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Peculiar Properties of Hot Plasma Formation under the Influence of Intense Femtosecond Pulses on the Surface of Molten Metal

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Abstract—The peculiar properties of plasma formation on the surface of different liquid metals by femtosecond laser radiation have been studied. It is shown that plasma formation and generation of hard X-radiation on the surface of molten metal depends substantially on the contrast and weakly on the polarization of laser radiation, which clearly distinguishes the plasma produced in our experiments from the plasma generated on the surface of a solid target.

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Plasma formed by laser radiation with an intensity ranging from 10^{16} to 10^{21} W cm^{-2}, is a very promising field of investigation [1]. In particular, this plasma can be used for developing efficient short pulse sources of X-ray and corpuscular radiation. Usually, solid-state targets [2–7], beams of atomic clusters [8, 9], microdroplets [10, 11], and jets of liquids [12] are involved in this type of experiment. As well, the surface of a liquid can be used as a target. Such a target is of great interest, since a liquid surface is capable of regeneration after laser exposure; thus, there is no need for the continuous displacement of the target during experiments. This opens interesting prospects for the development of plasma sources with a high pulse-repetition frequency.

Such an approach has been described in several publications [13–17]. Thus, in [14] the spectrum of the deceleration radiation of plasma that forms on an aqueous surface using a single femptosecond laser pulse with a 10 Hz frequency, or by using a pulse packet with a 10 ns period and a 10 Hz packet-repetition frequency, was studied. An attempt to obtain plasma under the influence of laser radiation with a 2 kHz pulse-repetition frequency was made in [13]. In our publications [16, 17] it has been shown that using molten gallium as a target and laser radiation with a 10 Hz pulse-repetition frequency it is possible to achieve stable operation of an X-ray plasma source during a time interval of more than one hour and a half.

The contrast of a femptosecond laser pulse, i.e., the presence of short prepulses or an extensive base for a pulse, influences to a great extent both the behavior of plasma formation as a whole and generation of the hot electron component of plasma [1]. In particular, the effective optimization of X-ray emission from a target and the average energy of hot electrons can be obtained by means of contrast modification. In the present work, we studied the peculiar properties of plasma formation using femtosecond laser radiation on the surface of different liquid metals, viz., how the polarization and nanosecond contrast of the radiation influence the parameters of the hot electron component of plasma. It was found that plasma formation and the generation of hard X-ray radiation on the surface of a molten metal depend substantially on contrast and weakly on the polarization of the laser radiation, which clearly distinguishes the plasma produced in our experiments from the plasma generated on the surface of a solid target.

A schematic view of the experimental rig is shown in Fig. 1. A small copper cell filled with a low-melting-point metal was placed in a vacuum chamber evacuated with a fore pump to a residual gas pressure of 10^{-2} Torr. The camera was heated with a resistance-type heater. The temperature of the molten metal was measured using a thermocouple dipped into the metal. During the experiments the maximum temperature of the metal target exceeded 600 K. Femtosecond radiation of the laser system on a Ti : Sa crystal (1 mJ, 60 fs, 805 nm, 10 Hz) was focused on the target surface using a special lens with a correction for chromatic and spherical aberrations. The focal length of the lens was 6 cm, this provided a beam 1 cm in diameter full width at half-maximum (FWHM). With this lens the laser beam was focused on a spot 4 μm in diameter with an intensity of up to 5×10^{16} W cm^{-2}. Laser radiation with *p*-polarization was directed at the target surface at an incident angle of 45°. The contrast of the laser radiation within the nanosecond time scale was measured with a fast photodiode and could be changed from 10 to 400 by means of accurate rotation of the polarizer in the Pockels cell of the regenerative amplifier. As well, the con-

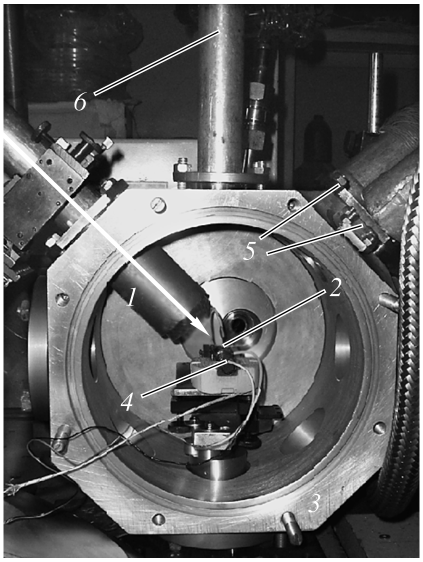

Fig. 1. Schematic view of the experimental rig: *1*–femtosecond laser pulse, *2*–liquid metal target, *3*–vacuum chamber, *4*–resistance-type heater, *5*–X-ray detectors, *6*–time-of-flight ion spectrometer.

trast could be increased by a factor of 10^3 using an additional Pockels cell maintained at the output of the amplifier.

The measurement of hard plasma X-radiation was carried out using two photoelectric multipliers equipped with an NaI(Tl) scintillator. Different attenuator assemblies made from aluminum, beryllium, and tantalum foils were placed forward of the input window of the detectors, which allowed measurement of hard X-ray emission in various spectral ranges. Estimation of the average energy of the hot electron component of the plasma was performed using methods we described previously [18]. For time-of-flight plasma diagnostics, an ion spectrometer with a 1.2 m drift was also attached to the vacuum chamber, which was placed in a position suitable for measuring ions accelerated along a normal to the target surface. The drift portion of the spectrometer, evacuating by an additional turbopump, was separated from the main vacuum chamber by a diaphragm with an orifice 0.9 mm in diameter. During experiments the working pressure in the drift portion of the spectrometer was maintained at a level of 5×10^{-5} Torr, which is essential for the proper operation of the VEU-7 microchannel plate detector. As well, high-quality vacuum conditions guarantee a nearly collisionless mode (with no more than one collision per ion) for the ions moving from the plasma to the detector.

Comparison of different physical–chemical characteristics over a great number of liquid species has shown that, due to their low saturated vapor pressure, high atomic number, etc., molten metals can be considered as an optimal choice for a liquid target [16].

We have found that the best results for stability of the laser-plasma source using liquid gallium target are obtained when the temperature of the liquid metal greatly exceeds its melting point [16, 17]. Thus, for gallium at 540 K, stable generation of X-radiation has been obtained during one hour and half of continuous operation of a laser system in the 10 Hz mode [17]. In these series of experiments the contrast of the femtosecond laser radiation was about 400. Histograms showing the number of laser shots needed to reach a certain electron temperature are presented in Fig. 2a. On average, the efficiency of the conversion of laser-pulse energy into deceleration X-ray radiation with energies of quanta greater than 2.5 keV was $(2.2 \pm 0.4) \times 10^{-4}$ %.

Expansion and optimization of the spectral range of the radiation produced by a laser-plasma source can be made by proper selection of the target material. In this connection, we studied indium and bismuth targets. Indium was heated to 510 K. The experimental results are shown in Fig. 2b. These data differed markedly from those obtained using a gallium target (Fig. 2a). First of all, a considerable data spread from one experiment to another is a characteristic feature of the results presented in Fig. 2b. Such a wide data spread cannot be connected with instability of the laser or recording systems. All our experiments can be classified in two groups. Those belonging to the first group (60% of the total number) had a 13 keV average energy of the hot electrons with a narrow width in the distribution, and those from the second group (40% of the total number) exhibit a higher average energy of the hot electrons, of 34 keV in magnitude, and a larger width of the experimental distribution. Hot electrons with an extremely high average energy, up to 100 keV, were observed in some experiments. The presence of two groups of experiments can be considered as a sign of the two physically differing mechanisms responsible for the interaction of femtosecond laser radiation with a liquid metal target.

Unusual results were obtained for the bismuth target. In this case the target temperature was 575 K. The experimental results are presented in Fig. 2c. As previously for the indium target, an increase in the atomic number of bismuth promotes further growth in the absolute yield of the hard X-ray emission, to a value of 10^{-3}%. The average energy of the hot electrons also increased to 40 keV. Our statistical analysis indicated that more than 80% of the experiments belonged in this case to the second (high-energy) distribution, which was observed for the indium target as well.

Under our experimental conditions (a contrast value of 400) the average energy of the hot electrons can be estimated in the framework of the resonant absorption model of laser radiation [1]. In that case

$$E_h \sim 12(I\lambda^2)^{1/3} \text{ keV},$$

where the intensity I is normalized to 10^{16} W cm^{-2}, and the radiation wavelength λ is normalized to 1 μm. In accordance to this estimate, the average energy of the

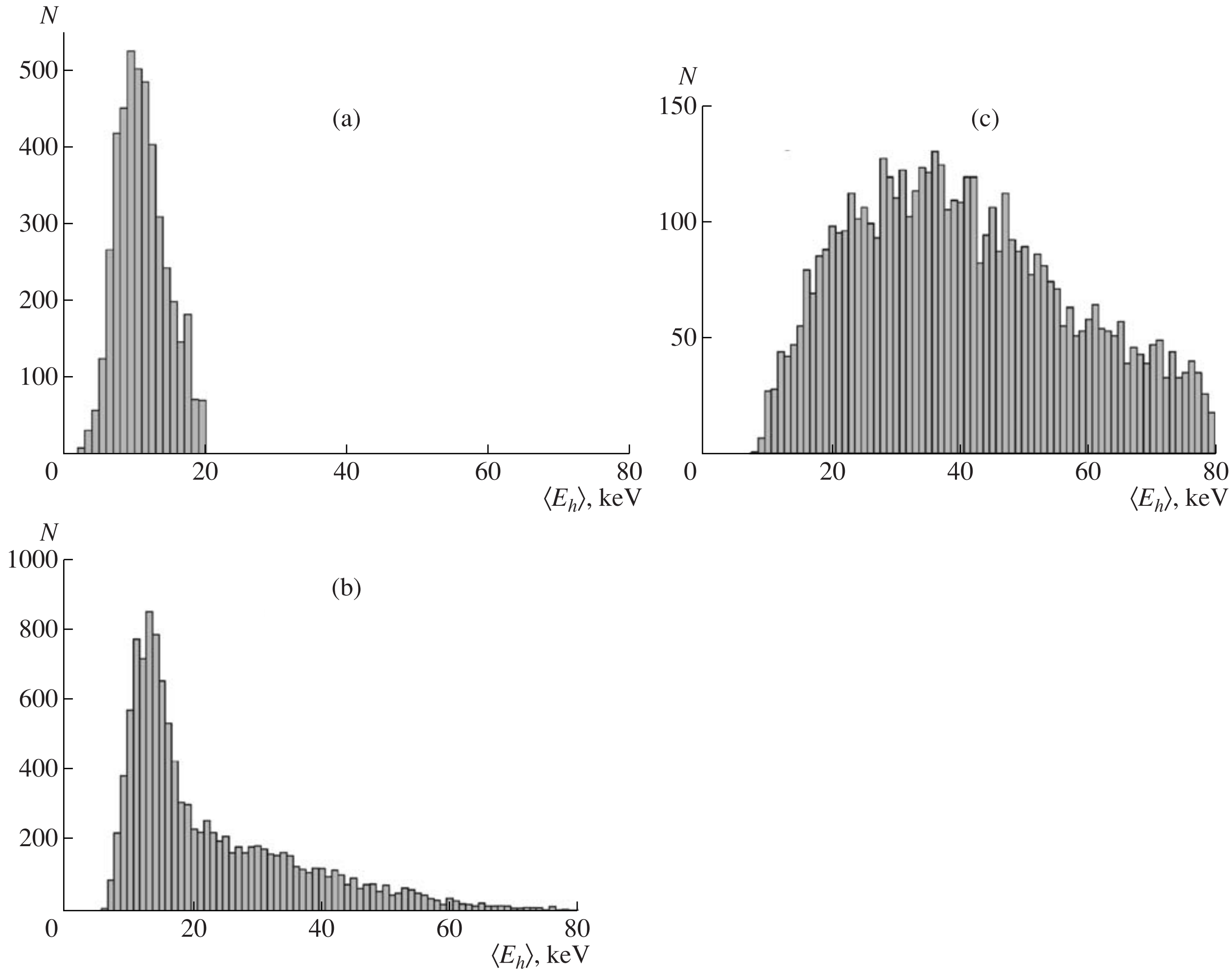

Fig. 2. Average energy distributions of the hot electrons for the plasma formed on the surface of the different metal targets: (a) gallium ($T = 540$ K), (b) indium ($T = 510$ K), and (c) bismuth ($T = 575$ K).

hot electrons should be 10 keV in our experiments. Such a value is in a good agreement with the results obtained for the gallium target, as well as with the data from the first group of experiments (with a lower average energy of the hot electrons) observed for the indium target (Figs. 2a, 2b). At the same time, the data from the second group of experiments for indium along with the data for the bismuth target are beyond the scope of given estimation. Taking these results into account, we once again think about the existence of additional physical processes responsible for the more highly efficient generation of hot electrons and their heating to a higher energy.

For more information about the peculiar properties of hot plasma formation under the influence of the femtosecond laser pulses on the surface of molten metal we performed a series of experiments aimed at clarifying how the contrast and polarization of laser radiation contribute to the observed effects. In fact, the generation of electrons via the interaction of laser radiation with a flat plasma-vacuum interface essentially depends on the contrast value (which determines, in particular, the mechanisms of physical processes responsible for the generation of the hot electrons) and the direction of polarization (since in the framework of nonrelativistic interactions hot electrons can be produced by optical radiation with only *p*-polarization).

These experiments were carried out with an indium target. Figure 3 shows the histograms of the average energy distributions of hot electrons measured for three different values of the contrast in the case of *p*- (Fig. 3a) and *s*- (Fig. 3b) polarized radiation. For *p*-polarization we found a clear increase in the average energy of the hot electrons and also found experiments with an extremely high average energy when the contrast value decreases. For *s*-polarization at a contrast value of 400 the generation of the hot electrons is observed; however, their average energy had a value of 3 keV, which is considerably lower than the 10 keV obtained for *p*-polarization. With a decrease in contrast, the average energies of hot electrons for both types of polarization come into close agreement, i.e., the generation of hot

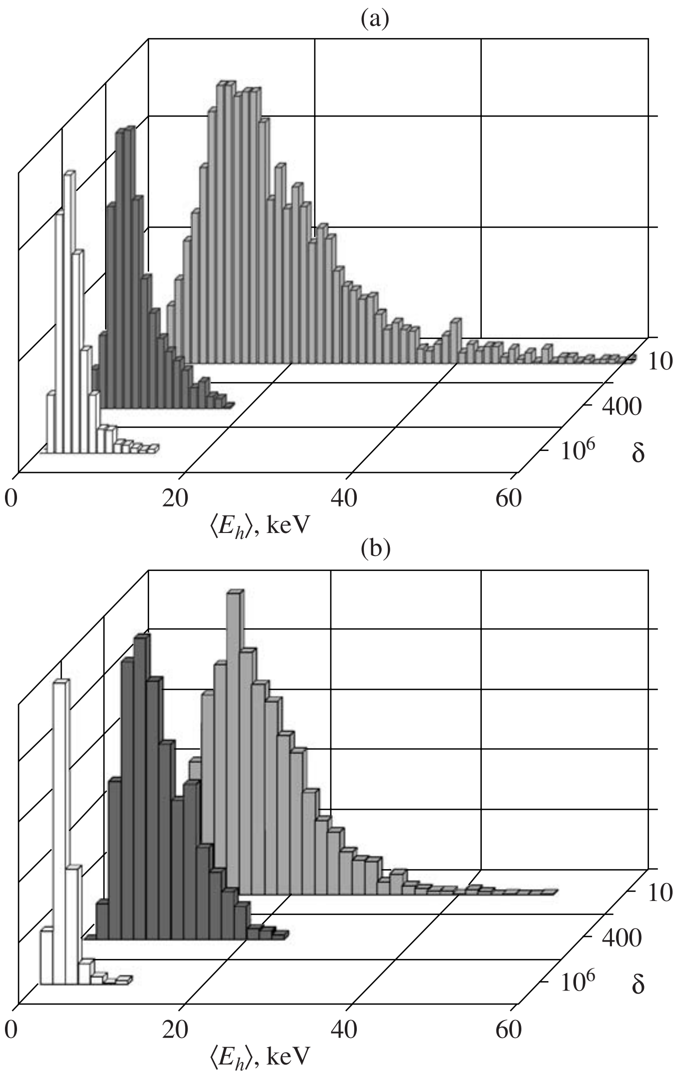

Fig. 3. Average energy distributions of the hot electrons for the plasma formed on the surface of the indium target by the laser pulse with (a)-*p* and (b)-*s* polarization, and different contrast values δ.

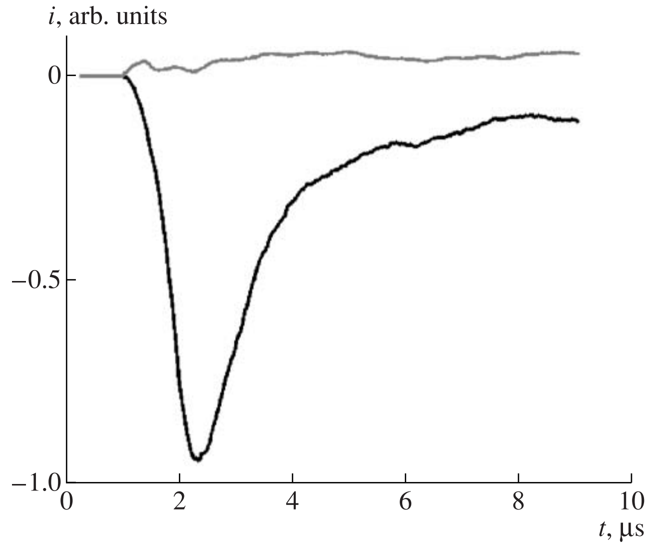

Fig. 4. Ion currents emitted from plasma at the surface of the indium target under the influence of laser radiation with *p*-polarization (the gray line was obtained for a contrast value of 10; the black line corresponds to a contrast value of 10^6).

electrons in such conditions does not depend on polarization.

To all appearance, the physical model that the most correctly explains the obtained results is described in Ref. [14]. It consists of: (i) formation of microdroplets via evaporation of a liquid by a prepulse, (ii) following condensation of evaporated species in the form of microdroplets, and (iii) interaction of the basic pulse with the microdroplets. In the cited papers the plasma was formed by femtosecond laser radiation on the water surface under atmospheric pressure. The authors observed a substantial increase in the yield and in the quasi-temperature of the hard X-ray emission using packets of femtosecond pulses with a 10 ns time delay between pulses into the packet instead of a single laser pulse. Special experiments also showed a that region with a highly heterogeneous optical density formed over the water surface. In spite of the fact that the experimental conditions are rather different (in our case the target is placed in a vacuum chamber, heavy metal with high boiling-point is used as a target, and some differences in the time structure of the radiation exist as well), the formation of metal microdroplets by the prepulse is the most reasonable explanation of the observed phenomena. In the framework of this model the weak influence of polarization is easily explained, since the generation of hot electrons via the interaction of laser radiation with spherical microdroplets takes place for every type of linear polarization of radiation. Moreover, the formation of microdroplets should occur in a different way, depending on the physical–chemical properties of the metal and increasing the target temperature over the melting-point of metal as well. This explains the observed differences in the results obtained for the gallium, indium, and bismuth targets.

To support of our hypotheses, the ion currents emitted from the plasma were measured. Actually, for plasma formation by a femtosecond laser pulse the acceleration of ions occurs mainly along a normal to the target surface. In the case of a flat target it results in a quasi-one-dimensional plasma expansion along this direction. For a spherical microdroplet the plasma expansion is three-dimensional. Taking this into account, we used a time-of-flight ion spectrometer along the normal to the target surface.

The results obtained for the indium target are shown in Fig. 4. For radiation with a high contrast value (in this case, exceeding 10^6) we observed a peak in the ion current distribution for ions with an energy of 150 keV at the peak, which corresponds to the acceleration of the ions by the ambipolar field on the plasma-vacuum interface. The maximum energy of these ions was 600 keV. A second peak (not shown in Fig. 4), corresponding to the evaporation of the target material, was recorded at a large time value. At a low contrast value (about 10) the intensity of energetic ions was below the noise-level, while the peak in the ion current distribution relating to the evaporated ion species appears approximately at the same time number as in the case of the high contrast value. Although a more detailed discussion of these findings is beyond the scope of the present publication, our time-of-flight ion current measurements confirmed the formation of microdroplets by a prepulse.

In this manner, we have shown that molten metals such as gallium, indium, and bismuth can be used as target materials for the development of a highly stable laser-plasma source operating at a 10 Hz laser pulse-repletion frequency. There is no need for target displacement during the functioning of such a source, which considerably simplifies its construction.

The time structure of the laser pulse influences the characteristics of this source to a great extent, such as the yield of hard X-ray emissions and the average energy of the hot electron component. The extremely high values of the average energy of the hot electrons observed at low contrast of the laser radiation can be explained via microdroplet formation by prepulse. The time structure of the ion currents studied in this work indicates a three-dimensional behavior of plasma expansion at a low contrast value of the laser radiation in comparison to the quasi-one-dimensional plasma expansion obtained for a high contrast value.

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