JLMN-Journal of Laser Micro/Nanoengineering Vol. 3, No. 1, 2008

Growth of Nd:$Gd_3Ga_5O_{12}$ Thin Films by Pulsed Laser Deposition for Planar Waveguide Laser

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Pulsed laser radiation of a KrF excimer laser was used for the deposition of thin Nd^{3+} doped $Gd_3Ga_5O_{12}$ (Nd:GGG) films on yttrium aluminium garnet (YAG) and sapphire single crystal substrates. By variation of PLD-parameters such as temperature and processing gas pressure, amorphous and single crystalline thin films were produced. The morphology and the composition of the grown films were investigated by optical microscopy, scanning electron microscopy and electron dispersive X-ray spectroscopy. Thickness and structural properties of the deposited films were determined by optical reflection spectroscopy and X-ray diffraction, respectively. The optical properties of grown films on different substrates were compared. An amorphous Nd:$Gd_3Ga_5O_{12}$ thin film on yttrium aluminium garnet was used for demonstration of an infrared waveguide laser. A planar wave guiding structure was formed in deposited film between two parallel grooves micromachined using laser radiation delivered by a femtosecond CPA-Laser-System. The resulting waveguides were polished and provided with resonator mirrors. With a 5% output coupler at 1064 nm, a laser threshold of 1080 mW and 0.2% slope efficiency were obtained.

DOI: 10.2961/jlmn.2008.01.0005

Keywords: Waveguide laser, pulsed laser deposition, fs-laser microstructuring, thin film, neodymium

1. Introduction

Waveguide lasers offer the potential for highly efficient and cost-efficient solid state light sources. Like fibre lasers, waveguide lasers exhibit a large intensity-length product and a good overlap of the pump and the signal mode, resulting in a reduced laser threshold compared to bulk lasers and increased conversion efficiency. Unlike in fibre lasers the numerical aperture and the geometry of waveguide lasers can easily be tailored to be optimally pumped using diode laser radiation. Therefore, a cost effective integration of a waveguide laser and the pump diode laser seems to be feasible in the future. In addition, a later integration of further optical and electronic components like polarisers, electro-optical modulators and gratings is possible, enabling further applications. For example, the development of cheap integrated pulsed waveguide lasers for marking, illumination or medical technology is highly desirable.

The integration of waveguide lasers with pump diode lasers seems to be possible, if the diode lasers can be monolithically joined with structured waveguides on a common mount. For this purpose a material suitable for low-temperature processing has to be identified. To develop an efficient integrated waveguide laser, a material with high conversion efficiency is required. Furthermore, the necessary production technologies with the flexibility to process those materials for prototypes have to be found. The benefits of the integration are a reduction of the cost for the assembly and the possibility to exploit mass production capabilities common in microelectronics for the production of the waveguide lasers. For the fabrication of planar waveguide structures diverse techniques such as liquid phase epitaxy, metalorganic chemical vapour deposition, molecular beam epitaxy, ion implantation, diffusion, sputtering or pulsed laser deposition have been used. Especially pulsed laser deposition (PLD) has been proven as an excellent method for the production of planar waveguides. Due to its flexibility regarding the desired composition and the structure of the resulting thin film, PLD has been successfully applied for the deposition of laser active materials such as Er:ZBLAN [1], Ti:Sapphire [2], neodymium doped YAP ($YAlO_3$) [3], YAG ($Y_3Al_5O_{12}$) doped with neodymium [1] and ytterbium [4], GGG ($Gd_3Ga_5O_{12}$) doped with praseodymium [5] and neodymium [6] etc.

For the thin films' deposition, sintered ceramics and single crystal materials were used. Furthermore, pulsed laser deposition enables the transfer of the stoichiometry of the used target material to the substrate. By variation of the process parameters such as temperature and processing gas pressure amorphous, polycrystalline and crystalline thin films have been achieved [6, 9].

Non-structured planar waveguide lasers from Nd:GGG films grown by PLD were demonstrated [6, 7, 8]. The lowest damping of 0.1db/cm has been achieved in Nd:GGG films deposited from single crystal target on YAG substrates [6]. The Nd:GGG waveguides on YAG substrates exhibited a numerical aperture of 0,75 and a slope efficiency of more than 10% has been achieved in the planar waveguide lasers.

In this paper we report on the growth of Nd: $Gd_3Ga_5O_{12}$ thin films using PLD as technique suitable for low-temperature processing and on the demonstration of an infrared waveguide laser fabricated by femtosecond laser micro structuring of the pulsed laser deposited amorphous film to define ridge wave guiding structures.

2. Experimental set- up

Amorphous and crystalline Nd:$Gd_3Ga_5O_{12}$ thin films were grown by ablation of a 1% Nd doped GGG crystalline target by means of KrF excimer laser radiation (wavelength λ = 248 nm, repetition rate f = 20 Hz, pulse duration τ = 20 ns). The target was polished to optical surface quality and was rotated during the deposition allowing the ablation of fresh material. The laser beam was formed by a mask, which was imaged by a telescopic lens system to a 1.1 x 1.7 mm^2 rectangular spot on the target surface at an angle of incidence of 45°. Using a repetition rate of f = 20 Hz and an energy density of ε_p = 2,5-3 J/cm² films up to 5 µm in thickness were grown on single crystal YAG ($Y_3Al_5O_{12}$) and sapphire, as determined by optical reflection spectroscopy. The deposition was carried out in a vacuum chamber using oxygen as processing gas. The oxygen pressure was varied in the range of P = 2-30 Pa. The substrate temperature was varied from room temperature to T_{sub} = 880°C and was monitored by a pyrometer (Kleiber). The distance between the target, positioned at an angle of 45°, and the substrate was varied from 3.5 to 5 cm.

To investigate the effect of the deposition parameters such as substrate temperature and processing gas pressure on the resulting optical properties of the deposited Nd:GGG films, two series of thin films were fabricated. The first series was grown at various processing gas pressures between 2 and 30 Pa and constant substrate temperature of T_{sub} = 850°C, whereas the second series was deposited at different substrate temperatures between 400°C and 880°C and constant processing gas pressure of P = 10 Pa.

The elemental composition of the deposited films was determined by the spatially resolved energy dispersion X-ray (EDX) technique. The structural characteristics of the Nd:$Gd_3Ga_5O_{12}$ films were determined by X-ray diffraction (XRD) using θ-2θ scans in Bragg-Brentano geometry with Cu Kα radiation (wavelength 0.15406 nm).

The ridge waveguide was structured in the pulsed laser deposited thin films using a commercial, multi-pass regeneratively amplified Kerr-lens mode-locked Ti:Sapphire laser (Concerto, Thales laser). The laser operates at a repetition rate of f = 1 kHz and a pulse duration of τ = 100 fs at a wavelength of λ = 810 nm with 25 nm bandwidth and a nearly diffraction limited beam quality ($M^2 \leq 1.5$). To reduce the roughness of the waveguides edges, the polarization of the light was oriented perpendicular to the scanning direction to prevent ripples formation rectangular to the waveguides edges [10]. A 63 x objective with NA=0.7 (Leica) and a 50 x objective with NA=0.55 (Olympus) were used for the focusing of the laser radiation.

Using 1:1 imaging optics the radiation of a diode laser (wavelength λ = 808 nm, power P_{pump} = 1.2 W, facet area 1 µm x 100 µm) was coupled into the polished edge of the neodymium doped films to investigate their spectroscopic properties. A monochromator (Chromex 500 IS) was used to disperse the emission from the films, which was detected by a CCD camera (Andor iDUS DU420A-OE).

For the damping measurements of the waveguides a CMOS camera was used to collect the scattered light from the waveguide and the lateral exponential decay of the intensity along the waveguide was fitted in terms of an exponential function.

3. Results and discussion

3.1 Composition of deposited Nd: $Gd_3Ga_5O_{12}$ thin films

The composition, e.g. the [Ga]:[Gd] atomic ratio, of the films deposited at 850°C as determined by EDX-measurement depends on the processing gas pressure (Fig. 1).

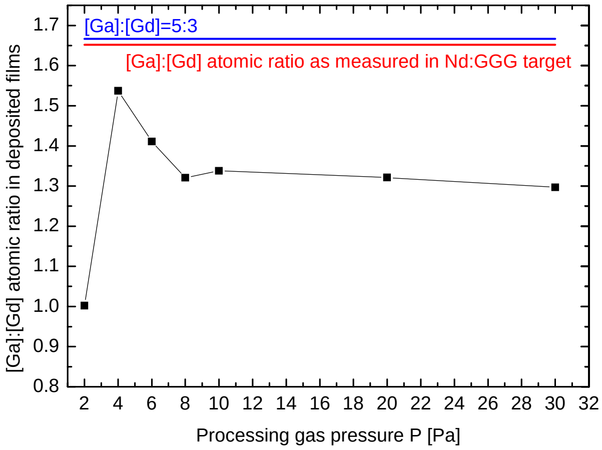

Fig. 1 Dependence of the [Ga]:[Gd] atomic ratio as a function of the processing gas pressure in the films deposited at T_{sub} = 850°C

The [Ga]:[Gd] atomic ratio measured in the 1% neodymium doped GGG single crystal target which was used for the deposition is similar to the theoretical stoichiometric $Gd_3Ga_5O_{12}$ composition. The [Ga]:[Gd] atomic ratio in deposited films is closest to the 5/3 stoichiometric value of $Gd_3Ga_5O_{12}$ at a pressure of P = 4 Pa. With a decrease of processing gas pressure, the deposited films became Ga deficient. This phenomenon has been explained by the decomposition of the Ga_2O_3 to Ga_2O and O_2 at low oxygen pressure. Because Ga_2O is a volatile compound it can sublimate from the surface of the growing film due to high substrate temperature or from the target surface due to the interaction with the laser beam [5]. However, with an increase of processing gas pressure, the deposited films become Ga deficient again and the [Ga]:[Gd] atomic ratio decreases from 1.53 to ~1.30 and saturates in the range from 10 to 30 Pa. The expectation that the deposited films become Ga deficient only at lower pressure was not confirmed here. For this phenomenon the recombination of target material due to interaction with the laser beam at higher pressure may be responsible. To investigate this behaviour the composition in the ablated target area was investigated and characterized by EDX.

The target was ablated using 45,000 pulses at P = 10 Pa of oxygen. To investigate the modification of the $Gd_3Ga_5O_{12}$ stoichiometric composition due to interaction with laser radiation at higher pressure, three line scans (10 points with a step of 40 µm) over non-ablated (edges of the track) and ablated target areas (middle of the track) were performed (Fig. 2 top photograph).

The $Gd_3Ga_5O_{12}$ stoichiometry very close to the bulk crystal was detected only at the edge of the track (area I and III) (Fig. 2 bottom diagram). A higher [Ga]:[Gd] atomic ratio in the area next to the ablated zone was detected in the re-deposited material (debris).

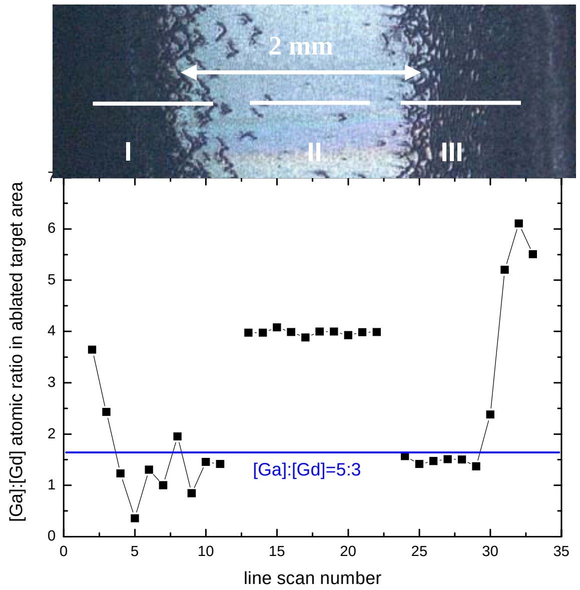

Fig. 2 Photograph of ablated target area (top photograph) and line scan of the [Ga]:[Gd] atomic ratio (bottom diagram) ratio

In the central part of the ablated area (II) an enrichment of Ga was detected, as well. The increased concentration of gallium in this area may be explained by increased diffusion of gallium to the surface due to the interaction with laser radiation at a higher pressure. Further experiments are necessary to prove this assumption.

Concluding the results of this experiment an influence of processing gas pressure on $Gd_3Ga_5O_{12}$ stoichiometric composition in deposited films was found. The [Ga]:[Gd] atomic ratio in the target is higher than measured in the films. In the deposited films Ga loss may occur due to the modification in the films after the deposition.

3.2 Structural and optical characteristics of deposited Nd: $Gd_3Ga_5O_{12}$ thin films

Use of a substrate temperature of T_{sub} = 750°C during PLD resulted in amorphous Nd:$Gd_3Ga_5O_{12}$ films and at T_{sub} = 865°C single crystalline Nd:GGG films were deposited on YAG substrate without any annealing process necessary as revealed by XRD (Fig. 3).

The fluorescence emission spectrum of a crystalline film excited at λ = 808 nm exhibits the individual lines from the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition (at 1060 nm) broadened but similar to the spectrum obtained from a bulk single crystal sample (Fig. 4). The spectrum of the amorphous film is broadened inhomogeneously and the maximum of the emission intensity is shifted to a larger wavelength of about 1066 nm (Fig. 4). The fluorescence life-time of the ${}^4F_{3/2}$ level was determined to be ~ 160 µs in amorphous films and 180 µs in single crystalline films in close agreement with the value of the bulk crystal.

The emission spectra of thin films grown on sapphire single crystal substrates are inhomogeneously broadened and the maximum of the emission intensity is shifted to a wavelength at about 1066 nm, similar to the amorphous thin films on YAG substrates (Fig. 5). The fluorescence intensity of the films deposited at 750 °C does not show much difference to the films ablated at 880°C (Fig. 5). Grown films did not show the typical crystalline lines due to a relatively large mismatch between sapphire single crystal and GGG. Thus, Nd:GGG films deposited on sapphire do not seem to be a promising material combination for a planar waveguide laser.

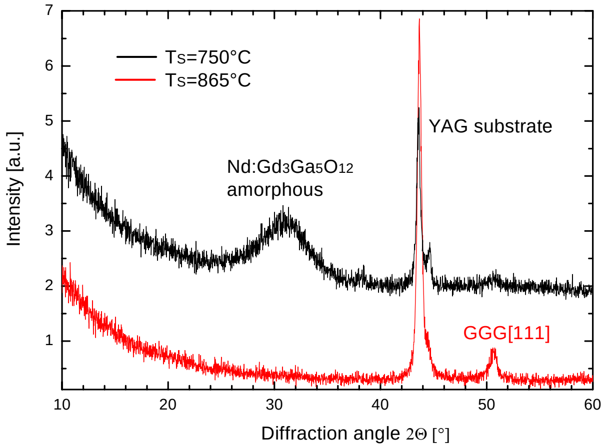

Fig. 3 X-ray diffraction spectra of Nd:$Gd_3Ga_5O_{12}$ grown on YAG single crystal substrate

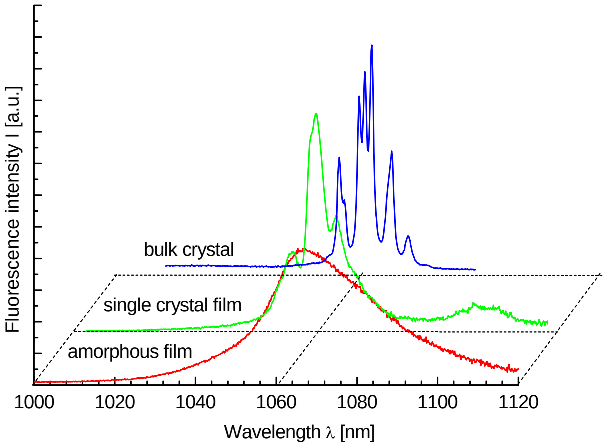

Fig. 4 Fluorescence spectra of an amorphous and a single crystalline Nd:$Gd_3Ga_5O_{12}$ film and a bulk crystal

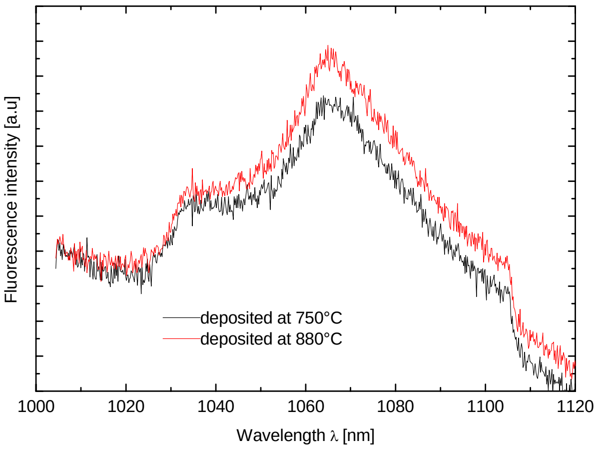

Fig. 5 Fluorescence spectra of Nd:$Gd_3Ga_5O_{12}$ films grown on sapphire single crystal substrate

To determine the correlation between processing parameters (e.g. processing gas pressure, energy density on the target, substrate temperature) and optical quality of deposited thin films different processing parameters were used for deposition of thin films.

Using a substrate temperature of $T_{sub} = 410°C$, processing gas pressure $P = 2$ Pa and an energy density of $\varepsilon_p = 3$ J/cm^2 films with $0.8 \pm 0.2 \times 10^6$ particles/cm^2 were fabricated. Using an energy density of $\varepsilon_p = 2.5$ J/cm² and a processing gas pressure of $P = 8$ Pa at a substrate temperature of $T_{sub} = 830°C$ resulted in very smooth films with very a low droplet density of $0.2 \pm 0.1 \times 10^4$ particles/cm^2. Due to different process parameters films with different quality values were produced. The use of high pressure during the deposition process enabled the deceleration of high-energetically species from the target on the way to the substrate and less damage in the deposited layer can be achieved. High pressure combined with high substrate temperature can result in a smooth film surface due to the given ability of deposited species for better movement and self organisation at higher temperatures.

Other than processing gas pressure, substrate temperature and energy density on the target, the optical quality of deposited films may depend on the condition of the target material, e.g. a polished or unpolished surface, sintered or single crystal. For the repeatability of deposited films with smooth surface further experiments are planned.

The amorphous thin film used for waveguide laser demonstration was deposited at $T_{sub} = 750°C$ and processing gas pressure of $P = 8$ Pa. Thin film 1.1 µm in thickness with $2 \pm 1 \times 10^{3}$ particles/cm² (Fig. 6) was grown on single crystalline [100] YAG substrate using a target-to-substrate distance of 4 cm and an energy density of $\varepsilon_p = 3$ J/cm^2.

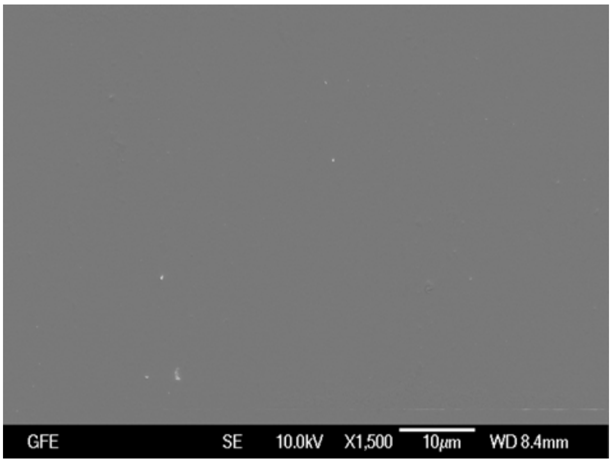

Fig. 6 Scanning electron micrograph of the amorphous $Nd{:}Gd_3Ga_5O_{12}$ thin film

3.3 Laser activity of an amorphous Nd: $Gd_3Ga_5O_{12}$ thin film

The thin films grown by PLD were micromachined by means of fs-laser radiation by generating two parallel grooves with an offset of 100 µm resulting in a ridge waveguiding structure. The ridge waveguide was cut to a length of 3 mm, lapped and polished to optical quality of the end facets. After cleaning with alcohol the in-coupling mirror (anti reflective at 808 nm and highly reflective at 1064 nm) and the out-coupling mirror (highly reflective at 808 nm and 5 % out-coupling at 1064 nm) were mounted directly to the waveguides. The pump light from a λ = 808 nm broad area diode laser was coupled into the waveguide using a 1:1 imaging optical system. At a pump power of 1080 mW measured in the collimated beam, the laser threshold was reached. The emission spectra changed from the broad fluorescence peak of the amorphous film to the typical narrowed laser emission peak with three longitudinal modes (Fig. 7).

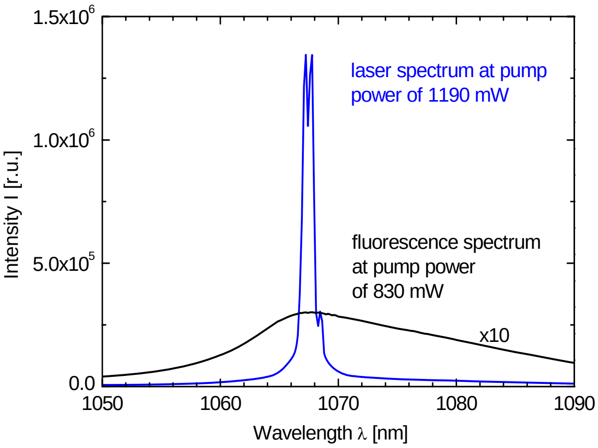

Fig. 7 The laser spectrum of an amorphous $Nd{:}Gd_3Ga_5O_{12}$ waveguide laser

Using a pump power of 1.2 W a signal of 1.35 mW was measured. A slope efficiency of 0.2 % in respect to the pump power of the collimated diode-laser was obtained (Fig. 8).

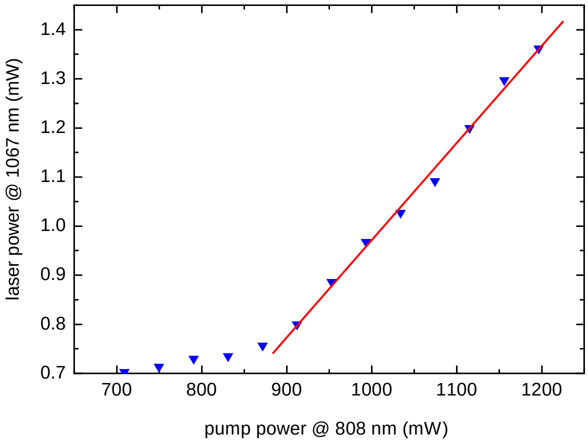

Fig. 8 Slope efficiency of an amorphous $Nd{:}Gd_3Ga_5O_{12}$ waveguide laser

To increase the slope efficiency of a planar waveguide laser, the coupling losses between waveguide and mirrors as well as the in-coupling losses of the diode laser radiation have to be minimized. Further investigations are planned to quantify the loss channels in the resonator such as scattering from edge roughness. Primary the losses due to particles have to be reduced by identification of suitable deposition process parameters such as processing gas pressure, substrate temperature and energy density on the target.

4. Conclusion and outlook

Using pulsed laser deposition amorphous and single crystalline films of Nd:$Gd_3Ga_5O_{12}$ were fabricated. Optical properties of single crystal thin films such as fluorescence emission spectra and life-times were similar to the target material.

An influence of the processing gas pressure on the [Ga]:[Gd] atomic ratio in deposited films was found. With decreasing processing gas pressure, the films became Ga deficient.

The laser action of an amorphous Nd:$Gd_3Ga_5O_{12}$ ridge waveguide fabricated by pulsed laser deposition and subsequent structuring of the thin film using femtosecond laser radiation was demonstrated. An amorphous Nd:$Gd_3Ga_5O_{12}$ ridge waveguide 1.1 µm in height, 100 µm in width and 3 mm in length pumped by diode laser radiation using 1:1 imaging optics showed the laser effect. Further experiments involve investigations to increase the ridge waveguide laser efficiency and the comparison of a waveguide consisting of single-crystal Nd:GGG as well as an investigation for deposition of other laser active materials and their potential for planar waveguide laser manufacturing.

Acknowledgment

The authors would like to thank Mr. Herbert Horn-Solle of the Lehrstuhl für Lasertechnik for his assistance in SEM and EDX measurements, Mrs. Ann-Cristin Dippel of the Institute für Werkstoffe der Elektrotechnik II for access to the X-ray diffractometer and Philips Innovative Applications NV for funding of the project.

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(Received: May 14, 2007, Accepted: November 26, 2007)