



Fabrication, properties and laser performance of Ho:YAG transparent ceramic

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ABSTRACT

Highly transparent Ho:YAG ceramic was successfully fabricated by solid-state reaction and vacuum sintering. The optical properties, the microstructure and the laser performance of the Ho:YAG ceramic were investigated. Ho:YAG ceramic with the average grain size of $\sim 15 \mu\text{m}$ was obtained by sintering at 1760°C for 20 h. The in-line transmittances in the visible region and the infrared region were both over 80%. No pores, impurities and secondary phases were detected in the grains and at the grain boundaries. The 1 at.% Ho:YAG ceramic slab ($1.5 \text{ mm} \times 10 \text{ mm} \times 18 \text{ mm}$) was end-pumped by a Tm-YLF laser at 1910 nm. The maximum output power of 1.95 W was yielded with a slope efficiency of 44.19% and Tm to Ho optical-optical efficiency was 24% at 2091 nm.

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1. Introduction

In recent years, an increasing interest has been focused on the solid-state lasers operating in the eye-safe spectral region near $2 \mu\text{m}$ because of their varied applications such as optical communications, coherent laser radar, atmospheric sensing and medical equipment [1–4]. Also, high-power quasi-continuous wave (QCW) $2 \mu\text{m}$ lasers are efficient pump sources of optical parametric oscillators (OPOs) and optical parametric amplifiers (OPAs).

Currently two technologies are in wide use—diode-pumped $1.9 \mu\text{m}$ thulium (Tm^{3+}) and $2.1 \mu\text{m}$ holmium (Ho^{3+}) lasers. The performance of the Tm^{3+} in $\text{Y}_3\text{Al}_5\text{O}_{12}$ (YAG)/ YAlO_3 (YAP)/ $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ (GGG) single crystal has been widely studied and the laser operation has been successfully realized [4–7]. Diode-pumped Tm:YAG ceramic laser also has been studied in our previous work [8]. Ho^{3+} lasers are used as pump sources for non-linear optics to generate mid-infrared radiation in the spectral range of $3\text{--}5 \mu\text{m}$, for lidar applications as their emission wavelengths are less affected by the atmospheric water vapor absorption than those of Tm^{3+} lasers, in medicine and for plastic welding [9]. Moreover, direct resonant pumping Ho $^5\text{I}_7$ manifold offers the advantages of high slope efficiency (70% in Ho:YLiF₄) [10], minimal heating due to low quantum defect of less than 10% between pump and laser.

The performance of the Ho:YAG single crystal has been widely studied [11,12] and the laser operation has been successfully realized [13–18]. Holmium-doped yttrium aluminum garnet (Ho:YAG) ceramic is the promising material of choice for $2.1 \mu\text{m}$ applications due to its well-known excellent thermo-mechanical properties and advantages compared with Ho:YAG single crystal (for example: ease of fabrication, less cost, mass production, feasibility of large size, high holmium concentration, etc.). The laser performance of Ho:YAG ceramic has been reported in Dr. Cheng's work with the specimen we provided [19]. And to the best of our knowledge, fabrication and properties of highly transparent Ho:YAG ceramic has not been reported yet.

Ho:YAG does not have any suitable absorption feature in the traditional diode laser wavelength window of 785–980 nm, so that Ho:YAG cannot be directly pumped by traditional diode lasers. Using Tm-doped laser operating at approximate $1.9 \mu\text{m}$ direct resonant pumping Ho $^5\text{I}_7$ manifold offers the advantages of high quantum efficiency, minimal heating due to low quantum defect between pump and laser of $\sim 10\%$, and reduced up-conversion losses caused by Tm sensitized. Therefore, this approach has the advantage of very low quantum defect heating with the result that high lasing efficiencies are attainable.

Available room-temperature pump sources for Ho are the Tm laser in the hosts YAG, YLiF₄ (YLF), and YAP. The absorption spectrum of Tm:YLF falls within the emission spectrum of commercially available laser diodes emitting peak of 792 nm. The emission spectrum of Tm:YLF matches well with the relatively broad absorption spectrum of Ho:YAG. Therefore, Tm:YLF laser allows relatively low

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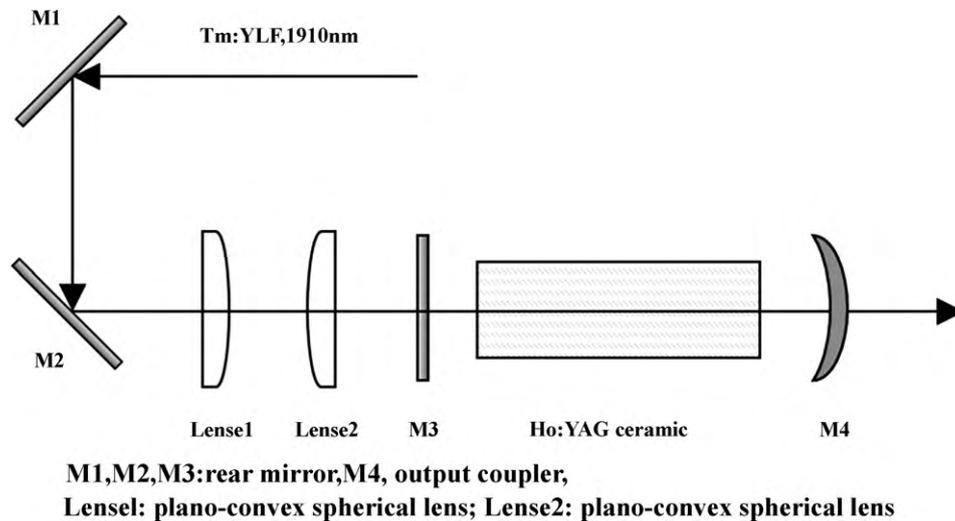


Fig. 1. The scheme of the Ho:YAG ceramic laser experiment.

brightness diode pump sources to be used and efficiently pumping Ho:YAG [20,21].

In this paper, highly transparent Ho:YAG ceramic was successfully fabricated by a simple solid-state reaction method and vacuum-sintering technology. The microstructure, the spectral characteristics and room-temperature laser actions of Tm:YLF laser pumped Ho:YAG ceramic in CW modes were reported in this work.

2. Experimental procedures

2.1. Ceramic fabrication

High-purity powders of α - Al_2O_3 (99.99%, Shanghai Wusong Chemical Co. Ltd., Shanghai, China), Y_2O_3 (99.99%, Shanghai Yuelong New Materials Co. Ltd., Shanghai, China), and Ho_2O_3 (99.99%, Conghua Jianfeng Rare-Earth Co. Ltd., Guangzhou, China) were used as starting materials. These powders were blended according to the stoichiometric ratio of 1 at.% Ho:YAG and ball-milled with high-purity corundum balls for in ethanol 10 h, with a binder and addition of 0.6 wt% tetraethyl orthosilicate (TEOS) as a sintering aid. Then, the alcohol solvent was removed by drying the milled slurry at 80 °C for 4 h in oven. The dried powder mixture was ground and sieved through 200-mesh screen. After removing the organic component by calcining at 500–800 °C for 2 h, the powder mixture was dry-pressed with a low pressure into $\Phi 25$ mm disks in a steel mold and then cold-isostatic-pressed at 250 MPa into green bodies. Then the green bodies were vacuum-sintered at 1760 °C for 20 h. After sintering, the specimens were annealed at 1400 °C for 1 h in air to relieve internal stresses and fill oxygen vacancies formed during

the vacuum-sintering process. Finally, highly transparent Ho:YAG ceramics were obtained.

The phase composition of the sample was identified by X-ray diffraction (Model D/MAX-2550V, Rigaku, Tokyo, Japan). The sample mirror-polished on both surfaces was used to measure optical transmittance and absorption spectra (Model U-2800 Spectrophotometer, Hitachi, Tokyo, Japan). For measuring the fluorescence spec-

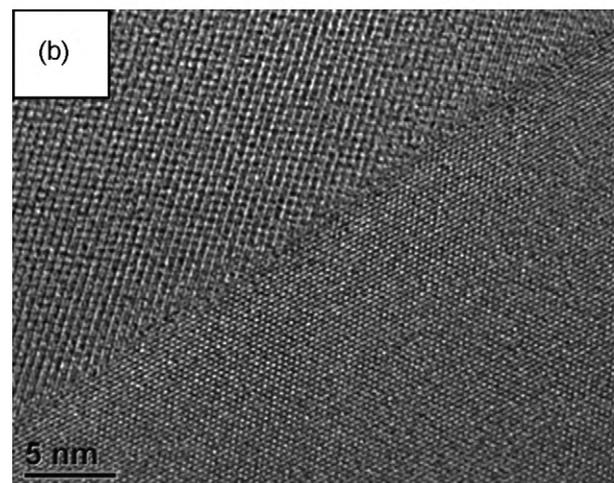
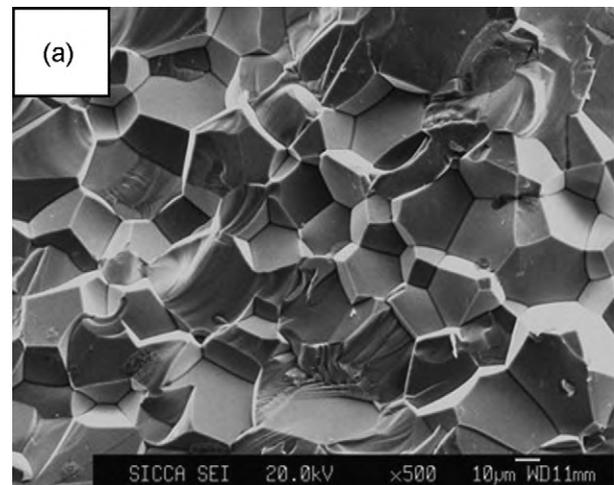


Fig. 3. The EPMA micrograph of the fractured surface (a) and the high-resolution TEM micrograph (HRTEM) of the grain boundary (b) of the Ho:YAG ceramic.

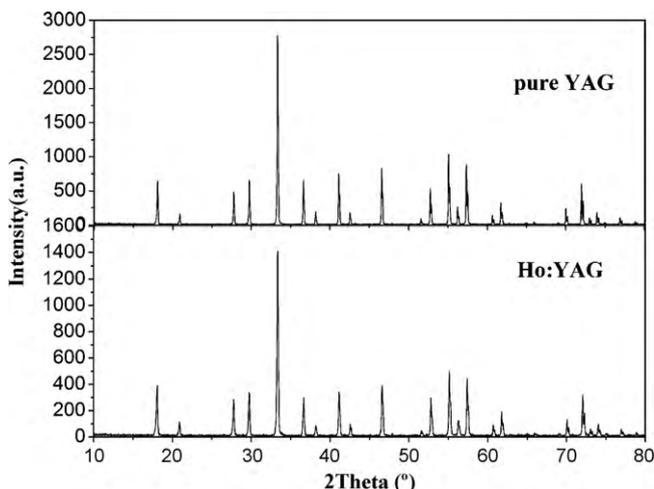


Fig. 2. XRD patterns of pure YAG and Ho:YAG samples.

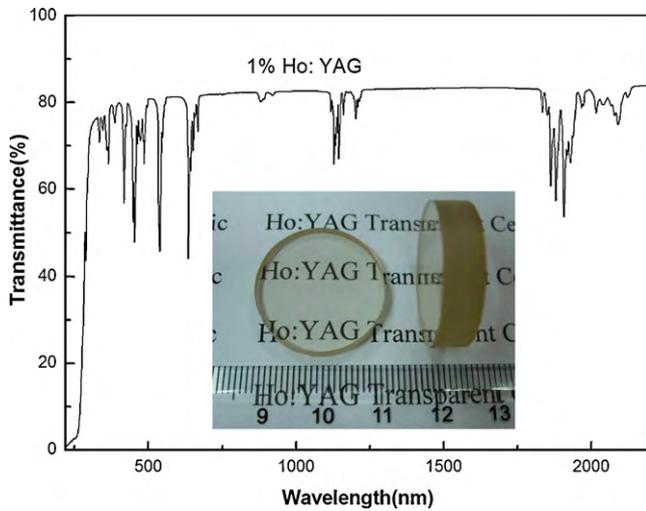


Fig. 4. The transmittance spectrum of mirror-polished 1 at.% Ho:YAG sample. The inset picture is the appearance of the sample.

trum (Model Fluorolog-3, Jobin Yvon, Paris, France), the specimen was excited at 640 nm by the flash lamp. The microstructure of the fractured surface of the sample was observed by electron probe micro-analyzer (EPMA, Model JXA-8100, JEOL, Tokyo, Japan). The microstructure of grain boundary was characterized by a field emission transmission electron microscopy (FETEM, Model EM 2100, JEOL, Tokyo, Japan).

2.2. Laser experiment

Fig. 1 shows a schematic diagram of the experimental setup. The dimension of the sample is 1.5 mm × 10 mm × 18 mm. Both surfaces (1.5 mm × 10 mm) of the Ho:YAG ceramic were mirror-polished, parallel, and coated with an anti-reflection at 1910 nm and 2100 nm. The sample was end-pumped by a Tm:YLF laser with an emission wavelength of around 1910 nm. M1 and M2 were two rear mirrors which were set at 45° angle with the Tm:YLF laser beam and anti-reflection coated at 1910 nm. The laser cavity consisted of two mirrors (M3 and M4), where M3 was anti-reflection coated at 1910 nm and high-reflection coated at 2100 nm, and M4 was the output coupler (OC). The transmittance at 2090 nm and curvature radius of the output coupler is 5%, 400 mm.

3. Results and discussion

Fig. 2 displays the XRD patterns of Ho:YAG and pure YAG ceramic at room temperature. The locations of Ho:YAG peaks are almost the same as that of pure YAG although the strength of peaks is different. That is because that the Ho:YAG ceramic has the same structure as the pure YAG ceramic. The calculated lattice constant for Ho:YAG ceramic is 1.2009 nm, which is quite similar to that of pure YAG ceramic (the calculated lattice constant for pure YAG is 1.2010 nm), because that Ho^{3+} takes the position of Y^{3+} in the lattice structure

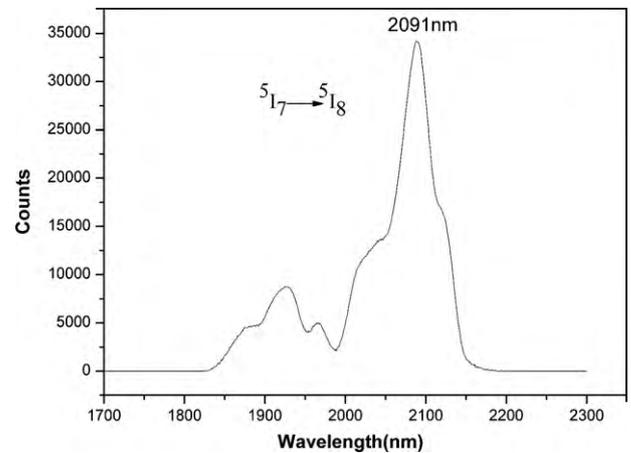


Fig. 6. The fluorescence spectrum of the 1 at.% Ho:YAG ceramic at room temperature.

and the radius of Ho^{3+} (89.4 pm) is just a little smaller than that of Y^{3+} (90 pm).

The EPMA micrograph of fractured surface (Fig. 3(a)) and the high-resolution TEM micrograph (HRTEM) of the grain boundary (Fig. 3(b)) show the microstructure of the Ho:YAG ceramic. As shown, the grain sizes are quite uniform and the average grain size is about 15 μm . The grain boundary is clear and clean. There are no pores, impurities and secondary phases in the grains and at the grain boundary.

From the in-line transmittance spectrum in Fig. 4, we can see that Ho:YAG laser wavelength (around 2.1 μm) is right in the absorption band (1800–2100 nm), which indicates that Ho:YAG laser has a self-absorption process. In this case, normally the Ho^{3+} doping levels should not be above 3% in the Ho:YAG laser material. The inset picture is the appearance of the samples. The samples are highly transparent and the thickness is 5 mm.

The emission spectrum of Tm:YLF (Fig. 5(a)) [22] coincides well with the absorption spectrum of Ho:YAG transparent ceramic (Fig. 5(b)). Both of the peak emission cross-section lines (1.880 and 1.907 μm) in Tm:YLF are well suited to pump Ho:YAG ceramic efficiently. For this work, we use the 1.9 μm Tm:YLF line as a pump source.

Fig. 6 is the room-temperature fluorescence spectrum excited by the flash lamp. The emission region with a peak at 2091 nm is from 1800 nm to 2200 nm which comes from the transition $5\text{I}_7 \rightarrow 5\text{I}_8$ of the 1% Ho:YAG ceramic. With the fluorescence spectrum, the sample was suitably coated and the laser experiment was well designed in Fig. 1.

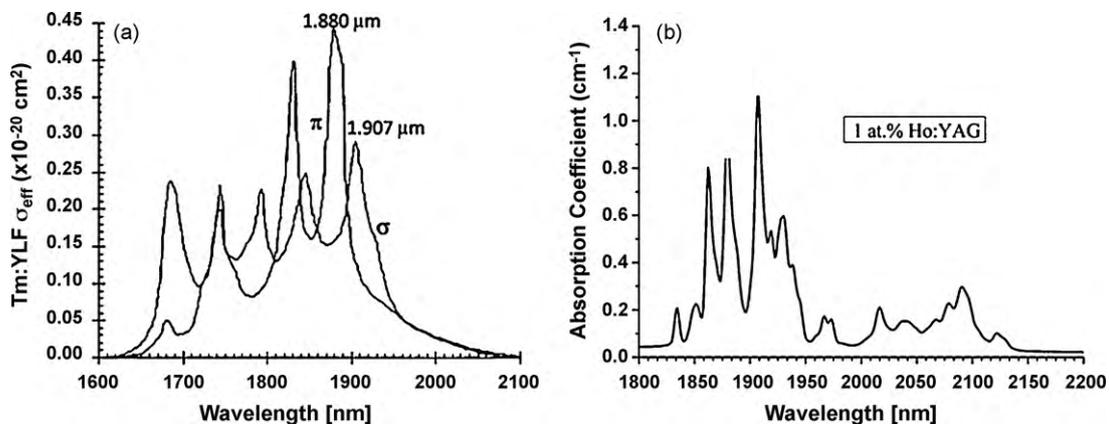


Fig. 5. The effective emission cross-section of Tm:YLF (a) [13] and the absorption spectrum of the Ho:YAG ceramic from 1800 nm to 2200 nm (b).

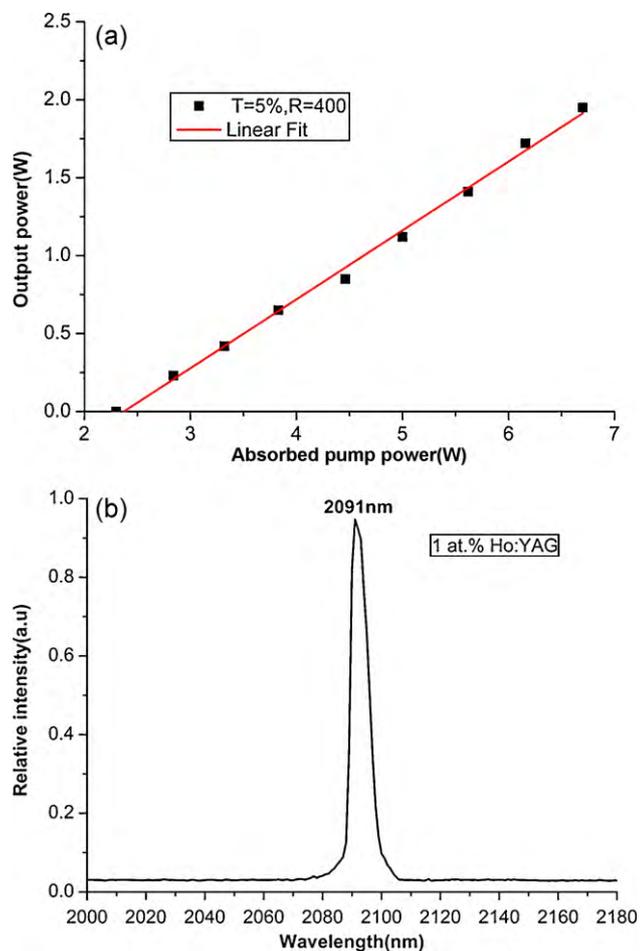


Fig. 7. The laser output power versus the absorbed pump power (a) and the laser spectrum (b) for the 1 at.% Ho:YAG ceramic.

To the best of our knowledge, highly transparent Ho:YAG ceramic is fabricated for the first time in this paper. The Ho:YAG ceramic slab was end-pumped by a Tm-YLF laser. The laser spectrum of the 1 at.% Ho:YAG ceramic is centered at 2091 nm, as shown in Fig. 7(b). Fig. 7(a) illustrates the laser output power versus the absorbed pump power for the 1 at.% Ho:YAG ceramic. The maximum laser output power of 1.95 W has been obtained with only 6.7 W of absorbed Tm pump power. A linear fit to the data yielded a slope efficiency of 44.19% with a threshold of approximately 2.3 W. The Tm:YLF to Ho:YAG optical-to-optical efficiency was 24%. The slope efficiency and optic-optic transformation efficiency of Ho:YAG ceramic yielded in this study are still lower than those of Ho:YAG crystal (the slope efficiency and optic-optic transformation efficiency of Ho:YAG crystal are 57% and 38%, respectively [23]), however, with improvement of ceramic fabrication process

and optical quality, Ho:YAG ceramic is a very promising substitute for Ho:YAG crystal in the foreseeable future.

4. Conclusions

Highly transparent Ho:YAG ceramic with average grain size of $\sim 15 \mu\text{m}$ was obtained by solid-state reaction and vacuum sintering. The grain boundary was clean and no secondary phase was observed. The 1 at.% Ho:YAG ceramic slab was end-pumped by a Tm-YLF laser at 1910 nm. The maximum output power of 1.95 W was obtained with a slope efficiency of 44.19% and Tm to Ho optical-to-optical efficiency of 24% at 2091 nm. The results of the laser experiment show that the quality of such kind of ceramics is good enough to be used as a highly efficient laser material.

Acknowledgments

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References

- [1] K. Scholle, E. Heumann, G. Huber, *Laser Phys. Lett.* 1 (2004) 285.
- [2] N.D. Vieira Jr., I.M. Ranieri, L.V.G. Tarelho, N.U. Wetter, S.L. Baldochi, L. Gomes, P.S.F. de Matos, W. de Rossi, G.E.C. Nogueira, L.C. Courrol, E.A. Barbosa, E.P. Maldonado, S.P. Morato, *J. Alloys Compd.* 344 (2002) 231.
- [3] M.J. Bader, J. Hocaoglu, S. Walther, M. Seitz, R. Sroka, C.G. Stief, O. Reich, *Med. Laser Appl.* 24 (2009) 132.
- [4] C.T. Wu, Y.L. Ju, Z.G. Wang, Q. Wang, C.W. Song, Y.Z. Wang, *Laser Phys. Lett.* 5 (2008) 793.
- [5] Y.L. Lu, J. Wang, Y. Yang, Y.B. Dai, A.P. Dong, B.D. Sun, *J. Alloys Compd.* 429 (2007) 296.
- [6] Y. Yang, Y.L. Lu, J. Wang, Y.B. Dai, B.D. Sun, *J. Alloys Compd.* 455 (2008) 1.
- [7] H. Zhou, X.H. Ma, G.T. Chen, W.C. Lv, Y. Wang, Z.Y. You, J.F. Li, Z.J. Zhu, C.Y. Tu, *J. Alloys Compd.* 475 (2009) 555.
- [8] W.X. Zhang, Y.B. Pan, J. Zhou, W.B. Liu, J. Li, B.X. Jiang, X.J. Cheng, J.Q. Xu, *J. Am. Ceram. Soc.* 92 (2009) 2434.
- [9] M. Eichhorn, *Appl. Phys. B* 93 (2008) 269.
- [10] F. Wang, D.Y. Shen, D.Y. Fan, Q.S. Lu, *Chin. J. Laser* 36 (2009) 1727.
- [11] A. Wnuk, M. Kaczkan, Z. Frucacz, I. Pracka, G. Chadeyron, M.-F. Joubert, M. Malinowski, *J. Alloys Compd.* 341 (2002) 353.
- [12] S. Edvardsson, D. Åberg, *J. Alloys Compd.* 303–304 (2000) 280.
- [13] N.G. Zakharov, O.L. Antipov, V.V. Sharkov, A.P. Savikin, *Quantum Electron.* 40 (2010) 98.
- [14] S.D. Jackson, A. Sabella, D.G. Lancaster, *Quantum Electron.* 13 (2007) 567.
- [15] A. Hemming, J. Richards, S. Bennetts, A. Davidson, N. Carmody, P. Davies, L. Corena, D. Lancaster, *Opt. Commun.* (2010), doi:10.1016/j.optcom.2010.05.078.
- [16] M. Schellhorn, *Appl. Phys. B* 85 (2006) 549.
- [17] S. So, J.I. Mackenzie, D.P. Shepherd, W.A. Clarkson, J.G. Betterton, E.K. Gorton, J.A.C. Terry, *Opt. Express.* 14 (2006) 10481.
- [18] C. Kieleck, A. Hirth, M. Schellhorn, *Proc. SPIE* 5989 (2005) 598905.
- [19] X.J. Cheng, J.Q. Xu, M.J. Wang, B.X. Jiang, W.X. Zhang, Y.B. Pan, *Laser Phys. Lett.* 7 (2010) 351.
- [20] M. Schellhorn, S. Ngcobo, C. Bollig, *Appl. Phys. B* 94 (2009) 195.
- [21] X.M. Duan, B.Q. Yao, C.W. Song, J. Gao, Y.Z. Wang, *Laser Phys. Lett.* 5 (2008) 800.
- [22] P.A. Budni, M.L. Lemons, J.R. Mosto, E.P. Chicklis, *Quantum Electron.* 6 (2000) 629.
- [23] K. Scholle, P. Fuhrberg, *Conf. Lasers and Electro-Optics*, San Jose, 2008, paper CtuAA1.