

INTERACTION OF LASER RADIATION WITH MATTER

PACS numbers: 47.80. –v; 42.62. –b; 42.55.Ks
DOI: 10.1070/QE2009v039n02ABEH013947

Temporal structure of an electric signal produced upon interaction of radiation from a HF laser with the bottom surface of a water column

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Abstract. **Generation of an electric signal is investigated when a HF-laser pulse interacts with the lower surface of a water column in a cell with a bottom transparent to laser radiation, while the upper surface of the water column remains open. The electric signal exhibits a temporal structure of two spikes spaced by time τ which is linearly dependent on the laser output energy. It is found that the value of τ (up to 1.3 ms) is an order of magnitude greater than the time during which the vapour pressure in a cavity produced due to the volume explosive boiling of water in the exposed area is greater than the atmospheric pressure. The second spike was determined to appear upon the collapse of the vapour cavity. A mathematical model is constructed that explains the motion of the water column above the vapour cavity taking into account the temporal evolution of the vapour pressure above it. It is shown that the prolonged lifetime of the vapour cavity after the decrease in the vapour pressure down to the atmospheric value is caused by the inertial motion of the water column acquiring the velocity at the initial stage of the cavity expansion. The calculated time of the water column motion agrees well with the experimental time interval between the spikes of an electric signal.**

Keywords: *nonchain HF laser, water, charge separation, volume explosive boiling, vapour cavity.*

1. Introduction

The effect of generation of an electric signal, when IR-laser radiation having the power density below the plasma-formation threshold interacts with a water surface, was discovered in [1]. In these experiments, a cell with water was placed between the plates of an uncharged capacitor. When the water surface was irradiated by a $Cr^{3+}:Yb^{3+}:Ho^{3+}$: YSGG laser emitting 10-mJ, 150-ns pulses at 2.92 μm, the potential difference across the capacitor plates was detected to be up to 10 mV. To explain the observed effect, the authors of [1] suggested that the electric signal is produced due to the growing degree of water dissociation in a thin (about 1 μm) overheated surface layer and charge separation in it caused by the difference in the diffusion rates of H^+ and OH^- ions. The authors of paper [2] used this assumption to construct a mathematical model of charge separation for the case when a laser pulse acts on a water surface that is in contact with a plate of a material transparent to laser radiation (‘closed’ water surface). The model takes into account explosive water boiling during which a vapour cavity is produced, the opposite sides of the cavity acquiring charges of different signs because of diffusive separation of charges during heating.

It follows from the calculations [2] that it is the potential difference at the boundaries of the vapour cavity that should contribute most to the observed electric signal. Pulsed IR lasers (much more powerful than those used in [1]) – a nonchain electric-discharge chemical HF laser and CO_2 laser pumped by a volume self-sustained discharge – were used in [3, 4] to investigate the generation of electric signals. Electric signals with the amplitude over 15 V were detected upon interaction of laser radiation with both an open (free) and closed water surface plate, which is transparent in the IR region. The research made it possible to find a relation between the generation of electric signals and the volume explosive boiling of water and to give explanation of the effect at a qualitative level.

It was found that the characteristics of the electric signal observed in the experiments with the closed water surface [3, 4] depend on many factors, in particular, the force with which the fused-quartz window used to deliver radiaiton was pressed against the water cell. This fact indicated the possible overlap of the electric signal generated directly in water by signals arising from, for example, striction effects in dielectric elements of the cell (e.g., in dielectric tie rods used for pulling the cell together) whose construction may experience pressure jumps of up to 150 atm [5]. To avoid this kind of noise, we investigated in this paper the effect of electric signal generation upon interaction of radiation from a nonchain HF laser with a closed water surface under conditions when the surface of the water column opposite the irradiated surface remains open.

2. Experimental

We studied distilled water with the resistivity $\rho \approx 10\ \mathrm{M\Omega\ cm}$ at temperature 20 °C. The scheme of the experimental setup is given in Fig. 1a. The cell was made of a fragment of a fused-quartz tube with the inner diameter 35 mm and length $l = 30 - 80$ mm. A 9-mm-thick window of IR-type quartz was welded to one end of the tube. The height d of the water column varied from 10 to 80 mm. Radiation from a nonchain electric-discharge HF laser was

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Received 29 July 2008; revision received 15 September 2008
Kvantovaya Elektronika **39** (2) 179 – 184 (2009)
Translated by M.V. Politov

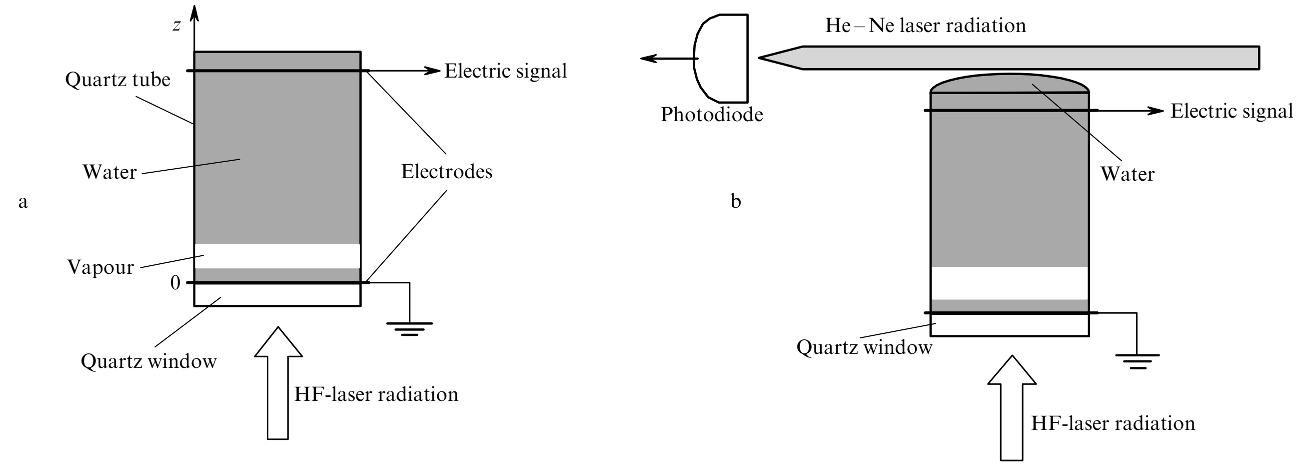

Figure 1. Scheme of the experimental setup (a) and scheme for monitoring the motion of the open water surface (b).

coupled into the cell from the bottom (closed-surface irradiation conditions [5]) and the top water surface remained open unlike the experimental conditions in [3, 4]. The energy density distribution W of laser radiation with respect to the radius r of the focusing spot on the surface of the object under study is well approximated by a Gaussian $W(r) = W_0 \exp(-r^2 a^{-2})$, where $a = 8.5$ mm. The pulse width at half maximum τ_p was 140 ns. The maximum energy E on the water surface immediately behind the quarts window was 1.3 J. The spectral selection of the HF-laser output was not performed. Two ring-shaped electrodes of width 3 mm, one round the bottom weld and the other round the upper rim, were used to read out the electric signal. As in [3], the electric signal was fed to an osillograph through a voltage repeater.

In some experiments we controlled the motion of the open water surface caused by the action of laser radiation. For this purpose (as shown in Fig. 1b) we directed a thin beam (about 1 mm in diameter) of a He – Ne laser above the cell rim so that it barely touched the surface of the convex water meniscus. The rise of the open water surface under the action of a laser pulse leads to a partial overlap of the He–Ne-laser beam and a decrease in the signal detected by a photodiode. We also measured the time profiles and amplitudes of a sound wave generated in the region where HF-laser radiation is absorbed (radiation is absorbed in a thin 1-μm water layer near the surface of the quartz window), at different distances from this region depending on the laser pulse energy. The method of such measurements is explained in detail in paper [5].

3. Experimental results and discussion

Figure 2 presents the electric signal oscillograms for different laser output energies E obtained in experiments with a 30-mm cell for the water column of height $d = 30$ mm. One can see that the electric signal has a rather complicated structure with two pronounced peaks. In this research the primary attention was paid to the nature of the second electric signal peak. The analysis of the processes leading to the appearance of high-frequency oscillations on the electric signal envelope and the processes determining the temporal structure of the first peak is the topic of our next paper.

It is interesting to note (Fig. 2) that the second peak is produced with a large time delay τ with respect to the laser pulse, the time delay increasing with increasing the pulse energy. For the maximum radiation energy $E = 1.3$ J in our experiments, the value of τ achieves 1.2 ms. As shown in the

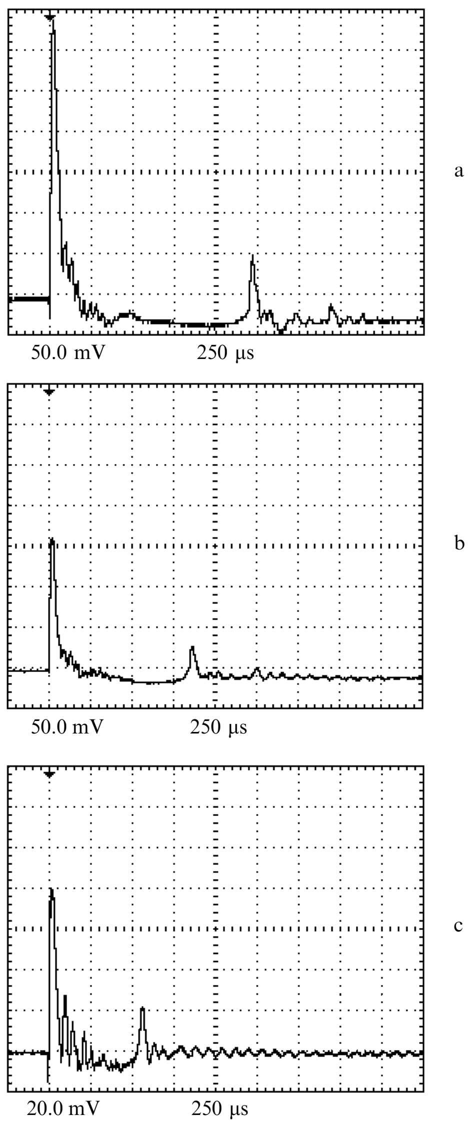

Figure 2. Oscillograms of the electric signal for $l = d = 30$ mm, $E = 1.3$ (a), 1.0 (b) and 0.75 J (c).

model described below, this value is manifold higher than the time during which the pressure in the vapour cavity produced upon volume explosive boiling of water in the irradiated region exceeds the atmospheric pressure.

Figure 3 shows the amplitude A of the first electric signal peak and time interval τ between the peaks as functions of the pulse energy E for $l = d = 30$ mm. One can see that the dependence of τ on E is described by a linear function and the minimum energy $E_{\min} \approx 0.23$ J at which the electric signal can still be detected, as in [3], is close to the threshold of explosive boiling of water measured under the same conditions of radiation focusing on the surface. It follows from Figs 2 and 3 that the change in the experimental conditions (irradiation of the closed water surface) compared to those used in [3] leads not only to the appearance of the second peak in the electric signal, but also to the variation in the sign of the signal (the first peak) and a decrease in its amplitude by almost an order of magnitude given the same E and the focusing spot size.

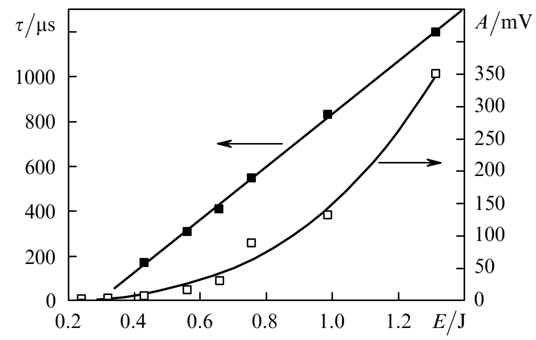

Figure 3. The amplitude A of the first electric signal peak and time interval τ between the peaks as functions of the laser output energy E for $l = d = 30$ mm.

It is natural to relate this specific features of the electric signal detected in this paper (as compared to that observed in [3, 4]) to the changed experimental conditions, namely, the appearance of the open water surface lying opposite the surface exposed to laser radiation. We can suggest that there is some correlation between the movements of the open surface, which accompany the interaction of laser radiation with water in a thin layer near the input window, and generation of the electric signal. Figure 4 shows oscillograms of the photodiode output (see Fig. 1b) and electric signal for $l = 50$ mm, $d = 51$ mm (with the convex meniscus taken into account) and $E = 1.25$ J. The decrease in the output of the photodiode corresponds to a rise of the water surface, while the increase in the output – to its lowering (reverse motion) (Fig. 1b). One can see from Fig. 4 that the surface begins rising when the sound wave reaches it, which is shown as a narrow dip in the photodiode output oscillogram (Fig. 4b). When the pulse energy $E = 1.25$ J, the surface achieves its maximum height within 500 μs (with respect to the laser pulse), this time interval decreasing with decreasing E. During the reverse motion (Fig. 4a) the signal detected by the photodiode demonstrates local maxima corresponding to the change in the direction of the open water surface motion. These maxima correlate with drastic changes in the electric signal. Apart from the strongly pronounced peak, called the second one above, we can also see irregular spikes after the second peak, which do not

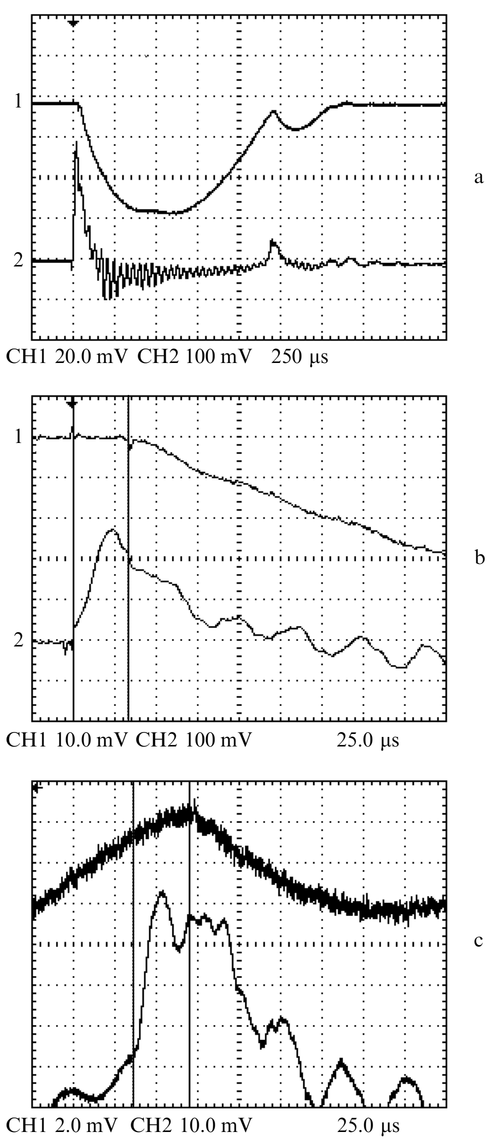

Figure 4. Oscillograms of the photodiode output (channel 1) and electrical signal (channel 2): the entire electrical signal, the first (b) and second (c) electric signal peaks.

necessarily accompany each laser pulse, if we compare the oscillograms of the photodiode output and electric signal.

Thus, all the observed peaks of the electric signal, which follow the first peak with a large time delay, are generated during the reverse motion of the open water surface. It is obvious that the arrival of the shock-wave distributions at the water surface is responsible for drastic changes in the direction of the surface motion, which are observed in this case, and their correlation with the peaks of the electric signal. It follows from Fig. 4a that the local maxima in the photodiode output are delayed with respect to electric signal spikes. The delay $t_{\mathrm{del}} \approx 34$ μs (Fig. 4c) is close to the time the sound travels from the bottom quartz window to the open water surface (Fig. 4b). Thus, the second peak in the electric signal is produced in the processes which develop near the quartz window surface, probably, during the collapse of the vapour cavity (the opposite boundaries of which, according to [2, 3], should have charges of different signs) formed at the early stage of the volume explosive boiling of water upon laser irradiation. This result indicates that the lifetime of this cavity is much longer than the time of vapour cooling down [2, 5] during which the pressure in the cavity exceeds the atmospheric pressure.

Consider the processes which can make the vapour cavity near the quartz window live longer and the second peak of the electric signal appear with a large delay after the first peak. The motion of the water column in the quartz cell open from above upon irradiation by HF-laser pulses can be tentatively divided in three stages.

The first stage involves the heating of a thin (about 1 μm) water layer near the surface of the quartz window (bottom) of the cell (see Fig. 1a) by HF-laser radiation. Because of heat dissipation through the quartz–water interface, the temperature profile $T(z,t)$ with the maximum not at the interface but some distance away from it (in the body of water) is produced in water. A rise of temperature in the overheated layer shifts the dissociation equilibrium of water towards growing concentrations of H^+ and OH^- ions. The presence of large temperature gradients during the heating of water by laser pulses leads to the appearance of concentration gradients and diffusion flows of H^+ and OH^- ions. Because the diffusion coefficient of H^+ ions is almost twice larger than that of OH^- ions, an initial separation of charges should take place in water: the area near the temperature maximum becomes depleted of H^+ ions, i.e. acquires a negative charge, while the areas near the interface and farther in the water body become positively charged [2].

When the threshold temperature is achieved $T_{\rm th} \approx 0.9T_{\rm c} = 583$ K [6] (where $T_{\rm c} = 647$ K is the critical temperature), the volume explosive boiling of water occurs in the maximum of the temperature profile and a vapour cavity with oppositely charged boundaries is produced. As the permittivity of water $\varepsilon_{\rm w} \approx 80$ exceeds tens times the permittivity of water vapour $\varepsilon_{\rm v} \approx 1$, the electric-field strength in the vapour cavity is more than an order of magnitude higher than the electric-field strength in the surrounding water. For this reason, the potential difference between the vapour cavity boundaries should make the greatest contribution to the evolution of the entire electric signal [2].

The pressure in the vapour cavity at the moment of its formation is equal to the pressure $p_{\rm s}$ of saturated vapour at temperature $T_{\rm th}$ and is 98 atm, i.e. about two orders of magnitude higher than the atmospheric pressure. Because of the difference between the pressures inside and outside the cavity, the vapour cavity starts expanding rapidly, pushing the water column up in the process (see Fig. 1a). The expansion of the vapour cavity is the second stage of the process accompanying the action of HF-laser radiation on water. If the duration of the first stage (initial heating of water) is no longer than the pulse duration (about 300 ns), the second stage lasts as long as the vapour pressure in the cavity remains higher than the atmospheric pressure (10–30 μs).

The motion of the water column and vapour cavity at times much longer than 10 μs is the third stage of the evolution of the water layer under the action of HF-laser radiation on the water surface. At this stage water can be regarded as an incompressible fluid and the vapour pressure in the cavity and the atmospheric pressure as external actions on the top and bottom boundaries of the water column in the cell. If we neglect the friction of water with the cell walls and the gravitational force of the water column as compared to the atmospheric pressure p_0 and consider the cell motionless, then the equation of motion for the water column of height d has the form

$$\rho_{\rm w} d \frac{{\rm d}^2 z_{\rm c}}{{\rm d}t^2} = p_{\rm v}(t) + p_{\rm pa}(t) - p_0, \qquad (1)$$

where $\rho_{\rm w}$ is the water density; $z_{\rm c}$ is the coordinate of the centre of gravity of the water column; $p_{\rm v}(t)$ is the vapour pressure in the cavity; $p_{\rm pa}(t) = \varepsilon v_{\rm s} I(t)/c_p$ is the photoacoustic pressure [7] caused by the expansion of a thin water layer heated by a laser pulse of intensity $I(t)$ near the cell bottom at the first stage of the interaction process; ε is the thermal expansion coefficient of water; $v_{\rm s}$ is the speed of sound in water; c_p is the heat capacity of water at constant pressure. In the case of explosive boiling of water, the relative contribution of the photoacoustic pressure in the acceleration of the water column is negligible because it is non-zero only during the action of a laser pulse. However, if the intensity of the laser pulse is not sufficient for explosive boiling of water, the contribution of the photoacoustic pressure is most decisive.

Unlike the short-lived action of pressures $p_{\rm pa}(t)$ and $p_{\rm v}(t)$, the atmospheric pressure is constant and determines the negative acceleration of the water column in the cell for the time ~ 100 μs and longer. Integrating equation (1) with respect to time, we find the velocity of the water column to be:

$$v(t) = v_0 - \frac{p_0}{\rho_{\rm w} d}\, t, \quad v_0 = \frac{1}{\rho_{\rm w} d} \int_0^{t_0} [p_{\rm v}(t) + p_{\rm pa}(t) - p_0]{\rm d}t, \qquad (2)$$

where t_0 is the time during which there is an excess pressure in the vapour cavity. Thus, the water column moves with a constant acceleration $a = -p_0/(\rho_{\rm w} d)$ and an initial velocity v_0.

The total time of the uniformly accelerated motion of the water column determined by the expression

$$t_{\max} = -2\frac{v_0}{a} = \frac{2}{p_0} \int_0^{t_0} [p_{\rm pa}(t) + p_{\rm v}(t) - p_0]{\rm d}t \qquad (3)$$

is independent of the height d of the water column. Thus, some time later $t_{\max}$ after the laser-pulse action the vapour cavity collapses. The collapse of the cavity whose surfaces, as was mention above, are charged, should affect the electric signal, which is, probably, observed in the experiment in the form of the second electric signal peak (i.e. $t_{\max} = \tau$ in Fig. 3).

It is necessary to find the evolution of the temperature profile in water and the dynamics of pressure in the vapour cavity to determine $t_{\max}$.

The mathematical model of interaction of laser radiation with the water layer in the cell (for a closed water surface) at the stage of initial heating and expansion of the vapour cavity is described in detail in [2] for a $Cr^{3+}:Yb^{3+}:Ho^{3+}$: YSGG laser emitting at 2.92 μm. Radiation from a HF laser represents a combination of lines with different intensities in the wavelength range from 2.7 to 3.1 μm [8] in which the absorption coefficient of water α varies from 3000 to 14000 cm^{-1} [9]. Thus, we followed the computation concept of paper [9] and chose three emission lines of approximately equal intensities with the wavelengths $\lambda_i = 2.74$, 2.83 and 2.87 μm to which the water absorption coefficient α_i of 3200, 8600 and 10000 cm^{-1}, respectively. The heat conduction equation for the water layer in this case takes the form:

$$\frac{\partial T}{\partial t} = \chi_w \frac{\partial^2 T}{\partial z^2} + Q(z,t) \quad Q(t,z) = \frac{I(t)}{c_p \rho_w} \frac{1}{3} \sum_{i=1}^{3} \alpha_i \exp(-\alpha_i z), \quad (4)$$

where χ_w is the water heat conductivity. The intensity $I(t)$ of HF-laser radiation on the water surface was given by the expression $I(t) = I_0 \exp[-(t - 2t_L)^2/t_L^2]$ with $t_L = 78$ ns, which well approximates the actual pulse shape. The intensity I_0 is related to the laser energy density W as $I_0 = W/\tau_p$. The thermophysical parameters of quartz and water used in our calculations did not differ from those used in [2].

In the numerical simulation of the interaction of laser radiation with water in the quartz cell, we determined the threshold energy density $W_{th} = 0.22$ J cm^{-2} at which the temperature of the overheated water layer achieves $T_{th} = 583$ K, and water boils explosively followed by the formation of a vapour cavity. Figure 5 shows the evolution of the pressure $p(t) = p_{pa}(t) + p_v(t) - p_0$ acting on the water column in the cell for the laser-pulse energy density W_{th} at the initial stage of the process (the total duration of the pressure pulse exceeds 20 μs). The first component of the pressure pulse $p_{pa}(t)$ (photoacoustic pressure) repeats the shape of the laser pulse. 250 ns later after the onset of the laser pulse (at the trailing edge of the pulse), the explosive boiling of water occurs and the pressure jumps to the saturated-vapour pressure corresponding to temperature T_{th}. Further expansion and cooling of the vapour in the cavity leads to an abrupt drop of pressure in it, which is illustrated by the second component $p_v(t)$ of the pressure pulse.

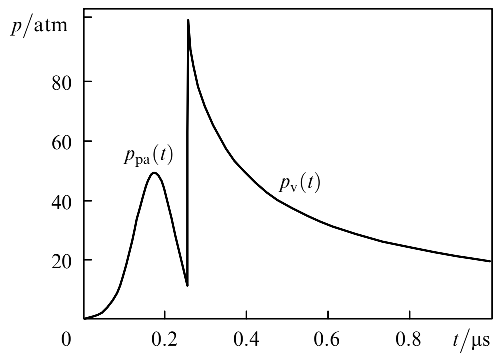

Figure 5. Time dependence of the pressure acting on the water column for $W_{th} = 0.22$ J cm^{-2}.

It follows from (3) that the total time of motion of the water column is equal to doubled area under the pressure curve normalised to p_0.

Figure 6 presents the total time t_{max} of motion of the water column as a function of the laser pulse energy density W [curve (*2*)] changing from W_{th} to $W_c = 0.31$ J cm^{-2} at which the water temperature achieves T_c. By way of comparison Fig. 6 also shows the experimental dependence [curve (*1*)] of the time interval τ between the electric signal peaks on the average energy density in the irradiated spot $\bar{W} = E/(\pi a^2)$. One can see that the theoretical dependence $t_{max}(E)$ agrees fairly well with the experimental one not only qualitatively [dependence $t_{max}(E)$ is close to a linear one], but also quantitatively (in view of impossibility to take into account a real Gaussian energy distribution of a laser pulse in the irradiated spot in the given one-dimensional model). Fairly good agreement between theoretical and experimental results allows us to suggest that the properties of water for the above-the-critical temperatures (when the energy density in a laser pulse is $W \geqslant W_c$) are a smooth continuation of the water properties for the below-the-critical temperatures because the behaviour of curve (*1*) in Fig. 6 does not change when $W \geqslant W_c$.

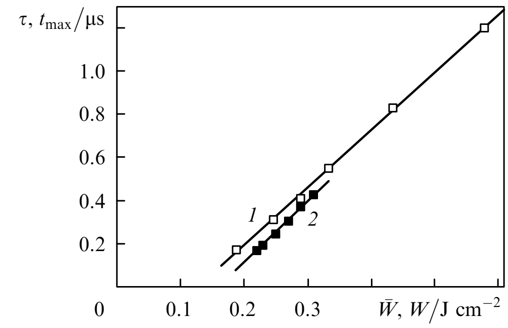

Figure 6. The time interval τ between the electric signal peaks as a function of the average energy density $\bar{W}$ in the irradiation spot (*1*) and the total time t_{max} of motion of the water column (*2*) as a function of the energy density W.

It follows from the model described above that the maximum time t_{max} of motion of the water column and time interval between the electric signal peaks should not depend on the height d of the water column. However, we observed this dependence in the experiment. Figure 7 presents the oscillograms of the electric signal for the 30-mm-long cell, laser output energy $E = 1.3$ J and different heights d of the water column. One can see that when d decreases from 30 to 9 mm, the amplitude of the first electric signal peak falls from 380 to 120 mV, and the amplitude of the second one – from 100 to 10 mV. This is caused by a decreased capacitive coupling between the electrode and the region of the electric signal generation. The time interval τ between the two electric signal peaks also decreases, though to a much less degree: from 1.22 to 1 ms, i.e. no more than by 20 %. We can suggest that the observed decrease in τ may result from some factors being neglected in the model, e.g. friction between the cell walls and liquid or mechanical vibrations of the cell. It is necessary to note that in the experiment the area of exposure is about one fourth the area of the cell bottom. This, in particular, may result in the vapour cavity collapse due to not only vertical motion of the water column, but also 'flowing' from the cavity sides. To take these processes into account, it is necessary to develop a complicated two-dimensional model. However, we believe that at the present stage of the research on the effect of the electric signal generation, simple models giving general notion of its mechanisms are preferable.

The change in the shape of the electric signal with decreasing the height of the water column is also of importance. Starting from $d = 15$ mm, a rapid growth in the negative component of the electric signal after the second peak is observed. In this case, water sputtering is observed. The full shape of the electric signal for $d = 9$ mm and a 5-ms div^{-1} sweep is shown in Fig. 7d. One can see that the amplitude of the negative component achieves 1.3 V. The polarity, amplitude, rise time duration and total duration of this electric signal component indicate that its

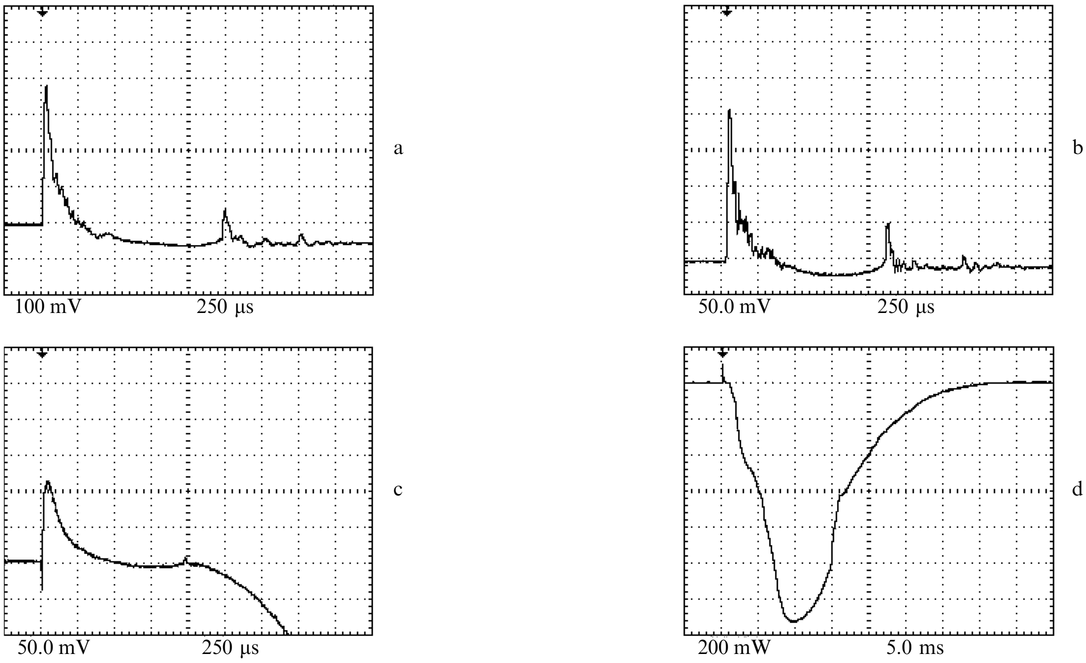

Figure 7. Oscillograms of the electric signal for $l = 30$ mm, $E = 1.3$ J, $d = 30$ (a), 24 (b) and 9 mm (c, d).

appearance is caused by the formation of a scattering layer of charged water drops (sputtering) [3, 4], which probably results from the development of splitting processes on the open water surface upon the shock-wave action on it.

The formation of splitting layers in water caused by the shock-wave action was investigated theoretically and experimentally in [10, 11]. The amplitudes and durations of the shock wave, which we measured with a piezoelectric pressure sensor [3 – 5], proved to be close to those realised in the given experiments. The authors of papers [10, 11] calculated the time of formation of splitting layers, which is well correlated in our experiments with the time delays between the onset of the laser-pulse action (accompanied by a pressure jump) and the beginning of the growth in the negative electric signal component.

It is also necessary to note that in [12] the splitting of an open water surface was investigated using a method for detecting the electric signal generated in the electric explosion of conductors in water. The absence of effects of splitting-layer formation in the electric signal detected in our experiments with the water column heights $d > 15$ mm seems to be caused by a considerable attenuation of the shock-wave perturbation produced during the explosive boiling of water at the cell bottom to the moment of its propagation to the open surface of the water column.

4. Conclusions

We have studied in this paper the generation of an electric signal when a pulse from a HF laser interacts with the lower surface of a water column placed in a cell with a transparent bottom, the upper surface of the water column being open. Given such conditions of irradiation, the electric signal demonstrates two pronounced peaks, and the time interval between them is linearly dependent on the laser output energy and over an order of magnitude greater than the time during which the pressure in the vapour cavity produced in the explosive boiling of water in the irradiated thin layer is higher than the atmospheric pressure. It has been found that the second electric signal peak is produced during the collapse of the vapour cavity. A theoretic model has been constructed to explain the motion of the water column above the vapour cavity with allowance for the evolution of the pressure in it. It has been shown that the calculated total time t_{max} of the water column motion, which accompanies the action of a laser pulse on the closed water surface, is close to the time interval τ between the electric signal peaks. Therefore, the prolonged lifetime of the vapour cavity after a decrease in the pressure in it to the atmospheric pressure is caused by inertial movement of the water column, which acquires its velocity at the initial stage of the cavity expansion.

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