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Deliverable Nature	
R = Report, P = Prototype, D = Demonstrator, O = Other	R
Dissemination Level	
PU = Public PP = Restricted to other programme participants (incl. the Commission Services) RE = Restricted to a group specified by the consortium (incl. the Commission Services) CO = Confidential, only for members of the consortium (incl. the Commission Services)	PU

A. Abstract / Executive Summary

The research activity within this period was concentrated on three main issues. The first one aimed obtaining of transparent Nd:YAG ceramic media by a solid-state phase synthesis method. Secondly, a disordered Yb:$Ca_3Li_{0.275}Nb_{1.775}Ga_{2.95}O_{12}$ (Yb:CLNGG) laser crystal was grown by the Czochralski method; laser emission around 1.03 μm was obtained under the pump with a fiber-coupled diode laser at 972 nm. In the third activity a new method of fabricating waveguides by direct writing with a fs-laser beam was developed and applied to Nd:YAG ceramic medium; efficient laser emission at 1.06 μm and 1.3 μm was achieved under the pump with a 808-nm emitting fiber-coupled diode laser. Such laser devices could be obtained in ceramic media. The research was performed in the Laboratory of Solid-State Quantum Electronics from INFLPR.

B. Deliverable Report

1 Introduction

The ceramic techniques allow nowadays obtaining of poly-crystalline laser media with very good optical quality, featuring easy manufacturability at competitive prices. Such ceramic materials can be obtained for the realization of good output performances in free generation mode, in passively Q-switched regime, but also for the generation of sub-ps laser pulses. The general purpose of our work is the fabrication of rare-earth ions doped ceramics and disordered crystals with wide fluorescence band suitable for generation of laser pulses close to 100-fs duration.

2 Objectives

The realization of a ceramic material depends on the employed technique, but there is no reported procedure in the literature that describes in details such a fabrication process. Therefore, a first objective was to establish a preparation method for a ceramic medium, and in the beginning we did the experiments for Nd:YAG. Once understood, such preparation techniques could be adapted and then applied to other ceramic media. Alternatively, a disordered Yb:CLNGG crystal was obtained by the Czochralski pulling growth method and efficient laser emission was obtained under the pump with a fiber-coupled diode laser. Mode-locking with a SESAM device is the next step within this direction; emission can be realized in the bulk material or in a more compact device, such as a waveguide. A method of writing waveguides with a fs-laser beam was applied to Nd:YAG, from which efficient laser emission at the fundamental wavelengths of 1.06 μm and 1.3 μm was obtained. Following the improvements of the Yb:CLNGG optical quality, the new technique will be used for inscribing waveguides in this laser medium.

3 Work performed / results / description

3.1. Obtaining of Nd:YAG ceramic

In order to realize Nd:YAG ceramic media we adopted the diagram procedure that is shown in Fig. 1. Powders of α-Al_2O_3 (99.99%, 20 to 50 nm in diameter), Nd_2O_3 (99.99%, 20 to 40 nm in diameter) and Y_2O_3 (99.99%, 20 to 50 nm in diameter) were dried at 250°C and weighted in respect to stoichiometric ratio of Nd:YAG (1.0 at %). Next, tetraethyl orthosilicate (TEOS) was added (as sintering aid) to these powders, followed by mixing that was done in ethyl alcohol using a mill with alumina balls; in some experiments mixing with a magnetic device was also performed. Polyethylene glycol (PEG-400) was added few hours before ending this process, in order to avoid clustering. The powder drying was made in nitrogen atmosphere using in a spray-drying Buchi 250 machine that was operated with 70°C temperature at the entrance and 40°C temperature at the output. Removal of some ethyl alcohol traces or water was done by keeping the mixed powders in an oven at 240°C for 24 hours.

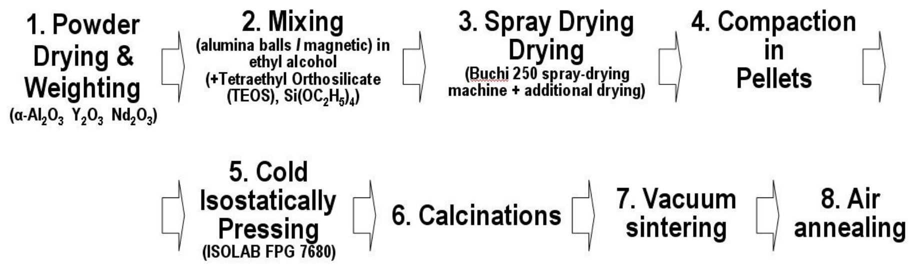

Fig. 1 The method of solid-state phase synthesis used at ECS-INFLPR for obtaining Nd:YAG ceramic.

Shaping was obtained by pressing the mixed powders at 100 bars and at a temperature of 100°C. Then, for removal of some organics residues the sample was annealed in air at 700°C. Next, a high pressure ISOLAB FPG-7680 installation (Stansted Fluid Power Ltd., UK) was employed to further press the sample at a pressure of 2450 bars for about 30 min. Sintering was performed in vacuum, using two devices: an oven, as well as an ADL crystal-growing equipment in which heating was obtained by induction. Finally, the samples were annealed in air at about 1300°C temperature.

The first samples were obtained using vacuum annealing in an oven. A heating rate of ~60°C/h was used to reach the working temperature of 1730°C; the ceramic samples were kept 16 hours at this temperature and then they were cooled down with a rate of 40°C/h. One of the samples was then polished; Fig 2a shows a photo of a 1.0-at.% Nd:YAG that was translucent. SEM photos of another sample are presented in Fig. 2b and Fig. 2c, at a scale of 10 µm and 4 µm, respectively. Analysis concludes that the granules dimensions had a diameter ranging between 5 to 20 µm, while some pores were observed.

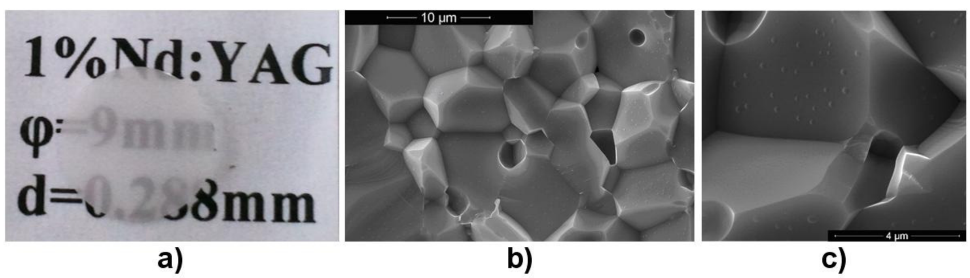

Fig. 2 **a)** A photo of a translucent 1.0-at.% Nd:YAG ceramic sample obtained by annealing in an oven is shown. SEM micrographs of the 1.0-at.% Nd:YAG ceramic cample are presented at scales of **b)** 10 µm and **c)** 4 µm.

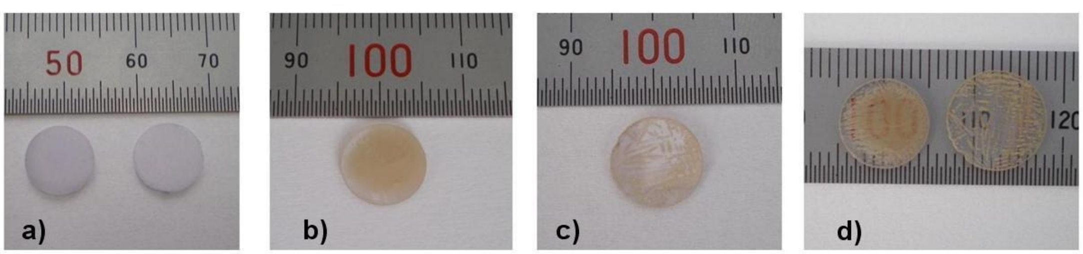

Fig. 3 Photos of 1.0-at.% Nd:YAG ceramic samples obtained by sintering in an ADL crystal-growt equipment are shown at various condition of sintering **a)** 1760°C/6h; **b)** 1740°C/12 h; **c)** 1760°C/16 h; **d)** a comparison between samples b) and c) is given.

Attempts to improve the quality of the Nd:YAG ceramics were made by performing the sintering process in an ADL crystal growth equipment (http://ecs.inflpr.ro/crystalgrowth.html). This installation could realize and keep the vacuum and temperature conditions better than the available oven used previously. As shown in Fig. 3, some Nd:YAG ceramics were translucet (Fig. 3a), and some exhibit zones with good transparency (Fig. 3b and Fig. 3c) but also has defects (the yellowish parts). An analysis performed with the help of our partners from Italy (Prof. Dr. Laura Esposito, CNR - Istituto di Scienza e Tecnologia dei Materiali

Ceramici Via Granarolo, 64 I-48018 Faenza, Italy) concluded that the Nd:YAG ceramics presents traces of some impurities (like Zn, Si, or W) or grains with composition close to YAM ($Y_4Al_2O_9$) phase. Further experiments aiming improving the Nd:YAG ceramic quality are under development.

3.2. Crystal growth and laser performances of an disordered Yb:CLNGG laser crystal

The partial disordered crystal of calcium niobium gallium garnet (CNGG) combines the advantages of both glasses and crystals, having wide absorption and emission bands and quite high thermal conductivity. An excess of Nb^{5+} ions and the formation of cationic vacancies are inevitable in the crystal, and the presence of such cationic vacancies might lead to some negative effects on laser action. The cationic vacancies can be eliminated by introducing Li^{+} ions into the lattice of CNGG, forming a new crystal of calcium lithium niobium gallium garnet (CLNGG) [1]. Laser emission in free-generation regime was reported from an Yb:CLNGG laser crystal under diode laser pumping [2], and very recently an Yb:CLNGG laser crystal was mode-locked by a single-walled carbon nanotube saturable absorber to obtain pulses at ~1.05 μm as short as 90 fs [3].

Disordered Yb:CLNGG laser crystals (with chemical formula Yb:$Ca_3Li_{0.275}Nb_{1.775}Ga_{2.95}O_{12}$) was obtained in our laboratory by the conventional Czochralski pulling method. A view of the boule crystal (with 4.3-at.% Yb doping) is shown in Fig. 4a, from which an uncoated, 3.5-mm thick Yb:CLNGG sample (as presented in Fig. 4b) was prepared for the experiments. The laser crystal was placed in a short, plane-plane resonator; the resonator rear mirror had wide HR coating at the laser wavelength of 1.04 μm and HT coating at the pump wavelength of 972 nm, whereas the out-coupling mirror (OCM) had a defined transmission (T) at the lasing wavelength. The pump was made in quasi-continuous wave mode (quasi-cw) at 972 nm with a fiber-coupled diode laser (JenOptik. Co., Germany); the fiber end (100-μm diameter, NA= 0.22) was imaged into Yb:CLNGG with a 1:1 optical system. The pump pulse duration was 1 ms; the repetition rate was maintained below 10 Hz in order to avoid thermal effects induced by optical pumping in the disordered Yb:CLNGG laser crystal.

a) b)

Fig. 4 a) A view of the boule Yb:CLNGG crystal is presented after growing. **b)** The 3.5-mm thick, 4.3-at.% Yb:CLNGG medium that was used in the laser emission experiments is shown.

The laser pulse characteristics are shown in Fig. 5. For example, when the resonator was equipped with an OCM of transmission T= 0.25, the laser crystal yielded pulses with maximum energy of 6.2 mJ for an absorbed pump pulse energy of 22.8 mJ; this corresponds to an overall optical-to-optical efficiency (with respect to the absorbed energy) of 0.27. The slope efficiency (with respect to the absorbed pump pulse energy) was η_{sa}= 0.33 (Fig. 5a). The spectra of emission recorded at the maximum energy of the pump pulse are shown in Fig. 5b. For an OCM with T= 0.03 the spectrum ranged from 1034.6 nm to 1040 nm (FWHM definition). An increase of the OCM transmission at T= 0.60 narrowed the emission spectrum band, from 1028.05 nm to 1031.3 nm, with the maximum at ~1030 nm. Further experiments will consider the improvements of the laser emission performances in free-generation regime (by growing Yb:CLNGG crystals with better optical quality, and by using coated crystals in optimized laser resonators), and realization of mode-locking operation. Realization of waveguide lasers in such a laser medium is considered by the technique described in Section 3.3.

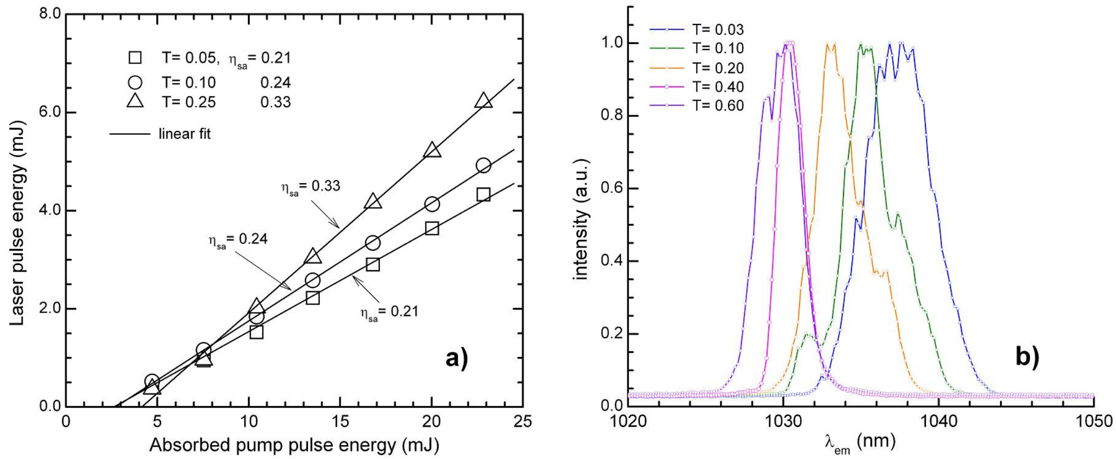

Fig. 5 a) Laser pulse energy versus the absorbed energy of the pump pulse. **b)** The emission spectra at the maximum energy of the pump pulse. T: the OCM transmission.

3.3. Waveguides realized in Nd:YAG ceramic medium by fs-laser writing with a helical moving technique

The waveguide lasers show interesting features, like compactness, low emission threshold and output performances close of those yielded by the bulk material; these characteristics makes them very attractive in optoelectronics. For fabrication of such a device, a powerful tool is the direct femtosecond (fs)-laser writing technique [4]. In the case of a tubular (cladding) waveguide, many parallel tracks are around a defined contour [5]. As shown in Fig. 6, within this translation method the Nd:YAG ceramic is moved transversally to the fs-laser beam, on direction Oy starting from surface S1. Once surface S2 is reached the focusing point of the fs-laser beam is changed to a new location (in the Oxz plane) and the writing continues with a new translation. Thus, an unmodified bulk material that is surrounded by many tracks with decreased index of refraction in the adjacent boundaries is obtained, and waveguiding is realized in the bulk region surrounded by the tracks. On the other hand, in order to avoid the medium fracture the tracks are inscribed at a distance of few μm between and consequently some unmodified material will remain between the tracks. These regions with unchanged refractive index can increase the waveguide propagation losses, decreasing thus the laser emission performances. A better overlap between the inscribed marks that build the waveguide walls can be achieved by moving the Nd:YAG medium on a helical trajectory, as shown in Fig. 6b [6]. Thus, the medium is moved circularly in the Oxz plane and translation is performed on direction Oy.

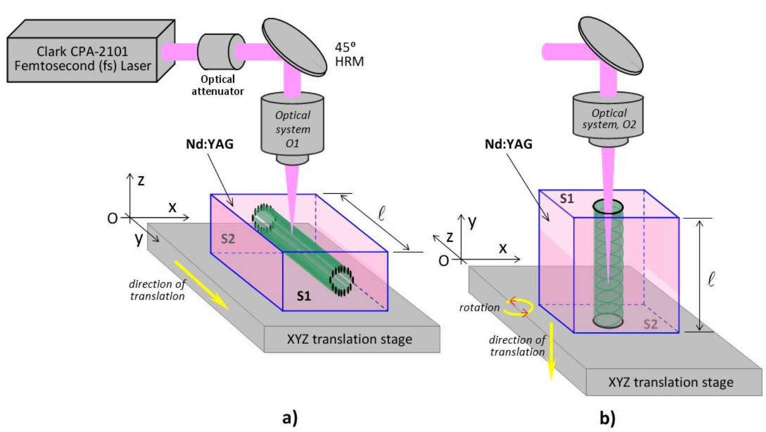

Fig. 6 The experimental setups used for inscribing cladding waveguides in Nd:YAG by direct writing with a fs-laser beam are shown: **a)** the step-by-step translation technique and **b)** the helical movement of the laser medium.

We have used this new writing scheme to inscribe circular waveguides in a 1.1-at.% Nd:YAG (5-mm thick) ceramic medium. For example, a 100-µm in diameter waveguide is presented in Fig. 7a (cross-section in the Oxz plane) and Fig. 7b (cross section in the Oxy plane). For comparison, a similar waveguide (i.e. diameter of 100 µm) was realized in the same Nd:YAG ceramic medium by the step-by-step translation technique (photos of this waveguide are shown in Fig. 7c and Fig. 7d). It can be clearly seen that an waveguide with continuous boundaries was realized by the helical movement of the Nd:YAG.

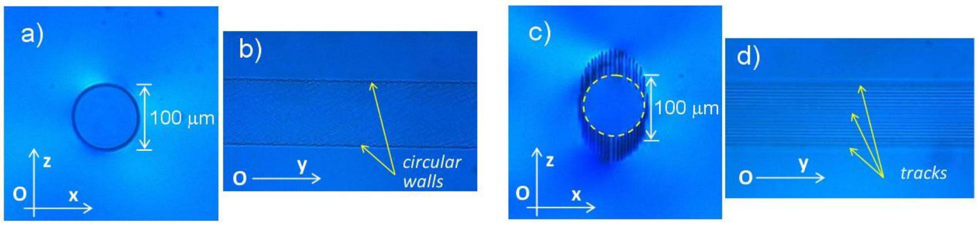

Fig. 7 Photos of waveguides with 100-µm diameter that were inscribed by the helical moving of the Nd:YAG: **a)** cross section (Oxz plane) view and **b)** side (Oxy) view and by the classical translation technique: **c)** cross section (Oxz plane) view and **d)** side (Oxy) view.

The performances of laser emission at 1.06 µm and 1.3 µm were investigated under the pump at 808 nm with a fiber-coupled (100-µm diameter, NA= 0.22) diode laser (LIMO Co., Germany). The diode was operated in quasi-cw mode, with 1 ms pulse duration and 10 Hz repetition rate. A good coupling of the pump beam into each waveguide was achieved by imaging the fiber end into Nd:YAG with a collimating lens of 50-mm focal length and a focusing lens of 30-mm focal length. A short, plane-plane resonator with the mirrors positioned closed of Nd:YAG was used.

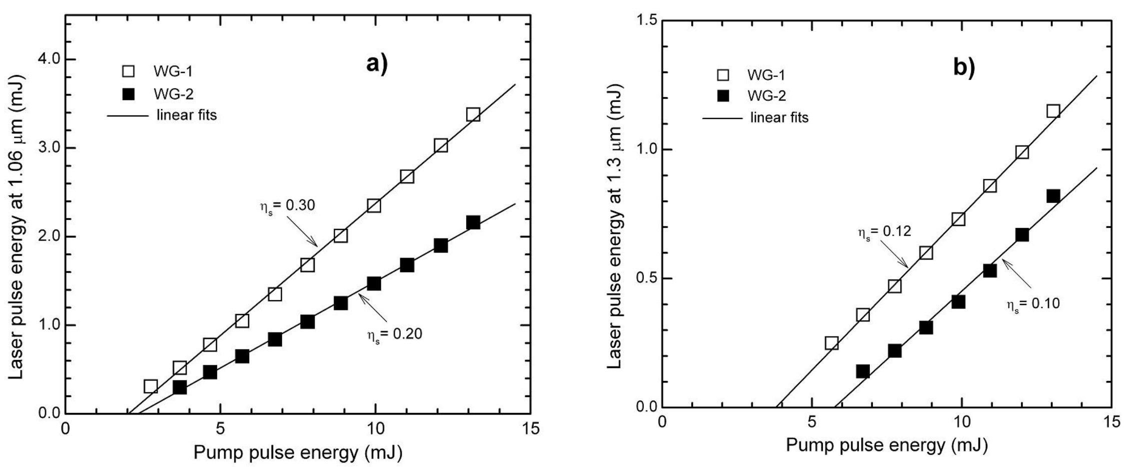

Fig. 8 Performances of laser emission at **a)** 1.06 µm, OCM with T= 0.10, and **b)** 1.3 µm, OCM with T= 0.03 obtained from the waveguides inscribed by the new helical movement scheme (WG-1) and by the classical step-by-step translation technique (WG-2).

It was found out that the waveguide realized by the helical moving of the Nd:YAG medium (WG-1) delivered laser pulses at 1.06 µm with 3.4 mJ energy (E_p), for the pump with pulses of energy E_{pump}= 13.1 mJ (i.e. overall optical-to-optical efficiency η_o= 0.26); the slope efficiency amounted at η_s= 0.30 (Fig. 8a). The waveguide WG-2 that was inscribed by the translation techniques yielded laser pulses with E_p= 2.2 mJ (E_{pump}= 13.1 mJ, η_o~0.17) and slope η_s= 0.20. The WG-1 waveguide improved the laser emission characteristics at 1.3 µm, too. Thus, as shown in Fig. 8b, laser pulses with E_p= 1.2 mJ (with η_o~0.09) at slope η_s= 0.12 were measured from this waveguide. On the other hand, the WG-2 device outputted pulses with maximum energy E_p= 0.8 mJ at 1.3 µm (at η_o~0.06), while slope efficiency was η_s= 0.10.

4 Conclusions

In summary, we applied a solid-state phase synthesis method to obtain Nd:YAG ceramic media. At this phase of the experiments the NdLYAG samples are trans-lucid; further experiments will be performed for optimization the sintering stage, which is believed to be responsible for the present results. A disordered Yb:CLNGG laser crystal was grown by the Czochralski pulling technique, and efficient laser emission around 1.03 μm was realized under the pump at 972 nm with a fiber-coupled diode laser. Furthemore, we have developed a new technique for obtaining circular waveguides in a laser crystal. This new method was applied to inscribe circular waveguides in Nd:YAG ceramic, from which laser emission at 1.06 μm and 1.3 μm was successfully obtained under the pump at 808 nm with a fiber-coupled diode laser. Such kind of waveguides are intended to be fabricated in the disordered Yb:CLNGG crystal and then mode-locking emission to be realized. The financing from the LaserLab-Europe, EuroLite project was acknowledged in a paper that was published in the Opt. Mat. Express journal.

5a. References

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5b. Publications

1. G. Salamu, F. Jipa, M. Zamfirescu, and N. Pavel, “Cladding waveguides realized in Nd:YAG ceramic by direct femtosecond-laser writing with a helical movement technique,” Opt. Mat. Express **4** (4), 790-797 (2014). DOI:10.1364/OME.4.000790 http://dx.doi.org/10.1364/OME.4.000790