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High-temperature plasma produced on a free liquid surface by femtosecond laser pulses

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***Abstract*. The plasma formed by femtosecond laser radiation with an intensity higher than 10^{16} W cm^{-2} at the free surface of a liquid (VM-1 vacuum oil) in vacuum has parameters which are similar to the parameters of the high-temperature plasma produced at the surface of a solid target. The hot-electron temperature (derived from X-ray and ion time-of-flight measurements) is 6 ± 3 keV for the VM-1 oil target and 4 ± 1 keV for a crystal silicon target. The optical diagnostics of the relaxation of the liquid target surface revealed that the limiting laser pulse-repetition rate whereby the interaction takes place with the unperturbed liquid surface may be as high as 10 Hz.**

Keywords: *high-temperature femtosecond laser plasma, X-rays, ion currents.*

1. Introduction

The plasma produced by high-intensity femtosecond laser radiation is a unique source of broadband incoherent X-ray radiation, fast electrons and ions [1, 2]. To date, extensive experimental material has been accumulated concerning the parameters of femtosecond laser plasmas produced by irradiating targets of different kind: solid [3], porous [4], 'clustered' [5], etc.

The papers on plasma production by high-intensity femtosecond laser irradiation of a substance in the liquid phase were published relatively recently. In the overwhelming majority of these papers, the jets of different liquids were employed as targets: copper nitrate or ethylene glycol solutions [6], liquid gallium [7] or nitrogen [8]. At the same time, the use of a free liquid surface is of interest from the standpoint of simplicity of experimental realisation, which makes it possible to work with an immobile target. We know only two papers [9, 10] in which measurements were made of the hard X-ray emission spectrum of the plasma produced by ultrahigh-intensity femtosecond laser irradiation of the free surface of liquid mercury and water. In the latter case, experiments were conducted in the air at atmospheric pressure, which was supposedly responsible for a significant reduction of the X-ray generation efficiency due to the unavoidable losses resulting from the ionisation of the surface air layer. Also, several papers were devoted to the study of the plasma emerging at a liquid surface irradiated by picosecond and femtosecond pulses with an intensity below 10^{13} W cm^{-2} [11, 12] as well as to the observation of the shock waves produced in the breakdown region [13].

Choosing the object of investigation presents a certain difficulty when staging experiments to investigate the interaction of high-intensity femtosecond laser radiation with a free liquid surface in vacuum. To produce a high-temperature laser plasma at the target surface with the subsequent efficient generation of hard X-ray radiation above 2 keV, the laser radiation intensity should exceed 10^{16} W cm^{-2}. To do this, the liquid should feature a low saturation vapour pressure to avoid the self-action of laser radiation and the breakdown of the surface gas layer. Viscosity, which defines the relaxation rate of the liquid surface after its interaction with the laser pulse, is also a characteristic of no small importance. This circumstance becomes fundamentally important in experiments when laser systems operating in pulse-periodic regimes are used. Metals in the liquid phase (gallium, mercury), some alloys (for instance, lead with bismuth), and vacuum oils comply with the above requirements.

The aim of our work is to investigate the properties of the high-temperature plasma produced by femtosecond laser pulses at the free surface of the liquid (vacuum oil).

2. Experimental

The high-temperature plasma was produced at the target surface by the radiation of a femtosecond dye laser system [4]. The femtosecond laser radiation (p-polarisation, 200 fs, 0.3 mJ, an intensity contrast ratio of $\sim 10^5$, the 45° angle of radiation incidence on the target) was focused with an aberration-free objective to produce an intensity of $\sim 2 \times 10^{16}$ W cm^{-2} at the target surface. The experimental setup is shown in Fig. 1. A cell 3 cm in diameter with a 5-mm thick layer of the VM-1 vacuum oil is in the vacuum chamber. In its chemical composition, the VM-1 oil constitutes a mixture of paraffinic, naphthenic, and aromatic hydrocarbons and of asphalto-resinous substances. The saturation oil vapour pressure is of the order of 10^{-7} Torr and its kinematic viscosity is equal to 0.7 cm^2 s^{-1}.

To determine the plasma parameters, we measured the yield of hard X-rays and ion time-of-flight spectra. To carry

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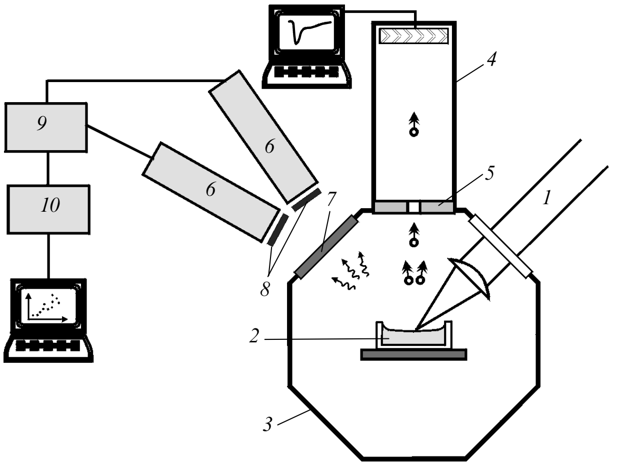

Figure 1. Scheme of the experiment: (*1*) laser radiation; (*2*) target (VM-1 vacuum oil); (*3*) vacuum chamber; (*4*) time-of-flight detector; (*5*) plate with a hole 2 mm in diameter; (*6*) X-ray detector; (*7*) beryllium filter; (*8*) removable band filters; (*9*) charge-sensitive amplifier; (*10*) analogue-to-digital converter.

out the time-of-flight measurements with the help of a chevron-type microchannel plate (MCP), the time-of-flight detector was differentially pumped to 10^{-5} Torr for a pressure of 10^{-4} Torr in the chamber. The yield of hard X-rays into the spectral ranges with energies above 8, 10, and 25 keV was measured with two detectors involving FEU-119 photomultipliers with NaI(Ti) scintillators. Different sets of band filters of the following composition were placed in front of them: 200 μm of Al + 100 μm of Be, 400 μm of Al + 100 μm of Be, and 100 μm of Al + 100 μm of Be + 39 μm of Ta. The laser radiation was accurately focused on the target surface by measuring the yield of X-rays from the plasma.

3. Experimental results

The yield of hard X-rays from the plasma measured in different spectral ranges is shown in Fig. 2. We observed a large scatter in the values of X-ray yield for the same intensity of laser radiation. In our opinion this may be due to the local production of new chemical reaction products on the oil surface upon laser irradiation, when the next laser pulse interacts not with oil, but with its modification.

Comparative data for X-ray yield into the photon energy range above 10 keV obtained with targets of crystal silicon and the VM-1 vacuum oil are given in Fig. 3. The X-ray yield for silicon is approximately eight times the X-ray yield for oil. Since this parameter depends on the atomic number of the material as $Z^{3/2}$ [14], from the silicon – oil X-ray yield ratio it is possible to estimate the effective atomic number Z_{eff} of the material of the VM-1 oil. Calculated from the data in Fig. 3 is a value $Z_{\text{eff}} \sim 3 \pm 1$, which corresponds to the chemical oil composition of paraffinic C_nH_{2n+1}, naphthenic C_nH_{2n+2}, and aromatic C_nH_{2n-6} hydrocarbons (the concentration of asphaltenes in the oil is low). Therefore, the efficiency of hard X-ray generation in the plasma at the surface of a liquid target does not substantially differ from the efficiency in the case of a solid target.

The measurements of the X-ray yield into two different spectral ranges make it possible to estimate the temperature

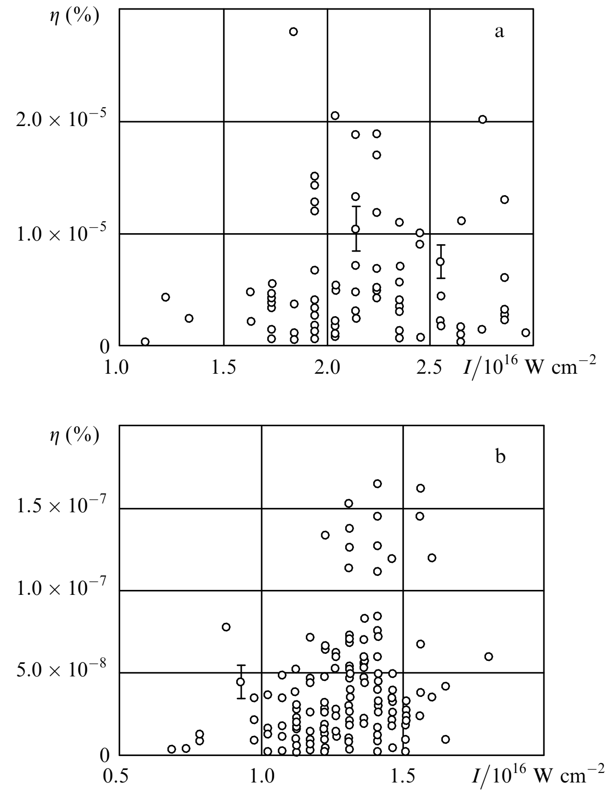

Figure 2. Yield η of X-rays into the spectral ranges above 10 (a) and 25 keV (b) as a function of the laser pulse intensity I.

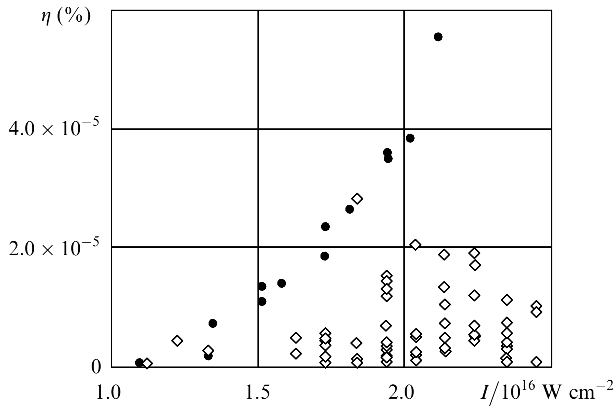

Figure 3. Yield η of X-rays from crystal silicon (●) and the VM-1 vacuum oil (◇) into the spectral range above 10 keV.

of hot electrons in plasma in every laser shot by taking advantage of the modified band filter technique [14, 15]. The result of processing the experimental data is shown in Fig. 4. The data for the crystal silicon target are also given for comparison in Fig. 4. The electron temperature of the plasma produced at the liquid surface exceeds, on the average, the electron temperature corresponding to solid targets. In particular, for a laser pulse intensity of 1.3×10^{16} W cm^{-2} the electron temperature in the plasma at the liquid surface is 6 ± 3 keV, while the electron temperature of the plasma at the crystal silicon surface is 4 ± 1 keV.

We also investigated the parameters of the high-temperature plasma produced at the free liquid surface employing the technique of ion time-of-flight measurements. Fig. 5

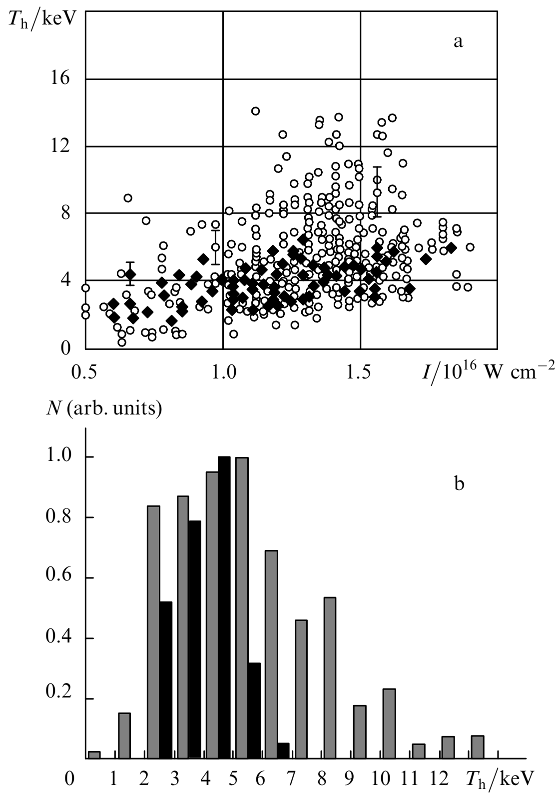

Figure 4. Temperature of hot electrons T_h as a function of the laser pulse intensity I (a) and electron statistical (shot-to-shot) temperature distribution (b). Silicon is represented by black columns and the VM-1 vacuum oil by the grey ones.

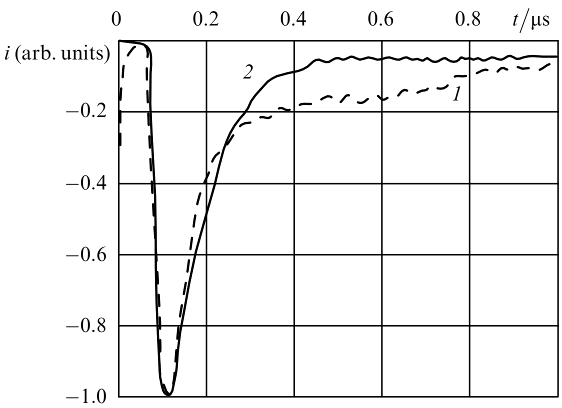

Figure 5. Time dependences of ion currents i for targets of crystal silicon (*1*) and the VM-1 oil (*2*).

shows the typical time dependences of ion current from the plasma produced at the VM-1 oil surface and at the crystal silicon surface. The extreme points of the ion currents correspond to the arrival of protons at the detector [16] and are practically coincident, which is indication that the temperatures of the hot electron plasma component are the same in these two cases.

It is noteworthy that the trailing edges of the ion currents behave differently. For a crystal silicon target, the signal corresponds to silicon ions which have a lower Z/M ratio in comparison with carbon, which makes up the basis of the vacuum oil. Consequently, the silicon ions arrive at the time-of-flight detector later in time. Furthermore, proceeding from a one-dimensional hydrodynamic model of plasma expansion it is possible to estimate the temperature of hot electrons from plasma ion currents [15]. Estimates obtained with the aid of this method agree, to within the error, with the estimates derived from the X-ray yield. In particular, the electron temperature in the same pulse estimated from the X-ray yield was equal to 4 ± 1 keV and from the ion currents to 3.5 ± 1 keV.

To estimate the limiting repetition rate of high-intensity femtosecond laser pulses, experiments were carried out to determine the relaxation time of liquid surface oscillations. We observed the reflection of probe He – Ne laser radiation from the target surface after its irradiation by a femtosecond laser pulse (Fig. 6). In the setup employed, the femtosecond laser pulse propagated coaxially with the probing beam. To do this, the probing laser beam entered from the rear side of the mirror through a hole drilled at an angle of 45°, while the principal beam was incident on the mirror at an angle of 45°.

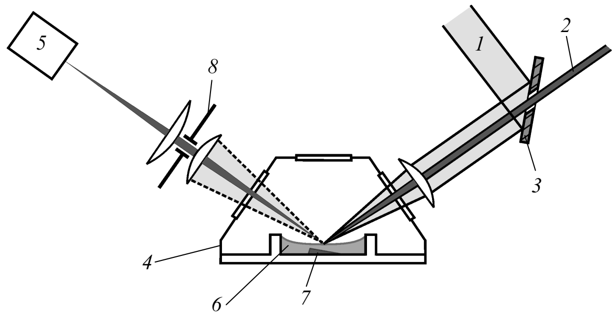

Figure 6. Setup for the optical diagnostics of the liquid surface: (*1*) femtosecond radiation; (*2*) probing He – Ne laser radiation; (*3*) mirror with a hole drilled at 45° to its surface; (*4*) vacuum chamber; (*5*) photomultiplier; (*6*) target (VM-1 vacuum oil); (*7*) reflecting wedge; (*8*) diaphragm.

A photomultiplier was used to observe the dynamics of the reflection of probing radiation from the target surface. Since the femtosecond pulse radiation had an intensity minimum at the centre of the beam (prior to arrival at the target it bounced from mirror (*3*) with an orifice), only the He – Ne laser radiation that reflected from the target surface could reach the photomultiplier through diaphragm (*8*). The photomultiplier signal was delivered to a digital oscilloscope and digitised with time resolutions of 41 μs, 1.3 μs, and 20 ns. Fig. 7 shows the time dependence of the signal recorded with the photomultiplier on the delay relative to the instant of plasma initiation. The positive large-amplitude spike near the abscissa zero arises from the detection of the plasma-scattered femtosecond laser pulse, which is incompletely suppressed by spatial filter (*8*).

The dynamics of He – Ne laser radiation reflection from the relaxing liquid surface is determined by the negative signal recorded by the photomultiplier at the times greater than 2 μs (see Fig. 7). This signal passes through a minimum to return, after a time, to the initial zero level. For the time of liquid surface relaxation to the initial state we assume the time interval from the instant of plasma initiation to the moment at which the photomultiplier signal recovers the e^{-1} level relative to the minimum of the signal. The relaxation time is 15 ± 5 ms and depends only slightly on the intensity

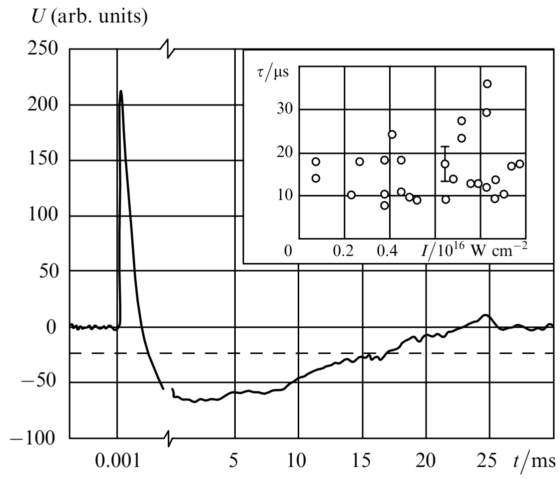

Figure 7. Signal recorded by the photomultiplier. The inset shows the dependence of liquid surface relaxation time on the intensity of femtosecond laser radiation. The dashed line shows the e^{-1} level of the photomultiplier signal amplitude.

of a laser pulse in the $10^{15} - 10^{16}$ W cm^{-2} intensity range (see the inset in Fig. 7). This is supposedly due to the fact that the surface relaxation over these periods of time is determined by the properties of the liquid. Assuming the liquid motion to be potential, the time of surface wave damping can be estimated as $\tau = g^2/(2\nu_k\omega^4)$ [17], where g is the free fall acceleration; ν_k is the kinematic viscosity; $\omega^2 = gk + (\alpha/\rho)k^3$ is the frequency of sound waves; α is the surface tension coefficient; ρ is the density; and k is the wave number of the sound wave defined by the cell geometry. The theoretical estimate (10 ms) agrees well with experiment. Therefore, the laser pulse repetition rate should not exceed 10 Hz.

During exposure to a pulse sequence, the liquid may heat up to the boiling temperature due to its low thermal conductivity, which will also result in the oscillations of its surface. We estimate the upper limit for the laser pulse repetition rate. It is well known that the coordinate of the thermal wave front in the one-dimensional case is defined by the expression $z(t) = (\chi t)^{1/2}$, where t is the time and χ is the thermal diffusivity. Then, the liquid volume heated in one 'shot' can be estimated as $V \sim z^3 = (\chi\Delta t)^{3/2}$, where $\Delta t = \nu^{-1}$; ν is the laser pulse repetition rate. Let us assume that a fraction α of the laser energy W is converted to heat in every laser 'shot'. This liquid volume will therefore heat up to a temperature $\Delta T = \alpha W/(c\rho V) \sim \alpha W \nu^{3/2}/(c\rho\chi^{3/2})$, which should be much lower than its boiling temperature ($\Delta T \ll \Delta T_b = T_b - T_0$, where T_b is the boiling temperature and T_0 is the room temperature). For the VM-1 oil, the parameters are $\chi \simeq 10^{-3}$ cm^2 s^{-1}, $c = 1.44$ J g^{-1} K^{-1}, and $\rho = 0.87$ g cm^{-3}, $\Delta T_b \sim 300$ K [18]. For typical values $\alpha \sim 10 - 30$ % we obtain the following condition for the laser pulse repetition rate: $W\nu^{3/2} \ll 10^{-3}$ J Hz$^{3/2}$. In our experiments the laser pulse energy W was equal to ~ 300 μJ, which resulted in the limitation of laser pulse repetition rate $\nu \ll 50$ Hz.

Therefore, it has been demonstrated for the first time that the high-temperature femtosecond laser plasma produced at a free liquid surface in vacuum possesses properties that are largely similar to the properties of the plasma produced at a solid target surface. For a laser pulse intensity of 10^{16} W cm^{-2}, the hot electron temperature of the plasma at the surface of the VM-1 vacuum oil is $T_h = 6 \pm 3$ keV and at the crystal silicon surface is $T_h = 4 \pm 1$ keV. The optical diagnostics of the relaxation time of the surface of the VM-1 vacuum oil showed that the laser pulse repetition rate should not exceed 10 Hz.

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