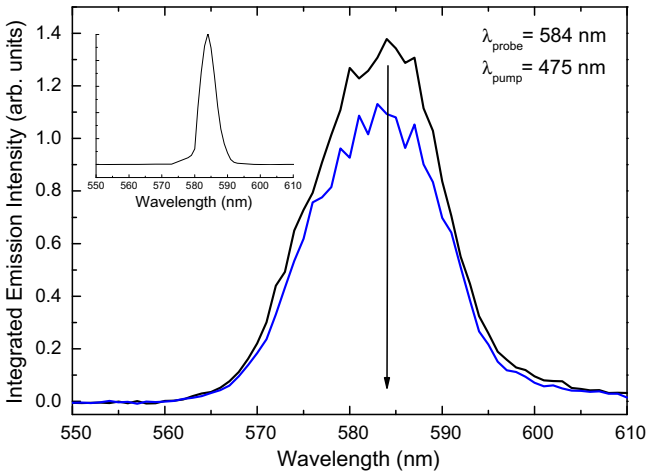

Fig. 3. Integrated emission intensity obtained using a pump at 475 nm and with or without a probe at 584 nm. The inset shows the probe spectra with a width of 5 nm.

associated with the $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transition that occurs at the probe wavelength and it is the physical basis of signal amplification. Under this pumping scheme population inversion for $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transition at 584 nm is expected at $t = 0$ s because $^6H_{13/2}$ is not initially populated. Moreover, to corroborate that this enhancement of the emission is due to the probe beam, the wavelength of the probe beam was shifted from 584 nm to the sides of the spectrum and it was found that the intensity enhancement was shifted accordingly to the probe wavelength. However, when the probe beam wavelength is chosen to lie outside of the emission band no changes in the emission spectrum are registered.

Here we explain how one can estimate the net optical gain in the sample from our experimental data. When a probe beam passes through a solid medium, its intensity decreases from the initial I_0 value to the final $I_{\text{probe}}(L)$ value according to the exponential law:

$$I_{\text{probe}}(L) = I_0 e^{-\alpha L} \tag{1}$$

where α and L are the absorption coefficient and the length of the sample, respectively ($L = 0.2$ cm). When the pump and the probe are simultaneously switched on, the intensity recorded after the sample at the signal wavelength, I_{pp}, is given by

$$I_{\text{pp}} = I_{\text{p}} + I_0 e^{(g_{\text{int}} - \alpha)L} \tag{2}$$

where I_{p} is the spontaneous emission intensity induced by the pumping radiation and g_{int} is the internal gain coefficient. Then, the net optical gain coefficient g_{net} can be expressed as

$$g_{\text{net}} = g_{\text{int}} - \alpha \tag{3}$$

Since α is negligible at 584 nm [11,13], I_{probe} is constant along the sample and equation, then $g_{\text{net}} = g_{\text{int.}}$ The signal enhancement (SE) is defined as

$$\text{SE} = \frac{I_{\text{pp}} - I_{\text{p}}}{I_{\text{probe}}} \tag{4}$$

By introducing Eqs. (2) and (3) in the expression (4), SE can be related directly with the net optical gain coefficient as follows:

$$\text{SE} = \exp(g_{\text{net}} L) \tag{5}$$

The intensities I_{p}, I_{pp} and I_{probe} can be experimentally measured. As we use a pulsed excitation source, I_{p} and I_{pp} are experimental curves that decay after the excitation pulse. Therefore, the values of SE and g are calculated by using Eqs. (4) and (5) from these experimental curves. The maximum of the detected intensity just

after the pump pulse was used in order to obtain the g value. The gain coefficient as a function of the pump energy density is given in Fig. 4. A continuous growth of the net gain coefficient as a function of the pump power density can be noticed in this figure. The maximum value for the net gain has been observed for a pump energy density of 0.72 J/cm^2 corresponding to 1.9 cm^{-1} (~8.2 dB/cm) during the maximum of the detected intensity.

In order to study the gain dynamics of this process, the rate equations that describe the excited state population changes have been analysed. A simplified scheme of the levels involved in the process under consideration is shown in Fig. 2. N_i (i = 1, 2, 3) represent the population of the $^6H_{15/2}$, $^6H_{13/2}$ and $^4F_{9/2}$ states, respectively. The rate equations for the populations of the involved levels are

$$\frac{dN_3}{dt} = \phi_{pump}\sigma_{ab}N_1 - W_3N_3 - \phi_{probe}\sigma_{em}(N_3 - N_2) \quad (6)$$

$$\frac{dN_2}{dt} = W_3b_{32}N_3 - W_2N_2 + \phi_{probe}\sigma_{em}(N_3 - N_2) \quad (7)$$

$$\frac{dN_1}{dt} = -\phi_{pump}\sigma_{ab}N_1 + b_{31}W_3N_3 + b_{21}W_2N_2 \quad (8)$$

The number of ions removed from ground state by the pump beam is expressed as $\phi_{pump}\sigma_{ab}N_1$, where σ_{ab} is the absorption cross section at 475 nm and ϕ_{pump} is the incident pumping flux. W_3 and W_2 are the $N_3 \rightarrow N_2$ and $N_2 \rightarrow N_1$ radiative relaxation probabilities, respectively and b_{ij} are the radiative branching ratios. The value of stimulated emission produced by the probe beam is expressed as $\phi_{probe}\sigma_{em}N_3$, where σ_{em} is the emission cross section of the $N_3 \rightarrow N_2$ transition and ϕ_{probe} is the probe beam flux. The temporal evolution of the gain has been simulated with the help of these rate equations.

Due to the pulse of the pump beam, Dy^{3+} ions are excited from the ground state N_1 to the upper state N_3. Then, population inversion for $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transition at 584 nm is expected at t = 0 s because $^6H_{13/2}$ is not initially populated. The large energy gap from the $^4F_{9/2}$ to the next lower energy level, around 7000 cm^{-1}, prevents high multiphonon deexcitation. Therefore, only radiative relaxation of this level is expected.

The rate Eqs. (6)–(8) are a set of coupled differential equations that have no analytical solution and, therefore, they have been numerically solved in order to simulate the optical amplification process. The parameters used in this simulation are taken from [11,13]; $W_3 = 2582\ s^{-1}$, $W_2 = 92.15\ s^{-1}$, $\sigma_{em} = 0.58 \times 10^{-20}$ cm^2, $\sigma_{ab} = 6.0 \times 10^{-22}$ cm^2. The flux of the probe has been measured experimentally and the obtained value is $\phi_{probe} = 195\ \mu W/cm^2$.

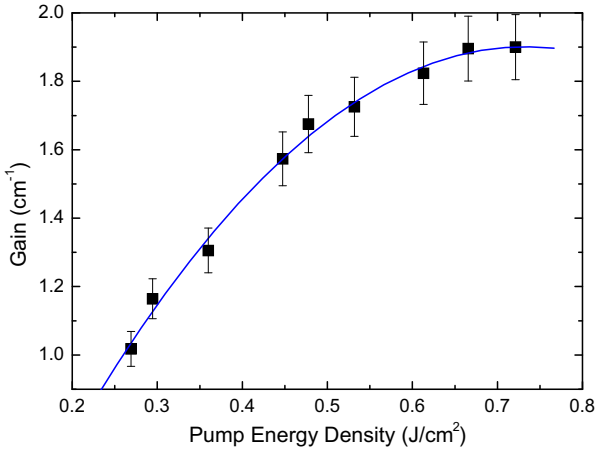

Fig. 4. Optical gain as function of the pump power density.

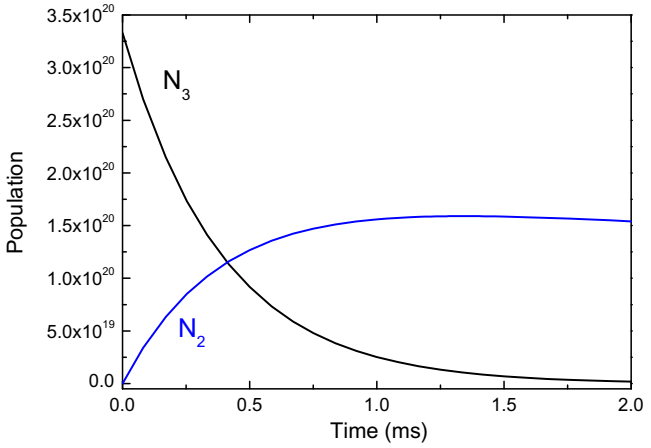

Fig. 5. Simulation of the temporal evolutions of the populations of the $^4F_{9/2}$ (N_3) and $^6H_{13/2}$ (N_2) levels.

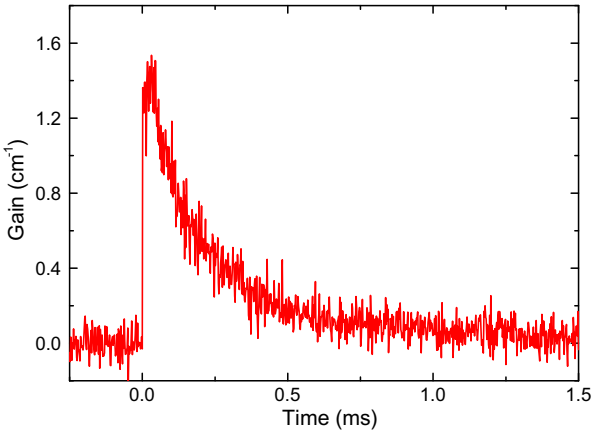

Fig. 6. Optical gain as function of the time for a pump power density of 0.45 J/cm^2.

The branching ratios b_{ij} are specified in Ref. [13]; $b_{32} = 0.54$, $b_{31} = 0.33$ and $b_{21} = 1$.

Fig. 5 shows the simulated temporal evolution of the N_2 and N_3 levels as a function of the time. At the initial time, N_3 is populated due to GSA and there are not any ions in the N_2 state. Then N_2 increases due to the ions that decay from N_3. The gain coefficient can be related to the N_3 and N_2 populations through the expression:

$$g = \sigma_{em}(N_3 - N_2) \quad (9)$$

According to Eq. (9) and Fig. 5 a positive optical gain is expected during the first 300 μs. This simulation is in good agreement with the experimental results as can be seen in Fig. 6. Under a pump of 0.45 J/cm^2 a net gain is obtained during this temporal interval.

A rough estimation of the population inversion that is achieved between the Dy^{+3}: $^4F_{9/2}$ and the $^6H_{13/2}$ levels can be made taking into account Eq. (9). Using the previous value obtained for σ_{em}, the population inversion for the maximum gain coefficient, 1.9 cm^{-1}, is estimated about $N_3 - N_2 \approx 3.27 \times 10^{20}$ ions/cm^3 which is very close to the total Dy^{3+} concentration, around 3.33×10^{20} ions/cm^3 and indicates that at this pump power most of the Dy^{3+} ions are excited.

4. Conclusions

Optical gain has been observed in Dy^{3+} lithium niobate crystal by using the pump and probe technique at 584 nm at room temperature. In this experiment the sample has been excited with a

pulse OPO laser at 475 nm and amplification has been obtained during the first 300 μs. This gain depends on the pump energy density, reaching its maximum value of about 1.9 cm^{-1} (~8.2 dB/cm) at a pumping intensity of 0.72 J/cm^2.

Acknowledgments

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References

[1] M.E. Lines, A.M. Glass, Principles and Applications of Ferroelectrics and Related Materials, Clarendon Press, Oxford, 1997.
[2] P. Gúnter, J.P. Huignard (Eds.), Photorefractive Materials and their Applications, vol. 6, Springer Verlag, Berlin, 1998.
[3] M.S. McPherson, I. Ostrovskii, M.A. Breazeale, Phys. Rev. Lett. 89 (2002) 115506.
[4] T. Volk, M. Wöhlecke, Lithium Niobate: Defects Photorefraction and Ferroelectric Switching, Springer, Berlin, Heidelberg, 2008.
[5] G. Dominiak-Dzik, S. Gołąb, I. Pracka, W. Ryba-Romanowski, Appl. Phys. A 58 (1994) 551.
[6] J.K. Jones, J.P. de Sandro, M. Hempstead, D.P. Shepherd, A.C. Large, A.C. Tropper, J.S. Wilkinson, Opt. Lett. 20 (13) (1995) 1477.
[7] R. Burlot, H. Moncorgé, G. Manaa, Y. Boulon, J. Guyot, D. Garcia-Solé, Opt. Mater. 6 (1996) 313.
[8] J. Zheng, Y. Lu, G. Luo, J. Ma, Y. Lu, N. Ming, J. He, Z. Xe, Appl. Phys. Lett. 72 (15) (1998) 1008.
[9] W. Ryba-Romanowski, I. Sokólska, G. Dominiak-Dzik, S. Gołąb, J. Alloys Compd. 300–301 (2000) 152.
[10] L.F. Johnson, H.J. Guggenheim, Appl. Phys. Lett. 23 (1973) 96.
[11] W. Ryba-Romanowski, G. Dominiak-Dzik, P. Solarz, R. Lisiecki, Opt. Mater. 31 (11) (2009) 1547–1554.
[12] A.A. Kaminskii, U. Hommerich, D. Temple, J.T. Seo, K. Ueda, S. Bagayev, A. Pavlyuk, Jpn. J. Appl. Phys. 39 (2000) L208.
[13] G. Dominiak-Dzik, W. Ryba-Romanowski, M.N. Palatnikov, N.V. Sidorov, V.T. Kalinnikov, J. Mol. Struct. 704 (1–3) (2004) 139–144.