Appl. Phys. B 73, 191–194 (2001)
DOI: 10.1007/s003400100640

Applied Physics B
Lasers and Optics

F. CORNACCHIA[1]
A. DI LIETO[1,✉]
P. MARONI[1]
P. MINGUZZI[1]
A. TONCELLI[1]
M. TONELLI[1]
E. SOROKIN[2]
I. SOROKINA[2]

A cw room-temperature Ho,Tm:YLF laser pumped at 1.682 μm

[1] INFM, Istituto Nazionale di Fisica della Materia, Sezione di Pisa, Dipartimento di Fisica dell'Università, Via Buonarroti 2, 56127 Pisa, Italy
[2] Photonics Institute, TU Wien, Gusshausstr. 27/387, 1040 Wien, Austria

Received: 14 March 2001/Revised version: 15 June 2001
Published online: 19 September 2001 •

ABSTRACT We present the results obtained with a Ho,Tm:YLF crystal grown at a new crystal growth facility in Pisa. The optical quality of the sample has been tested by studying its performance as the active medium of a laser operating at 2.06 μm. We employed three different pump laser sources: a Ti:sapphire, a diode (both tuned at 793 nm) and, for the first time, a continuous-wave Co:MgF_2 laser, tuned at 1.682 μm. At room temperature the best slope efficiency was 30 % in the case of "red" pumping, and 59 % in the case of "infrared" excitation. The typical lasing threshold is about 100 mW.

PACS 42.55.Rz; 42.55.Xi; 81.10.Fq

1 Introduction

The 2 μm laser emission of the transition $^5I_7 \rightarrow {}^5I_8$ of Ho^{3+} has been widely studied for the purpose of developing eye-safe compact devices, suitable for many applications, such as optical communications, coherent laser radar, and medical instrumentation [1–4]. Several authors have reported laser action of this ion in different host crystals, in both the pulsed and continuous-wave (cw) regimes, and at different operating temperatures (see [5] for an extensive bibliography).

In this paper we describe the growth of a codoped 0.5 % Ho, 5.2 % Tm $LiYF_4$ (YLF) sample at the Physics Department of Pisa and its characteristics as the active medium of a laser operating at room temperature. In a preliminary step we tested the laser efficiency of the crystal using the traditional pumping scheme with two different cw laser sources: by means of a Ti:sapphire laser and a diode laser we excited the $^3H_6 \rightarrow {}^3H_4$ Tm^{3+} transition at 793 nm wavelength. Later we employed a Co:MgF_2 laser to pump the $^3H_6 \rightarrow {}^3F_4$ transition of Tm^{3+} ions at 1682 nm wavelength; as far as we know, it is the first time this pumping scheme has been successfully demonstrated. With all three pump lasers we observed the 2 μm laser emission from the $^5I_7 \rightarrow {}^5I_8$ transition of Ho^{3+} ions and we measured the laser threshold and efficiency with different transmissions of the output coupler.

2 Crystal growth apparatus and spectroscopic investigations

In this section we describe the experimental apparatus for the crystal growth set up at the Dipartimento di Fisica, Università di Pisa. The apparatus consists of a homemade Czochralski furnace with conventional resistive heating; special care has been devoted to the quality of the vacuum system, which has an ultimate pressure limit below 10^{-8} Torr.

The raw material for the crystal was prepared by mixing LiF and YF_3 powders in a platinum crucible. The doping was obtained by adding suitable amounts of TmF_3 and HoF_3 powders. We selected a dopant concentration of 5.2 % Tm and 0.5 % Ho. To avoid OH^- contamination, the powders were purified at AC Materials (Orlando, Fla., USA). The growth process was carried out in a high-purity (99.999 %) argon atmosphere.

During growth, the rotational speed of the sample was 5 rpm, the pulling rate 1 mm/h, and the temperature of the melt was 809 °C. The average size of the YLF crystal was about 20 mm in diameter and 35 mm in length, and its mass was about 24 g. The sample was oriented using the X-ray Laue technique and cut with an edge parallel to the c-axis. The dimensions of the crystal sample inside the laser are 6×6 mm^2, with a thickness of 2.4 mm. The wider faces were optically polished using alumina powder to laser tolerance.

For diagnostic purposes we recorded the room-temperature polarized absorption spectra ($E \parallel c$ and $E \perp c$) from the UV up to the IR wavelength region. Figure 1 shows a selection of these spectra around 790 and 1700 nm, normalized to show the absorption coefficient, α, in units of cm^{-1}. The UV spectrum around 200 nm did not show the typical absorption bands of OH^- radicals in crystals, within the sensitivity of our CARY 500 spectrophotometer, so we are confident that our sample is free of any contamination that might compromise its optical properties.

The visible and infrared spectra in the $E \parallel c$ polarization show many absorption peaks suitable for optical pumping. Among them, we chose the peak at 793 nm for pumping with the Ti:sapphire and diode lasers, and the peak at 1.682 μm for pumping with the Co:MgF_2 laser. These lines come from the

✉ *Permanent address:* Scuola Normale Superiore, Piazza dei Cavalieri 7, 56126 Pisa, Italy
(Fax: +39-050/844-333, E-mail: dilieto@df.unipi.it)

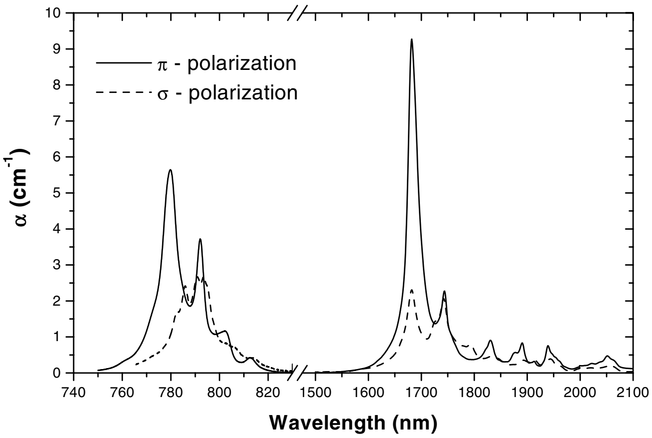

FIGURE 1 Absorption spectrum in the red and infrared wavelength regions for the $E \parallel c$ (*solid line*) and $E \perp c$ (*dashed curve*) conditions

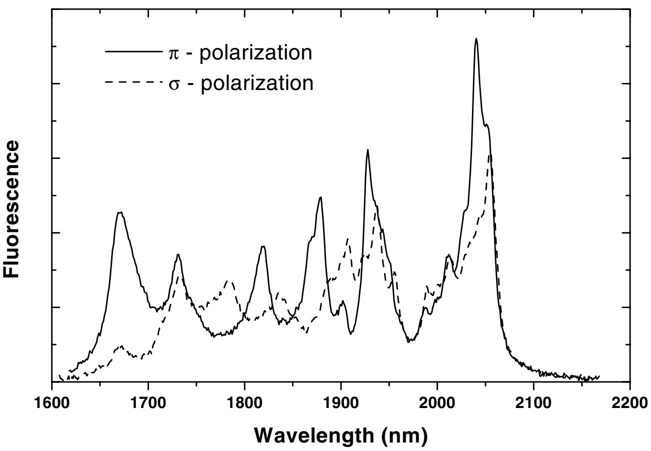

FIGURE 2 Fluorescence spectrum in the infrared wavelength region for the $E \parallel c$ (*solid line*) and $E \perp c$ (*dashed curve*) conditions

transition bands $^3H_6 \rightarrow {}^3H_4$ and $^3H_6 \rightarrow {}^3F_4$ of the Tm^{3+} ions and the absorption coefficients at room temperature are 3.7 and 9.3 cm^{-1}, respectively.

Another spectroscopic check on the crystal quality is related to the lifetime of the upper laser level of Ho^{3+}; we measured 14.5 ± 0.1 ms, and this value is in satisfactory agreement with that reported in the literature [6].

A last check was made by recording the room-temperature fluorescence in the infrared region near 2 μm when the crystal is pumped by red radiation. For this measurement the experimental setup was composed of the Ti:sapphire laser source, a focusing system, a 25-cm focal length monochromator to disperse the fluorescence, and a detector connected to a lock-in amplifier. Figure 2 shows a typical fluorescence spectrum recorded by using 210 mW of radiation emitted by the laser tuned at 793.8 nm; the spectral resolution of the monochromator was 4 nm. The curve has been normalized taking into account the spectral response of the PbS detector.

3 Laser experiments

The experimental setup for studying the laser performance of our crystal consisted of an astigmatically compensated X-folded four-mirror cavity. The crystal sample was placed at Brewster's angle, mounted on a copper or aluminum block for thermal dissipation, but without any further active cooling, and oriented with the crystallographic *c*-axis in the plane of incidence for both the pump and the emitted radiation. The cavity had two curved dichroic mirrors (radius of curvature 100 mm), placed at a distance approximately equal to their radius, a high-reflectivity plane mirror, and an output coupler. The folding angles and the length of the two arms of the cavity were chosen to compensate for the astigmatism produced by the crystal and to provide convenient matching of the resonator mode within the crystal.

The pump beam was focused by an uncoated lens to produce a waist size within the crystal comparable to the resonator spot. The focal length was different for each pump laser, to optimize the overlap of the beam diameters.

The first pump source we tried was a home-made cw Ti:sapphire laser; it could deliver up to 1 W in the spectral region of interest when pumped by a multiline Ar ion laser. The optical quality of this radiation was quite good with a nearly Gaussian output beam ($M^2 \approx 1.1$).

The second source was a commercial diode array (SDL-2432-P1) which could deliver up to 1.2 W of power with an emission bandwidth of approximately 2 nm. The astigmatism of its beam was compensated by an 8 mm focal length objective, placed in front of the laser diode output window and followed by an anamorphic prism pair with a 4× magnification in the plane parallel to the junction. With this arrangement the pump beam could be focused to an elliptical spot of approximately 100×25 μm at the entrance face of the laser crystal. When using both these pump sources, the best results were obtained by using a focusing lens of 70 mm focal length.

The last pump source we tested was a home-made cw $Co{:}MgF_2$ laser, pumped by a multimode Nd:YAG laser at 1.32 μm [7]. This laser is broadly tunable and if the active crystal is cooled at liquid nitrogen temperature it can deliver up to 1 W at 1.682 μm. The optical quality of the beam emitted by this source is good, and the pumping spot on the YLF crystal was circular. With this pump laser, the performance of the 2 μm laser was almost insensitive to the focal length of the focusing lens between 60 and 100 mm, and in the final measurement we used a 100 mm lens.

The input mirror could transmit about 95 % of the pumping radiation at 793 nm and about 84 % at 1.682 μm.

4 Results

In this section we describe the laser performances of our YLF crystal. In all the figures, the abscissae represent the input power, which is the laser pump power incident on the sample, and throughout this work all efficiencies refer to it and not to the power absorbed by the crystal, unless otherwise specified. The comparison with available literature results will be limited to fluoride crystals, for homogeneity with the host presented in this work.

Figure 3 shows the results obtained using the Ti:sapphire laser as the pumping source and with two different output couplers (OC). The best slope efficiency attained is about 30 % with the 3 % OC, and the threshold is always less than 100 mW. The maximum output power is about 250 mW and is limited by the available output of the Ti:sapphire laser.

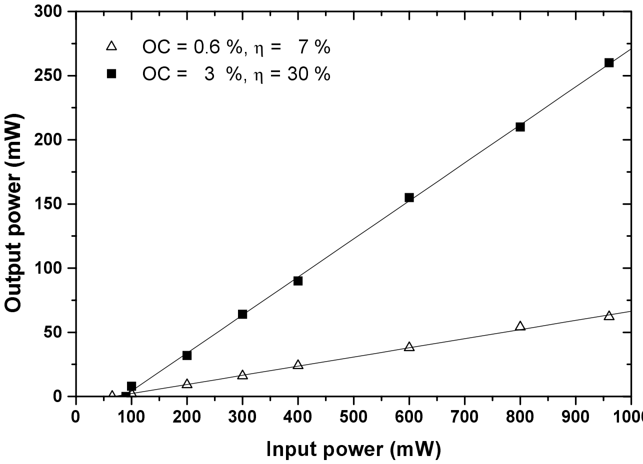

FIGURE 3 Output power as a function of input power when the pump is the Ti:sapphire laser tuned at 793 nm; the two sets correspond to different transmissions of the output coupler

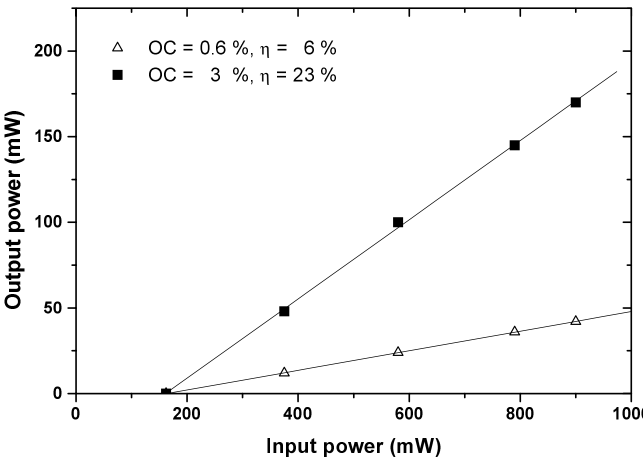

FIGURE 4 Output power as a function of input power when the pump is the diode laser tuned at 793 nm; the two sets correspond to different transmissions of the output coupler

Figure 4 shows the results obtained with the diode laser as the pumping source. In this case the best slope efficiency is 23 % with the 3 % OC, and the threshold is about 160 mW.

These results can be compared with those previously published in the literature. Many papers describe diode pumping of a YLF crystal kept at different temperatures, ranging from liquid nitrogen up to room temperature [8–12]. The best slope efficiency, η, is reached at 77 K [10]: the author quotes $\eta = 67\%$ relative to the absorbed power (AbsPw), which corresponds to ≈ 48 % when computed with respect to incident power (IncPw). At 234 K an η of about 25 % is reported in [12], at 248 K [11] quotes $\eta = 30\%$ (AbsPw, corresponding to ≈ 14 % IncPw), while at the subambient temperature of 275 K an η of about 60 % (AbsPw, ≈ 30 % IncPw) is quoted by [9]. For operation at room temperature we found three results in the literature, varying from 9 % [10] to 14 % [8], so the result reported here (23 %) compares very well and qualifies our crystal among the best ever produced.

The difference between Ti:sapphire and diode pumping at 793 nm should be ascribed to the much poorer beam quality of the diode radiation, which causes problems for proper focusing; in [8] the authors report a ratio between Ti:sapphire and diode pumping very similar to what we found here, even with a completely different optical layout. It is also worth pointing out that the structure of our optical cavity was optimized under Ti:sapphire pumping, so the configuration was not the best possible for diode pumping. We believe that about 30 % of the slope efficiency under diode pumping at 793 nm should be quite feasible at room temperature, when a high-quality laser beam is employed.

Figure 5 shows the results obtained with the $Co:MgF_2$ laser as the pumping source. The slope efficiencies show values that are substantially larger than for previous pumping conditions: the maximum efficiency is as high as 59 % with the 5 % OC, which is almost twice that obtained with the Ti:sapphire laser.

The cw lasing of a Ho,Tm system under pumping directly into the $^3F_4$ band has not been published in the literature. However a similar scheme, where only the first excited multiplet is involved, was demonstrated in [13], but in a different host (YAG) and without Tm doping. The results obtained with this pumping scheme seem to be reasonably good if compared with the traditional 793 nm pumping. The most important difference between the two pump mechanisms is that the infrared pump radiation directly populates the first excited state $^3F_4$ of Tm^{3+} ions, which can transfer its energy to the $^5I_7$ upper laser level of Ho^{3+}, while the 793 nm pumping route needs an additional cross-relaxation of the excited $^3H_4$ level into two excited $^3F_4$ levels, involving a second Tm^{3+} ion. There are additional losses in the 793 nm pump channel most probably related to the upconversion processes [14], resulting in the visible luminescence, that is present only under pumping at this wavelength.

A comparison can also be interesting in terms of the slope quantum efficiency, η_e, defined as in [15]:

$$\eta_e = \eta \frac{\lambda_L}{A\lambda_P}, \tag{1}$$

where λ_L and λ_P are respectively the laser and pump wavelengths, A is the fraction of the incident power absorbed in the

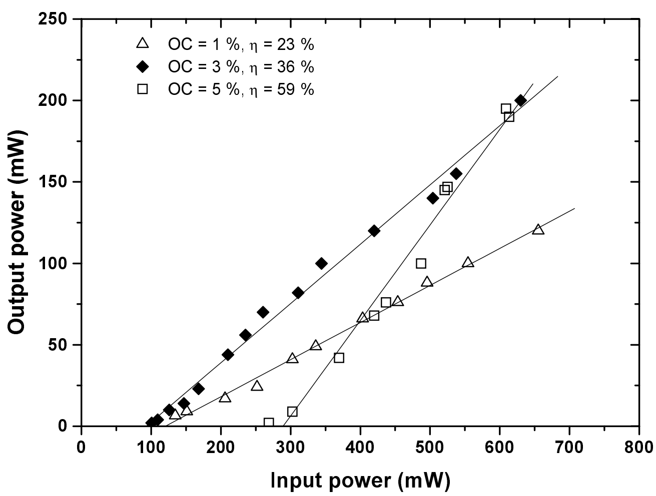

FIGURE 5 Output power as a function of input power when the pump is the $Co:MgF_2$ laser tuned at 1.682 μm; the three sets correspond to different transmissions of the output coupler

crystal, and η is the measured slope efficiency. For the 793 nm pumping case, using the best Ti:sapphire slope efficiency, we obtain $\eta_e = 1.32$, which should be compared with the maximum theoretical value $\eta_t = 2$. For the 1.682 μm pumping case, the measured quantum efficiency is at best $\eta_e = 0.81$, to be compared with 1 (for the different scheme of levels involved in the lasing effect), so the normalized slope quantum efficiency η_e/η_t in this condition is ≈ 20% larger than for the "classical red" pumping scheme. If otherwise the slope efficiency is compared for the condition of equal output coupling (3 %) the "red" and "infrared" pumping show about the same performance.

When the Ti:sapphire laser measurements were carried out, the 5 % OC was not available, so this comparison is not truly conclusive, but the comparison of our results with the literature points out that only a small increase in performance can be expected by increasing the output coupling for the red-pumping situation.

To prove the consistency of our efficiency measurements, we calculated the intrinsic losses when the crystal is pumped at 1.682 μm, using both the Findlay–Clay [16] and the Caird [17] analyses, based on the threshold values and on the slope efficiencies respectively. Both methods give a similar value for the intrinsic losses: about 4.5 %/cm in a single pass.

The 1.682 μm pumping scheme was suggested a few years ago [18] as a promising scenario in order to obtain superior performance by decreasing upconversion losses. The results obtained here confirm those predictions, even if the Tm concentration in our crystal was optimized for 793 nm pumping and not for the infrared. In fact, according to the authors of [18], the best performance with 1.682 μm pumping should be achieved with a much lower Tm concentration. So further work is in progress to optimize the laser performance of a Ho,Tm:YLF crystal with respect to dopant concentration and thickness of the sample for this infrared pumping scheme.

5 Conclusions

The results we report here show that the growth apparatus developed at the Physics Department of Pisa can produce samples of very high optical quality, suitable for room-temperature laser systems. The slope efficiency values we measured with both the Ti:sapphire and the diode laser pumping conditions are comparable or better than the best values reported in the literature.

For the first time the 1.682 μm pumping channel has been demonstrated in a Ho,Tm:YLF crystal, producing a source of very high efficiency. Thus we confirmed the theoretical prediction that this pumping channel can be advantageous, and this result is important because nowadays high-power diode lasers emitting in the 1.6–1.8 μm region begin to be commercially available. These results confirm the high versatility of Ho,Tm:YLF crystals, and the possibility of producing a simple and efficient 2 μm room-temperature laser system in an "all solid state" configuration, with many different options for the pumping scheme.

ACKNOWLEDGEMENTS The authors wish to thank Hans P. Jenssen and Arlete Cassanho for helpful discussions and important suggestions. Moreover, they are grateful to Ilaria Grassini, Alessandro Masetti and Fabio Torri for their support in developing the experimental setup.

REFERENCES

1 S.W. Henderson, C.P. Hale, J.R. Magee, M.J. Kavaja, A.V. Huffeker: Opt. Lett. **16**, 773 (1991)
2 S.W. Henderson, K. Ota: Rev. Laser Eng. **25**, 19 (1997)
3 H.P. Weber, M. Frenz, H. Pratisto, M. Ith, R. Hausler, P. Schar: Laser Phys. **8**, 785 (1998)
4 K.E. Murray, N.P. Barnes, B.M. Walsh, R.L. Hutcheson, M.R. Kotka: In *OSA Trends in Optics and Photonics*, Vol. 34, Advanced Solid-State Lasers, ed. by H. Injeyan, U. Keller, C. Marshall (Optical Society of America, Washington, DC 2000) p. 170
5 A.A. Kaminski: *Crystalline lasers: physical processes and operating schemes* (CRC Press, Boca Raton 1996)
6 H. Chou: *Energy transfer in codoped Ho, Er:YLF and its potential for improving the efficiency of* 3 μm *laser*, MS thesis (MIT, Cambridge 1983)
7 A. Di Lieto: Opt. Laser Eng. (November 2001), to be published
8 M. Tonelli, M. Falconieri, A. Lanzi, G. Salvetti, A. Toncelli: Opt. Commun. **129**, 62 (1996)
9 B.T. McGuckin, R.T. Menzies: IEEE J. Quantum Electron. **QE-28**, 1025 (1992)
10 H. Hemmati: Opt. Lett. **14**, 435 (1989)
11 G.J. Koch, J.P. Deyst, M.E. Storm: Opt. Lett. **18**, 1235 (1993)
12 A. Finch, J.H. Flint, D.M. Rines: *OSA TOPS on Advanced Solid-State Lasers*, Vol. 1, ed. by S.A. Payne, C. Pollock (Optical Society of America, Washington, DC 1996) p. 312
13 C.D. Nabors, J. Ochoa, T.Y. Fan, A. Sanchez, H.K. Choi, G.W. Turner: IEEE J. Quantum Electron. **QE-31**, 1603 (1995)
14 A. Brenier, J. Rubin, R. Moncorge, C. Pedrini: J. Phys. (Paris) **50**, 1463 (1989)
15 A. Sanchez, R.E. Fahey, A.J. Strauss, R.L. Aggarwal: Opt. Lett. **11**, 363 (1986)
16 D. Findlay, R.A. Clay: Phys. Lett. **20**, 277 (1966)
17 J.A. Caird, S.A. Payne, P.R. Staver, A.J. Ramponi, L.L. Chase, W.F. Krupke: IEEE J. Quantum Electron. **QE-24**, 1077 (1988)
18 S.A. Payne, L.K. Smith, W.L. Kway, J.B. Tassano, W.F. Krupke: J. Phys: Condens. Matter **4**, 8525 (1992)