Nonlinear bulk absorption in $Y_{3-x}Er_xAl_5O_{12}$ crystals

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A nonlinear bulk absorption by impurity particles of submicron size has been observed for the first time in yttrium-erbium-aluminum garnet crystals. The local absorption coefficients of these particles reach values $\geqslant 10^3$ cm^{-1}.

The bulk absorption (α) has been measured in crystals, glasses, and fibers by the method of adiabatic laser calorimetry with cw lasers in many studies.[1–3] It was suggested in Ref. 4 that periodic pulsed lasers be used to measure α in $Y_{3-x}Er_xAl_5O_{12}$ crystals. When crystals are heated by a pulse with a length of 50 μs or more, the value of α does not change; it is the same as the value in the case in which the crystal is heated by a cw laser. When we reduced the pulse length by an order of magnitude (to 1 μs or less), we observed a nonlinear bulk absorption of laser light in $Y_{3-x}Er_xAl_5O_{12}$ crystals for the first time.

Let us assume that a crystal contains defects with large local absorption coefficients. In this case we need to consider the relationship between the length of the laser pulse, t_l, and the time scale of the heat transfer between the crystal and the particle, t_x. Specifically, if the condition

$$t_l < t_x = R^2/g \tag{1}$$

holds, where R is the defect radius, and g is the thermal diffusivity, the defect may undergo a pronounced selective heating with respect to the crystal if the energy of the laser pulse is sufficiently high.

Hopper and Uhlmann[5] have studied the process by which an absorbing defect in a glass is heated by laser light through a solution of the heat-conduction equation. Heat evolution in the material was taken into account. They assumed that the thermal properties of a defect remain the same as it is heated. Danileĭko *et al.*[6] studied the effect of a change in the thermal properties of a defect in a crystal with a temperature on the thresholds for laser damage. To obtain some specific estimates of the laser damage thresholds, they built into the theory, in a phenomenological way, a parameter serving as a measure of the nonlinearity of the absorption of a heated defect.

The value of α in the $Y_{3-x}Er_xAl_5O_{12}$ crystals is measured by the method of adiabatic laser calorimetry with the help of a periodic-pulse $Y_3Al_5O_{12}$-Nd^{3+} laser ($\lambda = 1.06\ \mu$m). The experimental layout is shown in Fig. 1. The laser beam is directed along the axis of the cylindrical sample (6 mm in diameter and $L = 70$ mm long). The length of the laser pulse is varied from 200 μs to 10 ns; the experimental geometry, the average power of the light, and the pulse repetition frequency are all held constant. The sample temperature rises after the laser is turned on because of bulk and surface absorption of the crystal. The measurements are taken on the initial part of the $d\theta/dt$

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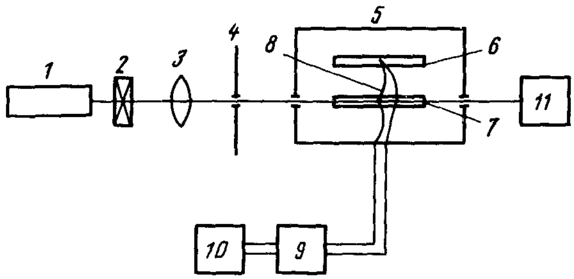

FIG. 1. Experimental layout for measuring the bulk absorption coefficient in $Y_{3-x}Er_xAl_5O_{12}$ crystals by the method of adiabatic laser calorimetry. 1—Laser; 2—mechanical shutter; 3—lens with $f = 200$ mm; 4—diaphragm; 5—heat-insulating chamber; 6—control crystal; 7—test crystal; 8—copper-constantan thermocouple; 9—dc amplifier; 10—chart recorder; 11—calorimeter.

curve, before the arrival of the thermal wave from the ends of the sample, at[1] $t < (L/2)^2(1/6g)$.

The ends and lateral surface of the samples are treated to the point at which they are of purity classes 14 and 12, respectively.

The bulk absorption coefficient is found from the formula[3]

$$\alpha = \frac{c\rho\pi R^2}{P}\left(\frac{d\theta}{dt}\right)_{t=0}, \tag{2}$$

where ρ is the specific weight of the sample, $P = 1/2(n + 1/n)P_{\rm meas}$, $P_{\rm meas}$ is the laser power, n is the refractive index of the material, $(d\theta/dt)_{t=0}$ is the initial slope of the time dependence of the sample temperature θ, R is the radius of the sample, and c is the specific heat of the yttrium-erbium-aluminum garnet.

The error in the measurement of α is $\pm 5\%$.

Figure 2 is a plot of α against the length of the laser pulse. It can be seen from this

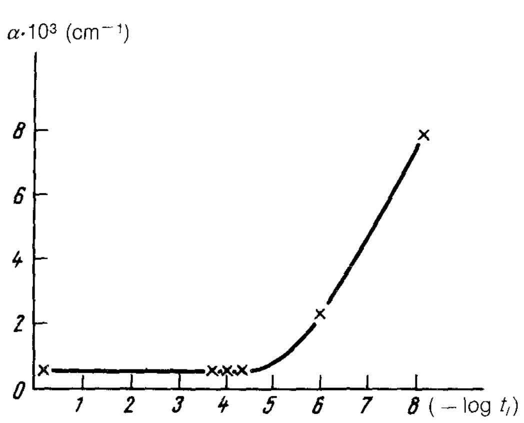

FIG. 2. Bulk absorption coefficient of the $Y_{3-x}Er_xO_{12}$ crystal versus the length of the pulse from the $Y_3Al_5O_{12}-Nd^{3+}$ laser.

figure that in the interval $t_l = 200$–$500\ \mu s$ the value of α remains essentially constant at $6\times10^{-4}\ cm^{-1}$. When the crystals are heated by light pulses with a length of $1\ \mu s$ and 10 ns, we observe an increase in α. The bulk absorption coefficients at $t_l = 1\ \mu s$ and $t_l = 10$ ns are 2.5×10^{-3} and $8\times10^{-3}\ cm^{-1}$, respectively.

The dependence of α on the length of the laser pulse is evidence that these crystals contain strongly absorbing defects.

If we assume that the thermal diffusivity of the defect and that of the crystal are comparable in magnitude ($g\cong10^{-2}\ cm^2/s$ for $Y_3Al_5O_{12}$), an estimate of the size (R) of the local defect from expression (1) yields $R\cong0.5$–$0.1\ \mu m$.

Let us estimate the average local absorption coefficient of the defects. The additional component of the bulk absorption due to the heated defects can be written as

$$\Delta\alpha_{meas} = \alpha_{loc} VN, \qquad (3)$$

where α_{loc} is the average local absorption coefficient of the defects, V is the volume of a defect ($V\cong8\times10^{-15}\ cm^3$), $\Delta\alpha_{meas} = \alpha(t_l = 10\ ns) - \alpha(t_l = 50\ \mu s)$, and N is the concentration of defects.

Danileĭko[7] estimated the concentration of clusters of a foreign phase which might be responsible for the laser destruction of $Y_3Al_5O_{12} - Nd^{3+}$ crystals. According to that study, the concentration of defects of this sort could be $\sim10^9\ cm^{-3}$. The local absorption coefficient of the defects can therefore reach values of $10^{-3}\ cm^{-1}$ or more. If we assume that the defects in $Y_{3-x}Er_xAl_5O_{12}$ crystals are metallic inclusions (with an absorption coefficient[5] $\sim10^5\ cm^{-1}$), then we estimate the defect concentration to lie in the range 10^6–$10^7\ cm^{-3}$.

This has been the first observation of absorption by impurity particles of submicron size in laser crystals. The technique developed here for measuring α, as the crystals are heated by laser pulses of various lengths, makes it possible to identify such defects and to analyze the nature of the nonlinear absorption without destroying the crystal.

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