EXCITED STATE ABSORPTION MEASUREMENTS AND 1064 nm LASER EMISSION OF $YVO_4:Nd^{3+}$ SINGLE CRYSTAL FIBER GROWN BY THE LHPG TECHNIQUE

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Abstract

High quality neodymium doped YVO_4 single crystal fibers were grown by the Laser Heated Pedestal Growth (LHPG) technique and characterized by XRD, absorption, fluorescence, lifetime and excited state absorption measurements. Laser action at 1064 nm was achieved for a 1.0 atm% doped sample adapted to an end-pump cavity with output power efficiency comparable to a bulk crystal indicating the potentiality of the fiber for the construction of diode-pumped compact lasers.

Introduction

For decades, great interest has been demonstrated for neodymium doped crystals that can be used as laser active media with applications in the near infrared region. Among these crystal hosts, the orthovanadates YVO_4 and $GdVO_4$ have attracted special attention due to their excellent structural, mechanical and spectroscopic properties [1]. The laser emissions of $YVO_4:Nd^{3+}$ at 1064 and 1342 nm for instance, present cross sections 2.7 and 18 times larger than the well know $YAG:Nd^{3+}$ respectively, and a four times larger absorption coefficient for the ideal pumping level at 808 nm. Most of the spectroscopic and structural studies in $YVO_4:Nd^{3+}$ have been done on large bulk crystals grown by Czochralski and other techniques but more recently, the increasing demand for compactness of optical devices such as diode-pumped, high efficiency miniature lasers, has also raised the interest for single crystal fibers (SCFs). One advantage of compact lasers is the ability to obtain single mode oscillation without the need of introducing wave plates or etalons in the laser cavity. The use of small crystals or fibers enable shortening of the cavity so that its mode spacing is greater than the gain bandwidth and that way only one mode oscillates. Besides, compact lasers allow the pumping with low cost diode lasers, rather than lamps, and it is easier to stabilize their short cavity to minimize thermal and mechanical perturbations.

Among the techniques that can be used for the growth of SCFs, the Laser Heated Pedestal Growth (LHPG) has proved to be a very efficient one. It can be described as a modified floating zone technique in which the molten zone is supported by the surface tension between two solid precursor rods referred to as "seed" and "nutrient". The rods can be crystalline, polycrystalline or simply a mixture of powder precursors, and the heating is done by a high power CO_2 laser (λ = 10.6 μm) eliminating the need of crucibles. These characteristics make LHPG very advantageous due to its low cost operation and the possibility of growing fibers in much faster processes than the usual ones. Such processes are also adequate to obtain $YVO_4:Nd^{3+}$ SCFs, but the growth of this crystal has proved to be a difficult task, not only by LHPG, but also using other techniques. Apparently, oxygen deficiencies due to vanadium oxide evaporation cause valence changes of vanadium ions during growth and that results in the formation and inclusion of dark colored phases that compromise the optical quality of the crystals. The high melting point of YVO_4 (~1850 °C) adds further difficulty to its growth. Therefore, several attempts have been made to improve growth conditions and we have also made a contribution by achieving the successful growth of transparent $YVO_4:Nd^{3+}$ SCFs employing isostatic oxygen atmosphere (5-10 atm) during the melt and pulling processes of LHPG at high pulling rates (~0.5 $mm.min^{-1}$) [2]. More recently, we have also noticed and reported that the inclusion of La^{3+} ions in the vanadates composition cause a controlled structural disorder that brings significant stability to the molten zone during growth [3]. As for the spectroscopic features of these $RE_{1-x}La_xVO_4$ (RE = Y, Gd) fibers doped with Nd^{3+}, they are not much different than those of YVO_4 and $GdVO_4$ except for a slight broadening of spectral lines. This broadening is an advantageous characteristic if one considers that diode lasers usually have a linewidth of 2-3 nm (larger than the absorption lines of neodymium doped into crystals), and therefore, temperature control of the diode is often needed to increase the pump efficiency.

Regarding solid-state lasers, absorption from excited states (ESA) can be a considerable loss mechanism for stimulated emissions, therefore, an evaluation of these transitions strengths and their implication on the laser efficiency is of great importance. Gain-ESA spectrum has been measured for bulk $YVO_4:Nd^{3+}$ and it was concluded that at 1064 nm, the loss via such mechanism is negligible when compared to the high emission cross section of the laser transition [4]. In this paper, we present the gain-ESA spectra of the SCFs of $YVO_4:Nd^{3+}$ in

addition to their general spectroscopic characterization via absorption, fluorescence and lifetime measurements, in comparison with a commercial laser bulk crystal grown by the Czochralski method. Moreover, by adapting a 1.0 atm% Nd^{3+} doped SCF and the bulk crystal to an end-pump (808 nm) configuration laser cavity, stimulated emission was observed at 1064 nm for both samples and their slope efficiency was compared.

Experimental Setup

Single crystal fibers of YVO_4 doped with 0.1 to 1.0 atm% Nd^{3+} were grown from precursor cylindrical rods as described elsewhere [2]. The rods were prepared from the homogeneous mixture of optical graded Y_2O_3, V_2O_5 and Nd_2O_3 oxides with a polymeric reagent, followed by extrusion of the resultant ceramic paste. After drying in air, the rods were used as both seed and nutrient, in the melt and pulling processes of LHPG. The growths were done in a closed chamber, under controlled 5.0 atm isostatic oxygen atmosphere at the pulling rate of 0.5 $mm.min^{-1}$. The inspection of crystal structure and presence of secondary phases was done by X-ray diffraction.

The spectroscopic measurements were done at room temperature. Absorption spectra were obtained in the range of 670 to 950 nm, using a Nicolet Magna IR 850 spectrophotometer equipped with a Si detector and a quartz beamsplitter. Fluorescence measurements were done from 850 to 1430 nm under diode excitation at 808 nm and using an InGaAs detector. The average 1/e lifetime values of excited $^4F_{3/2}$ were obtained as a function of Nd^{3+} concentration, from the fluorescence decay curves in time, pumping with an OPO laser at around 808 nm. The gain-ESA spectra were taken using a pump-probe experimental setup [5]. The samples were pumped by a 14 Hz modulated Ti:sapphire laser at 808 nm and the probe radiation was provided by a broadband tungsten lamp modulated at 600 Hz. Appropriate spectral filters were used to prevent second-order effects and the signal was collected by a Ge detector. Taking the normalized difference in transmitted intensities I_p of the pumped crystal sample and I_u of the unpumped sample and assuming that $I_p \approx I_u$, it can be shown that:

$$\frac{I_p(\lambda) - I_u(\lambda)}{I_p(\lambda)} = n_e AL\left[\sigma_{GSA}(\lambda) + \sum_i \left(\frac{n_i}{n_e}\right)\left(\sigma_{SE,i} + \sigma_{ESA,i}\right)\right] \quad (1)$$

where n_e is the overall excited population, A is the amplification factor of the lock-in and L is the sample thickness, n_i / n_e is the ratio of populations in level i and the total density of excited ions. σ_{GSA}, σ_{SE} and σ_{ESA} are the ground state absorption (GSA), stimulated emission (SE) and excited state absorption (ESA) cross sections, respectively. The effective stimulated cross section is defined as $\sigma_{SE} - \sigma_{GSA}$. The spectra is calibrated in regions where only GSA and SE are present and ESA is not expected. Absorption cross sections are determined from independent measurements and the emission cross sections around 1064 nm were determined by the Fuchtbauer Lundenburg expression. Since there isn't ESA at 1064 nm and the only metaestable level is $^4F_{3/2}$ the ratio n_i / n_e is 1, and the calibration was done by equaling the σ_{SE} of $^4F_{3/2} \rightarrow {}^4I_{11/2}$ to the $(I_p\text{-}I_u)/I_p$ spectrum.

The laser experiments at 1064 nm were performed in a polished YVO_4 fiber doped with 1.0 atm% Nd^{3+} using an end-pump configuration laser cavity. The cavity consists of the fiber, a 50 mm radius of curvature output mirror, with 5% transmission around 1064 nm, and a plane mirror (90% transmission at 800 and total reflection at 1064 nm). Due to the ease in tuning, and control of the pump mode, a Coherent Ti:sapphire laser was used as the pumping source but a diode laser could have been used as well.

Results and Discussions

The growth of YVO_4:Nd^{3+} single crystal fibers, with typical diameters of 500 μm and length of 1 cm, by LHPG was very successful. The oxygen atmosphere in the growth chamber played an important role in minimizing O^{2-} deficiencies during the melt, and consequently the presence of V^{5+} ions was favored in detriment to V^{3+} and V^{4+}. That way, the fibers grew completely transparent and free from secondary phases or dark inclusions, as verified by optical microscopy. The characteristic tetragonal zircon structure of the compound, space group I41/amd, was determined and the unit cell parameters are $a = b = 7.120$ Å and c = 6.290 Å. A detailed Raman study of the fibers studied in this work was performed and it was verified that the SCFs and bulk crystal spectra are practically identical, with maximum phonon energy of ~890 cm^{-1}.

From the room temperature ground state absorption and fluorescence spectra of the fibers and bulk crystal, it can be noted that the characteristic transitions of Nd^{3+} present practically identical lineshapes and positions for both crystal forms. The spectra were obtained with π-polarization and the absorption band around 808 nm ($^4F_{5/2}$,$^2H_{9/2}$ levels) in the fibers' spectra present a full width at half maximum (FWHM) of ~5 nm, while that corresponding to the bulk crystal is ~2 nm. Because the optical quality and structure of the fibers is comparable to those of the bulk crystal, the difference in FWHM may be attributed to slight structural distortions or strains, resultant from

the high thermal gradient during growth, or lack of perfect fiber orientation. As for the π-polarized fluorescence spectra obtained with excitation at 808 nm, transitions are observed at around 900, 1060 and 1340 nm. The fluorescence line at 1064 nm ($^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition) presents a FWHM of ~3 nm for both crystal forms. The radiative lifetime value of $^4F_{3/2}$, obtained from the fluorescence decay curve of the lowest doped fiber (0.1 atm%) and for the bulk crystal is approximately 90 μs, according to the literature. For the 1.0 atm% nominally doped SCF the lifetime drops to 52 μs due to ion-ion interactions as the $^4F_{3/2}, {}^4I_{9/2} \rightarrow {}^4I_{15/2}, {}^4I_{15/2}$ cross relaxation.

The gain-ESA spectrum of a SCF presented in Figure 1 is also very similar to that reported for a bulk crystal [4]. In this spectral range the stimulated emissions at 1064 nm ($^4F_{3/2} \rightarrow {}^4I_{11/2}$) and at 1342 nm ($^4F_{3/2} \rightarrow {}^4I_{13/2}$) can be observed in addition to ESA transitions between 980 and 1063 nm ($^4F_{3/2} \rightarrow {}^2D_{3/2}, {}^4G_{11/2}, {}^2G_{9/2}, {}^2K_{15/2}$), around 1260 nm ($^4F_{3/2} \rightarrow {}^2G_{9/2}, {}^2K_{13/2}$) and around 1340 nm ($^4F_{3/2} \rightarrow {}^4G_{7/2}$), according to the energy levels partial diagram in the inset. The stimulated emissions at 1064 and 1342 nm present lower cross section values than those reported in the literature for $YVO_4:Nd^{3+}$, due to low resolution of our experimental apparatus but ESA results are in very good agreement with the literature [4]. At 1056 nm the maximum ESA cross section is less than 4×10^{-20} cm^2 and even if there is an overlap of a wing of this transition (with smaller σ_{ESA}) with the stimulated emission at 1064 nm, it is evident that the ESA loss is negligible in comparison to the emission cross section of 59×10^{-20} cm^2. The laser line at 1342 nm however, lies much closer to a strong ESA transition and considering its linewidth (typically 2 nm), we can infer that the laser performance at this wavelength is strongly compromised due to SE-ESA spectral overlap. Since these values are very similar to those reported for an $YVO_4:Nd^{3+}$ laser bulk crystal, we expected that the fiber has the same laser potentiality at 1064 nm as does the bulk.

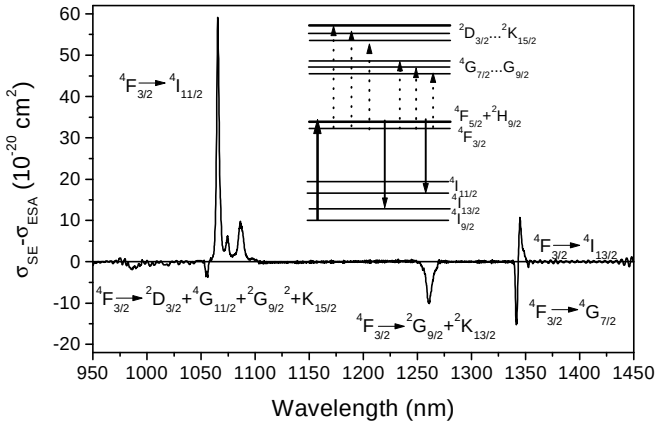

Figure 1: Gain-ESA spectra of a $YVO_4:Nd^{3+}$ (1.0 atm%) SCF obtained with π-polarization. The SE and ESA transitions from level $^4F_{3/2}$ are indicated in the partial energy levels diagram in the inset of the figure.

Figure 2 presents the curves of measured output power at 1064 nm, as a function of pumping power, for a 0.1 cm long fiber doped with 1.0 atm% Nd^{3+}, in comparison to the commercial bulk crystal. The laser threshold for the SCF is approximately 10 mW and the slope efficiency is 42%, in contrast to 48% obtained for the BSC with even lower emission threshold. These results are in good agreement with those presented in the literature, taking into account experimental differences, Nd^{3+} concentrations and optical quality of $YVO_4:Nd^{3+}$ samples [6]. The slope efficiency values obtained for commercial Czochralski grown $YAG:Nd^{3+}$ crystals are around 53% whereas for the crystal of $GdVO_4:Nd^{3+}$ 57% efficiency has been obtained [1].

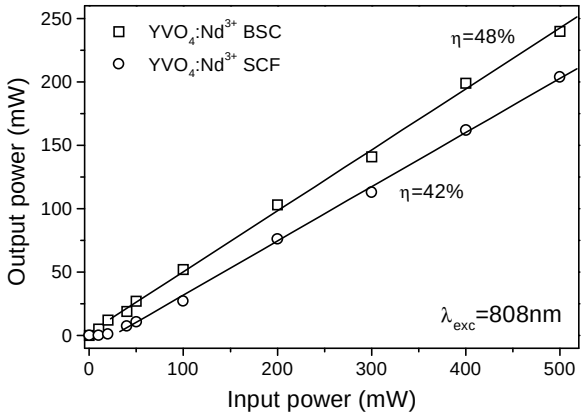

Figure 2: Output power curves of 1064 nm laser emission, as a function of 808 nm launched pumping power: (a) $YVO_4:Nd^{3+}$ SCF and (b) $YVO_4:Nd^{3+}$ bulk single crystal (BSC).

Conclusions

Neodymium doped (0.1-1.0 atm%) YVO_4 single crystal fibers with ~500 μm diameter and 1-2 cm long were successfully grown by LHPG under controlled O_2 atmosphere and at high pulling rates. The fibers present very good optical quality and were characterized by XRD, optical absorption, fluorescence, lifetime and excited state absorption measurements. The spectroscopic results are in very good agreement with those of a commercial bulk $YVO_4:Nd^{3+}$ laser crystal. A 0.1 cm long fiber doped with 1.0 atm% Nd^{3+} was tested in an end-pump cavity and laser action was achieved at 1064 nm with an efficiency of 42% in comparison to 48% for the bulk crystal. These results are very motivating for the confection of $YVO_4:Nd^{3+}$ compact fiber lasers considering the significant lower cost and time-saving characteristics of LHPG when compared to other growth techniques.

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