PACS numbers: 42.65.Re; 52.50.Jm; 52.57.Bc; 52.38.Ph DOI: 10.1070/QE2007v037n07ABEH013491

# Highly stable plasma source produced on the liquid-gallium surface by a femtosecond laser pulse

V.M. Gordienko, M.V. Kurilova, E.V. Rakov, A.B. Savel'ev, D.S. Uryupina

Abstract. Experimental results are presented which demonstrate that a plasma produced on the melted-gallium surface by a femtosecond laser pulse of intensity above  $10^{16}$  W cm<sup>-2</sup> is an efficient and stable source of incoherent hard X-rays with a pulse repetition rate of 10 Hz. For gallium heated up to 270 °C, the X-ray yield decreased by ~ 25% [from  $(2.2 \pm 0.4) \times 10^{-4}$ % to  $(1.7 \pm 0.4) \times 10^{-4}$ %] after 50000 laser shots, while the average energy of hot electrons decreased from  $9.3 \pm 0.9$  to  $9.0 \pm 1.1$  keV.

Keywords: laser plasma, hard X-ray source, liquid targets.

#### 1. Introduction

During the last decade, the properties of the plasma produced by femtosecond laser pulses of intensities  $10^{16} - 10^{21}$  W cm<sup>-2</sup> have been investigated in many papers. First of all, this plasma is of interest as the efficient source of picosecond X-rays, beams of fast multiply charged ions and protons, neutrons, etc. (see, for example, reviews [1, 2]). Targets studied in experiments are usually plates made of different materials [3–8]. Clusters [9, 10], microdroplets [11, 12] or liquid jets [13] were used in some papers, which considerably modifies the plasma properties and eliminates the necessity for the displacement of a target after each laser pulse. The same goals can be achieved by using the liquid surface as a target under the condition that it has time to recover between the two successive laser pulses.

In papers [14-16], a free liquid surface was used as a target for producing a hot plasma by a femtosecond laser pulse of intensity above  $10^{16}$  W cm<sup>-2</sup>. The authors of paper [14] studied the spectrum of X-ray bremsstrahlung from a plasma produced on the water surface by single laser pulses or a train of laser pulses separated by the 10-ns time interval. The laser pulse (or laser-pulse train) repetition rate in these experiments was 10 Hz. It was found that X-rays emitted by the plasma produced by a pulse train were harder than X-rays emitted by the plasma produced by the plasma produced by

V.M. Gordienko, A.B. Savel'ev, D.S. Uryupina International Laser Center & Department of Physics, M.V. Lomonosov Moscow State University, Vorob'evy gory, 119992 Moscow, Russia; e-mail: ab\_savelev@phys.msu.ru;

M.V. Kurilova, E.V. Rakov Department of Physics, M.V. Lomonosov Moscow State University, Vorob'evy gory, 119992 Moscow, Russia

Received 15 December 2006; revision received 24 January 2007 *Kvantovaya Elektronika* **37** (7) 651–655 (2007) Translated by M.N. Sapozhnikov single laser pulses. This increase in the energy of X-ray quanta is caused by the appearance of microdroplets upon irradiation by a pulse train. The authors of paper [15] studied the interaction of laser pulses, emitted at a pulse repetition rate of 2 kHz, with the liquid mercury surface. They measured only the X-ray spectrum of the plasma and did not report other plasma parameters. The principal problem of the stability of the X-ray yield as a function of the number of laser pulses (the interaction time) was also not studied.

Note that experiments with liquid targets have been performed in the above-mentioned studies in the air at the atmospheric pressure, which considerably reduced the X-ray yield due to inevitable losses caused by the air ionisation in the laser beam waist, the radiation self-action, etc. Thus, liquids with a low saturation vapour pressure, which can be used as targets in vacuum, are of the most interest. In [16], experiments were performed in which a target representing a cell with a VM-1 vacuum oil was placed in a chamber evacuated down to a pressure of  $10^{-2}$  Torr. It was shown that the properties of the plasma produced by a  $\sim 10^{16} \ {\rm W \ cm^{-2}}$  femtosecond laser pulse did not differ from those of the plasma produced on a solid target. However, the oil boiling in the repetitively pulsed regime (the pulse repetition rate was 1 Hz) prevented the development of a stable plasma source. To suppress this effect, a liquid with the high heat conduction and high boiling point is required, for example, a liquid (melted) metal. In this paper, we present the experimental data demonstrating the possibility of the development of a highly stable laserplasma X-ray source by using a liquid metal (gallium) as a target.

In experiments on the interaction of high-power femtosecond laser pulses with liquid targets the choice of the target material is very important. To produce a hightemperature laser plasma on the target surface generating efficiently hard X-rays in the region above 2 keV and fast ions, it is necessary to provide the laser radiation intensity above  $10^{16}$  W cm<sup>-2</sup>. Therefore, to avoid the self-action of laser radiation and breakdown of the surface gas layer, the saturation vapour pressure of the liquid should be low. The important parameters of the liquid are also its viscosity, surface tension, and heat conduction - the cooling rate of the irradiated region.

The choice of gallium as the target material was determined, first of all, by its physical properties such as

(i) the low melting point  $(30 \,^{\circ}\text{C})$  allowing the preparation of a liquid-metal target without considerable restrictions on the vacuum-chamber design;

(ii) the extremely low saturation vapour pressure  $(9.31 \times 10^{-36} \text{ Pa} \text{ at the melting point})$  and a rather high boiling point (2300 °C);

(iii) the high heat conduction  $(1.4 \times 10^{-5} \text{ m}^2 \text{ s}^{-1} \text{ at the temperature 30 °C})$  providing the efficient heat outflow from the interaction region during the time between two laser pulses.

## 2. Experimental results

Figure 1 shows the scheme of the experimental setup. Plasma was produced by a 110-fs,  $350-\mu J$ ,  $1.24-\mu m$  pulse from a Cr : forsterite laser (the pulse repetition rate was 10 Hz) [17].

A liquid-gallium target was placed in a heated cell of diameter 5 mm and depth 2 mm. The gallium temperature



**Figure 1.** Scheme of the experimental setup: (1) femtosecond laser pulse; (2) vacuum chamber; (3) liquid-gallium target; (4) X-ray detectors; (5) X-ray filter (Be or Al); (6) resistive heater; (7) thermocouple.

could be varied from room temperature ( $\sim 20 \,^{\circ}$ C) up to 270  $^{\circ}$ C. The temperature was controlled with a thermocouple. The system (the gallium target, heater, and thermocouple) was placed into a vacuum chamber evacuated down to  $10^{-2}$  Torr with the help of a backing pump. Radiation was focused to a spot of diameter 4 µm on the target with the help of an aberration-free objective with a focal distance of 6 cm. This provided the power density on the target surface up to  $\sim 10^{16} \text{ W cm}^{-2}$ . The angle of incidence of radiation on the target was 45°. The integrated yield of hard X-ray bremsstrahlung was measured with two X-ray detectors based on an NaI(Tl) scintillator and a FEU-119 photomultiplier. Band-pass X-ray filters mounted in front of the detectors transmitted X-rays with energies above 2.5 keV (Be 200 µm) and above 7.5 keV (Be  $200 \ \mu m + Al \ 300 \ \mu m$ ). Both detectors were absolutely calibrated [18]. Measurements of the X-ray yield in two different spectral ranges allowed us to estimate the average energy of hot electrons in each laser shot [18, 19], thereby controlling the stability of plasma parameters.

Because the physical properties of the liquid strongly change in the region of laser irradiation, we performed a series of measurements of the X-ray yield of the plasma irradiated by single pulses or by laser pulses with a pulse repetition rate of 10 Hz. The gallium temperature in both cases was 270 °C. Figure 2 presents the results of these experiments obtained upon irradiation by 100 successive pulses. One can see that the X-ray yield in different spectral regions and the average energy of hot electrons in both cases are the same. The conversion of the laser pulse energy to Xray bremsstrahlung (the X-ray yield  $\eta$ ) for E > 2.5 keV was  $(2.2 \pm 0.4) \times 10^{-4}$  % and for E > 7.5 keV -  $(4.2 \pm 0.5) \times$  $10^{-5}$  %. The average energy of hot electrons was  $9.3 \pm 0.9$  keV. Thus, we have shown experimentally that the gallium target has time to recover within 100 ms after laser irradiation, and parameters of the plasma produced in the repetitively pulsed regime at 10 Hz do not differ from parameters of the plasma produced upon irradiation by a single pulse.

Note also that the X-ray yield in test experiments performed in air at the atmospheric pressure was lower approximately by a factor of 15  $[(1.5 \pm 0.4) \times 10^{-5} \%$  for E > 2.5 keV]. This suggests that the presence of vacuum is the important condition for the development of the efficient X-ray source.



Figure 2. X-ray yield  $\eta$  in the spectral regions above 2.5 and 7.5 keV and the average energy  $\langle E_h \rangle$  of hot electrons as functions of the laser shot number N upon irradiation by a single pulse (a) and pulses emitted at a repetition rate of 10 Hz (b).

We studied the long-term stability of the plasma source by irradiating the target by laser pulses for a long time. First of all, we investigated the time dependence of the X-ray yield. We performed  $\sim$  50000 laser shots, which corresponds to one and a half hours of continuous irradiation by pulses with a pulse repetition rate of 10 Hz. The target was not displaced during the experiment and no additional focusing was performed. Figure 3 presents the X-ray yield in the spectral region above 2.5 keV and the average energy of hot electrons as functions of the laser shot number.



**Figure 3.** X-ray yield  $\eta$  in the spectral range above 2.5 keV (a) and the average energy  $\langle E_h \rangle$  of hot electrons (b) as functions of the laser shot number *N* upon irradiation by laser pulses at a repetition rate of 10 Hz at the gallium temperature  $T_{\rm Ga} = 270$  °C. The white curve corresponds to the adjacent average value.

For gallium heated up to 270 °C during 50000 laser shots, the X-ray yield decreased approximately by 25 % – from  $(2.2 \pm 0.4) \times 10^{-4}$ % down to  $(1.7 \pm 0.4) \times 10^{-4}$ %, while the average energy of hot electrons changed from  $9.3 \pm 0.9$  keV to  $9.0 \pm 1.1$  keV. In this case, the X-ray yield and the average energy of hot electrons were virtually the same in approximately 45% of cases. The decrease rate of the average X-ray yield in the spectral range above 2.5 keV was  $(1.2 \pm 0.2) \times 10^{-9}$ % per pulse, and the root-meansquare deviation for any subsequent 100 laser shots did not exceed 24% of the average value. Because the average total energy of X-ray bremsstrahlung for E > 2.5 keV was ~ 0.5 nJ, we estimated the average power of the plasma Xray source as 5 nW in the repetitively pulsed regime with a pulse repetition rate of 10 Hz.

Our experiments also revealed a strong dependence of the long-term stability of the plasma source on the melted gallium temperature. Thus, for  $T_{Ga} \sim 50$  °C, the long-term stability of the hard X-ray yield and the average energy of hot electrons proved to be considerably lower than at 270 °C. In this regime, the X-ray yield decreased by 25 % already after the first 4000 laser shots.

The decrease in the hard X-ray yield and the average energy of hot electrons in the plasma observed in experiments is obviously explained by the decrease in the laser radiation intensity. This is caused by the lowering of the liquid level during experiments due to the mass removal and deviation of the average surface of the liquid from the position corresponding to the exact focusing of radiation on the target.

Let us estimate how the laser radiation intensity should decrease and how the liquid level should lower after 50000 laser pulses. The contrast of laser pulses in our experiments was high enough for a prepulse to produce no plasma, and the main pulse was absorbed at the sharp plasma – vacuum interface. Then, the average energy  $\langle E_h \rangle$  of hot electrons, the intensity *I*, and the laser wavelength  $\lambda$  are related by the expression [18]

$$\langle E_{\rm h} \rangle = 7.5 (I \lambda^2)^{2/3} \text{ keV}$$

By using the experimental data for  $T_{Ga} = 270 \,^{\circ}\text{C}$  (Fig. 3b), we can obtain the relation for the initial laser pulse intensity and its intensity at the end of the experiment (after 50000 laser shots):

$$I_{\rm end} = I_{\rm initial} \left( \frac{\langle E_{\rm h} \rangle_{\rm end}}{\langle E_{\rm h} \rangle_{\rm initial}} \right)^{3/2} = 0.95 I_{\rm initial}.$$

Such a change in the Gaussian beam intensity [20] corresponds under our experimental conditions ( $\lambda = 1.24 \ \mu m$ , the beam diameter at FWHM  $\rho_0 = 0.7 \ cm$ , the focal distance of a lens  $f = 6 \ cm$ ) to the lowering of the liquid level by 3  $\mu m$ .

The liquid-level lowering can be also estimated by measuring the gallium volume evaporated by a laser pulse. Because the typical absorption coefficient of the plasma does not exceed 50 % and up to 90 % of the absorbed energy is spent to accelerate ions in a thin layer, no more than 5 % of the incident pulse energy is spent for target heating due to the heat outflow [21]. For the pulse energy of 350 µJ, this amount of heat is sufficient to evaporate approximately  $6 \times 10^{-16}$  m<sup>3</sup> of gallium (a cube with edge 8.5 µm). Then, the liquid volume should decrease by  $3 \times 10^{-11}$  m<sup>3</sup> after 50000 laser shots. The liquid surface area in our cell is  $\sim 2.5 \times 10^{-5}$  m<sup>2</sup>, and therefore, the liquid metal level should lower approximately by 1  $\mu$ m, which is smaller than the value obtained above  $(3 \mu m)$ . This can be explained by the fact that in the repetitively pulsed regime (10 Hz), a crater is formed on the surface whose depth increases with the number of pulses. This assumption is confirmed by our observations at lower temperatures of the gallium target. Thus, at  $T_{Ga} = 50$  °C, the same change in the average energy of hot electrons occurs an order of magnitude faster, after less than 5000 laser pulses. Because the gallium temperature does not affect the rate of mass removal from the target, we can assume that the gallium viscosity increases with decreasing temperature and, therefore, the relaxation time of the liquid metal surface increases after the next laser pulse. This accelerates the growth of the crater depth.

A small lowering of the liquid metal level due to formation of a shallow crater can be easily compensated

by displacing a focusing lens. This allows us to stabilise the parameters of the plasma source for considerably longer times, which is demonstrated by our experimental results presented in Fig. 4. After irradiation of one point on the target by approximately 12000 laser pulses, the X-ray yield decreased approximately by half (the experiment was performed at  $T_{Ga} = 234$  °C, which explains a rapid decrease in the X-ray yield). To recover the X-ray yield, the objective was drew closer to the target by 20 µm.



**Figure 4.** X-ray yield  $\eta$  in the spectral range above 2.5 keV as a function of the laser shot number N upon irradiation by laser pulses at a repetition rate of 10 Hz at the gallium temperature  $T_{\text{Ga}} = 234 \,^{\circ}\text{C}$ . The white curve corresponds to the adjacent average value; the black arrow indicates the instant of additional radiation focusing.



**Figure 5.** X-ray yield  $\eta$  in the spectral range above 7.5 keV (a) and the gallium temperature (b) as functions of the laser shot number *N*. The white curve corresponds to the adjacent average value.

The liquid metal level can be also controlled without the objective displacement. A heater in our setup was a copper rod, which experiences a considerable temperature expansion. Therefore, by heating or cooling the copper rod near some average temperature, we can rise or lower the liquid metal level. The corresponding experimental results are presented in Fig. 5. At the initial instant the liquid metal surface is in the objective focus, which corresponds to the maximum X-ray yield. Then, the heater and target temperature was increased until the disappearance of X-rays. At

this moment the objective focus was inside the target. After approximately 110 laser shots, the heating was terminated, and the gallium temperature began to decrease. At  $T_{Ga} \sim 130$  °C, the X-ray intensity was recovered. The difference of the initial target temperature (110 °C) from the end temperature (130 °C) is explained by the liquid-level lowering due to the removal of the target material. Note that the target level rises namely due to the expansion of the copper heater rather than the target material because the coefficient of thermal expansion of gallium is considerably lower than that of copper.

### 3. Conclusions

We have developed a stable X-ray plasma source by producing a hot plasma on the melted gallium surface irradiated by  $10^{16}$  W cm<sup>-2</sup> laser pulses at a pulse repetition rate of 10 Hz. To create the X-ray source stable for several hours, it is necessary to heat gallium up to the temperature considerably exceeding its melting point (30 °C). The X-ray yield for the gallium target heated up to 270 °C decreased after 50000 laser shots by  $\sim 25\%$  [from  $(2.2\pm0.4)\times$  $10^{-4}$  % to  $(1.7 \pm 0.4) \times 10^{-4}$  %], while the average energy of hot electrons decreased from  $9.3 \pm 0.9$  keV to  $9.0 \pm 1.1$  keV. We have also shown that the change in the parameters of the X-ray plasma source during an hour is caused by the lowering of the average level of the liquid metal in the cell and can be eliminated by a small displacement of the focusing lens or a small change in the heater temperature.

The analysis of the physical and chemical properties of various materials shows that hot dense plasmas can be also produced on the surfaces of liquid low-melting-temperature metals such as indium, bismuth, tin, lead, etc. In particular, this allows one to control the spectra of X-ray bremsstrahlung and X-ray line emission of plasmas. The average power of the X-ray source can be further increased by using femtosecond lasers with a pulse repetition rate of several kilohertz. However, it is necessary to study the stability of the X-ray source at such high pulse repetition rates.

*Acknowledgements.* This work was supported by the Russian Foundation for Basic Research (Grant No. 04-02-16341a and partially Grant Nos 05-02-16476a and 07-02-00724a) and by the ISTC (Grant No. 2651p).

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