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Transparent layered YAG ceramics with structured Yb doping produced *via* tape casting

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A B S T R A C T

The flexibility of the ceramic production process, in particular in terms of shaping and spatial control of distribution of active ions, is one of the strong points in favor of transparent ceramics. In high power lasers in particular, where thermal management is a critical issue, the finely controlled design of spatial distribution of the doping ions within the laser gain media can reduce undesired thermally induced effects and large temperature gradients, and thus enhance the efficiency and laser beam quality especially under increased thermal load. In the present work transparent structured YAG ceramics with Yb doping were produced by tape casting followed by thermal compression of assembled tapes and sintered under high vacuum. The thermal compression of variously doped tape cast layers is a very promising method because it allows a high precision and good control over dopant distribution in the sintered material. After sintering, the distribution of Yb across the layers was characterized by SEM-EDX and the thickness of Yb diffusion zones between the layers with different Yb content was measured. Optical homogeneity was assessed by means of optical transmittance mapping of the samples and by 2D scanning of laser output. The effect of structured dopant distribution on laser performance was measured in quasi-CW and CW regime with different duty factors. Slope efficiency values higher than 50% were measured both in quasi-CW and in CW lasing conditions. The results are in good agreement with previously calculated predictions, confirming the beneficial effect of structured doping on laser performances and enlightening the impact of the residual scattering losses. Compared to other processing methods, such as the pressing of granulated powders, tape casting followed by thermal compression leads to straight and narrow interfaces between layers with different composition and allows to build structures composed of extremely thin layers with defined dopant content.

1. Introduction

Yb:YAG is one of the most important gain media for diode-pumped solid state laser (DPSSL) systems providing advantages such as low quantum defect, long storage lifetime and wide absorption bandwidth suitable for diode pumping with InGaAs high power laser diodes emitting around 940 nm. The full miscibility of the YAG-YbAG system means that materials with high concentrations of dopant can be produced, increasing the absorption and consequently the output power. On the other hand, the thermally-induced effects due to the lower thermal conductivity after adding the dopant are still a barrier to the achievement of a high-energy extraction efficiency with the Yb:YAG system. The control in the distribution of the dopant can be successfully exploited to manage and reduce the thermal and thermomechanical effects (e.g. thermal lens, stress-induced depolarization, surface deformation) deriving from the laser pumping process. In addition, a suitable non-uniform dopant distribution can help to reduce Amplified Spontaneous Emission (ASE) effects with respect to uniform doping [1]. The positive effect of dopant content variation in Yb:YAG laser gain media has been studied in the recent years, in particular for applications in high-power lasers [2–4] and as thin disk [5,6] or microchip [7] laser media composed of doped and dopant-free layers, with results showing that a well-designed dopant distribution in the active medium can provide significant advantages. The benefit of such structure is twofold: the non-absorbing part without dopant is not directly heated by the pump absorption, and its presence helps in decreasing temperature gradients, reducing

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peak temperatures and consequently leads to the limitation of thermally induced effects such as the change of the index of refraction, surface deformations or thermomechanical stresses. As the thermal conductivity decreases with increasing dopant content, the introduction of a dopant-free part is also favorable from the viewpoint of heat flow and cooling in general. Thermal lensing and other thermally induced effects in Yb:YAG and other Yb doped garnets have been studied in a number of works [8–12] and especially under higher thermal load they are very important and can deteriorate the laser output.

For single crystals the only possibility to produce such structured laser gain media is diffusion bonding [5,7,13–15]. This requires above all a high-quality polishing of both surfaces that are to be bonded together (usually to surface flatness ~λ/10 [16]), a demanding and costly process, especially with increasing bonded surface area. Even more complicated is the case of bonding of multiple pieces. As transparent ceramics have moved from the position of a possible material to that of a competitor to single crystals during the last two decades, it is important to seek the advantages of ceramic technology for the production of laser gain media. Among others it is worth mentioning the wide choice of shaping techniques providing routes for the production of internally structured parts *in situ* [17]. One of the possible approaches is the use of tape casting.

Tape casting is a well-established ceramics shaping method [18], above all where thin sheets are required, viz. membranes [19], substrates for electronics [20], SOFCs [21], etc. However, apart from the possibility to provide thin sheets with defined thickness and high homogeneity, tape casting may be also used for the production of bulk ceramics. The high content of organic additives (binders, plasticizers), giving the cast and dried “green” tapes high elasticity, makes it also possible to assemble and stack various tapes, and then consolidate them into one piece. This is done by the application of pressure at elevated temperature, when the organic additives soften and the tapes are bonded together. The possibility to stack the tapes (of same or different composition) and form layered materials has been particularly useful for the production of PZT with controlled porosity [22] or various multilayered composites [23,24].

The use of tape casting has been recently proposed also for transparent ceramics: YAG or Mg-Al spinel [25]. Two research groups reported on the production of transparent rare-earth doped YAG *via* tape casting and a two-step sintering, vacuum sintering followed by hot isostatic pressing of Er:YAG [26] and Nd:YAG [27]. The use of tape casting has been reported also for Yb:YAG [28], where a structure with Yb doping increasing towards the center was reported.

In this work we report on the design, production and characterization of transparent YAG ceramics with multi-layered structures and one-dimensional controlled Yb doping profiles. The samples were produced by tape casting and sintered under vacuum. The dopant distribution profiles were designed by means of Finite Elements Analysis for the evaluation of the thermal and thermomechanical behaviour resulting in an end-pumped, end-cooled configuration, with the aim to reduce the peak temperature and the surface deformation with respect to a uniform dopant distribution with the same overall gain.

2. Experimental

2.1. Production

Two different layered structures were produced: “0–10” and “1-3-5-7”, where the numbers relate to the Yb doping level in the respective layers. The selection of these structures was based on Finite Elements Analysis described in [2]. The materials were prepared by solid state reaction sintering of a mixture of high-purity oxide powders. The following powders were used: Al_2O_3 (TM – DS-6, Taimei Chemicals, Japan; purity > 99.99%, mean grain size 0.2 μm), Y_2O_3 (REacton®, Alfa Aesar, MA, USA; purity > 99.99%, mean grain size 3.9 μm), Yb_2O_3 (REacton®, Alfa Aesar, MA, USA; purity > 99.99%, mean grain size 5.6 μm). The oxide powders were mixed with solvents (ethanol and methyl ethyl ketone), sintering additive (tetraethyl orthosilicate, TEOS), a binder, dispersant and plasticizers, and mixed by ball milling for 48 h. Afterwards, it was cast on a mylar sheet and left to dry for 24 h. The dried tape sheets, approximately 100 μm thick, were cut into desired shapes (discs 26 mm in diameter), which were assembled and pressed at elevated temperature (68 MPa, 100 °C) into compact green bodies (see Fig. 1).

The green compacts were treated in air at 600 °C in order to remove the organic additives. The samples were then sintered under high vacuum (10^{-4} Pa) in a clean Mo-W furnace at 1735 °C for 16 h and afterwards annealed in air for the removal of oxygen vacancies and reoxidation of Yb^{2+} ions to Yb^{3+}. Annealing temperature of 1100 °C was selected in order to prevent silicon segregation at grain boundaries. The sintered and annealed samples were mirror polished with diamond pastes and their final thickness was 1.9 mm and 2 mm (sample 0–10 and 1-3-5-7, respectively). The difference in thickness is due to the polishing process.

2.2. Characterization

2.2.1. Microstructure

Morphology of starting powders and microstructure of sintered samples were analyzed by SEM (FE-SEM, SIGMA, Zeiss, Oberkochen, Germany, with GEMINI column) coupled with EDX (INCA Energy 300, Oxford Instruments, UK). No conductive coating was applied on the samples. EDX line scans were performed across the polished sections of samples in order to provide information about the thickness of layers with different Yb content and of interdiffusion zones.

2.2.2. Optical transmission

Optical transmittance was measured with a Lambda 1050 (Perkin Elmer, USA) spectrophotometer in the range 200–1100 nm. Furthermore, the samples were characterized by a transmittance mapping setup shown in Fig. 2. In this setup, the sample is illuminated by a fiber coupled LED emitting at 623 nm. The illuminating beam is collimated by the lens L1. The lens L2 reimages the sample on a monochromatic CCD camera (6.6 μm pixel pitch, 1360 × 1024 pixels, 12 bit AD conversion) with a magnification factor of about 1.65. This arrangement was used to obtain transmission images of the sample, to detect the presence of internal defects which appear as dark spots against the illuminated background. The overall resolution is about 5 μm. By means of a proper spatial filtering, realized with a knife edge placed on the optical axis in the focal plane of the lens L2, the same setup provides parallel-light Schlieren images (see for instance [29]). This imaging method evidences the gradients of the refractive index in the direction perpendicular to the knife edge. This arrangement was used to investigate the possible presence of internal refractive index inhomogeneities in the samples, as well as to evidence the boundaries of the layers with different doping.

2.2.3. Laser performance

Sintered and mirror polished samples were characterized in an end pumped laser cavity illustrated in Fig. 3. The cavity was longitudinally pumped through its flat end mirror EM, which was

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