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Highly doped Nd:YAG crystal used for microchip lasers

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Abstract

The highly doped Nd:YAG (Nd concentration as high as 3 at.%) crystals were grown by the temperature gradient technology. The spectral properties of these highly doped Nd:YAG crystals were reported and compared with that of the traditional Nd:YAG (1.1 at.% Nd) grown by the Czochralski method. Although concentration quenching exists in highly doped Nd:YAG crystals, the laser performances have demonstrated that highly doped Nd:YAG crystals have the advantages over that of the traditional grown Nd:YAG such as efficiencies, especially used in microchip lasers.

Keywords: Highly doped Nd:YAG; Temperature gradient technology; Microchip lasers

1. Introduction

Recent developments of laser diodes (LD) pumped solid-state microchip lasers have stimulated the development of high gain mediums. Among the high gain mediums, the neodymium (Nd) doped yttrium aluminum garnet (YAG) is the most available laser material used for microchip lasers and supports huge area of laser applications. There are also other laser materials such as Nd-doped yttrium vanadate (YVO_4) used as microchip lasers, although Nd:YVO_4 crystal is a suitable material for high efficient microchip laser owing to its large absorption coefficient (31.2 cm^{-1} for 1 at.% Nd:YVO_4) and large absorption cross-section ($\sim 25 \times 10^{-19}$ cm^2) [1], high power operation is difficult because its thermal–mechanical properties are poor and the grown of such high-quality crystal with highly doping concentration is difficult. The high-quality Nd:YAG crystal can be obtained by the traditional Czochralski (CZ) method, but one of several causes that limits the use of Nd:YAG crystal as microchip lasers is the Nd ions concentration in YAG is very low and requires longer crystal to obtain enough gain. When Nd-doped YAG is grown, yttrium in YAG is substituted by Nd ions. But the Nd ion concentrations is limited to less than 1.1 at.% because the distribution coefficient of Nd in YAG is only 0.2. The high concentration of laser active ion in a laser crystal absorbs power of pump laser effectively with short crystal length and it makes the laser system very compact. So the high concentration laser crystals is the goal of development of microchip lasers and many efforts have been taken

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to search such high concentration laser materials. Recently some studies have been reported on highly Nd-doped crystals [2,3] or ceramics [4,5], compared Nd-doped YAG crystals with Nd-doped YAG ceramics, we can clearly see that Nd:YAG crystals are prior to Nd-doped YAG ceramics, because Nd-doped YAG crystal is a single crystal and has a perfect structure, and the structure of Nd-doped YAG ceramics is not perfect, when pump light is insert into Nd-doped YAG ceramics, there exists scattering, so the absorption of pump power is not effective. So, highly Nd-doped YAG would become more excellent laser material in all solid-state age with several fine characteristics which essentially YAG possesses, for instance, relatively long fluorescence lifetime, high heat conductivity, chemical and physical stability, and other isotropic natures. These characteristics act effectively especially in high power or pulsed operations.

We had earlier successfully grown high-quality sapphire crystal by the temperature gradient technique (TGT) and sapphire crystals [6] and Nd: YAG [7] were also grown by the TGT. Here, we report the growth of highly doped Nd:YAG (Nd concentration as high as 3 at.%) with TGT and spectral properties and laser performance of such promising laser material used for microchip lasers.

2. The grown of highly doped Nd:YAG crystal with TGT

Highly doped Nd:YAG crystals were grown in tapered molybdenum crucibles with a lower seed end. A cylindrical seed crystal with ⟨1 1 1⟩ direction was placed in the seed position. Oxide powders of high purity Al_2O_3, Y_2O_3 and Nd_2O_3 used for crystal growth are readily available from the chemical reagent factories at grades of 8 purity. The three powders must be ignited for several hours at a temperature of 1300 °C to remove moisture. After weighing according to the crystal stoichiometry with excess of 0.5 wt.% Al_2O_3, the powders were thoroughly mixed and then pressed to form blocks with diameter close to the inner diameter of the crucible. These blocks were sintered at 1400 °C for 48 h in air and loaded into crucible. The furnace was then loaded for the growth process (crucible, heat shields, and heating element), outgassed to 10^{-5} Torr and the material was melted and kept molten for several hours. After the temperature field was stabilized, crystallization was started by slow cooling at a rate of ~0.6 °C/h with a high precision temperature program controller. The whole crystallization process was completed automatically. During the growth process, it is essential that the temperature and the thermal field are very stable and an important factor is the flow stability of the circulation cooling water so that the solid–liquid interface advance with linear velocity. With stabilized thermal field and sufficiently slow stable cooling rate, scattering centers and second phase particles will be eliminated.

A highly doped Nd:YAG crystal by TGT, 60 mm in diameter and 100 mm in length, is shown in Fig. 1. The as-grown Nd:YAG crystals were slightly brown in color. This color could be removed by a high temperature annealing, the

Fig. 1. Highly doped Nd:YAG crystal grown by TGT (ϕ60 × 100 mm).

annealing process is as following: in oxygen atmosphere the Nd:YAG crystals were fired at about 1350 °C for 50 h, and cooled to room temperature at a rate of 10 °C/h. After annealing, the color of highly doped Nd:YAG was changed to pink.

The distribution of Nd along the growth axis was determined by X-ray fluorescence method, and at the same time the fluorescence lifetime (τ) was also measured along the growth axis of highly doped Nd:YAG crystal. Compared the Nd concentration along the growth axis and fluorescence lifetime, there is a relationship between Nd concentration and fluorescence lifetime, such as the fluorescence lifetime of 230 μs is corresponding to 1 at.% Nd-doped Nd:YAG measured by X-ray fluorescence method, and 175 μs vs. 2 at.% Nd-doped Nd:YAG crystal, etc. And the accuracy of measuring Nd concentration of Nd:YAG through measuring fluorescence lifetime is within 10% compared to X-ray fluorescence. Although X-ray fluorescence method can measure Nd concentration quantitatively, it is complex and we are short of this equipment, and the measurement of fluorescence lifetime is simple and non-destruction to Nd:YAG crystals, for convenience, Nd concentration along the growth axis of highly doped Nd:YAG crystals can be approximately measured through measuring the fluorescence lifetime. The decay of fluorescence intensity were measured using pulsed Ti:sapphire laser. Fluorescence from the Nd:YAG samples were focused on a photodiode with a convex lens through a filter which passed only around 1064 nm wavelength. A Nd:YAG crystal sample doped with 1 at.% Nd grown by the CZ method was also prepared for comparison. So from the comparison of fluorescence lifetime of highly doped Nd:YAG crystals and 1 at.% Nd:YAG crystal grown by CZ, the concentration distribution of highly doped Nd:YAG is shown in Fig. 2. From Fig. 2, the length of 3 at.% Nd:YAG is about 10 mm and the length of greater than 2 at.% Nd:YAG is about 15 mm. And the fluorescence lifetime along the radius direction was also measured and found the concentration along the radius direction is nearly unity. So highly doped Nd:YAG (>2 at.% Nd) is about a quarter of the whole Nd:YAG crystal and a lot of microchip wafers can be produced.

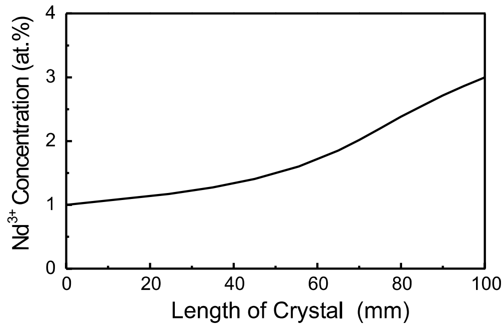

Fig. 2. The distribution of Nd along growth axis of highly doped Nd:YAG single crystal grown by TGT.

3. The spectral and laser properties of highly doped Nd:YAG crystals grown by TGT

3.1. The absorption and luminescence properties of highly doped Nd:YAG

The highly doped Nd:YAG samples used in measuring spectral properties are two pieces $5 \times 5 \times 1\ mm^3$ crystals doped with 2 and 3 at.% Nd respectively. First, we measured the absorption coefficient at 808 nm for pump wavelength, the YAG sample doped with 2 at.% Nd (HD2) and the sample doped with 3 at.% Nd (HD3) absorb 81% and 90% respectively. The spectral optical density $OD(\lambda) = 0.435\alpha(\lambda)L$ of Nd:YAG crystal at room temperature was measured using a Lambda Perkin–Elmer 9 UV/VIS/NIR spectrometer and converted to absorption coefficient $\alpha(\lambda)$ by measuring the sample thickness L, and the resolution of the measured absorption coefficient is $\pm 0.15\ cm^{-1}$. Regarding of the loss on the surface, the absorption coefficients were 14.9 cm^{-1} for HD2 and 20.9 cm^{-1} for HD3. We also measured the absorption coefficient of 1 at.% Nd:YAG crystal grown by the traditional CZ method, and found that the coefficients of HD2 and HD3 are two and three times of the typical absorption coefficient of 1 at.% doped Nd:YAG crystal. From the point of absorption of highly doped Nd:YAG grown by TGT, we can see that the Nd:YAG crystals used in microchip lasers may be shorter than that of traditional grown Nd:YAG crystals and can obtain high output efficiency. 20.9 cm^{-1} is the absorption coefficient for

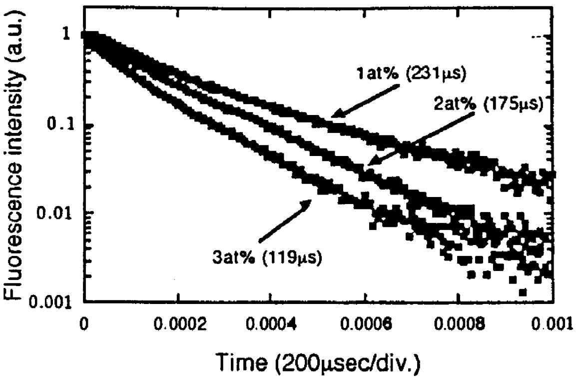

Fig. 3. Decay of fluorescence intensities of Nd:YAG crystals.

3 at.% Nd:YAG, it is slightly low, this may be caused the error of the Nd concentration measured by measuring the fluorescence lifetime.

Secondly, we measured the time decay of fluorescence intensity using pulsed Ti:sapphire laser. Fluorescence from the Nd:YAG samples were focused into a photodiode with a convex lens through a filter which passed only around 1064 nm light. A sample of traditional grown 1 at.% Nd:YAG crystal was also prepared for the comparison. Fig. 3 shows the time dependence of the fluorescence intensities. Each slope of the curves exhibits that time constants of decay are 231 μs for 1 at.% Nd:YAG, 175 μs for HD2, and 120 μs for HD3, respectively. When the Nd ion concentration is increased, the fluorescence lifetime is shorten, so there is concentration quenching in highly doped Nd:YAG crystals. The concentration quenching may be ascribed to more-short-range mechanism of the Nd^{3+}–Nd^{3+} quenching interaction. This phenomenon plays an increasingly important role with rising Nd ion concentration (Nd^{3+} concentration > 1.5 at.%). Although there is concentration quenching in highly doped Nd:YAG crystals, the crystal quality is still excellent, this is demonstrated by the laser experiments in Section 3.2. It should be noted that, taking into account the effects of the short-range quenching interaction of Nd^{3+}–Nd^{3+}, the Nd–Nd fluorescence decay of highly doped Nd:YAG crystal is a highly complicated problem, and is a subject for future investigations.

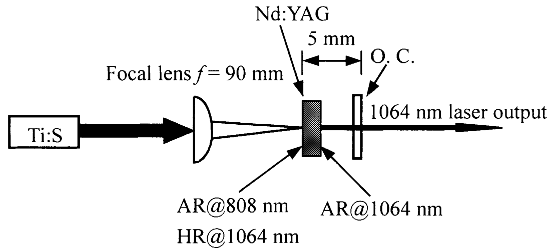

Fig. 4. The schematic of Ti:sapphire laser pumped highly doped Nd:YAG laser: Ti:S—Ti:sapphire laser, OC—output coupler.

3.2. The pulse and cw laser performance of highly doped Nd: YAG crystals

Fig. 4 shows the schematic of Ti:sapphire laser pumped highly doped Nd:YAG laser. High-quality YAG crystals doped with 2 and 3 at.% Nd were used as laser gain mediums, they were cut into $5 \times 5 \times 1$ mm^3 pieces. One of the square surfaces was anti-reflection (AR) coated at 808 nm and totally reflective coated at 1064 nm. The other side was AR coated at 1064 nm. Ti:sapphire lasers in pulsed (1 kHz, 100 ns, 40 mW) and cw (120 mW) operation were used as the pump sources. The beam from the Ti:sapphire laser, after beam shaping with a plano-convex lens of $f = 90$ mm, was focused onto a spot with a diameter of 50 μm, and the beam transversal distribution was TEM_{00} mode, and the transverse mode of the Ti:sapphire laser is $M^2 = 2.05$. According to the pump beam parameters, the focal length is about 2.4 mm, and the Nd:YAG active medium with 1 mm thickness is in this range, so the effect of focal length can be negligible. Flat mirrors with reflectivities of 96%, 90%, and 80% were used as output couplers to construct the plane–plane laser cavity. The laser cavity is about 5 mm length. Best results were obtained with the output coupler with 90% reflectivity. Fig. 5 shows the average output power as a function of the absorbed pump power under pumping with the pulsed Ti:sapphire laser. Using HD2 sample, a threshold pump power of 5.8 mW and a slope efficiency of 64% were obtained, and the optical efficiency (the ratio of output power and absorbed pump power) is as high as 50% when the maximum pump power is as high as 27 mW. The output power of 12.5 mW was limited only by

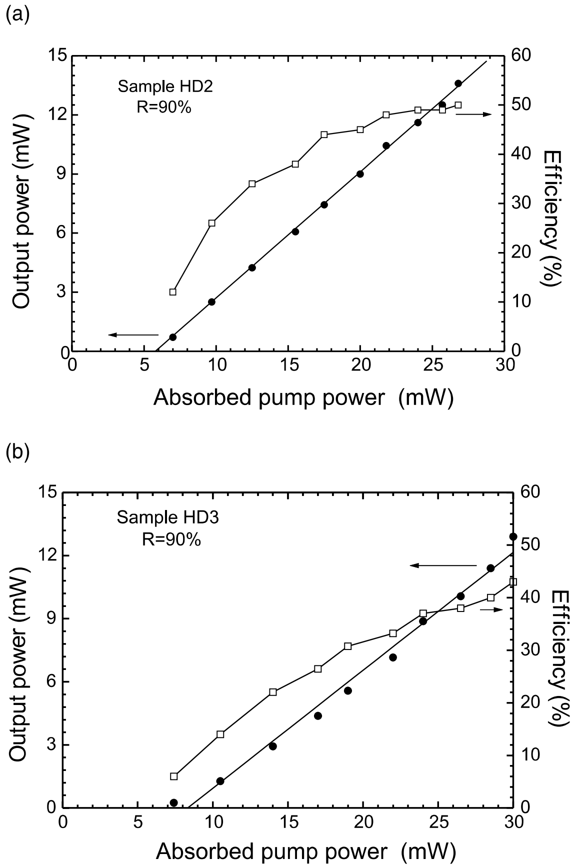

Fig. 5. Output power as a function of absorbed pump power in pulse operation: (a) HD2, (b) HD3.

the short of high power pump source and the maximum output power could be scalable with high pump power. On the other hand, HD3, showed a threshold pump power of 8.0 mW and a slope efficiency of 55%, the optical efficiency is as high as 43% when the maximum pump power is 30 mW. Fig. 6 shows the average output power as a function of absorbed pump power under pumping with cw Ti:sapphire laser. HD2 had a threshold pump power of 14.5 mW and a slope efficiency of 47%. HD3 showed a higher threshold pump power of 17.7 mW and a lower slope efficiency of 25%. At the same time, with the increase of the pump power, the output power of HD3 in cw operation becomes flattened, this may be caused by the heating of the crystal. From the laser experimental results (Figs. 5 and 6), we can see that 2 at.% Nd-doped YAG crystal shows more superior laser properties than 3 at.% Nd-doped YAG whenever they are pumped by pulsed or cw Ti:sapphire laser. It may be due to drastic decrease of decay constant in 3 at.% Nd-doped YAG crystal or the length of 3 at.% Nd-doped YAG. It is difficult to infer that the behavior of decay time is caused by whether intrinsic quenching or degradation of the crystal with excessive doping. Although the time constant decreases, it is still longer than that of Nd-doped YVO_4 crystal even for 3 at.% Nd-doped YAG (90 μs for 1 at.% Nd-doped Nd:YVO_4 crystal vs. 120 μs for 3 at.% Nd-doped YAG crystal). To optimize the concentration of Nd ions in YAG crystal or the length of 3 at.% Nd-doped YAG and using high power LD as pump source, high performance Nd:YAG crystal with higher output power and higher efficiency should be realized.

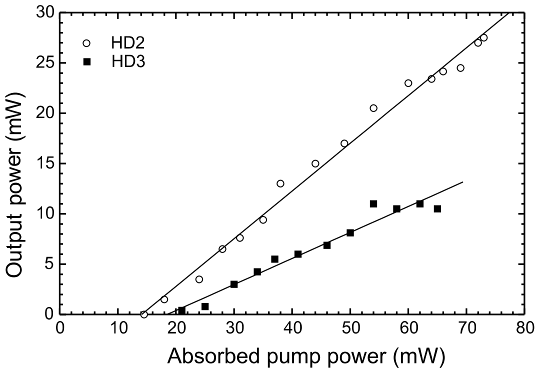

Fig. 6. Output power as a function of absorbed pump power in cw operation.

4. Conclusions

The highly doped Nd:YAG crystals were grown by TGT successfully and the distribution Nd ions along the growth axis is no unity and the highly doped Nd:YAG (>2 at.% Nd) is about a quarter of the whole Nd:YAG crystal, so a lot of Nd:YAG microchip wafers can be cutoff the Nd:YAG crystal. The absorption coefficient and the decay time were measured and showed that although the absorption coefficient of Nd:YAG crystals is lower than that of Nd:YVO_4 crystals (20.9 cm^{-1} for

3 at.% Nd-doped Nd:YAG crystal, 31.2 cm^{-1} for *c*-cut 1 at.% Nd-doped Nd:YVO_4 crystal), the decay time of highly doped Nd:YAG crystals is longer than that of Nd:YVO_4 crystals (120 μs for 3 at.% Nd:YAG crystal, 90 μs for 1 at.% Nd:YVO_4). And laser performance showed that high efficiency can be obtained with highly doped Nd:YAG as gain medium. So using highly doped Nd:YAG crystals as gain medium is superior than that of Nd:YVO_4 crystal owing to good thermal–mechanical properties of Nd:YAG crystals. Although that 3 at.% Nd:YAG is inferior than that of 2 at.% Nd:YAG, through optimization of the length of 3 at.% Nd-doped YAG and using high power pump source, high power and high efficiency should be obtained. And these properties of highly doped Nd:YAG crystals show that they can be used in most useful microchip laser fields and will hold an important position in microchip solid-state laser area.

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