Fabrication and thermal effects of highly transparent polycrystalline Nd:YAG ceramics

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ABSTRACT

Highly transparent polycrystalline 2.0 at.% Nd:YAG ceramics were fabricated by a solid-state reactive sintering method using commercial α-Al_2O_3, Y_2O_3 and Nd_2O_3 powders as starting materials. The in-line transmittances of the Nd:YAG ceramics vacuum sintered at 1750 °C for 50 h with the thickness of 5.8 mm are 83.9% at 1064 nm and 82.5% at 400 nm. The thermal effects in the Nd:YAG ceramics were mainly investigated in detail. It is found that the thermal focal length decreases with the increase of pump power. The experimental results of thermal focal lengths are in accordance with the theoretical calculations. The observed depolarized beam patterns and depolarization phenomena illustrate the detailed change of thermally induced birefringence in Nd:YAG ceramics. The depolarization shows a obvious nonlinear change tendency at low pump power.

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1. Introduction

As a promising candidate for high power laser materials, Nd^{3+}-doped $Y_3Al_5O_{12}$ have attracted much attention since last century because of its remarkable advantages such as excellent spectral characteristic, structural stability and high thermal conductivity [1–5]. Compared with Nd:YAG single crystals, polycrystalline Nd:YAG ceramics are more and more attractive due to higher doped concentration, available large size and shorter fabrication period [6–9]. Two typical methods are usually used to prepare YAG transparent ceramics. One is a solid-state reactive sintering method [10] and the other is vacuum sintering of YAG powers synthesized by a co-precipitation method [11]. As to the solid-state reactive sintering route, numerous parameters of the fabrication process should be well controlled in order to prepare highly transparent Nd:YAG ceramics [12]. So the choice and treatment of starting powders [13,14,8,15], the category and the amount of sintering aids [16,17], the ball milling process [18], the forming method [19–23], the sintering trajectory [24,25], the post-treatment process [26,27], and the characterization of laser ceramics [28,29] have been widely studied. Most reported researches employed increased laser output power and improved laser beam quality to enhance the laser properties of Nd:YAG ceramics, however, their thermal effects have been seldom studied.

Thermal effects are believed to be the main factor to limit the development of high power solid-state laser, including the Nd:YAG ceramic lasers [30–32]. Thermal effects have significant influences on all the major aspects of solid-state power, such as laser efficiency, output beam quality, resonator stability and mode matching [33–37]. The temperature difference caused by the generated heat in laser gain medium and the removed heat by cooling can result in the inhomogeneous temperature within the specimen, which subsequently gives rise to the variation of refractive index and the thermal distortion of the laser beam. There are some investigations about the thermal effects in Nd-doped glass, Nd:YAG single crystal and ceramic [38–44]. It is demonstrated that the thermal lens effect and thermally induced birefringence are the main thermal effects in laser materials [45–48]. It is highly desirable to comprehensively investigate the thermal effects in Nd:YAG ceramics, which is beneficial to design and fabricate the Nd:YAG ceramics and laser resonator with high performance.

In this work, we prepared the Nd:YAG ceramics with high optical quality by a solid-state reactive sintering method. Then the related physical properties were investigated. More importantly, the detailed change of thermal lens effect with the resonant cavity length, thermally induced depolarized pattern in Nd:YAG ceramics at various pump powers were also studied. We tried to explain the obvious nonlinear change tendency at low pump power in our result.

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2. Experimental procedure

The 2.0 at.% Nd^{3+}-doped YAG ceramics used in the experiment were fabricated by a solid-state reactive sintering method which was reported in our previous work [49]. Commercially available high-purity powders of α-Al$_2$O$_3$ (99.98%, Alfa Aesar, USA), Y$_2$O$_3$ (99.999%, Alfa Aesar, USA) and Nd$_2$O$_3$ (99.99%, Alfa Aesar, USA) were used as starting materials. Tetraethoxysilane (TEOS, 99.999%, Alfa Aesar, USA) and magnesium oxide (MgO, 99%, Alfa Aesar, USA) were used as sintering aids. These powers were blended together with the stoichiometric ratio of (Nd$_x$Y$_{(1-x)}$)$_3$Al$_5$-O$_{12}$ ($x = 0.02$) and mixed in ethanol for 10 h. After ball milling, the slurry with the solid loading of 1.8 g/ml was dried at 80 °C in the oven and sieved through a 200-mesh screen. After calcining at 800 °C, the powders were uniaxially pressed into a 75 mm diameter disk at low pressure and further pressed by cold isostatic pressing (CIP) at 250 MPa. The as-obtained green body was sintered under vacuum at 1750 °C for 50 h. After sintering, the specimen was annealed at 1450 °C for 10 h in air to eliminate the oxygen vacancies. Then the element was processed into a slab of $3 \times 3 \times 6$ mm^3 in size for the laser experiments.

The microstructures of the power mixture, the polished surface and the fracture surface of the specimen were observed by the field emission scanning electron microscopy (FESEM, S-4800, Hitachi, Japan). Mirror-polished samples on both surfaces were used to measure the in-line transmittances and absorption spectra by a UV–VIS–NIR spectrometer (Cary-5000, Varian, USA). The emission spectrum and fluorescence decay curve of the post-annealed specimen at 1064 nm were recorded using a spectrophotometer (Edinburgh, FLS920), with a microsecond flash Xe lamp (Edinburgh, F900) as the exciting source at 355 nm. The signals were detected with a NIR PMT (Hamamatsu, R5509).

The pump-induced thermal focal length of Nd:YAG ceramics was measured by a parallel planar cavity method based on the stability theory of resonators. Magni [50] has demonstrated that the thermally induced focus can be regarded as a thin lens. This assumption has been widely accepted to be proved exactly for most practical situations. Nd:YAG ceramic is regarded as an ideal thin lens with a focal length of f_T located at the center of the specimen. The distances between the center of specimen and the mirrors M_1 and M_2 are L_1 and L_2, respectively. According to the standard ABCD matrix approach, the resonator stability parameter g_1 and g_2 can be easily obtained, given by

$$g_1 = 1 - L_2/f_T, \tag{1}$$

$$g_2 = 1 - L_1/f_T, \tag{2}$$

Then the conditions for which the resonator is stable are

$$0 < (1 - L_2/f_T)(1 - L_1/f_T) < 1, \tag{3}$$

The extreme situation is that the ceramic is put very close to one mirror. Thus the length of L_1 is limited to several millimeters and the condition $0 < (1 - L_1/f_T) < 1$ can always be met. The inequality (3) can be simplified into the following form:

$$(1 - L_2/f_T) > 0 \tag{4}$$

From inequality (4) we can get that there exists a critical point, determined by the condition $L_2 = f_T$, at which the laser resonator begins to be unstable. This value L_2 is the same as the thermal focal length at some pump power level. So we can measure the thermal focal lengths directly at various pump power levels by changing the value L_2.

The experimental setup to measure the thermal lens focal length is shown in Fig. 1. A diode pump laser at 808 nm was used as the pump source, which was focused onto the specimen with the radius of 0.5 mm through fiber coupling. One surface of Nd:YAG ceramic was AR-coated at 808 nm and HR-coated at 1064 nm. The other surface was AR-coated at 1064 nm. The output mirror with 10% transmission at 1064 nm was used to realize the laser output.

The pump–probe method was adopted to measure the depolarization caused by thermally induced birefringence, which was defined by the ratio of the depolarized power to the total pumped power as $D_{pol} = P_\perp/P_{total}$. The setup schematic is shown in Fig. 2. A He–Ne laser was used as the probe which was collimated to a 0.3 mm radius. The probe was reflected by a dichroic beam splitter after passing the polarizer. The polarizer and analyzer were set to be orthogonal. The depolarization was measured under the non-lasing condition.

3. Results and discussion

The average particle sizes of Y$_2$O$_3$ and α-Al$_2$O$_3$ powders are about 3 μm and 0.3 μm, respectively. The aggregated Y$_2$O$_3$ particles are consisted of many primary particles with the diameter of about 100 nm. It illustrates that the powders are effectively crushed and homogeneously mixed by the plenary ball milling process. The specific surface area of the ball-milled powder mixture is 10.3 m^2/g. The size of the specimen is 57 mm in diameter and 5.8 mm in thickness. The main physical properties of the specimen is listed in Table 1.

Fig. 3 shows the in-line transmittance spectrum of the 2.0 at.% Nd:YAG ceramics with thickness of 5.8 mm. The transmittances of the specimen can achieve 83.9% at 1064 nm and 82.5% at 400 nm, which are highly approaching to the theoretical values of Nd:YAG. It demonstrates that the Nd:YAG ceramics used in this work is of high optical quality.

We measured the laser output powers for different cavity lengths, as shown in Fig. 4. The output power is directly related to the gain and loss in cavity. In different cavities, the output power changes a lot, not only the maximum output power but also the slope efficiency. However, all the curves have the similar characteristic. The output power increases linearly with the pump power increasing at first, then saturates, and decreases to zero with a slight increase of pump power.

It is shown that with the increase of pump power, the focal length decreases, accompanied by increased cavity losses in the mean time. It illustrates the enhancement of the optical distortion. In the end, it leads to saturation and a decrease of output power to zero.

The calculation of focal length was carried out on the basis of the thermal lens model developed by Innocenzi et al. [51]. The thermal focal length is expressed by the following equation:

$$f_T = \frac{\pi K_c \omega_p^2}{\eta P_{in} dn/dT} \left[\frac{1}{1 - \exp(-\alpha l)} \right] \tag{5}$$

The calculation parameters of Nd:YAG ceramic are shown in Table 2.

We measured the thermal focal lengths in different cavities, as shown in Fig. 5. With the increase of pump power, the focal length decreases significantly. It is noted that the experimental results ($f_T = 4.5P_{in}^{-1}$) have a certain agreement in comparison with that of the theoretical calculations ($f_T = 4.9P_{in}^{-1}$). It proves the accuracy of our measured data. The slight difference between the measurements and calculations derives from the approximation during the derivations of formulas and the artificial errors.

Fig. 6 shows the depolarized beam patterns for the Nd:YAG ceramics, which were observed with a CCD camera. The change of depolarized beam pattern is obvious. With the increase of pumping power, the depolarized beam pattern reveals as a four-leaf-like pattern and then changes into a ring-like pattern.