INTERACTION OF LASER RADIATION WITH MATTER

PACS numbers: 42.65.Sf; 42.55.Rz; 81.05.Je; 81.05.Kf
DOI: 10.1070/QE2009v039n01ABEH013750

Generation of a bleaching wave in an ST-50-1 glass ceramics induced by a Nd : YAG laser

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***Abstract*. It is shown that upon exposing glass ceramics to laser radiation for which the initial polycrystalline phase is opaque, whereas the corresponding glass phase is transparent, the transparency oscillations can be produced due to the laser-induced phase transitions from the crystalline to amorphous state and vice versa, resulting in the propagation of a bleaching and darkening wave.**

Keywords: *glass ceramics, laser-induced phase transition, bleaching wave*.

1. Introduction

It is known that laser heating of glass ceramics induces variations in their structural phase state [1]. The local amorphisation (vitrification) of glass ceramics in the crystalline phase leads to local variations in its physical (density) and optical (transparency in the visible and near-IR ranges) properties.

These phenomena have been investigated in detail in works of Skiba's group (see, for example, [2]) and [1], where the amorphisation of a ST-50-1 polycrystalline devitrified glass exposed to the CO_2 laser radiation and its crystallisation upon less intense irradiation were analysed. The CO_2 laser wavelength (10.6 μm) lies within the fundamental absorption band of silicate glass ceramics and therefore they strongly absorb this radiation both in amorphous and crystalline states.

The situation is substantially different when glass ceramics is exposed to visible and near-IR radiation. The absorption ability of polycrystalline glass ceramics is quite high due to strong scattering and therefore it is rapidly heated in the irradiated region. As the melting temperature of glass ceramics is achieved, the polycrystalline structure disappears (crystals are melted) and scattering and absorption drastically decrease. As a result, the region where radiation is absorbed is displaced to the boundary of the leading melting front. Thus, the change in the optical properties of the material leads to a drastic change in the distribution of the absorbed light energy over depth, resulting in the nonmonotonic variation of the temperature and optical properties of the material.

Figure 1 presents the general view of the absorption region and the type of the motion of the phase-transition boundary in a glass ceramic plate exposed to radiation from CO_2 and Nd : YAG lasers.

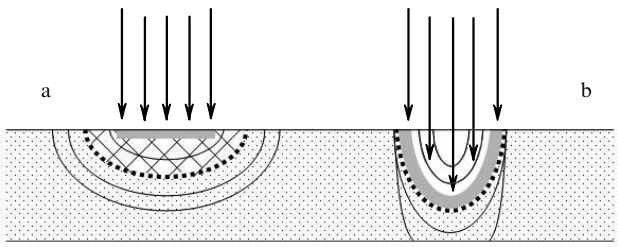

Figure 1. Principal scheme of the growth of the amorphous region in a ST-50-1 devitrified glass irradiated by CO_2 (a) and Nd : YAG lasers (b): positions of the amorphisation (transparency) front at different times (solid curves); the boundary of the region where temperature is equal to the melting temperature (squares); the region where temperature exceeds the melting temperature (hatched); and regions absorbing radiation (shown by grey).

This paper is devoted to the study of phase transitions in a CT-50-1 devitrified glass induced by a cw Nd : YAG laser.

2. Experiment

We investigated spatiotemporal variations in the transparency of glass ceramics caused by the formation and propagation of a bleaching (and darkening) wave in it. Processes proceeding in the region of glass ceramics irradiated by a laser were recorded with a video camera. Figure 2 shows the scheme of the setup [3]. Radiation from laser (*1*) (Nd : YAG or CO_2 laser) propagated through open shutter (*2*) and aperture (*3*) and was directed with the help of mirror (*4*) and ZnSe lens (*5*) to CT-50-1 devitrified glass sample (*6*) mounted on 2-axis stage (*7*) equipped with a heater. A part of laser radiation was directed to power meter (*8*). Probe radiation from He – Ne laser (*9*) passed through a modified region and was focused to photodiode (*11*). Variations in the probe radiation intensity gave information on temporal variations in the glass ceramics structure. The temperature and heating and cooling rates of the material irradiated by the laser were

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Received 1 November 2007; revision received 10 July 2008
Kvantovaya Elektronika **39** (1) 59 – 62 (2009)
Translated by M.N. Sapozhnikov

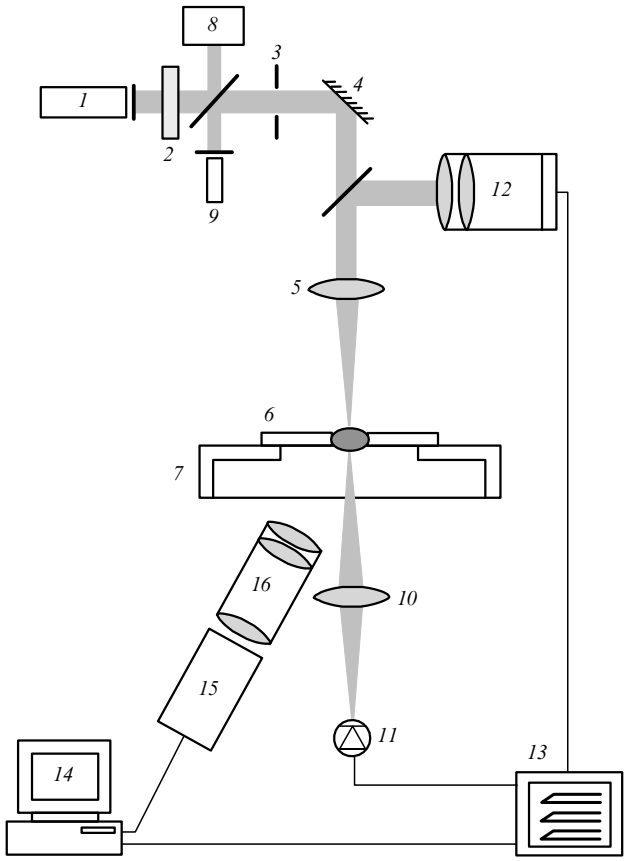

Figure 2. Scheme of the experimental setup for studying the motion of the bleaching front in glass ceramics: (*1*) Nd : YAG or CO_2 laser; (*2*) shutter; (*3*) aperture; (*4*) mirror; (*5, 10*) lenses; (*6*) sample; (*7*) 2-axis stage; (*8*) power meter; (*9*) He–Ne laser; (*11*) photodiode; (*12*) pyrometer; (*13*) oscilloscope; (*14*) computer; (*15*) video camera; (*16*) microscope.

measured with fast digital optical pyrometer (*12*). Photodiode (*11*) and pyrometer (*12*) were connected with oscilloscope (*13*) to transfer data from these devices to the monitor of computer (*14*). Video recording was performed with video camera (*15*) by using microscope (*16*).

We found that the bleaching of glass ceramics induced by radiation from the cw Nd : YAG laser principally differs from that caused by the CO_2 laser. One of the most important differences is the possibility of the formation of amorphous regions of large depth (exceeding their radius) upon irradiation by the Nd : YAG laser. This is determined by the displacement of the absorption zone inside the material to the melting-front boundary, unlike the case of irradiation by the CO_2 laser, when absorption occurs in the surface layer.

The most interesting effect observed upon exposing glass ceramics to radiation from the Nd : YAG laser was the appearance of oscillations of transmission in the irradiated region (Fig. 3). Such bleaching waves were observed in 0.6-mm-thick CT-50-1 plates irradiated by the 6×10^3 W cm^{-2} Nd : YAG laser beam of diameter 300 μm. Samples were heated up to 450 °C before irradiation. Note the presence of periods t_{st} of the stable phase state of the material between periods of the amorphisation (t_{am}) and crystallisation (t_{cr}).

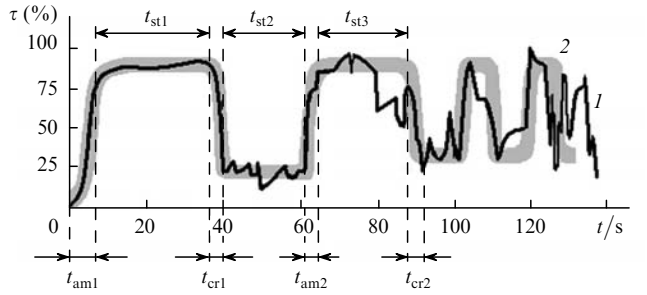

Figure 3. Change in the transmission of a ST-50-1 devitrified glass plate irradiated by a cw Nd : YAG laser (*1*) and the corresponding smoother dependence (*2*).

Our experiments gave the picture of variations in the transparency in glass ceramics induced by radiation from the cw Nd : YAG laser.

Radiation from the 10.6-μm CO_2 laser is always absorbed, both in the polycrystalline and amorphous phase, in a thin surface layer, and the melting front moves inside glass ceramics due to heating caused by the heat conduction. The temperature of the melt increases during irradiation, the maximum temperature being achieved on the glass ceramic plate surface. The melted region is also expanding in the radial direction during melting. After compete melting of the plate (its bleaching), the plate heating continues until the stationary temperature is achieved.

Irradiation by the 10.6-μm cw Nd : YAG laser causes the self-induced bleaching of a glass ceramic plate. First radiation is absorbed in the surface layer of the plate and produces its heating. The surface layer becomes transparent for incident radiation after melting. The region absorbing radiation moves from the surface to the polycrystalline part of glass ceramics behind the melting front. In this case, temperature in the melt and outside it does not exceed the melting temperature, achieving the maximum values (the melting temperature) directly on the bleaching front. As the deeper layers of glass ceramics are melted, the region absorbing radiation moves downwards. When the thickness of the remaining crystal layer of the plate under the melt becomes comparable with the penetration depth of radiation into glass ceramics ($1/\alpha = 105$ μm [1], where α is the absorption coefficient), the process slows down due to the decrease in absorption.

After melting of the glass ceramic plate over the entire depth, it becomes transparent for incident radiation and heating ceases. Then crystalline structures begin to form in the glass ceramic melt due to the gradual radial outflow of heat from the heated (melted) region. In this case, absorption drastically increases and transparency decreases. Glass ceramics again begins to absorb radiation and the process is repeated. Thus, the transmission of the plate oscillates.

3. Calculation

As shown above, we observed oscillations of the transmission of the glass ceramic plate, which we explain by phase transitions of the medium from the amorphous to crystalline state and vice versa. We studied this process analytically by using a physical model of processes of heating and cooling a glass ceramic plate with the changing absorption ability. We assumed in calculations that the

phase transition from the amorphous to crystalline state occurred abruptly (similar transitions were observed in GeSb films irradiated even by femtosecond laser pulses [4], although under our experimental conditions for a 0.6-mm-thick plate the transition occurs, of course, during some time, which can be estimated as $h^2/a \sim 1$ s (a is the thermal diffusivity of glass ceramics). This value was noticeably lower than the transparency oscillation period and therefore was neglected in calculations.

Analysis of the possibility of appearing of transparency oscillations in a glass ceramic plate showed that the most important factor determining the temperature kinetics was the radial heat removal from the irradiated region. Preliminary estimates showed that radiative and convective heat removal, the energy expenditure and heat release during phase transitions play only a minor role.

Calculations were performed by using the following assumptions:

(i) The temperature of a glass ceramic plate was assumed constant over the plate thickness. This is justified because the plate is thermally thin: our preliminary calculations showed that the temperature drop along the plate thickness after 10 s did not exceed 17 %. The plate was assumed melted in calculations if the thickness-averaged temperature at the centre of the irradiated region achieved the melting temperature (upon irradiation by the Nd : YAG laser, the temperature in the melted region is approximately equal to the melting temperature, while outside this region the temperature is lower than the melting temperature). It was assumed similarly that the plate was crystallised if its thickness-averaged temperature at the centre of the irradiated region was decreased down to the crystallisation temperature.

(ii) The heat spent for melting and the heat release upon crystallisation were neglected. This assumption, of course, somewhat distorts quantitative characteristics, but the qualitative picture of the process is preserved. We used this assumption because of the lack at our disposal of reliable data about specific melting and crystallisation energies and about variations of these quantities during a periodic melting-crystallisation process related to the effect of incomplete crystallisation observed in experiments.

(iii) The absorption ability was assumed constant during each stage of the process. In reality, it varies somewhat due to a rather large penetration depth ($\sim$100 μm) of the 1.06-μm radiation into glass ceramics and the bulk melting and crystallisation.

(iv) The radiation power density was assumed constant within the irradiated region. This assumption does not reduce the generality of the results obtained. In addition, the method of calculation allows one to take into account any spatial distribution of the radiation power density.

(v) Heat losses to the environment were neglected.

(vi) The size of the plate was assumed large enough.

Thus, variations in the structural and optical properties of the irradiated glass ceramic plate (changes in its phase state and absorption ability) are determined by the temperature T of the plate averaged over its thickness at the centre of the irradiated region.

Temperature T can be found by solving the problem of heating the matter absorbing radiation up to the melting temperature T_m. After achieving the temperature T_m, absorption ceases (due to the amorphisation of the devitrified glass) and the matter is cooled down to the crystallisation temperature T_{cr} due to radial heat removal, then it crystallises, absorption drastically increases (up the previous or close value), and the process repeats. Therefore, this process is nonlinear, i.e. the action of radiation, determined by the absorption ability, depends on temperature, while the kinetics of the absorption ability depends on the time during which temperatures T_m and T_{cr} are achieved.

The problem was solved by the method of sources [5]. The results of the solution are presented in Table 1, where

$$F = 2\pi \int_{r=0}^{r_*} \frac{q(1-R)}{\rho c h} \frac{\exp\{-r^2/[4a(t-t')]\}}{4\pi a(t-t')} r\mathrm{d}r;$$

$q = P/(\pi r_0^2)$ is the incident radiation power density; P is the radiation power; r_0 is the radius of the irradiated region; R is the reflection coefficient of the plate; ρ, c are the density and heat capacity of the glass ceramic plate; r is the distance from the centre of the irradiated region; r_* is the radius of the heat release region in the irradiated plate; γ is the

Table 1. Results of the solution of the problem on the local heating of a glass ceramic plate.

Cycle number	Process stage	Integral expression for temperature	Time interval t	Temperature
1	heating and melting (absorption of radiation)	$T = \int_0^t F\mathrm{d}t' + T_0$	$0 \leqslant t \leqslant t_{m1}$	$T = \frac{q(1-R)t}{\rho c h}\left[1 - E_2\left(\frac{r_0^2}{4at}\right)\right] + T_0$
	cooling and crystallisation (glass ceramic is transparent)	$T = \int_0^{t_{m1}} F\mathrm{d}t' + T_0$	$t_{m1} \leqslant t \leqslant t_{cr1}$	$T = \frac{q(1-R)t}{\rho c h}\left\{1 - E_2\left(\frac{r_0^2}{4at}\right) + \frac{t-t_{m1}}{t} E_2\left[\frac{r_0^2}{4a(t-t_{m1})}\right]\right\} + T_0$
2	heating and melting (absorption of radiation)	$T = \int_0^{t_{m1}} F\mathrm{d}t' + \int_{t_{cr1}}^{t} F\mathrm{d}t' + T_0$	$t_{cr1} \leqslant t \leqslant t_{m2}$	$T = \frac{q(1-R)t}{\rho c h}\left\{1 - E_2\left(\frac{r_0^2}{4at}\right) + \frac{t-t_{m1}}{t} E_2\left[\frac{r_0^2}{4a(t-t_{m1})}\right] + \frac{t-t_{cr1}}{t}(1-\gamma) - \frac{t-t_{cr1}}{t}(1-\gamma) E_2\left[\frac{r_0^2}{4a(t-t_{cr1})}\right]\right\} + T_0$
	cooling and crystallisation (glass ceramic is transparent)	$T = \int_0^{t_{m1}} F\mathrm{d}t' + \int_{t_{cr1}}^{t_{m2}} F\mathrm{d}t' + T_0$	$t_{m2} \leqslant t \leqslant t_{cr2}$	$T = \frac{q(1-R)t}{\rho c h}\left\{1 - E_2\left(\frac{r_0^2}{4at}\right) + \frac{t-t_{m1}}{t} E_2\left[\frac{r_0^2}{4a(t-t_{m1})}\right] + \frac{t-t_{cr1}}{t}(1-\gamma) - \frac{t-t_{cr1}}{t}(1-\gamma) E_2\left[\frac{r_0^2}{4a(t-t_{cr1})}\right] + \frac{t-t_{m2}}{t}(1-\gamma) E_2\left[\frac{r_0^2}{4a(t-t_{m2})}\right]\right\} + T_0$

residual transmission of the plate; T_0 is the initial temperature; t_m is the time during which the melting temperature is achieved; and $E_2(z)$ is the integral exponential function. Due to strong scattering of radiation from the Nd : YAG laser in glass ceramics, we assumed in calculations that $r_* = r_0 + (1 \div 2)\delta$, where δ is the penetration depth of radiation to the crystalline phase of glass ceramics.

Figure 4 shows the characteristic kinetics of temperature. The calculation was performed for $P = 4.2$ W, $T_0 = 450\,^\circ$C, $r_* = 0.45$ mm, and $h = 0.6$ mm. The residual transmission $\gamma = 0.1$ of the plate after the previous cycle was taken into account.

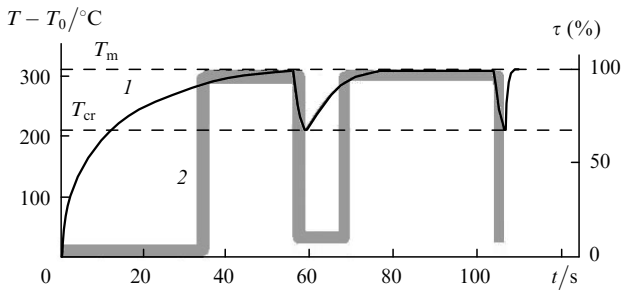

Figure 4. Calculated change in the temperature T of the ST-50-1 plate averaged over the plate thickness at the centre of the irradiated region (*1*) and the corresponding qualitative time dependence of the plate transmission (*2*).

The calculations have shown that in the certain range of acting parameters the abrupt change in the absorption ability of the plate leads to oscillations of transmission. The kinetics of this process, i.e. the duration of absorption-transmission cycles is determined to a great degree by the radiation power, the scattering efficiency, and the residual transparency observed after the first crystallisation. The duration of the cycles varies. For experimental parameters used in calculations, it decreases from cycle to cycle.

4. Conclusions

The results of the action of the cw 1.06-μm radiation from a Nd : YAG laser on glass ceramics considerably differ from the action produced by a CO_2 laser. This is explained by the fact that the optical properties of glass ceramics in the near-IR region are determined by its phase state: the crystal phase absorbs radiation, whereas the melted phase transmits it. As for far-IR radiation of a CO_2 laser, its absorption is almost the same in the crystal and melted phases of glass ceramics.

Thus, optical and thermal effects observed upon irradiation of glass ceramics by the Nd : YAG laser substantially differ from those observed upon irradiation by the CO_2 laser. The main properties of the process in this case are determined by nonlinear heating due to interrelation between the absorption ability and temperature of glass ceramics.

One of the most dramatic examples of such nonlinearity is oscillations of the transmission of a glass ceramic plate irradiated by a Nd : YAG laser, which were simulated mathematically.

Note that such effects should be also observed in other glass ceramics during crystal-amorphous state phase transitions, which are accompanied by the formation of a transparent phase, for example, in Foturan glass ceramics and semiconductors during laser-induced melting [6], etc.

Acknowledgements. This work was supported by the program 'Leading Scientific Schools of the Russian Federation' (Grant No. 02.512.11.2136), the Russian Foundation for Basic Research (Grant No. 07-02-00887-a), State Contract RNP 2.1.1784 and partially by NATO Grant No. CBP.EAP.CLG 982748.

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