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Optical properties and highly efficient laser oscillation of Nd:YAG ceramics

J. Lu[1,*], **M. Prabhu**[1], **J. Song**[1], **C. Li**[1], **J. Xu**[1], **K. Ueda**[1], **A.A. Kaminskii**[2], **H. Yagi**[3], **T. Yanagitani**[3]

[1] Institute for Laser Science, University of Electro-communications, 1-5-1 Chofugaoka, Chofu, Tokyo 182-8585, Japan
(Fax: +81-424/85-8960, E-mail: lu@ils.uec.ac.jp)
[2] Institute of Crystallography of the Russian Academy of Sciences, Crystal Laser Physics Laboratory, Leninski pr. 59, Moscow 117333, Russia
[3] Takuma Works, Konoshima Chemical Co., Ltd., 80 Kouda, Takuma, Mitoyo-gun, Kagawa 769-11, Japan

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Abstract. Optical absorption, emission spectra have been measured for polycrystalline Nd-doped $Y_3Al_5O_{12}$ ceramics. Fluorescence lifetimes of 257.6 μs, 237.6 μs, 184.2 μs and 95.6 μs have been obtained for 0.6%, 1%, 2% and 4% neodymium-doped YAG ceramics, respectively. For the first time, highly efficient laser oscillation at 1064 nm has been obtained with this kind of ceramics. Slope efficiency of 53% has been achieved on a uncoated 4.8-mm thick 1% Nd:YAG ceramics sample. Optical to optical conversion efficiency is 47.6%. Laser oscillation has also been obtained with a 2% Nd:YAG ceramics. The optical properties and laser output results have been compared with that of Nd:YAG single crystal grown by the Czochralski method. Almost identical results have been achieved including laser experiments results. But fabrication of Nd:YAG ceramics is much easier compared to the single-crystal growth method. And also large size (now of about 400 mm diameter×5 mm is available) and high-concentration (> 1%) Nd:YAG ceramics can be fabricated. The results show that this kind of Nd:YAG ceramics is a very good alternative to Nd:YAG single crystal.

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Nd-doped YAG single crystal fabricated by the Czochralski method has been widely used as solid-state laser material [1–3]. Although this method provides relatively large single crystals for different applications, it still has several disadvantages. For example, an expensive Ir crucible is required for the growth of single crystal, and contamination from it is hard to avoid. And it is also extremely difficult to dope > 1% neodymium homogeneously as a luminescence element in a YAG single crystal because the effective segregation coefficient of elemental neodymium for the host material (the YAG single crystal) is ≈ 0.2 [4, 5]. The growing technique for YAG single crystals requires great skill.

Compared to the growing technique of Nd:YAG single crystals, the fabrication of polycrystalline transparent Nd:YAG ceramics does not require any special technique, large size (now of about 400 mm diameter×5 mm sample is available) and high concentration (> 1%) neodymium-doped samples can be fabricated. And also multi-layer active elements and multi-functional ceramics can be easily fabricated together other than by the single-crystal growth method. These advantages give much more freedom in laser designs. Attempts have been made to make the YAG ceramics become translucent by hot press [6] and a wet chemical method [7–11]. However, the absorption coefficient of the YAG ceramics was larger than 7 cm^{-1}. In the last decade, investigations have been made to obtain high-quality, high-transparency Nd-doped YAG ceramics that can be used as a laser material to compete with Nd:YAG single crystals. In 1990, Sekita et al. developed a urea precipitation method to obtain a translucent or almost transparent YAG ceramics with background absorption coefficients of 2.5 to 3 cm^{-1} [12]. Optical properties of the ceramics were almost identical to those of single crystals grown by the Czochralski method. But laser oscillation could not be obtained because of the large background absorption. In 1995, Ikesue and Kinoshita fabricated high-transparency Nd:YAG ceramics using powders of Al_2O_3, Y_2O_3 and Nd_2O_3 with particles smaller than 2 μm as starting materials [13]. The scattering loss (0.009 cm^{-1}) for this sample was low enough to obtain laser output for the first time. Slope efficiency of 28% was reported.

Recently, the Konoshima company developed Nd:YAG ceramics successfully with a different method [14, 15]. The ceramics formation process and sintering process have been optimized and high-transparency, high-quality Nd:YAG ceramics have been fabricated. The average diameter of grain size is about 10 μm. The transmittance microscope photograph shows that no obvious defects exist in such samples. The pore volume in this kind of ceramic is only 1 ppm level which is about two orders lower than results in [16]. High laser efficiency that is comparable to that of Nd:YAG single crystal has been obtained.

*Corresponding author.

1 Measurement of optical properties

Optical absorption and emission measurements were carried out using a ANDO AQ-6315A optical spectrum analyzer. ANDO AQ-4303B white-light source was used for absorption measurement and a Mitsui MLD100 1-W 807-nm laser diode (LD) was used for fluorescence and laser experiments. The ceramics samples used in these measurements were $\phi 12 \times 2$ mm 0.6%, $\phi 12 \times 2$ mm 1%, $\phi 12 \times 2.5$ mm 2%, $9 \times 22 \times 0.86$ mm 4% Nd:YAG ceramics acquired from Konoshima Inc. $\phi 3 \times 2$ mm 0.9% Nd:YAG single crystal procured from Litton-Airtron Inc. was used for comparison.

The room-temperature absorption spectra of 1% Nd:YAG ceramics and 0.9% Nd:YAG single crystal are shown in Fig. 1. The Fresnel reflections have been subtracted from the absorption spectrum. If the absorption coefficient is assumed to be proportional to the neodymium concentration (it is reasonable in Nd:YAG with neodymium concentration $\leq 1\%$), the absorption coefficient of 0.9% single crystal should be 10% less than that of 1% ceramics. From Fig. 1, we see that the absorption coefficient of 0.9% Nd:YAG single crystal is about 15% less than that of 1% Nd:YAG ceramics. It means the actual neodymium concentration in single crystal may be a little less than 0.9% because it is hard to control the exact concentration in single crystals in the growing process. It is one of the advantages of Nd:YAG ceramics laser material that the rare-earth ions concentration can be precisely controlled. For single crystal and ceramics, the highest absorption peaks are both centered at 808.6 nm, the FWHMs (full width at half maximum) of absorption coefficient are both 1.04 nm. Room-temperature absorption spectra for 0.6%, 2% and 4% Nd:YAG ceramics also have been measured. No much difference has been observed except that the absorption coefficient increases with increase in neodymium concentration. Figure 2 shows the peak absorption coefficient of Nd:YAG ceramics around 808.6 nm versus neodymium concentration. From this figure, one can see the relationship between peak absorption coefficient and neodymium concentration is almost linear. For 0.6%, 1% and 2% samples, the experimental data are a little above the curve fitted line, for the 4% sample, the experimental datum is a little below the curve fitted line. It means the absorption coefficient decreases a little off the linear curve because of the interaction between ion to ion becomes stronger at high concentration.

Figure 3 shows the room-temperature fluorescence spectrum for ${}^4F_{3/2}$ to ${}^4I_{11/2}$ transition of 1% Nd:YAG single crystal and ceramics, respectively. In order to have a clear comparison, the fluorescence spectra for single crystal and ceramics were normalized and put together. From this figure, these two spectra are almost identical. The main emission

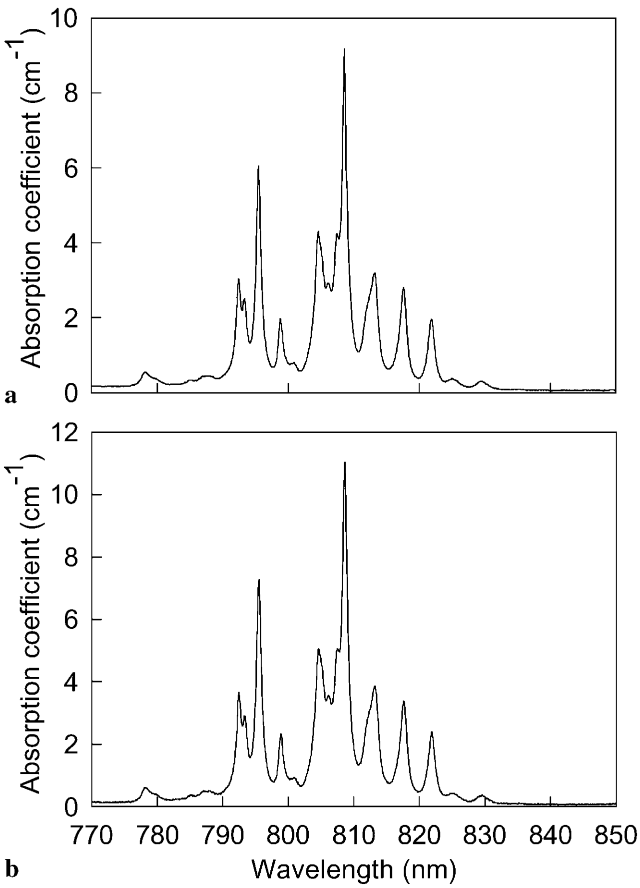

Fig. 1a,b. Absorption spectrum from 770 nm to 850 nm; **a** 0.9% Nd:YAG single crystal, **b** 1% Nd:YAG ceramic

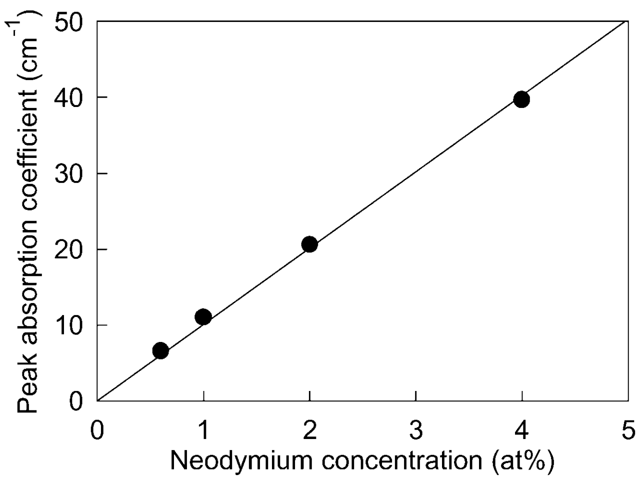

Fig. 2. Peak absorption coefficient around 808 nm ceramics versus neodymium concentration of Nd:YAG

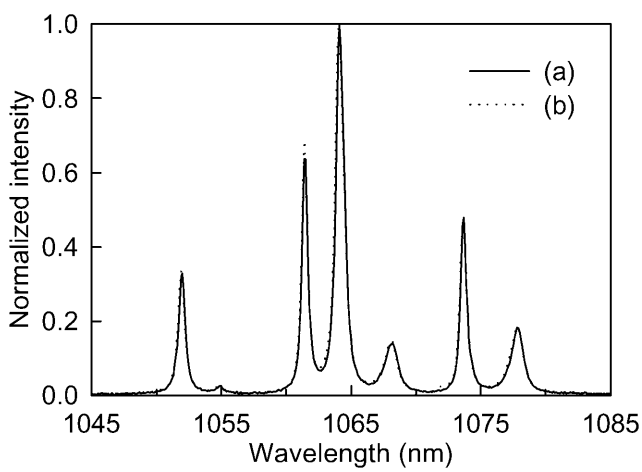

Fig. 3a,b. Fluorescence spectrum from 1045 nm to 1085 nm; **a** 1% Nd:YAG ceramic, **b** 0.9% Nd:YAG single crystal peak is at 1064.18 nm. FWHM is 0.78 nm. Emission spectrum for 0.6%, 2% and 4% Nd:YAG ceramics have also been measured. A little wavelength redshift has been observed with increasing the concentration because of a little bit change in the crystal field. Figure 4a shows the main fluorescence peak spectra near 1064 nm for 0.6%, 1%, 2% and 4% Nd:YAG ceramics, respectively. The four emission peaks are centered at 1064.15 nm, 1064.18 nm, 1064.24 nm and 1064.30 nm for 0.6%, 1%, 2% and 4% Nd:YAG ceramics, respectively. The redshift from 0.6% to 4% Nd:YAG ceramics is 0.12 nm. Because of the fluorescent quenching effect, the fluorescence emission line width at 1064 nm is also a little broadened with the concentration increases greater than 1%. Figure 4b shows the FWHM of the 1064-nm fluorescence peak. The FWHMs are 0.78 nm, 0.78 nm, 0.81 nm and 0.85 nm for 0.6%, 1%, 2% and 4% Nd:YAG ceramics, respectively. The line widths for 0.6% and 1% Nd:YAG ceramics are identical. It means that the fluorescent quenching effect is very weak for neodymium concentration less than 1%, which is similar to that of single crystal.

Using a quasi-cw LD produced by Hamamatsu Inc., the fluorescence lifetime for single crystal and ceramics have been obtained through curve fitting on the fluorescence decay curve. Figure 5 shows the fluorescence lifetime of Nd:YAG ceramics and single crystal versus neodymium concentration. The fluorescence lifetime for 0.6% Nd:YAG single crystal (also procured from Litton-Airtron Inc.) and 0.9% Nd:YAG single crystal are 256.3 μs and 248.6 μs, respectively, which agrees well with the earlier reports [17]. Fluorescence lifetimes of 257.6 μs, 237.6 μs, 184.2 μs and 95.6 μs have been measured, respectively, for 0.6%, 1%, 2% and 4% Nd:YAG ceramics. These data also agree well with the results in [17]. The fluorescence lifetime decreases dramatically when neodymium concentration exceeds 1%. The fluorescence lifetimes for 0.6% doped single crystal and ceramics are almost identical (only 1.3 μs difference). The fluorescence lifetime difference between 0.9% Nd:YAG single crystal and 1% Nd:YAG ceramics is 11 μs. We can predict that for the same concentration of Nd:YAG single crystal and ceramics, for example, 0.9% concentration, the lifetime difference should be less than 11 μs. From the fitted curve for ceramics fluorescence lifetime, the lifetime for 0.9% Nd:YAG ceramics is 244.2 μs, which is only 4.4 μs different from that of 0.9% Nd:YAG single crystal. Here we assume the neodymium ions inside the grain have the same conditions as those of single crystal, and we propose that the fluorescence lifetime difference is caused only by the neodymium ions in the vicinity of grain boundaries. Then, it means whatever distorted layers or grain boundary phase exist in the vicinity of grain boundaries have little influence on fluorescence lifetime in such ceramics samples.

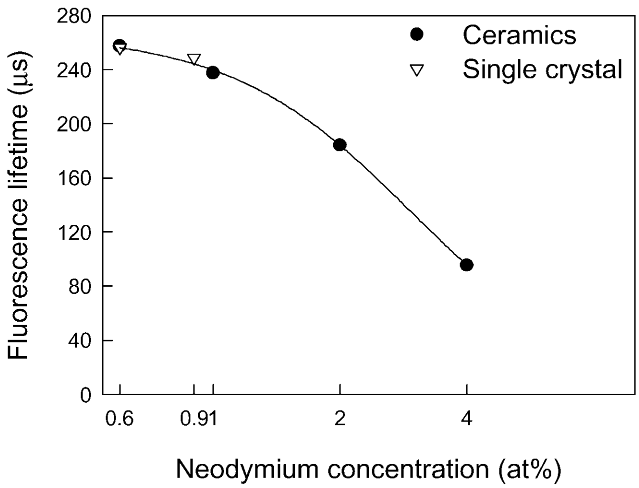

Fig. 5. Fluorescence lifetime of Nd:YAG ceramics and single crystal versus neodymium concentration. *Solid line* is the *fitted curve* for ceramic fluorescence lifetime

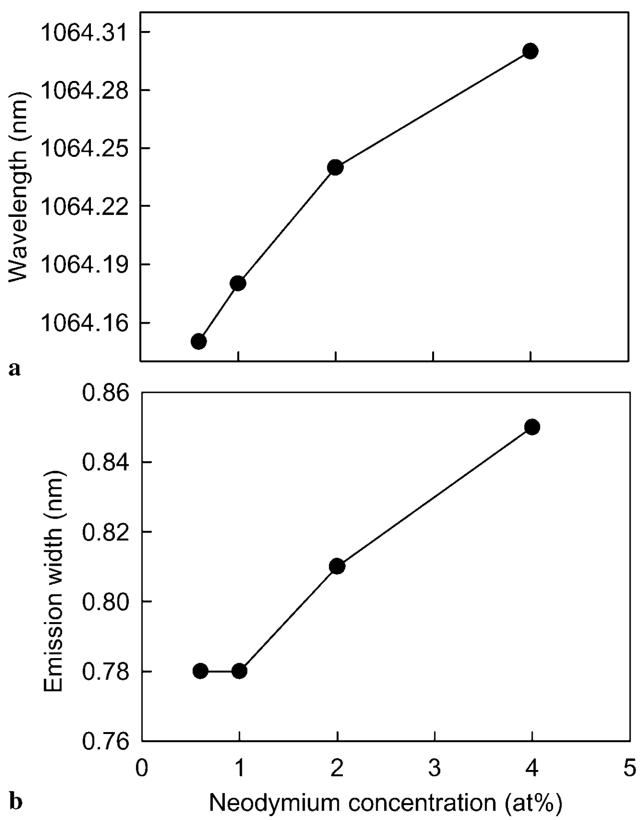

Fig. 4. a Fluorescence redshift versus neodymium concentration. **b** FWHM at 1064 nm versus neodymium concentration

2 Laser experiment

Laser experiments have been done on 0.6%, 1% and 2% and Nd:YAG ceramics samples without any coatings on either of the ends. Laser experiments have not been done with uncoated 4% Nd:YAG ceramics because of poor polishing quality. Such high-concentration Nd:YAG ceramics was fabricated for microchip lasers. Later, such 4% ceramics will be polished and coated with HR and AR coatings at 1064 nm to generate single-frequency oscillation. Laser output has been obtained for 0.6% Nd:YAG ceramics, but since the sample's thickness is only 2 mm, most of the pump power has not been absorbed by the sample and resulted in a very low slope efficiency. This kind of low-concentration sample was fabricated for high-power pump lasers, later we will test this kind of low-concentration samples with a high-power laser system. Here we only report the laser experiment results for 4.8-mm-thick 1% and 2.5-mm-thick 2% Nd:YAG ceramics.

A 5-mm-thick 0.9% Nd:YAG single crystal procured from Litton-Airtron Inc. was used for comparison with ceramics. Though the concentrations are different for single crystal (0.9%) and ceramics (1% and 2%), since the samples' thickness is long enough, the absorbed pump power is almost the same.

Figure 6 shows the schematic diagram of the laser experimental setup. A 1-W Hamamatsu 2901 LD with 50-μm emission profiles was used as a pump source. Input mirror is a flat high-reflection mirror at 1064 nm. Output mirror is a concave mirror with 250-mm radius and the reflectivity is 95% at 1064 nm. When the LD output is 1 W, about 750 mW pump power can be focused on the end of sample. The cavity length is about 20 mm.

Figure 7 shows the laser output versus pump power for 0.9% Nd:YAG single crystal, 1% and 2% Nd:YAG ceramics at 1064 nm. The threshold for 1% Nd:YAG ceramics is 65 mW which is only 15 mW higher than that of 0.9% Nd:YAG single crystal (50 mW) and the slope efficiency is 53% which is only 1.5% lower than that of single crystal (54.5%). The optical-to-optical conversion efficiencies for 0.9% Nd:YAG single crystal and 1% Nd:YAG ceramics are 49.3% and 47.6%, respectively. These are the best laser results obtained from 1% Nd:YAG ceramics to date. For the laser result on 2% Nd:YAG ceramics, the threshold is 104 mW which is larger than that of single crystal and 1% Nd:YAG ceramics. The slope efficiency is 34% which is much less than that of single crystal and 1% ceramics. The main reason may be due to the quenching effect in high-concentration Nd:YAG ceramics. The fluorescence lifetimes for 1% and 2% Nd:YAG ceramics are 237.6 μs and 184.2 μs, respectively. The difference in lifetime of 53.4 μs shows that obvious quenching effect had occurred. But high-concentration Nd:YAG ceramics have much higher absorption coefficients which in turn reduce the required length of laser materials. High-concentration ceramic laser materials can be polished into microchips and used to efficiently generate single-frequency laser output which may compete with microchip Nd:YVO4 single-frequency lasers. *This is our next step work.*

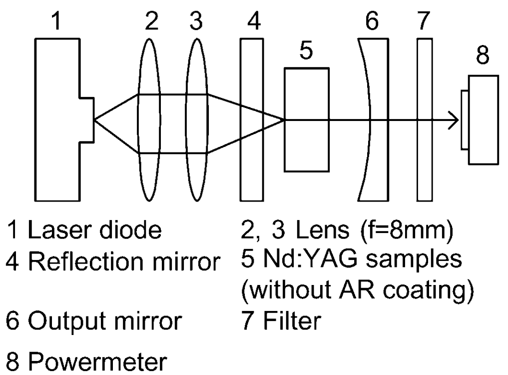

Fig. 6. Schematic diagram of the laser experimental setup

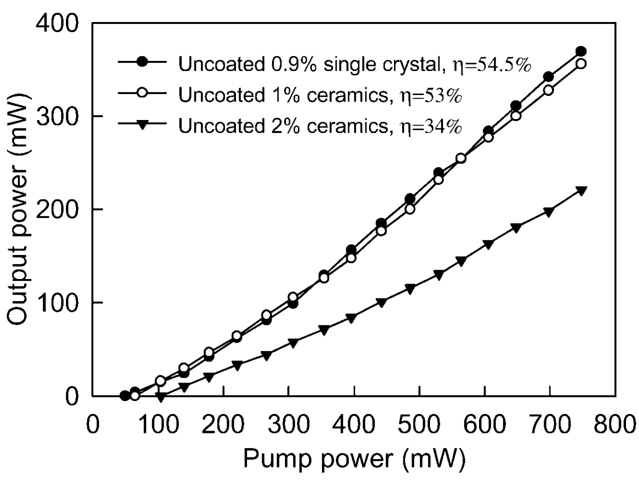

Fig. 7. Laser output at 1064 nm versus pump power

The laser results show that the quality of such kind of ceramics is good enough to be used as a highly efficient laser material. If ceramic samples are coated with antireflection coatings to reduce the cavity loss, more than 60% slope efficiency for 1% Nd:YAG ceramics and more than 40% slope efficiency for 2% Nd:YAG ceramics are predicted. The corresponding laser threshold will be much lower than those of uncoated ceramic lasers too.

The most important thing concerned in ceramics is the scattering loss caused by grain boundaries. According to the laser experimental result for 0.9% Nd:YAG single crystal and 1% Nd:YAG ceramics, it implies that the scattering loss at 1064-nm lasing wavelength in ceramics should be similar to that of single crystals. Since the grain boundary width (only sub-nanometer level) is much smaller than the lasing wavelength, the scattering intensity can be determined by the Rayleigh equation [18]. According to the Rayleigh equation, the scattering intensity is proportional to d^6/λ^4, where d and λ are radius of the scattering body and the measuring wavelength. From such relationship between scattering intensity and scattering body size, it can be predicted that the size of scattering center near Nd:YAG ceramics grain boundaries is smaller enough than 1 μm, so although the lasing light in a laser cavity travels tens of thousand times through grain boundaries, the total scattering loss caused by the grain boundaries is still very low. The detailed mechanism between the Nd:YAG ceramics microstructure and scattering loss will be studied later.

3 Conclusions

Optical absorption, emission spectra and fluorescence lifetime for Nd:YAG ceramics have been measured and compared to that of single crystal. Almost identical results have been obtained. Optical properties have also been investigated for different concentration Nd:YAG ceramics samples. The same variation as that of single crystal has been observed.

Laser experiment results show that the 1% Nd:YAG ceramics has almost the same laser efficiency as that of single crystal. It means the scattering loss at the lasing wavelength of 1.064 μm is similar to that of single crystal. High slope efficiency (53%) and low threshold (65 mW) have been obtained for an uncoated 4.8-mm thick 1% Nd:YAG ceramics which is the best laser oscillation result on ceramics laser materials to date. Apart from having the same level laser lasing efficiencies, fabrication of Nd:YAG ceramics is much easier compared to the single-crystal growth method, and also large-size and high-concentration samples can be fabricated easily.

This kind of Nd:YAG ceramics is a very good alternative to Nd:YAG single crystals.

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