

Spectroscopic Characteristics and Laser Performance of Nd:Y_{1.8}La_{0.2}O₃ Transparent Ceramics

Shenzhou Lu, Qihong Yang, Haojia Zhang, Yonggang Wang, Dongdong Huang, Qing Wang, and Zhiyi Wei

Abstract—(Nd_xY_{0.9-x}La_{0.1})₂O₃ ($x = 0.005\text{--}0.04$) transparent ceramics were fabricated by conventional solid-state processing. The radiative spectral properties of the ceramic samples were evaluated by fitting the Judd–Ofelt model with the absorption and emission data. (Nd_xY_{0.9-x}La_{0.1})₂O₃ ceramics have broad absorption and emission bands with a radiative decay time of 328 μs . The absorption cross section at 806 nm and stimulated emission cross section at 1078 nm are calculated to be 1.53×10^{-20} and 5.22×10^{-20} cm^2 , respectively. The product of quantum efficiency and the ionic concentration (ηN) exhibited a peak value at 1.5 at% Nd³⁺ ion concentration, while the lifetime decreases dramatically from 300 μs (0.5 at% Nd) to 49 μs (4.0 at% Nd). With 1.0 at% Nd:Y_{1.8}La_{0.2}O₃ ceramics acting as a laser medium, continuous-wave output power of 1.03 W was obtained at 1079.5 nm under an absorbed pump power of 7.2 W, corresponding to a slope efficiency of 18.4%.

Index Terms—Emission cross section, laser oscillation, Nd:Y_{1.8}La_{0.2}O₃, quantum efficiency, transparent ceramics.

I. INTRODUCTION

IN RECENT years, Nd³⁺-doped solid state laser materials have attracted much attention. The Nd³⁺-doped solid-state laser hosts with four-level system usually have large stimulated emission cross section, low laser output threshold, and strong absorption peak at 808 nm, which matches the emission wavelength of commercial laser diode (LD). Among these materials, the cubic Y₂O₃ is an ideal laser host because it has good optical, thermal, and chemical properties [1]–[3]. In particular, Y₂O₃ possesses a high thermal conductivity of 13.6 W/mK, which is larger than that of YAG [4]. However, the growth of high-quality Y₂O₃ single crystals is a challenging task because of its high melting point (2430 °C) and structural phase transition from cubic to hexagonal phase at ~ 2280 °C [5].

Many recent studies have focused on polycrystalline ceramic lasers since the first report of laser oscillation in

Nd:YAG transparent ceramics in 1995 [6]. Compared with single crystals, ceramic laser materials have many advantages such as ease of fabrication, low cost, high doping concentration and mass production in a large size. The first laser oscillation in Nd:Y₂O₃ ceramic materials was reported in 1973 [7], and the laser material is Nd³⁺ doped Y₂O₃-ThO₂ (NDY) solid-solution transparent ceramics. However, all researches of this material ground to a halt due to the radiological hazard of ThO₂. Since 2000, the optical quality of Y₂O₃ transparent ceramics has improved greatly and is good enough to obtain laser oscillations. In 2001, 1.5 at% Nd:Y₂O₃ transparent ceramics fabricated by nanocrystalline and vacuum sintering technology realized laser output under LD pumping, a maximum output power of 160 mW with a slope efficiency of 32% was obtained [8]. Nowadays, Nd:Y₂O₃ transparent ceramics can be fabricated at a relatively low sintering temperature of 1700 °C, which is about 700 °C lower than its melting point.

Our previous work shows that the sintering temperature of Y₂O₃ transparent ceramics could be further decreased by adding La₂O₃ as an additive [9]. Y_{2-2x}La_{2x}O₃ is a solid solution of Y₂O₃ and La₂O₃. It has cubic glass-like structure, which make it possible to have broad absorption bandwidth [10]. This is of great benefit to lower dependency on temperature control of laser device. In 2009, 1.5 at% Nd:Y_{1.8}La_{0.2}O₃ transparent ceramics fabricated by solid-state reaction method realized laser output, a maximum output power of 62 mW was obtained under diode pumping [11].

In this work, (Nd_xY_{0.9-x}La_{0.1})₂O₃ ($x = 0.005\text{--}0.04$) transparent ceramics were fabricated by conventional solid-state processing. The effects of Nd³⁺ concentration on the spectroscopic properties were investigated systematically, and a diode pumped CW laser oscillation with a maximum output power of 1.03 W was obtained in 1.0 at% Nd:Y_{1.8}La_{0.2}O₃ ceramic laser.

II. EXPERIMENT DETAILS

The starting materials Y₂O₃, La₂O₃ and Nd₂O₃ nanopowders with purity of 99.99% were weighted according to the desired composition of (Nd_xY_{0.9-x}La_{0.1})₂O₃ ($x = 0.005\text{--}0.04$), and then mixed with ZrO₂ balls in anhydrous alcohol for 5–10 h. The mixed powders were dried and calcined at 1100–1200 °C for 10 h, followed by 5–10 h ball-milling in anhydrous alcohol, dried and sieved. Finally disks with 23 mm in diameter and 6–8 mm in thickness were

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isostatically pressed at 200 MPa and sintered at 1650–1680 °C for 40–50 h in H₂ atmosphere.

Disc ceramic samples were double mirror polished for spectroscopic analysis. Optical microscopy (Model BX60, OLYPMUS, Japan) was used to observe the microstructures. The transmittance and absorption spectra in the UV-VIS-NIR were measured with a spectrophotometer (Model V-570, JASCO, Japan) using Xe light as pump source. The fluorescence spectra and fluorescence lifetime excited with 808 nm LD were measured with a fluorescence spectrum analyzer (Fluorolog-3, Jobin Yvon Spex, France). All spectroscopic analysis were made at room temperature.

III. THEORETICAL METHODS

The spectroscopic properties of Nd³⁺ in yttrium lanthanum oxide transparent ceramics were investigated by Judd–Ofelt (J-O) theory. In the J-O theory, the oscillator strength of an electronic transition of average frequency ν from a level aJ to another level bJ' is given by [12], [13]

$$f_{\text{cal}} = \frac{8\pi^2 m \nu}{3(2J+1)h e^2 n^2} \times [\chi_{\text{ed}} S_{\text{ed}}(aJ, bJ') + \chi_{\text{md}} S_{\text{md}}(aJ, bJ')]. \quad (1)$$

Here h is the Planck's constant, J is the total angular momentum of the initial state, e and m are the charge and the mass of the electron, respectively. The factors χ_{ed} and χ_{md} are the terms for local field corrections, which are functions of the medium refractive index n : $\chi_{\text{ed}} = n(n^2 + 2)^2/9$ and $\chi_{\text{md}} = n^3$. S_{ed} and S_{md} represent the line strengths for the induced electric and magnetic dipole transition, respectively. They are expressed by [14]

$$S_{\text{ed}}(aJ, bJ') = e^2 \sum_{t=2,4,6} \Omega_t \left| \langle aJ \| U^{(t)} \| bJ' \rangle \right|^2 \quad (2)$$

$$S_{\text{md}}(aJ, bJ') = \left(\frac{e^2 h^2}{16\pi^2 m^2 c^2} \right) \left| \langle aJ \| L + 2S \| bJ' \rangle \right|^2 \quad (3)$$

where $\langle \| U^{(t)} \| \rangle$ are the doubly reduced matrix elements of the unit tensor operator of rank $t = 2, 4$ and 6 , which are calculated from the intermediate coupling approximation. $\langle \| L + 2S \| \rangle$ are the magnetic dipole operator matrix elements.

The Ω_t parameters ($t = 2, 4$ and 6), known as the J-O parameters, are determined by a least-squares fit of the theoretical oscillator strengths (f_{cal}) to the values of experimentally measured oscillator strengths (f_{exp}) calculated from optical absorption spectrum using the equation [15]

$$f_{\text{exp}} = \frac{m c^2}{\pi e^2 N} \int \frac{\alpha(\lambda)}{\lambda^2} d\lambda \quad (4)$$

where c is the velocity of light in vacuum, N is the number density of Nd³⁺ ions. $\alpha(\lambda) = 2.303 D_0(\lambda)/d$ is the measured absorption coefficient at a given wavelength λ , and $D_0(\lambda)$ is the optical density ($\log I_0/I$), λ is the mean wavelength of the specific absorption band, d is the thickness of the sample.

The Ω_t parameters were further used to evaluate the radiative transition rates (A_{rad}) from a particular emitting level



Fig. 1. Photograph of (Nd_xY_{0.9-x}La_{0.1})₂O₃ transparent ceramics. From left to right, $x = 0.005, 0.01, 0.015, 0.02, 0.025,$ and 0.04 , respectively.

according to the equation [16]

$$A_{\text{rad}}(aJ, bJ') = \frac{64\pi^4}{3h(2J+1)\lambda^3} \times \left[\frac{(n^2+2)^2}{9} S_{\text{ed}} + n^3 S_{\text{md}} \right]. \quad (5)$$

The radiative lifetime (τ_{rad}) of an emitting state and the emission branching ratio ($\beta_{JJ''}$) of a transition can be defined as:

$$\tau_{\text{rad}}(J) = \frac{1}{\sum_{J'} A_{\text{rad}}(aJ, bJ')} \quad (6)$$

$$\beta_{JJ''} = \frac{A(aJ, b''J'')}{\sum_{J'} A(aJ, b'J')}. \quad (7)$$

IV. RESULTS AND DISCUSSION

A. Physical Properties and Absorption Spectra

Fig. 1 is the photograph of (Nd_xY_{0.9-x}La_{0.1})₂O₃ ($x = 0.005-0.04$) transparent ceramics. Each pellet is 18 mm in diameter and ~ 3 mm in thickness. All the samples are completely transparent and the letters behind the ceramics can be seen distinctly.

Fig. 2(a) shows the transmittance spectrum of Nd:Y_{1.8}La_{0.2}O₃ transparent ceramics. The transmittance of 3 mm thick sample is 80.0% at the lasing wavelength of 1078 nm, which is close to the theoretic transmittance value of Y₂O₃ at this wavelength. Fig. 2(b) is the optical micrograph of the polished surface after etched in HCl. It reveals that the sample has uniform grains with average size of 20–30 μm and there are almost no pores or impurities between the grain boundaries.

Fig. 3(a) is the room-temperature absorption spectrum of 1 at% Nd:Y_{1.8}La_{0.2}O₃ transparent ceramics. The absorption band suited for LD pumping is in the wavelength range from 780 to 850 nm corresponding to the $^4I_{9/2} \rightarrow ^4F_{5/2} + ^2H_{9/2}$ transition. Similar to Nd:Y₂O₃ ceramics [17], the strongest peak in this range is centered at 820 nm, of which the absorption coefficient is 4.27 cm⁻¹ and the full width at half-maximum (FWHM) is 4.0 nm. The FWHM of Nd:Y_{1.8}La_{0.2}O₃ ceramics at LD pump wavelength 806 nm is about 7.8 nm, which is two times that of Nd:Y₂O₃ ceramics (4 nm), and

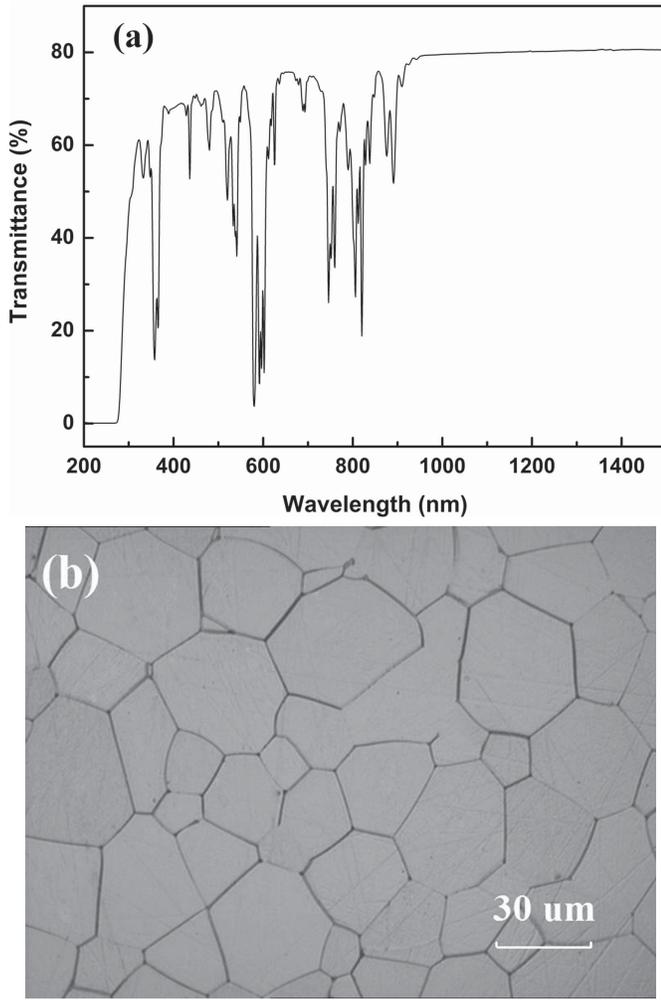


Fig. 2. (a) Transmittance spectrum and (b) optical micrograph of (Nd_xY_{0.9-x}La_{0.1})₂O₃ transparent ceramics.

nearly eight times that of Nd:YAG ceramics (~1 nm) [18]. It may be induced by a disorder glass-like structure of Y_{1.8}La_{0.2}O₃ transparent ceramics. On the other hand, as Nd³⁺ ion (98 pm) can substitute both the lattice sites of La³⁺ (103 pm) and Y³⁺ (89 pm) ions, two different groups of optical centers are present, and the absorption peaks are further inhomogeneously broadened [10]. Broad absorption bands of Nd:Y_{1.8}La_{0.2}O₃ ceramics are advantageous for lower dependency on pump wavelength and temperature control of laser device. The absorption cross section (σ_{abs}) of Nd³⁺ at wavelength λ can be calculated as follow:

$$\sigma_{\text{abs}} = \frac{2.303 \log(I_0/I)}{L \cdot N} \quad (8)$$

where $\log(I_0/I)$ is optical density, N is numbers of active ions in unit volume and L is thickness of the specimen. The calculated absorption cross sections at 806 and 820 nm are 1.53×10^{-20} and 1.96×10^{-20} cm², respectively. With increasing Nd³⁺ concentration, the positions of all spectral bands remain practically the same, whereas the absorption coefficient varies linearly with the concentration, as shown in Fig. 3(b).

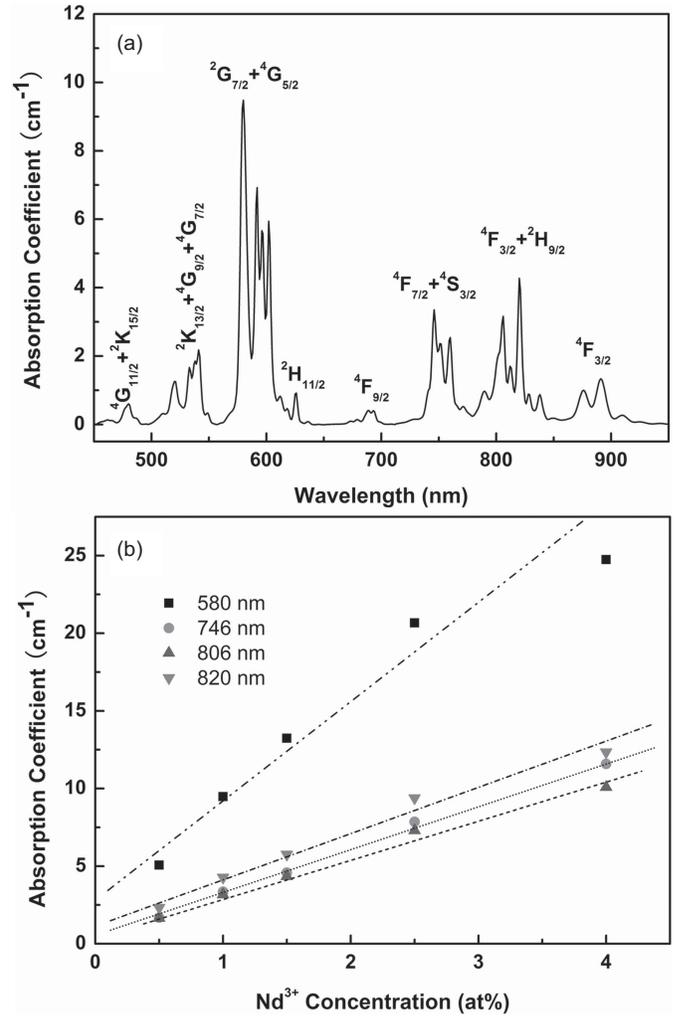


Fig. 3. (a) Room-temperature absorption spectrum of 1 at% (Nd_xY_{0.9-x}La_{0.1})₂O₃ transparent ceramic and (b) dependence of absorption coefficient at 580, 746, 806, and 820 nm from Nd³⁺ concentration.

B. Radiative Properties and Stimulated Emission Characteristics

The theoretical (f_{cal}) and experimental (f_{esp}) oscillator strength of the transitions were calculated by using equation (1) and (4), respectively. The results are given in Table I. Three intensity parameters Ω_t ($t = 2, 4, 6$) can be calculated from the measured values of oscillator strength for different transitions by using the least square fitting procedure. Theoretically, Ω_2 is strongly dependent on the local environments of rare earth ions sites and proportional to the covalence of Nd³⁺-O²⁻. The calculated results show that three intensity parameters (Ω_2 , Ω_4 , Ω_6) in our fabricated ceramics are 3.79×10^{-20} , 2.98×10^{-20} , and 2.24×10^{-20} cm², respectively. The Ω_2 value of Nd:Y_{1.8}La_{0.2}O₃ ceramics is smaller than that of Nd:Y₂O₃ ceramics ($\Omega_2 = 4.09 \times 10^{-20}$ cm²) [17], which indicates that Nd:Y_{1.8}La_{0.2}O₃ ceramics possesses stronger covalence than Nd:Y₂O₃ ceramics. Additionally, the root-mean-square error deviation (δ) of the intensity parameters is 0.11×10^{-6} , indicating the validity of the Judd-Ofelt theory for predicting the spectral intensities and the reliable calculations.

TABLE I
OBSERVED ABSORPTION BANDS WITH THE EXPERIMENTAL AND
CALCULATED OSCILLATOR STRENGTHS IN 1 at%
Nd:Y_{1.8}La_{0.2}O₃ TRANSPARENT CERAMIC

Excited States (Ground State ⁴ I _{9/2})	Wavelength (nm)	Oscillator Strength ($\times 10^{-6}$)	
		f_{exp}	f_{cal}
⁴ G _{11/2} + ² K _{15/2}	461.5	0.305	0.373
² G _{9/2} + (² D, ² P) _{3/2}	479	0.82	0.599
² K _{13/2} + ⁴ G _{9/2} + ⁴ G _{7/2}	532.5	4.958	4.715
² G _{7/2} + ⁴ G _{5/2}	589.9	20.123	20.134
² H _{11/2}	625.5	0.365	0.107
⁴ F _{9/2}	687.3	0.537	0.383
⁴ F _{7/2} + ⁴ S _{3/2}	752.3	4.342	4.546
⁴ F _{5/2} + ² H _{9/2}	812.3	5.209	4.996
⁴ F _{3/2}	887.2	1.743	1.924

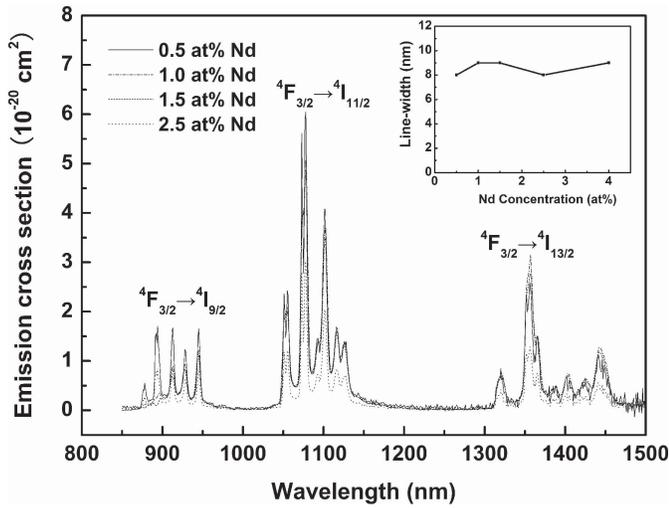


Fig. 4. Emission spectra of (Nd_xY_{0.9-x}La_{0.1})₂O₃ ($x = 0.005-0.04$) transparent ceramics at room temperature. Inset: dependence of Nd³⁺ concentration versus line-width of the emission band centered at 1076 nm.

Fig. 4 shows the room-temperature three-band emission spectra of Nd³⁺ ions (⁴F_{3/2} → ⁴I_J, J = 9/2–13/2) in (Nd_xY_{0.9-x}La_{0.1})₂O₃ ($x = 0.005-0.04$) ceramic. The inset is the dependence of Nd³⁺ concentration versus line-width of the emission bands centered at 1076 nm. All spectra present the same shape and the intensity of these emission bands decreases with concentration due to the quenching effect of Nd³⁺ ions. The strongest emission peak is at 1078 nm, corresponding to the ⁴F_{3/2} → ⁴I_{11/2} transition of Nd³⁺, with the FWHM of 5.3 nm, which is wider than that of Nd:Y₂O₃ (2.63 nm) [17] and Nd:YAG (~0.7 nm @ 1064 nm) [18]. Moreover, the broadening of the spectrum makes the two peaks centered at 1074 and 1078 nm to overlap and form a wider composite peak with FWHM of ~9 nm (As shown in Fig. 5). So compared with Nd:Y₂O₃ ceramics, Nd:Y_{1.8}La_{0.2}O₃ ceramics are more likely to achieve widely tunable and mode-locked pulse laser.

The stimulated cross sections σ_e of the ceramics is determined by the equation below

$$\sigma_e(\lambda) = \frac{1}{8\pi n^2 c} \frac{1}{\tau_{\text{rad}}} \frac{\lambda^5 I(\lambda)}{\int \lambda I(\lambda) d\lambda} \quad (9)$$

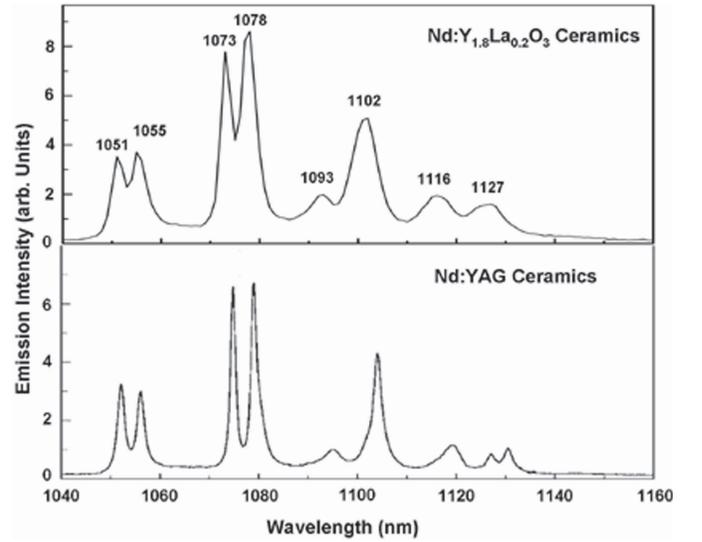


Fig. 5. Emission spectra of Nd³⁺ in Y_{1.8}La_{0.2}O₃ and YAG ceramics for the ⁴F_{3/2} → ⁴I_{11/2} transition at room temperature.

where the $I(\lambda)$ is the intensity of emission spectrum, n is refractive index of the material and c is light velocity. The stimulated emission cross section of the main laser transition is calculated to be $5.22 \times 10^{-20} \text{ cm}^2$ at 1078 nm. This value is smaller than the reported values of 7.63×10^{-20} and $6.35 \times 10^{-20} \text{ cm}^2$ at 1074.6 and 1078.6 nm wavelengths in 1.5 at.% Nd:Y₂O₃ ceramics, respectively [17].

Three intensity parameters (Ω_t) can be substituted in formula (5) to get the radiative transition probabilities of metastable level ⁴F_{3/2}. The calculated line strength (S_{ed}), transition probability (A), radiative decay time (τ_{rad}), and fluorescence branching ratio (β) of Nd:Y_{1.8}La_{0.2}O₃ ceramics are summarized in Table II along with the corresponding data of Nd:Y₂O₃ ceramics [17] and Nd:Y₂O₃ single crystal [19]. A radiative decay time of 328 μs is obtained for Nd:Y_{1.8}La_{0.2}O₃ ceramics, which is 7.5% higher than that for single crystal. In addition, for Nd:Y₂O₃ ceramics, the branching ratio $\beta_{J,11/2}$ of ⁴F_{3/2} → ⁴I_{11/2} intermanifold transition is about 1.2 times larger than $\beta_{J,9/2}$ of ⁴F_{3/2} → ⁴I_{9/2} channel. But for Nd:Y_{1.8}La_{0.2}O₃ ceramics, the value of $\beta_{J,9/2}$ is nearly equal to that of $\beta_{J,11/2}$. This may be induced by the radius difference between La³⁺ ($R = 1.03 \text{ \AA}$) and Y³⁺ ($R = 0.89 \text{ \AA}$), which will cause the distortion of lattice and the change of the crystal field. As a result, the $\beta_{J,9/2}$ of ⁴F_{3/2} → ⁴I_{9/2} channel in Nd:Y_{1.8}La_{0.2}O₃ ceramics becomes large after adding La₂O₃ as an additive.

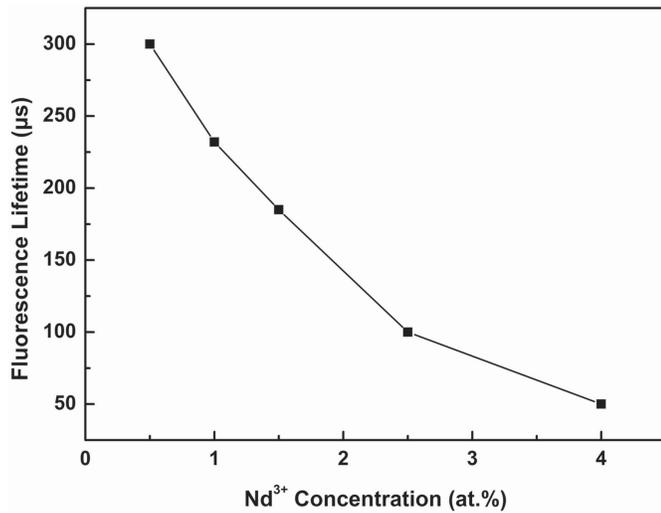
Table III summarizes the calculated results of fluorescence branching ratio (β), quantum efficiency (η), and nonradiative decay rates (W_{nr}) of the ⁴F_{3/2} → ⁴I_J (J = 9/2, 11/2, 13/2) emission transition in Nd:Y_{1.8}La_{0.2}O₃ ceramics. The W_{nr} values were obtained by subtracting the radiative decay rate from the fluorescence decay rate ($1/\tau_s - 1/\tau_r$). For 1.0 at% Nd³⁺-doped Y_{1.8}La_{0.2}O₃ ceramics, a decay time of 232 μs was obtained, which is very close to that of Nd:Y₂O₃ ceramics [17] and 11% shorter than that for Nd:Y₂O₃ single crystal [20]. In order to estimate the fluorescence quenching

TABLE II
 CALCULATED RADIATIVE TRANSITION CHARACTERISTICS OF Nd:Y_{1.8}La_{0.2}O₃ CERAMIC, Nd:Y₂O₃ CERAMIC, AND Nd:Y₂O₃ SINGLE CRYSTAL

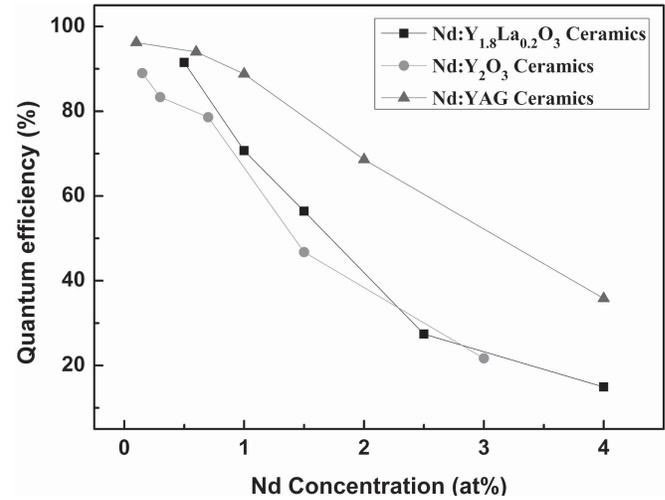
Sample	S _{ed} (×10 ⁻²⁰ cm ²)			A (s ⁻¹)			β (%)			τ _{rad} (μs)	Ref.
	⁴ I _{9/2}	⁴ I _{11/2}	⁴ I _{13/2}	⁴ I _{9/2}	⁴ I _{11/2}	⁴ I _{13/2}	⁴ I _{9/2}	⁴ I _{11/2}	⁴ I _{13/2}		
Nd:Y _{1.8} La _{0.2} O ₃ Ceramics	0.811	1.335	0.475	1418	1412	229	46.36	46.14	7.5	328	This work
Nd:Y ₂ O ₃ Ceramics	1.359	2.99	1.229	1125	1406	282	39.82	49.7	9.9	354	[17]
Nd:Y ₂ O ₃ Single crystal	1.36	1.92	0.61	1703	1358	212	–	–	–	304	[19]

 TABLE III
 FLUORESCENCE BRANCHING RATIO (β), QUANTUM EFFICIENCY (η), AND NONRADIATIVE DECAY RATES (W_{nr}) OF THE ⁴F_{3/2} → ⁴I_J (J = 9/2, 11/2, 13/2) EMISSION TRANSITION

Concentration (at%)	β _{9/2} (%)	β _{11/2} (%)	β _{13/2} (%)	τ _s (μs)		η (%)	W _{nr} (s ⁻¹)
				Nd:Y _{1.8} La _{0.2} O ₃ Ceramics	Nd:Y ₂ O ₃ Ceramics		
0.5	44.1	47.2	8.7	300	–	91.5	285
1.0	45.5	47	7.5	232	221	70.7	1262
1.5	45.4	46.5	8.1	185	175	56.4	2357
2.5	45.5	46.2	8.2	90	–	27.4	6951
4.0	45.2	46.6	8.2	49	–	14.9	17359
Ref.	This work			This work	[17]	This work	This work


 Fig. 6. Concentration dependence of decay time of Nd³⁺ ion in Y_{1.8}La_{0.2}O₃ ceramics.

process in Nd:Y_{1.8}La_{0.2}O₃ ceramics quantitatively, the decay times of ⁴F_{3/2} emission band for different Nd³⁺ concentrations were also measured. Fig. 6 shows the dependence of the decay time of Nd³⁺ ions in Y_{1.8}La_{0.2}O₃ ceramics for various concentrations. With the concentration of Nd³⁺ ion increasing from 0.5 to 4.0 at%, the decay time of Y_{1.8}La_{0.2}O₃ ceramics is decreased dramatically from 300 μs to ~50 μs. This can be ascribed to concentration quenching of Nd³⁺ ions. The very rich energy levels of Nd³⁺ ion enables a variety of energy transitions inside the system of Nd, which would induce a self-quenching of emission from the metastable energy level ⁴F_{3/2}. Especially when Nd³⁺ ions concentration is increased,


 Fig. 7. Dependence of the quantum efficiency (η) on Nd³⁺ concentration (N) for Nd³⁺-doped transparent ceramics.

the length between two adjacent Nd³⁺ ions is decreased and the transfer becomes easier.

Fig. 7 is the dependence of the quantum efficiency (η) on Nd³⁺ concentration (N) for Nd:Y_{1.8}La_{0.2}O₃, Nd:Y₂O₃ [17] and Nd:YAG ceramics [20]. As shown in Fig. 7, the η values of all three kinds of ceramics decrease dramatically with the increase of Nd³⁺ content. The trend of the quantum efficiency versus Nd³⁺ concentration for Nd:Y_{1.8}La_{0.2}O₃ ceramics is close to that of Nd:Y₂O₃ ceramics. For Nd:Y_{1.8}La_{0.2}O₃ and Nd:Y₂O₃ ceramic, the maximum values of η are 91.5% and ~89% when Nd³⁺ content is 0.5 at% and 0.1 at%, respectively. Although Nd:YAG ceramics show a larger η value, the

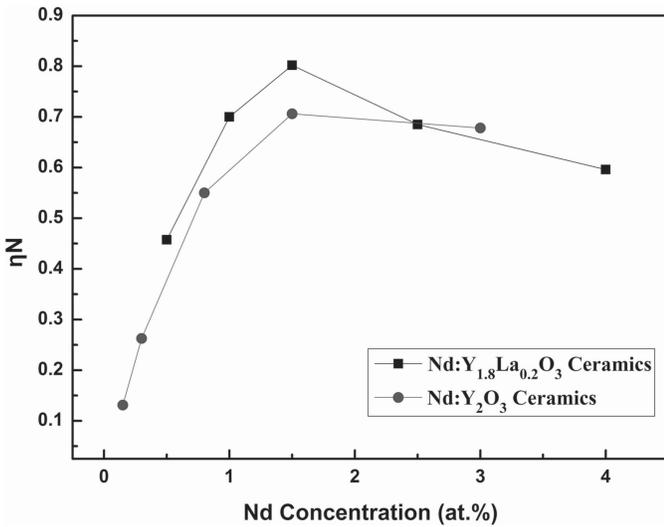


Fig. 8. Dependence of the quantum efficiency and ionic concentration product (ηN) on active ion concentration (N) for Nd:Y_{1.8}La_{0.2}O₃ and Nd:Y₂O₃ ceramics.

quantum efficiency of Nd:Y_{1.8}La_{0.2}O₃ ceramics (91.5%) is still comparable with that in Nd:YAG ceramics (94%) when Nd³⁺ concentration is lower (0.5 at%).

In transparent ceramics, the active ion concentration can be increased more than that of the single crystal without any appreciable structural deformations, so the pump absorption can be enhanced so as to neutralize the quenching effect. Thus, we calculated the product of quantum efficiency and the ionic concentration (ηN) as a merit of Nd:Y_{1.8}La_{0.2}O₃ ceramics. The results are shown in Fig. 8. The ηN values of Nd:Y₂O₃ ceramics were also given for comparison [17]. For both kinds of ceramics, the curves exhibit a decreasing tendency after 1.5 at% dopant concentration due to the concentration quenching effect of Nd³⁺ ions. The curves show a maximum value of 0.71 in Nd:Y₂O₃ ceramics when Nd concentration reaches 1.5 at%, whereas in Nd:Y_{1.8}La_{0.2}O₃ ceramics, the peak value is 0.84 at the same Nd concentration. Furthermore, in Nd:Y_{1.8}La_{0.2}O₃ ceramics, the ηN value is 0.68 even the Nd concentration up to 2.5 at%, which is still comparable with the maximum value of Nd:Y₂O₃ ceramics [17].

C. Laser Experiments

Fig. 9 shows the schematic diagram of the laser experimental setup. A 1.0 at% Nd:Y_{0.18}La_{0.2}O₃ ceramic with a dimension of $5 \times 3 \times 3$ mm³ was used for lasing test (without any antireflection coatings on both surfaces). The transmittance of this sample is 80.0% at the lasing wave band of 1078 ~ 1080 nm. The ceramic sample was placed in a 17 mm length plane-concave resonator. The pump source was a fiber-couple LD emitting at 808 nm with a core diameter of 200 μ m which was focused on the ceramic surface by two coupling lenses (L1 and L2). The input mirror was a flat high reflection mirror (HR) at 1020 ~ 1200 nm and had high transmission at the pumping wavelength. Output coupler (OC) was a concave mirror with 80 mm radius. OC with transmissions of 2%, 6% and 10% at 1080 nm were used in the experiment.

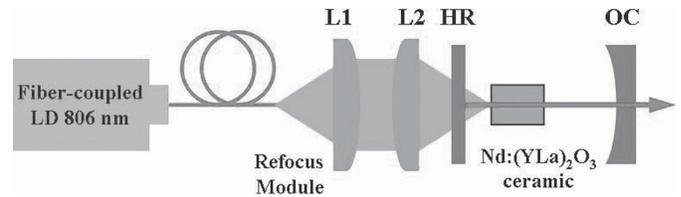


Fig. 9. Schematic diagram of the laser experimental setup. LD: laser diode. L1 and L2: lens. HR: high reflection plate mirror. OC: output coupler.

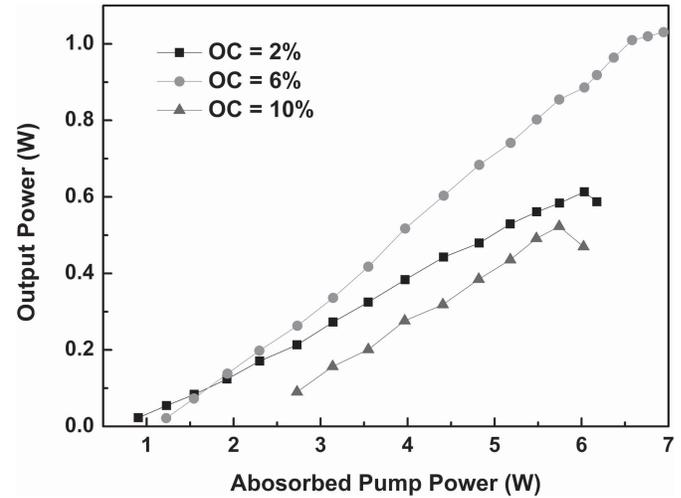


Fig. 10. Output power of ceramic sample versus absorbed pump power.

Fig. 10 is the dependence of the output power versus the absorbed pump power. By using the 6% OC, a maximum output power of 1.03 W was obtained at 1079.5 nm under an absorption pump power of 7.2 W, corresponding to a slope efficiency of 18.4% and an optical-to-optical efficiency of 14.8%. To the best of our knowledge, this is the highest output power achieved in an Nd³⁺-doped sesquioxide ceramic laser up to now, and the slope efficiency of 18.4% is second only to the reported highest slope efficiency for the Nd:Y₂O₃ ceramic lasers (32%) so far [8]. Based on CW laser running, an 8.1 ps pulse at a repetition rate of 98.4 MHz was also obtained in the subsequent passive mode-locking operation [21]. The results of the laser experiment confirmed the potential application of Nd:Y_{0.18}La_{0.2}O₃ transparent ceramics for high power and ultrashort pulse laser devices.

V. CONCLUSION

(Nd_xY_{0.9-x}La_{0.1})₂O₃ ($x = 0.005 - 0.04$) transparent ceramics were fabricated by conventional solid-state processing using high-purity nanopowders. The transmittance spectra and optical micrograph reveal the excellent optical quality of Nd:Y_{1.8}La_{0.2}O₃ ceramics with dense and porous-less structure. The absorption spectra of the samples are similar to that of Nd:Y₂O₃ ceramics. When the Nd³⁺ content is 1.0 at%, the FWHM and absorption cross section at the LD pumping wavelength of 806 nm are 7.8 nm and 1.53×10^{-20} cm², respectively. The strongest emission peak is at wavelength 1078 nm with the stimulated emission cross section of 5.22×10^{-20} cm², FWHM of about 7 nm and the decay

lifetime of 232 μ s. Compared with Nd:Y₂O₃ ceramics and single crystals, the broad absorption and emission bandwidths of Nd:Y_{1.8}La_{0.2}O₃ ceramics offer better possibility of achieving the miniaturized LD pumping apparatus and ultrashort mode-locked pulse laser output. By using 1.0 at% Nd:Y_{1.8}La_{0.2}O₃ ceramics as a laser medium, CW output power of 1.03 W was obtained at 1079.5 nm under an absorbed pump power of 7.2 W corresponding to a slope efficiency of 18.4 %, and this is the highest power for the Nd-doped sesquioxides ceramics laser up to now.

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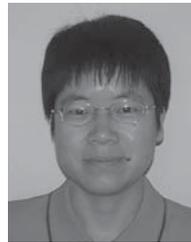
REFERENCES

- [1] H. Hoskins and B. H. Soffer, "Stimulated emission from Y₂O₃:Nd³⁺," *Appl. Phys. Lett.*, vol. 4, no. 1, pp. 22–23, 1963.
- [2] J. Stone and C. A. Burrus, "Nd: Y₂O₃ single-crystal fiber laser: Room-temperature cw operation at 1.07- and 1.35- μ m wavelength," *J. Appl. Phys.*, vol. 49, no. 4, pp. 2281–2287, 1978.
- [3] A. Dienning, B. M. Dicks, E. Heumann, J. P. Meyn, K. Petermann, and G. Huber, "Continuous-wave lasing of Tm³⁺ doped Y₂O₃ near 1.5- μ m," *OSA Topics Adv. Solid-State Lasers*, vol. 10, pp. 194–196, Jan. 1997.
- [4] K. Takaichi, H. Yagi, J. Lu, J. Bission, A. Shirakawa, K. Ueda, T. Yanagitani, and A. A. Kaminskii, "Highly efficient continuous-wave operation at 1030 and 1075 nm wavelengths of LD-pumped Yb³⁺:Y₂O₃ ceramic lasers," *Appl. Phys. Lett.*, vol. 84, no. 3, pp. 317–319, 2004.
- [5] V. Peter, A. Bolz, K. Petermann, and G. Huber, "Growth of high-melting sesquioxides by the heat exchanger method," *J. Cryst. Growth*, vols. 237–239, pp. 879–883, Apr. 2002.
- [6] A. Ikesue, T. Kinoshita, K. Kamata, and K. Yoshida, "Fabrication and optical properties of high-performance polycrystalline Nd:YAG ceramics for solid-state lasers," *J. Amer. Ceram. Soc.*, vol. 78, no. 4, pp. 1033–1040, 1995.
- [7] C. Greskovich and J. P. Chernoch, "Polycrystalline ceramic lasers," *J. Appl. Phys.*, vol. 44, no. 10, pp. 4599–4606, 1973.
- [8] J. Lu, J. Lu, T. Murai, K. Takaichi, T. Uematsu, K. Ueda, H. Yagi, T. Yanagitani, and A. A. Kaminskii, "Nd³⁺:Y₂O₃ ceramic laser," *Jpn. J. Appl. Phys.*, vol. 40, no. 12A, pp. L1227–L1279, 2001.
- [9] J. Ding, Q. Yang, Z. Tang, J. Xu, and L. Su, "Investigation of the spectroscopic properties of (Y_{0.92-x}La_{0.08}Nd_x)₂O₃ transparent ceramics," *J. Opt. Soc. Amer. B*, vol. 24, no. 3, pp. 681–684, 2007.
- [10] Q. H. Yang, C. G. Dou, J. Ding, X. M. Hu, and J. Xu, "Spectral characterization of transparent (Nd_{0.01}Y_{0.94}La_{0.05})₂O₃ laser ceramics," *Appl. Phys. Lett.*, vol. 91, no. 11, pp. 111918-1–111918-2, 2007.
- [11] Q. Yang, S. Lu, B. Zhang, H. Zhang, J. Zhou, Z. Yuan, Y. Qi, and Q. Lou, "Preparation and laser performance of Nd-doped yttrium lanthanum oxide transparent ceramic," *Opt. Mater.*, vol. 33, no. 5, pp. 692–694, 2011.
- [12] B. R. Judd, "Optical absorption intensities of rare-earth ions," *Phys. Rev.*, vol. 127, no. 3, pp. 750–761, 1962.
- [13] G. S. Ofelt, "Intensities of crystal spectra of rare-earth ions," *J. Chem. Phys.*, vol. 37, no. 3, pp. 511–520, 1962.
- [14] T. Kushida, H. M. Marcos, and J. E. Geusic, "Laser transition cross section and fluorescence branching ratio for Nd³⁺ in yttrium aluminum garnet," *Phys. Rev.*, vol. 167, no. 2, pp. 289–291, 1968.
- [15] A. A. Kaminskii, *Laser Crystals, Their Physics and Properties*, 2nd ed. Berlin, Germany: Springer-Verlag, 1981, p. 19.
- [16] W. F. Krupke, "Radiative transition probabilities within the 4f³ ground configuration of Nd:YAG," *IEEE J. Quantum Electron.*, vol. 7, no. 4, pp. 153–159, Apr. 1971.
- [17] G. A. Kumar, J. Lu, A. A. Kaminskii, K. Ueda, H. Yagi, and T. Yanagitani, "Spectroscopic and stimulated emission characteristics of Nd³⁺ in transparent Y₂O₃ ceramics," *IEEE J. Quantum Electron.*, vol. 42, no. 7, pp. 643–650, Jul. 2006.
- [18] H. X. Ma, Q. H. Lou, Y. F. Qi, J. X. Dong, and Y. R. Wei, "Spectrum characteristics of the polycrystalline transparent ceramic," *Laser Technol.*, vol. 28, pp. 488–491, May 2004.
- [19] M. J. Weber, "Radiative and multiphonon relaxation rare-earth ions in Y₂O₃," *Phys. Rev. B*, vol. 171, no. 2, pp. 281–291, 1968.
- [20] G. A. Kumar, J. Lu, A. A. Kaminskii, K. Ueda, H. Yagi, T. Yanagitani, and N. V. Unnikrishnan, "Spectroscopic and stimulated emission Characteristics of Nd³⁺ in transparent YAG ceramics," *IEEE J. Quantum Electron.*, vol. 40, no. 6, pp. 747–758, Jun. 2004.
- [21] Q. Wang, Z. Wei, Z. Wang, J. Zhu, D. Li, Z. Zhang, Q. Yang, H. Zhang, and S. Lu, "Continuous-wave and passively mode-locked ceramic Nd:Y_{0.18}La_{0.2}O₃ laser," unpublished.



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