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Investigation of thermal diffusivity dependence on temperature in a group of optical single crystals doped with rare earth ions

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ABSTRACT

The group of YAG, YVO_4 and GdCOB single crystals was examined to determine the thermal diffusivity as a function of temperature in range from 30 °C to 300 °C. Further investigations concerned on analysis of the influence of dopants on these dependencies. The experimental setup based on thermal wave method with mirage detection was used. The samples represented different crystallographic systems such as cubic (YAG) tetragonal (YVO_4) and monoclinic (GdCOB). The anisotropy of thermal conductivity of investigated samples was taken into account in the investigations. The crystals were doped with calcium ions, rare earth ions such as ytterbium, neodymium, and thulium, and also with transition metal vanadium. The results confirmed that influence of doping on the thermal diffusivity of investigated materials strongly depends on temperature. In general the thermal diffusivity decreases with increasing of sample temperature from 30 °C to 300 °C, however the drop in thermal diffusivity is the highest for pure single crystals. Doping is another factor reducing the heat transport in single crystals. Introduction of dopant ions into a crystal lattice leads to a significant decrease in the thermal diffusivity at lower temperatures in comparison with pure crystals. However, the influence of dopants becomes less pronounced with increasing temperature, and in case of weakly doped crystals it becomes negligible at higher temperatures. The interpretation of thermal diffusivity dependence on temperature for single crystals was based on the Debye model of lattice thermal conductivity of solids. The results allowed to conclude that the decrease of thermal diffusivity with temperature and increasing concentration of impurities is caused by shortening of the phonons mean free path due to phonon–phonon and phonon-point defect scatterings.

1. Introduction

Optically active crystals are key parts of solid-state laser systems [1,2]. In order to obtain crystals with required optical properties dopant ions are introduced into the pure single crystal lattice. A modification of physical and chemical properties of crystal used as lasing medium is possible thanks to proper selection of type and a concentration of dopant ions. Especially spectral properties can be improved by creation of beneficial structure of energy levels in the crystal, which results in an increase of pumping efficiency [3–6]. However, introduction of even small amount of dopant into the crystal structure deteriorates the heat transport properties of the crystal. Parameters such as the thermal conductivity and the thermal diffusivity are very sensitive to defects of crystal structure, including these created by doping ions incorporated into the crystal lattice [7–11]. It should be noted that the thermal properties are very important in selection of crystal for use in high-power laser systems. Effective heat abstraction from the medium is necessary to avoid overheating of the device which typically leads to deterioration of its functional parameters. In addition, local heating of low thermal conductivity materials causes high temperature gradients and high thermal stresses. It can cause damage to the device as a result of cracks of active medium [12]. During designing of active optical system operating at elevated temperatures the dependence of the thermal properties on temperature must also be taken into account. Therefore the problem of the influence of dopants and temperature on the heat transport in optical active crystals is particularly important.

The heat transfer in dielectric crystals occurs through collective vibrations of the crystal lattice represented by phonons. Treating the phonons as particles in the ideal gas, thermal conductivity κ can be described according to the kinetic-molecular theory by the formula:

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

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where C is the volume heat capacity, u is the phonon velocity and Λ is its mean free path.

Taking into account the relationship between the thermal conductivity κ and the thermal diffusivity α

$$\kappa = \alpha C, \tag{2}$$

Eq. (1) can be rewritten in the form:

$$\alpha = \frac{1}{3} u \Lambda. \tag{3}$$

The influence of temperature and dopants on phonon velocities, both longitudinal and transverse, can be neglected [7,13–15]. Thus, changes in the thermal diffusivity, caused by these factors, are almost completely defined by the changes in the mean free path of phonons. The mean free path is determined by phonon scattering processes: the phonon–phonon scattering and the scattering on crystal defects, among them on dopants. These scattering mechanisms exhibit different temperature dependences. Taking this fact into consideration joint analysis of the thermal diffusivity dependence on temperature is justified.

This paper is aimed at experimental determination of the temperature dependences of the thermal diffusivity of pure and doped YAG, YVO_4 and GdCOB single crystals with consideration of their anisotropy. In addition to experimental research, the results were subject to theoretical analysis. An interpretation of results was carried out in the frame of the phonon theory of heat transport.

2. Experiment

2.1. Samples

Investigated crystals represent three different crystallographic systems. The yttrium aluminum garnet $Y_3Al_5O_{12}$ (YAG) crystals have a cubic structure, the yttrium orthovanadates YVO_4 – a tetragonal structure, and the gadolinium calcium oxoborates $Ca_4GdO(BO_3)_3$ – a monoclinic structure. All crystals were grown by the Czochralski method. Details on the growth of individual crystals were described elsewhere [13,16,17]. The produced single crystals were of good optical quality with no visible defects. Each of the crystals was cut into a rectangular prism and properly oriented, which was confirmed in earlier structural investigations [7]. Single crystals were doped with different concentrations of rare earth ions such as ytterbium, neodymium, and thulium. Two of YAG crystals were doped with vanadium ions. Basic information about investigated samples is presented in Table 1.

2.2. Experimental method

The thermal diffusivity was measured by the thermal wave method with mirage detection. A one-dimensional model of the plane thermal wave propagating along x direction in a sample was assumed. The assumed geometry of measurement is schematically shown in Fig. 1.

Initial temperature of the entire sample was constant and equal to T_0

$$T(x,0) = T_0. \tag{4}$$

Then the modulation around the set temperature with a given frequency f and an amplitude θ_0, according to formula

$$T(0,t) = T_0 + \theta_0 \cos(2\pi f \cdot t) \tag{5}$$

was applied at the surface $x = 0$. The opposite surface $x = d$, where d is the thickness of the sample, remained constant and equal to T_0

$$T(d,t) = T_0. \tag{6}$$

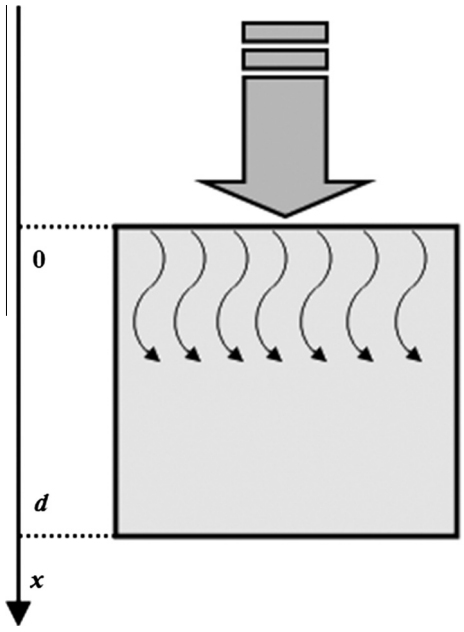

Fig. 1. Scheme of a temperature field disturbance propagating in the sample.

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
	Crystallographic directions		[1 0 0]	[0 1 0]	[0 0 1]
#7	YVO_4	–	10.09	10.18	10.31
#8	YVO_4	1.0 at% of Nd	10.09	9.88	10.16
#9	YVO_4	2.0 at% of Nd	10.12	9.90	9.10
#10	YVO_4	5.0 at% of Tm 0.4 at% of Ca	10.02	9.81	10.21
	Directions of axes of optical indicatrix		*X*	*Y*	*Z*
#11	$Ca_4GdO(BO_3)_3$	–	9.21	8.15	9.98
#12	$Ca_4GdO(BO_3)_3$	4.0 at% of Nd	9.21	8.31	10.00
#13	$Ca_4GdO(BO_3)_3$	7.0 at% of Yb	9.20	8.36	9.95