Int J Thermophys (2012) 33:707–715
DOI 10.1007/s10765-012-1177-1

Determination of Thermal-Diffusivity Dependence on Temperature of YAG Single Crystals with Different Concentrations of Yb^{3+} and V^{3+} Doping Ions

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Received: 4 May 2011 / Accepted: 21 February 2012 / Published online: 11 March 2012

Abstract Thermal diffusivities of pure and doped yttrium aluminum garnet single crystals were measured as a function of temperature. Samples doped with rare earth ions (3 at% and 25 at% of Yb^{3+}, and 0.8 at% and 2.1 at% of V^{3+}) were investigated in the temperature range from 34 °C to 300 °C. Determination of the thermal diffusivity was based on an analysis of propagation of a thermal wave in the sample. The frequency of the thermal wave was 100 mHz. A temperature disturbance connected with the thermal wave propagating in the sample was detected using the mirage effect. The results showed that the thermal diffusivity of all investigated samples decreases with an increase of sample temperature. A drop in the thermal diffusivity is more pronounced for pure and low-doped crystals.

Keywords Optical crystals · Temperature dependence · Thermal diffusivity · Thermal wave measurement

1 Introduction

The thermal diffusivity is a very important quantity determined in dynamic thermal measurements. It characterizes the speed of reaching thermal equilibrium between a sample and its surrounding. This parameter plays a key role in designing high power laser systems and other electronic devices with regard to the necessity of efficient heat transfer. The thermal diffusivity varies with temperature. Thus, in order to select

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a proper material for any devices working at elevated temperature (e.g., lasers), it is essential to characterize the thermal-diffusivity dependence on temperature.

Garnet crystals are frequently used in high power diode-pumped solid-state lasers. The yttrium aluminum garnet (YAG) is a high quality host crystal with excellent physical and chemical properties [1–4]. It can be easily doped with rare earth ions like Yb or V. Yb:YAG single crystals possess many advantages, such as high thermal conductivity [5], that are desirable for high-average-power laser systems. A complete characterization of selected properties of Yb:YAG single crystal and its applications was mentioned in our previous article [6]. Another popular crystal applied in nonlinear optics is YAG doped with vanadium ions. V:YAG crystal used in a laser technique assures a Q-switched operation and can be used as a saturable absorber. The group of YAG was studied in many arrangements, i.e., for switching of Nd:YAP, Nd:YAG, Nd:YVO_4, or Nd:KGW lasers with the flash lamp [7,8] or diode pumping [9].

This article presents the results of investigations of the thermal-diffusivity dependence on temperature for pure YAG ($Y_3Al_5O_{12}$) single crystals and crystals doped with different concentrations of trivalent ions: Yb and V. The influence of temperature and doping concentration on the thermal diffusivity is also discussed. In the next section, samples and the measuring method used for the investigation are described. Results of measurements carried out in the temperature range from 34 °C to 300 °C are shown in Sect. 3. Final conclusions end the paper.

2 Experiment

2.1 Samples

Pure and Yb^{3+}- and V^{3+}-doped YAG crystals were grown by the Czochralski method. Details related to the growing process were described elsewhere [6]. Five YAG samples were examined: pure, doped with 3.0 at% and 25 at% of Yb^{3+}, and doped with 0.8 at% and 2.1 at% of V^{3+}. All samples were cut into rectangular prisms and oriented along crystallographic directions [1 1 0], [1 1 1], and [2 2 4]. Basic information about samples is collected in Table 1. Investigated crystals are shown in the photo presented in Fig. 1.

Table 1 Basic information about samples

Sample		Dopant	Dimension (10^{-3} m)		
Crystallographic directions:			[1 1 0]	[1 1 1]	[2 2 4]
#1	$Y_3Al_5O_{12}$	–	10.64	10.62	10.63
#2	$Y_3Al_5O_{12}$	3.0 at% of Yb	10.59	10.63	10.68
#3	$Y_3Al_5O_{12}$	25 at% of Yb	10.63	10.47	10.64
#4	$Y_3Al_5O_{12}$	0.8 at% of V	10.15	10.14	10.13
#5	$Y_3Al_5O_{12}$	2.1 at% of V	10.15	10.02	10.13

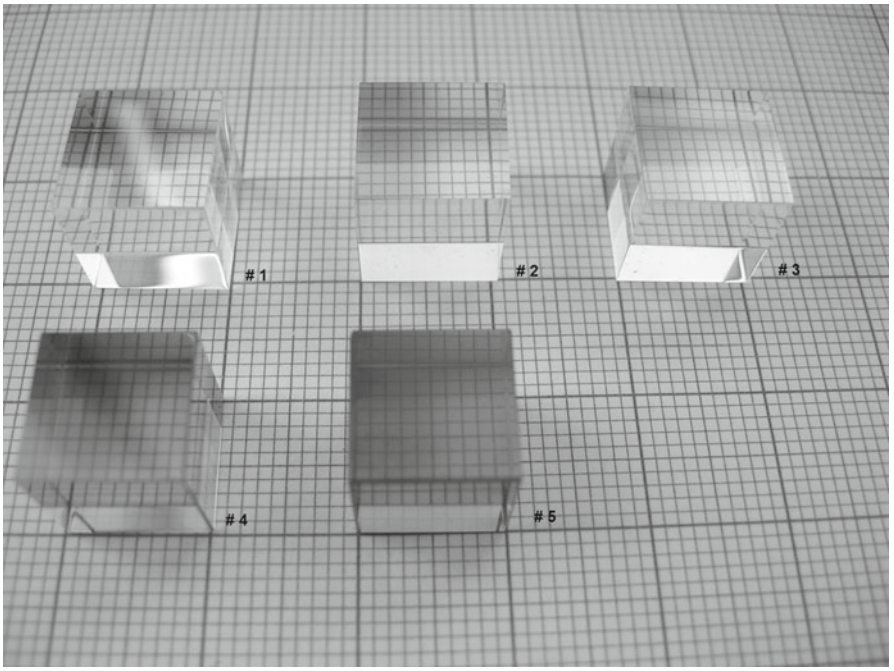

Fig. 1 Photo of YAG single crystals: *#1* pure, *#2* doped with 3 at% of Yb, *#3* doped with 25 at% of Yb, *#4* doped with 0.8 at% of V, and *#5* doped with 2.1 at% of V

2.2 Thermal Wave Measurements

The thermal diffusivity was measured by the thermal wave method with mirage detection. In previous experiments, the thermal wave was generated by a Peltier's module, on which the sample was placed [6]. It allowed continuous heating and cooling of the sample in such a way that its average temperature remained constant and equal to ambient. The modification of the experimental setup consisted of placement of the investigated sample in a chamber between two resistance heaters, which allowed sample heating and ensured the same temperature in the whole volume of the sample. In addition, it was possible to modulate the current of a selected heater with a constant frequency. This modulation caused a periodic temperature disturbance in the sample, i.e., it generated the thermal wave. In experiments described in this article, the modulation frequency was 100 mHz. In order to monitor the temperature of upper and lower sample surfaces, copper plates with connected thermocouples were placed between the sample and the heaters. To assure low heat resistance, thin layers of heat conducting paste were applied at all contacts.

The geometry of measurement is shown in Fig. 2. In order to obtain good thermal insulation of a sample and heaters, the system was placed in a chamber evacuated to a pressure of a few Pa. At the beginning of each measurement, the temperature of the sample was stabilized at a set value. Then the temperature modulation around the set

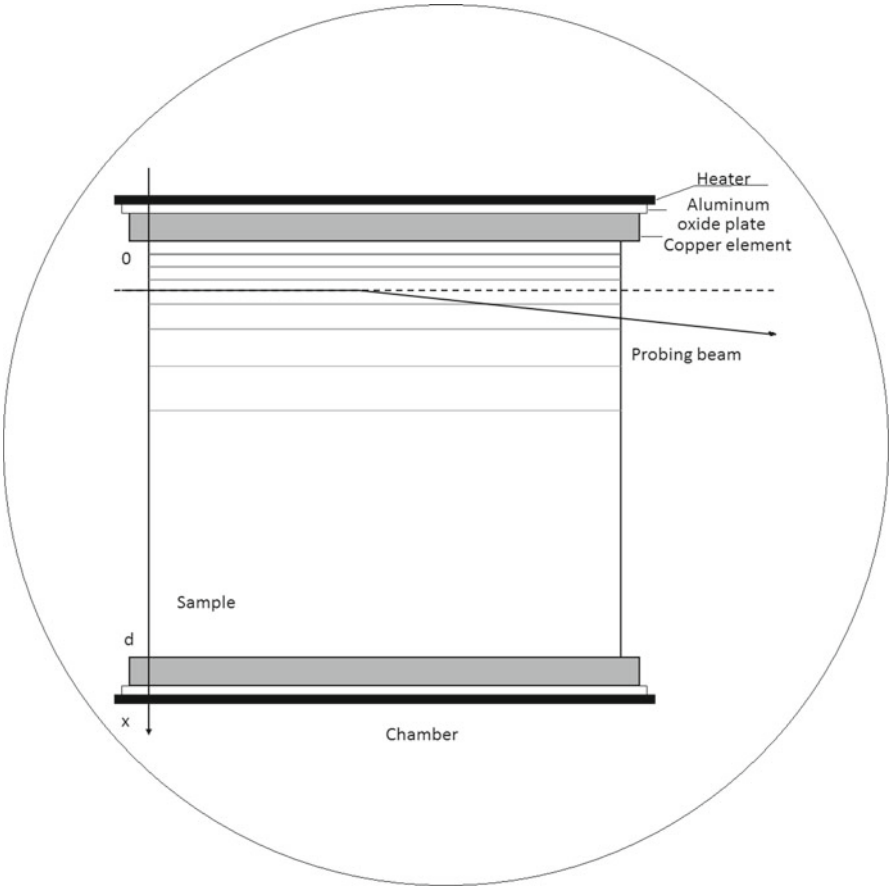

Fig. 2 Geometry of measurement

point was applied. Measurements were carried out for the range of temperature from 34 °C to 300 °C.

Detection of the temperature disturbance, which propagated in the sample, was based on the mirage effect. A detecting light beam from a He–Ne laser (LASOS 7672) was passed through the sample. The deflection of this beam was registered by a position sensor (Silicon Sensor DL400-7PCBA). The signals from the detector and from both thermocouples were fed to a data acquisition unit (Agilent 34970A) and collected on a PC via an RS-232 interface.

Assuming a one-dimensional model of heat propagation in a thermally thick sample, the phase delay $\Delta\varphi$ of the thermal wave at a distance h from a source is given by the formula [10],

$$\Delta\varphi = \sqrt{\frac{\pi f}{\alpha}}h + \varphi_0 \tag{1}$$

where f is the modulation frequency and φ_0 is a constant. The phase delay can be determined experimentally from measurement of the time delay Δt between the temperature measured by the thermocouple placed between the heater and the sample, and the deflection signal obtained from the position detector for a given position of the probing beam. The relation of these values is

$$\Delta\varphi = 2\pi f \Delta t. \tag{2}$$

By measuring the $\Delta\varphi$ dependence on h, one can determine the thermal diffusivity α from a simple linear fit.

3 Experimental Results and Discussion

The thermal diffusivity of each sample was determined for seven different temperatures from 34 °C to 300 °C. The thermal-diffusivity dependence on temperature was studied for only one crystallographic direction because YAG crystals are thermally isotropic [2]. As was mentioned in the previous section, the determination of the thermal diffusivity was based on the measurement of the time delay between the harmonic temperature component at the heater surface and the deflection signal from the position detector. Representative signals registered for pure YAG at 34 °C are shown in Fig. 3. The time delay is determined from the positions of neighboring maxima or minima on both curves. The time delay was measured for at least five different pairs of extrema, and then the obtained values were averaged. The phase delay was calculated from Eq. 2. The phase delay dependence on the probing beam position obtained for pure YAG at 34 °C is shown in Fig. 4. The thermal diffusivity was obtained by fitting the data with a straight line. In Figs. 5 and 6, dependences of the thermal diffusivity on the temperature determined for all investigated samples are shown. Results of measurements are also collected in Table 2.

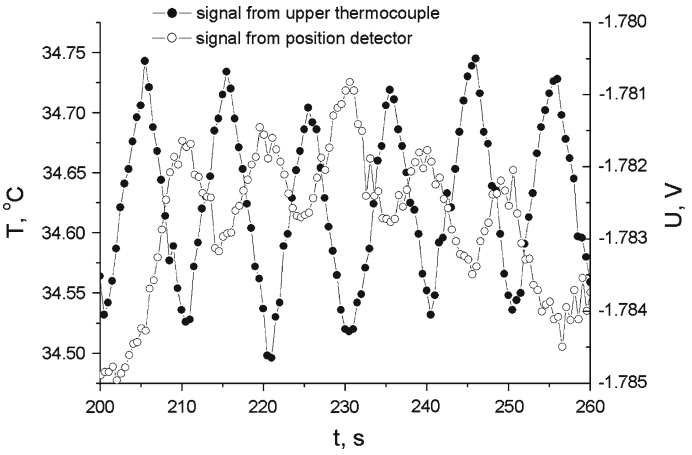

Fig. 3 Time delay of temperature disturbance propagating along the pure YAG. The signal from the position detector was measured for a selected distance of the probing beam from the modulated upper heater

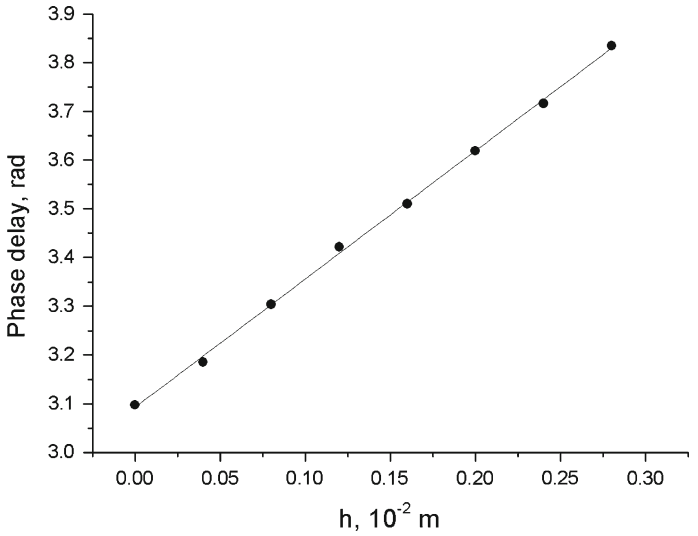

Fig. 4 Phase delay of the signal as a function of distance *h* from the sample surface measured in pure YAG crystal at 34 °C

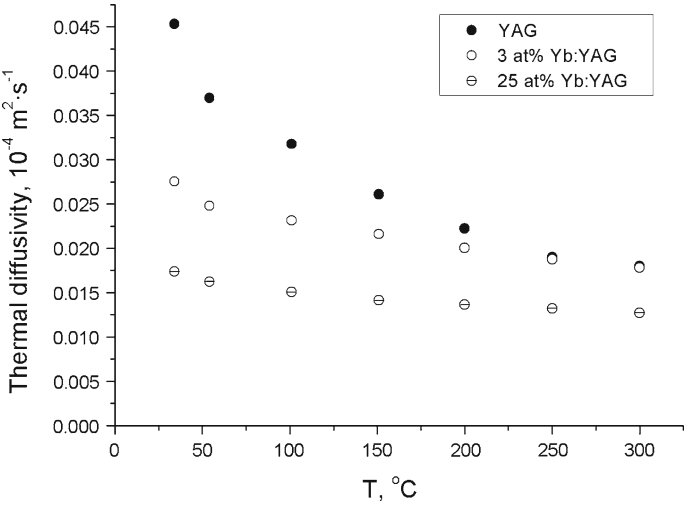

Fig. 5 Thermal-diffusivity dependence on temperature of pure YAG and doped with 3 at% and 25 at% of Yb

In the simplest approach, the thermal conductivity of dielectric crystals can be described by the formula known from the kinetic theory of gases [11],

$$\kappa = \frac{1}{3} C u \lambda \tag{3}$$

where u is the velocity of phonons, λ is their mean free path, and C is the heat capacity per unit volume. The thermal diffusivity α and the thermal conductivity κ are related by the formula,

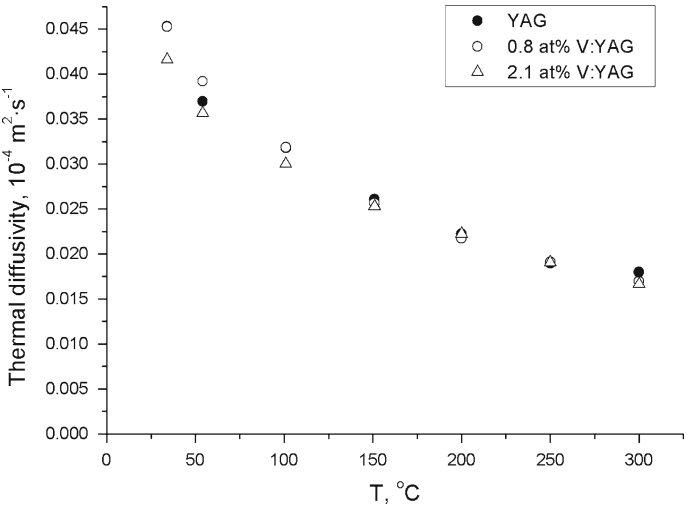

Fig. 6 Thermal-diffusivity dependence on temperature of pure YAG and doped with 0.8 at% and 2.1 at% of V

Table 2 Thermal diffusivities obtained for all investigated samples from 34 °C to 300 °C

T (°C)	Thermal diffusivity with uncertainties (10^{-6} m$^2 \cdot$ s^{-1})				
	Yb^{3+} concentration (at%)			V^{3+} concentration (at%)	
	0	3	25	0.8	2.1
34	4.53 ± 0.12	2.75 ± 0.18	1.74 ± 0.11	4.52 ± 0.29	4.16 ± 0.24
55	3.69 ± 0.12	2.48 ± 0.15	1.62 ± 0.18	3.92 ± 0.24	3.57 ± 0.10
101	3.18 ± 0.12	2.31 ± 0.13	1.51 ± 0.13	3.17 ± 0.16	3.00 ± 0.17
151	2.61 ± 0.19	2.16 ± 0.19	1.41 ± 0.20	2.57 ± 0.18	2.53 ± 0.10
201	2.22 ± 0.26	2.00 ± 0.10	1.36 ± 0.10	2.17 ± 0.10	2.22 ± 0.10
250	1.90 ± 0.27	1.88 ± 0.10	1.32 ± 0.17	1.90 ± 0.10	1.90 ± 0.10
300	1.79 ± 0.11	1.78 ± 0.10	1.27 ± 0.10	1.70 ± 0.10	1.66 ± 0.10

$$\alpha = \frac{\kappa}{C} = \frac{1}{3} u \lambda. \tag{4}$$

It means that changes in the thermal diffusivity are determined by changes in the velocity and the mean free path of phonons. So, the thermal diffusivity is directly related to quantities describing phonon kinetics in a system. It followed from our previous investigations that the influence of dopants on the ultrasound velocity in crystals is rather low [12]. So, changes in the thermal diffusivity are mainly caused by changes in the phonon scattering rate.

The thermal diffusivity decreases with increasing temperature for all samples. Such an effect can be explained by shortening of the phonon mean free path caused by a growing concentration of phonons, which results in more intense phonon–phonon

scattering. This effect is more pronounced in pure and low-doped crystals, where the phonon–phonon interactions play a key role in the heat transport. In the highly doped samples, the phonon scattering on impurities dominates at room temperature and leads to a decrease of the thermal diffusivity. However, a contribution of the phonon–phonon interactions to the phonon scattering becomes more important with increasing temperature, while the phonon-impurity scattering ratio remains constant. It may explain the fact that the thermal diffusivity of almost all samples, except the one containing 25 at% of Yb, is practically the same at 300 °C. Changes of the thermal diffusivity of the 25 at% Yb:YAG sample with increasing temperature are relatively low, but they still exceed 25 % in the analyzed temperature range. This sample can be treated as a mixed crystal (a solid solution) of YAG and YbAG with a high concentration of scattering centers, in which the phonon scattering on impurities at 300 °C is still the most important scattering mechanism.

4 Conclusion

The results of measurements carried out for pure and doped YAG single crystals revealed a significant decrease of the thermal diffusivity caused by increasing temperature from 34 °C to 300 °C. The thermal-diffusivity drop changed from about 60 % for a pure sample to about 25 % for YAG containing 25 at% of Yb. An important fact is that although the thermal diffusivity of the sample doped with 3.0 at% of Yb is considerably lower than that of pure YAG at 30 °C, their thermal diffusivities are practically the same at 300 °C. It shows that differences in the thermal properties measured at room temperature can be negligible at higher temperatures.

A general conclusion is that the influence of doping on the thermal diffusivity becomes less pronounced with increasing temperature, and above a certain temperature, it can be omitted.

Measured dependences confirm the assumption that dopant ions create additional scattering centers of phonons, and this mechanism is responsible for the decrease in the thermal diffusivity of doped crystals in comparison with the pure one [6]. When the temperature increases, the ratio of phonon–phonon scattering also increases, and at higher temperatures, this scattering mechanism prevails compared to the scattering on the defects. It means that the influence of doping on thermal properties of a material depends strongly on the temperature.

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