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Transmission of laser radiation by absorbing liquids

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***Abstract*. Transmission of radiation from a free-running 2.94-μm Er^{3+}:YAG laser by strongly absorbing liquids is studied. It is shown that transmission of radiation is caused by the development of a channel in the liquid, which is formed by a laser pulse itself. The transmission in water, ethanol, and glycerol is studied as a function of the laser pulse energy and the liquid thickness.**

Keywords: *radiation propagation, radiation self-action, water, erbium laser*.

1. Introduction

The effect of transparency of optically thick liquids (H_2O, C_2H_5OH) irradiated by IR pulses from a 2.94-μm Er^{3+}:YAG laser was first observed in paper [1]. The absorption coefficient α of hydroxyl-containing liquids, to which water and ethanol belong, at 2.94 μm amounts to 10^4 cm^{-1} and is determined by the concentration of hydrogen bonds in the liquid. Although this radiation should be almost completely absorbed by in a layer of thickness smaller than 1 μm (α^{-1}), the transparency of liquids was experimentally observed in layers of thickness up to 100 μm [1–3]. Three possible mechanisms of transparency were proposed [1]: (i) absorption saturation in a two-level quantum system caused by the action of intense laser radiation; (ii) a change in the medium density upon its laser evaporation; and (iii) a shift of the absorption spectrum upon heating water to high temperatures caused by the rupture of hydrogen bonds. However, neither of these mechanisms was experimentally confirmed, and the mechanism of transmission of intense laser radiation by absorbing liquids still remains open.

The transmission of laser radiation through thin water layers was studied experimentally in papers [3, 4]. It was assumed in both these papers that laser pulses produce a channel along which radiation propagates in absorbing water. However, channels produced by laser pulses propagating in water were not directly observed. It was assumed that the channels are formed only due to evaporation of water upon absorption of the laser pulse.

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At the same time, the formation of channels (whose length is much greater than the radiation absorption depth) upon irradiation of absorbing media by intense laser radiation is a well known experimental fact. For example, the production of a channel in glycerol upon its irradiation by a cw CO_2 laser was observed in paper [5]. The question about the threshold conditions of formation of a channel in a melt (for example, in laser cutting of metals) caused by the pressure of the recoil vapour of a material being evaporated on the liquid–vapour interface (the so-called dagger melting) was considered in detail in review [6].

The dynamics of interaction of intense laser radiation with absorbing liquids upon laser ablation was studied in many papers [7–9], in particular, the penetration depth of laser radiation in liquids was measured [10]. The dynamics of the channel development in water and gelatine irradiated by a free-running pulsed erbium laser was studied in paper [11], where the rates of formation of channels achieving a few tens metres per second were investigated. However, the mechanism of transparency of liquids caused by laser radiation has not been related so far to the formation of channels in the liquid, and has not been experimentally studied from this point of view.

In this paper, we investigated the action of radiation from an erbium laser on absorbing liquids and showed for the first time that the radiation can propagate through optically thick liquid layers due to the formation of through channels in them. We studied experimentally the transmission of radiation in water as a function of the laser pulse energy and the liquid thickness and compared this transmission with those in ethanol and glycerol. The dynamics of interaction of the laser beam with the liquids was studied using a video camera.

2. Experimental

We used in our experiments a free-running 2.94-μm Er^{3+}:laser with a pulse energy of 0.5 J and a pulse duration (FWHM) of 150 μs. The laser radiation was focused with a fluorite lens into a spot of diameter ~ 200 μm (at the 1/e level), which changed insignificantly over the layer thickness. The radiation intensity at the focus was 10 MW cm^{-2}. The liquids were irradiated in a cell with the IR silica bottom, which was transparent at the laser wavelength. We studied the transmission of radiation in water, ethanol, and glycerol layers of different thickness. The liquids were irradiated from above, through the free liquid surface, and from below, through the transparent cell bottom, normally to the surface.

We measured the energy of the transmitted laser pulse as a function of its initial energy and the liquid layer thickness. The shape of the incident and transmitted pulses was detected with an avalanche photodiode and a storage oscilloscope. The shape of the laser spot before and after propagation of radiation through the liquid layer was studied by the contour of an imprint produced by the laser beam on a thermosensitive paper.

The dynamics of action of laser radiation on the liquid was recorded with a video camera (the exposure time was 20 ms), with the subsequent processing of video images with the help of standard graphic editors. Although the laser radiation was invisible, the processes under study were visualised due to scattering of radiation from a pump flashlamp by vapours and inhomogeneities of liquids. The flashlamp for pumping an erbium laser emits a characteristic spectrum in the visible region, which allows the timing of processes under study with the laser pulse.

3. Experimental results

A laser pulse irradiating the absorbing liquid through its free surface or through the transparent bottom of a cell is partially transmitted by the liquid. We studied in most detail the transmission of a laser pulse through a water layer irradiated from above. Figure 1 shows the dependences of the water transmission on its layer thickness for different energies of the laser pulse. We observed under our experimental conditions the transmission of laser radiation through a water layer of thickness up to 8 mm. A fraction of the laser energy transmitted through the water layer increases with decreasing layer thickness and with increasing laser pulse energy (Fig. 2). A comparison of the water transmission with those of ethanol and glycerol shows that water is less transparent under the same experimental conditions (Fig. 3). Within the experimental error, the transmission of laser radiation in ethanol and glycerol is the same.

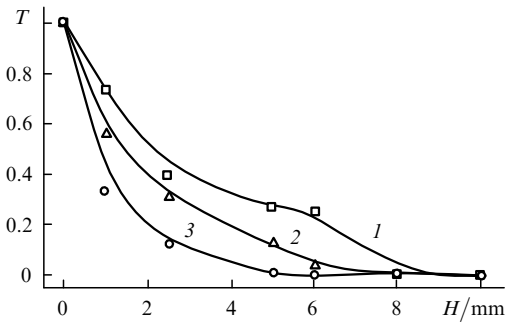

Figure 1. Dependences of the transmission of water on its layer thickness for laser-pulse energies equal to 0.5 (*1*), 0.3 (*2*), and 0.1 J (*3*).

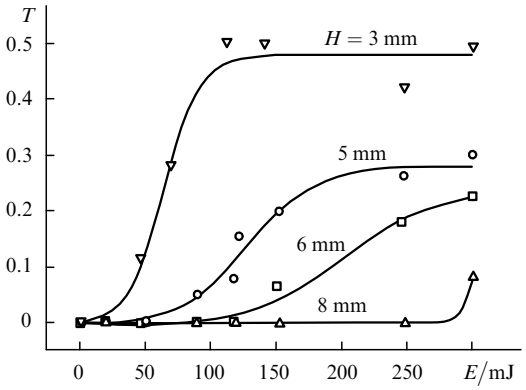

Figure 2. Dependences of the transmission of water on the laser-pulse energy for water layers of different thickness.

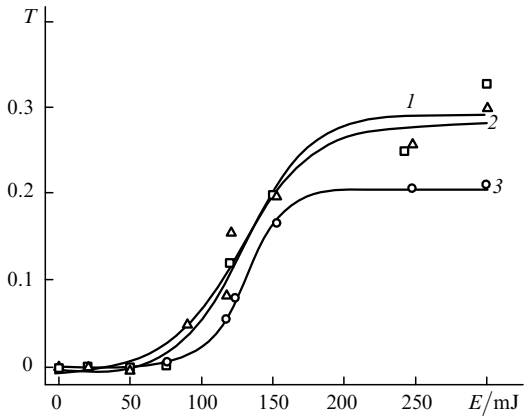

Figure 3. Dependences of the transmission of glycerol (*1*), ethanol (*2*), and water (*3*) on the laser-pulse energy for the liquid layer thickness of 5 mm.

The oscillograms of the incident and transmitted laser pulses are presented in Fig. 4. One can see that the duration of the transmitted laser pulse decreases with decreasing energy. The shape of the laser spot after the propagation of the laser beam through a liquid layer does not change significantly.

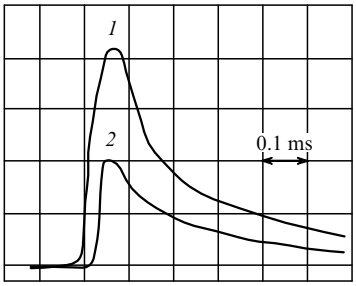

Figure 4. Oscillograms of the laser pulse before (*1*) and after (*2*) its propagation through a water layer of thickness 5 mm. The pulses are arbitrarily located along the time axis.

According to the video data, the laser beam incident on the liquid normally to its free surface produces a channel in the liquid. Figure 5 shows the channels produced by a 0.5 J-laser pulse in water and glycerol. The characteristic diameter of channels produced in liquids by a laser beam of diameter ~ 200 μm was 1 mm. Similar channels appeared upon irradiation of liquids from below. After the laser pulse termination, a channel decomposes into many bubbles of diameter 1–2 mm. The propagation of the laser pulse through the liquid layer was accompanied, both upon irradiation from above and from below, by the formation of a liquid jet directed from the free surface of the liquid (Fig. 6).

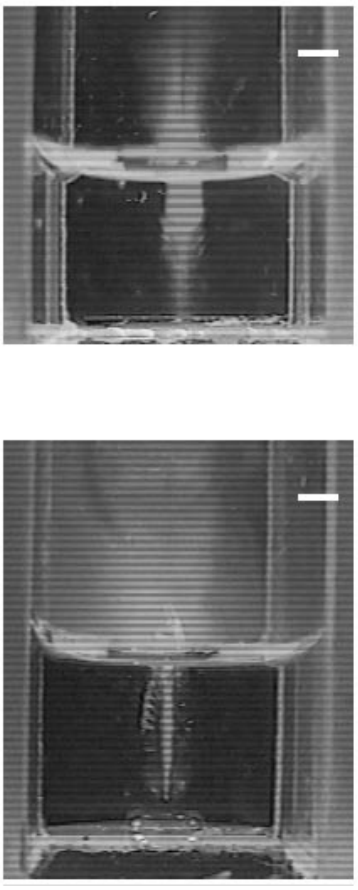

Figure 5. Channels formed during the propagation of the laser pulse through water (a) and glycerol (b). The mark corresponds to 3 mm.

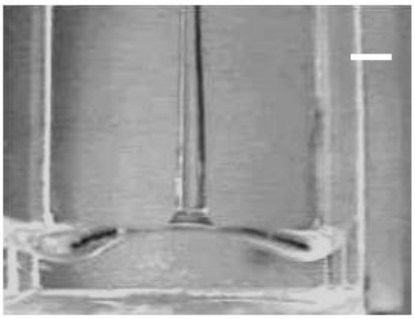

Figure 6. Jet formed upon irradiation of water through the transparent bottom of the cell. The mark corresponds to 3 mm.

Upon irradiation of the free liquid surface from above, the jet appears after the pulse propagation during the channel collapse, whereas upon irradiation of the liquid from the transparent bottom of the cell, the jet appears during the development of the channel. The jet continues to move upward after the laser pulse termination. Liquid jets with the highest ratio of the height to diameter were observed upon irradiation of glycerol from the bottom. In this case, the height of the jet was 10 cm (Fig. 7), whereas the height of the jet in water or ethanol did not exceed 1–2 cm. At the final stage of its development, the jet decomposed into drops falling on the liquid surface.

During the action of the laser pulse on the liquid, a directed vapour jet appeared over the liquid surface. In water and ethanol, this jet propagates predominantly perpendicular to the liquid free surface. The intense scattering of radiation from the pump flashlamp by the vapour jet shows that the vapour is supersaturated. Such vapour noticeably absorbs the laser pulse, which is confirmed by the formation of a channel in it (Fig. 5a).

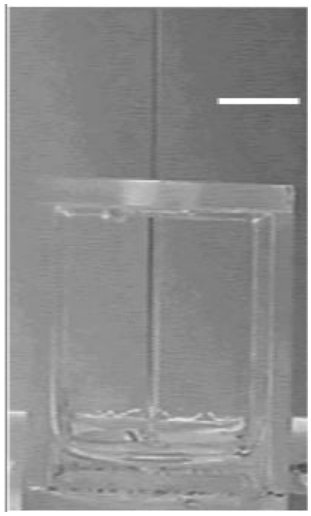

Figure 7. Jet formed upon irradiation of glycerol through the transparent bottom of the cell. The mark corresponds to 1 cm.

4. Discussion

The results obtained above demonstrate that the propagation of a laser pulse through strongly absorbing liquids is directly related to the formation of a channel in them. Long (1-ms) laser pulses that we employed in our experiments produced rather long channels of length up to 1 cm. The energy that is required for the heating and evaporating of liquids (water and ethanol) within a volume irradiated by a laser beam is greater than the laser pulse energy. Therefore, the liquid volume that can be evaporated by the laser pulse is incomparable with the volume of the channel being formed, and, hence, the mechanism of the channel formation differs from trivial evaporation.

The action of high-power laser radiation on a strongly absorbing liquid is related to the intense surface evaporation, resulting in the motion of the liquid under the reactive action of recoil vapours [6, 12]. A similar mechanism lies at the basis of dagger melting, which consists in a sharp increase in the melting depth of a metal when the laser radiation intensity exceeds a certain threshold [6]. The intense evaporation from the liquid surface caused by laser radiation results in the instability of a plane evaporation front [13] and in a rapid decrease in the surface curvature during the development of the channel deep inside the liquid.

Using the concepts developed in paper [12], we can estimate the threshold intensities q at which a channel can be formed in various substances due to the action of recoil vapours. The values of q for liquids are several orders of magnitude smaller than for metals. For example, $q \sim 3 \times 10^2$ W cm^{-2} for water, whereas $q \sim 10^5$ and 10^6 W cm^{-2} for low- and high-melting metals, respectively. This means that in liquids the channel is formed and radiation

propagates in it at moderate laser radiation intensities compared to metals.

The rate of the channel propagation in liquids is determined by the radiation intensity at the liquid-vapour interface, so that it should decrease with increasing channel depth. On the other hand, the evaporation conditions change as the channel is developing deep inside the liquid, which can lead to the stationary development of the channel in the liquid [11].

Let us estimate the mean rate V of the channel propagation in the liquid under the action of a laser pulse. The maximum energy that can be spent for the propagation of a liquid layer of thickness h is close to the total energy of the beam. Therefore, the development time of the channel in a layer is close to the total duration of the laser pulse $\tau \sim 1$ ms. For $h \sim 1$ cm, the mean propagation rate is ~ 10 m s^{-1}. The theoretical estimate of the channel development rate, taking into account both the dynamics of the evaporation front and the dynamics of the liquid motion is complicated and requires a combined solution of the thermal conductivity equation and the Navier–Stokes equation. Such calculations are not available in the literature known to us. If we assume that the rate of the channel development is equal to the propagation rate $V(T)$ of the evaporation front, then this rate is described by the expression [6]

$$V(T) = \frac{P(T)}{\rho}\left(\frac{A_\mathrm{m}}{2\pi N_\mathrm{a} T}\right)^{1/2},$$

where $P(T)$ is the vapour pressure; ρ is the liquid density; A_m is the atomic weight; and T is the liquid temperature. The estimate of the velocity of propagation of the evaporation front of water for $P(T) = 1$ atm and $T = 373$ K gives $V \sim 0.1$ m s^{-1}, which differs by two orders of magnitude from the experimental propagation velocity of the channel in liquids. Such a difference of the evaporation rate from the mean velocity of channel propagation along the beam axis suggests that a change in the curvature of the liquid surface caused by recoil vapours substantially affects the dynamics of the liquid motion.

One can see from the oscillogram in Fig. 4 that a fraction of the energy of a beam propagating through a layer of strongly absorbing liquid is spent for the formation of the channel, while the rest of the beam energy propagates along this channel. A lower transmission of radiation in water compared to other liquids studied here is explained by its high specific vaporisation heat, which equals 2.26×10^3 J g^{-1} compared to 0.84×10^3 J g^{-1} for ethanol and 0.82×10^3 J g^{-1} for glycerol. For this reason, the energy that is required for producing a channel in a layer of water is greater than that in other liquids, all other conditions being equal.

Note that the formation of the channel in liquids irradiated by intense laser pulses results in a change in the optical characteristics in the irradiated region. Unlike liquids, water vapours, which fill the channel during its development, are virtually transparent for radiation from an erbium laser because of the dissociation of hydrogen bonds upon the liquid-vapour phase transition and a decrease in their concentration upon further heating of vapours [14, 15]. For this reason, we can formally explain the transparency of liquids by a change in the density of liquids upon their laser evaporation [1]. Dense vapours formed in the channel and consisting of liquid microdrops can noticeably absorb laser radiation, thereby contributing to the total transmission of a liquid layer.

The results obtained above suggest that the formation of the channel can play an important role, resulting in the transparency of liquids observed upon their irradiation by shorter pulses, for example, nanosecond and picosecond pulses. In particular, in experiments performed in papers [1, 3, 16], a cavity, which is similar to a channel, can be produced in liquids upon their irradiation by laser pulses. When the thickness of the liquid layer (the cell thickness) is smaller than the beam diameter, a channel is degenerated to a vapour bubble through which laser radiation propagates.

Thus, we have shown in this paper that the propagation of laser radiation through strongly absorbing liquid is caused by the formation of a channel in the liquid produced by the laser pulse itself. The channel is filled by vapours, which are transparent for laser radiation. The channel is produced due to pressing through the liquid surface caused by the vapour recoil momentum (the dagger regime). This mechanism of the channel formation is energetically more efficient than evaporation of the entire liquid in a volume irradiated by the laser beam.

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