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Determination of specific losses in highly transparent Nd:YAG ceramics by laser calorimetry

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Abstract. **Specific absorption losses in five samples of (1%–4%) Nd:YAG optical ceramics are measured by laser calorimetry. The measurement results show that the specific losses increase from 7×10^{-3} to 13×10^{-3} cm^{-1} as the neodymium concentration increases above 2 at %. The principal possibility of separate determination of scattering and absorption losses using additional measurements of total losses in ceramic material is shown.**

Keywords: *laser ceramics, specific losses, laser calorimetry, optical homogeneity.*

1. Introduction

In recent years, significant progress has been achieved in technologies of synthesis of oxide laser ceramics, which made it possible, in particular, to obtain large (up to 50 mm in diameter) samples of Nd:YAG ceramics with comparatively low intrinsic losses and residual porosity [1, 2] based on domestic pressing and sintering technologies [3, 4]. The slope efficiency in domestic Nd:YAG ceramics was increased by several times, from 20% [5] to 36% [6] and 60% [7]. In [8], lasing with a slope efficiency of 40% at wavelength $\lambda = 2090$ nm was achieved in 1%Ho:YAG ceramics under intracavity pumping. Complex analysis of spectral lasing characteristics led to the conclusion that the specific losses in the studied ceramics are $(1-5) \times 10^{-2}$ cm^{-1} [6, 8].

In general, the microstructural homogeneity of domestic ceramics is somewhat inferior to that of the best international samples, which inevitably leads to lower laser parameters, including slope efficiency and threshold pump power [5, 6, 8, 9]. In this connection, of considerable interest are comparative studies of specific losses in ceramics, which strongly affect both laser parameters and efficiency of optical amplifiers. In the present paper, we publish the first results of systematic investigations of specific absorption losses in domestic Nd:YAG ceramics [Kotel'nikov Institute of Radio Engineering and Electronics (Fryazino Branch), Russian Academy of Sciences (FIRE)] by laser calorimetry [10–12] depending on the neodymium concentration, as well as the estimates of absorption and scattering losses in 1%Nd:YAG ceramics synthesised in the Institute of Electrophysics, Ural Branch, Russian Academy of Sciences (Ekaterinburg) (IEP).

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2. Laser calorimetry method

Calorimetry is a simple and reliable method for measuring low absorptions in optically transparent materials [10–12]. This method is based on measuring the temperature of a sample under study for some time before, during, and after laser irradiation.

The basic equations describing the change of the sample temperature with time have the form [12]

$$C\frac{\mathrm{d}T}{\mathrm{d}t} = \alpha LP - \beta(T - T_0), \tag{1}$$

$$P(t_1 \leqslant t \leqslant t_2) = P, \quad P(t < t_1, t > t_2) = 0, \tag{2}$$

where α is the specific absorption (in cm^{-1}); L is the sample length (in cm) along the laser beam axis; P is the probe radiation power (in W); C is the heat capacity of the sample (in J K^{-1}); β is the coefficient of heat transfer to the environment (in W K^{-1}); T and T_0 are the temperatures of the sample and the environment, respectively (in K or °C); and $t_1 - t_2$ is the laser irradiation time (sample heating time).

Equations (1) and (2) are valid under the following conditions: the time of measurement in the heating–cooling regime considerably exceeds the time of temperature stabilisation in the sample, i.e., the sample at each instant of time is in quasi-stationary conditions; the sample overheating $T - T_0$ is insignificant, which allows one to consider the heat transfer coefficient β as a constant value; and the environment temperature T_0 is constant (does not drift).

If the sample temperature at the time moment t_1 coincides with the temperature of the environment, then the solution of Eqns (1), (2) is described by the expressions

$$T(t) = T_0, \quad t \leqslant t_1, \tag{3}$$

$$T(t) - T_0 = \frac{\alpha LP}{\beta}\left\{1 - \exp\left[-\frac{\beta(t - t_1)}{C}\right]\right\}, \quad t_1 \leqslant t \leqslant t_2, \tag{4}$$

$$T(t) - T_0 = [T(t_2) - T_0]\exp\left[-\frac{\beta(t - t_2)}{C}\right], \quad t \geqslant t_2. \tag{5}$$

The C/β parameter determines the characteristic sample heating time at which the measurement accuracy of specific absorption α is maximal. Indeed, at small heating times, ($t_2 - t_1 \ll C/\beta$), overheating $T - T_0$ will be much smaller than the maximum possible value ($\alpha LP/\beta$), which will lead to deterioration of the measurement contrast, i.e., to a decrease in the useful signal with respect to the noise level. On the other hand, prolonged heating ($t_2 - t_1 \gg C/\beta$) makes it possible to achieve the maximum overheating ($T - T_0 = \alpha LP/\beta$), but the measurement error in this case increases due a possible drift of the environmental temperature T_0. Note also that in typical situations, when the sample volume is $\sim$0.1 cm^3, the C/β parameter is $\sim$100 s (see also Table 1).

3. Experiment

Specific absorption was measured for five disk-shaped samples of highly transparent (1%–4%) Nd:YAG laser ceramics. Four samples 7.3 mm in diameter and 1 mm thick were synthesised in FIRE [13], while the fifth sample 11 mm in diameter and 1 mm thick was synthesised in IEP [3, 4]. Two faces of ceramics produced in FIRE were polished and uncoated in contrast to ceramics made in IEP, whose faces were coated with anti-reflection and high-reflection films for the probe wavelength (1064 nm).

For calorimetric measurements, we developed a fast-response measuring cell based on two successively connected chromel–copel thermocouples with a total thermoelectric power of 125 μV K^{-1}, which were mounted on a copper base with a heat capacity of 50 J K^{-1}. The cell design allows one to measure the temperature of both disk and cylindrical optical elements. A highly stable cw Nd:YAG laser with an output power up to 2 W at $\lambda = 1064$ nm was used as a probe radiation source. The drift of the output power of this laser did not exceed 0.2% per hour. The temperature of samples under study was measured using a precision preamplifier with intermediate averaging; the data were read out and recorded on a computer with a frequency of 1 Hz. The instrumental temperature resolution including the intrinsic preamplifier noise was ±0.5 mK. At this accuracy, the limiting resolution of the calorimetric measurement of α without allowance for the laser source stability is $\sim 10^{-4}$ cm^{-1} at a probe radiation power of 1 W.

The measurement procedure was identical in all the cases and consisted in the following. A Nd:YAG laser beam was focused into the centre of the sample to a spot 300 μm in diameter, after which the beam was blocked and the sample temperature approached the copper base temperature until the drift became smaller than 5 mK min^{-1}. For the first minute of the measurement, the sample was not irradiated; from the second to the fourth minute, the sample was heated by a $\sim$2-W laser beam; beginning from the fourth minute, the laser beam was blocked and the sample cooled for the next two minutes. Then, for about half an hour, the sample temperature approached the base temperature, after which the second measurement was performed. The second measurement results coincided with the first ones within one–two percent in all cases, which testifies to a good reproducibility and a corresponding reliability of the obtained data. The measurement results were processed taking into account the Fresnel losses for ceramics produced by FIRE and the double passage of the probe radiation through the 1%Nd:YAG ceramics synthesised by IEP.

4. Results and discussion

Figure 1 shows the typical experimental curve for one of the ceramic samples from FIRE, while the solid grey lines are its approximations by dependences (4) and (5).

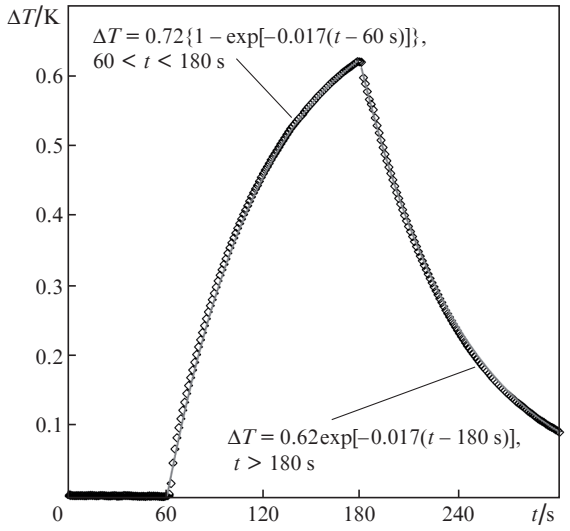

Figure 1. Time dependence of sample overheating $\Delta T = T - T_0$ (points) and its approximation by formulas (4) and (5) (grey curves). The ceramics is synthesised by FIRE, C_{YAG} = 0.1126 J K^{-1}, P = 1.71 W. Approximation parameters are as follows: heat transfer power αLP = 1.38 mW, $\alpha = 8.1 \times 10^{-3}$ cm^{-1}.

The main measurement results for all ceramics are listed in Table 1. In general, exponential dependences (4) and (5) describe the experimental data with an error not exceeding 1%–2%, which results from the fulfilment of the conditions given in Section 2 for initial Eqns (1) and (2) and confirms the above estimate of calorimetry resolution [$(1\%-2\%)\alpha \approx 10^{-4}$ cm^{-1}]. Taking into account the errors in the thermoelectric power calibration, in the determination of the heat capacity of samples by their geometric dimensions, and in the measurement of the probe radiation power by an OPHIR L30A power meter, the absolute accuracy of determination of specific absorption α can be estimated to be ±5% of the measured value. Note also that the heat transfer coefficients β at the heating and cooling stages almost completely coincide, which

Table 1. Main results of calorimetry data processing for the studied Nd:YAG ceramics.

Sample number	Sample	α/cm^{-1}	$\beta_\uparrow$C^{-1}/s^{-1}	$\beta_\downarrow$C^{-1}/s^{-1}
1547	1%Nd:YAG (FIRE)	8.1×10^{-3}	0.0170	0.0170
1546	2%Nd:YAG (FIRE)	7.0×10^{-3}	0.0161	0.0161
1545	3%Nd:YAG (FIRE)	1.14×10^{-2}	0.0167	0.0169
1544	4%Nd:YAG (FIRE)	1.29×10^{-2}	0.0169	0.0168
1570	1%Nd:YAG (IEP)	8.9×10^{-3}	0.00997	0.00998

Note. Subscripts ↑ and ↓ denote the heating and cooling regimes, respectively.

indicates the absence of a noticeable drift of the copper base temperature.

The obtained results show that the specific absorption of ceramics from FIRE increases approximately by a factor of 1.5 as the neodymium concentration increases above 2%, while the specific absorption of 1%Nd:YAG ceramics synthesised by FIRE and IEP is almost the same. It should be noted that the total specific losses for ceramics 1570 synthesised at IEP was previously determined from laser experiments [8] to be $\alpha_\Sigma = 1.6 \times 10^{-2}$ cm^{-1}. Thus, the specific scattering losses for this ceramics can be estimated to be 7.1×10^{-3} cm^{-1}, which is somewhat lower than the specific absorption losses (8.9×10^{-3} cm^{-1}). This value was determined without taking into account the ceramic material inhomogeneities, which may considerably affect the final result.

It should be noted that the specific absorption in all cases was measured in the central part of ceramic disks, and the probe beam diameter (300 mm) considerably exceeded the characteristic size of granules, which was usually below 10 μm [5]. For more detailed study of possible large-scale inhomogeneities in ceramics, it is necessary to scan the probe radiation over the sample cross section. Of particular interest is to compare the obtained data on specific absorption with the local lasing characteristics, which makes it possible to completely characterise ceramics as a material for active elements of high-power solid-state lasers.

5. Conclusions

Laser calorimetry is a simple and reliable method of determination of specific absorption of highly transparent ceramic materials. Within the available experimental facilities, the calorimetry resolution is 10^{-4}–10^{-5} cm^{-1} at a probe radiation power of 1–10 W, which is quite enough for testing the best domestic and international samples. Despite the fact that the specific loss in domestic laser ceramics is approximately an order of magnitude higher than that in the best foreign samples, this parameter demonstrates a stable tendency to decreases to 10^{-3} cm^{-1} and lower [7]. This value will be acceptable for producing large-scale active elements of multikilowatt laser systems including multi-cascade optical power amplifiers.

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