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Femtosecond plasma in dense nanostructured targets: new approaches and prospects

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1. Introduction

The recent advent of a new generation of terawatt femtosecond laser systems (FLS) (1 TW = 10^{12} W, 1 fs = 10^{-15} s) opens up fundamentally new possibilities for studying the interaction of radiation with matter and numerous applications in various fields of science and engineering. FLSs provide terawatt powers even at quite low light pulse energies from 10 to 100 mJ and the 10 – 100-fs pulse duration, which allows one, by focusing laser radiation, to obtain a huge power density light field exceeding 10^{16} W cm^{-2} (of the order of the Coulomb field in a hydrogen atom). Such superintense laser radiation, which cannot be obtained by any other method under laboratory conditions, permits the study of fundamental properties of matter under extreme conditions.

The development of this new tool for studies has a revolutionary significance for science, which can be compared with the creation of energy sources based on nuclear reactions. However, note here the fundamental difference related, first, to the miniaturization of the region of energy concentration (about 10^{-11} cm^3), which excludes the influence of the social factor on these studies despite the huge energy flux of the order of 10^{11} J cm^{-3}, and second, to the comparatively low-cost experimental setups. The advent of new types of FLSs has stimulated studies of the behavior of substances under conditions far from equilibrium. Such a statement of the problem is typical not only for fundamental studies in the fields of physics, chemistry, and biology, but also for applied studies devoted to the development of new promising technologies, which is reflected in an avalanche-like increase in the number of papers in this field and in the use of FLSs in main universities and scientific laboratories abroad. FLSs of the new generation have become basic instruments for multidisciplinary investigations.

The use of ultrashort light pulses fundamentally changes the process of interaction of laser radiation with matter, because the energy is absorbed in a thin solid layer before its hydrodynamic expansion to vacuum takes place. For femtosecond laser radiation intensities above 10^{15} W cm^{-2}, electrons acquire an energy of hundreds and thousands of electronvolts during the laser pulse, independent of the target material, resulting in the formation of a femtosecond laser plasma (FLP). The ion temperature does not vary noticeably, and after termination of the laser pulse, a strongly nonequilibrium plasma is produced in the interaction region, in which the electron subsystem has an energy that is, on the one hand, sufficient for generation of intense ultrashort X-ray radiation, stimulation of laser-controlled nuclear processes, etc. [1 – 3], and on the other hand, for efficient and precision evaporation of the irradiated volume, which forms the basis for the new femtotechnology of material processing [4 – 6].

In this paper, we report the main results obtained in the Laboratory of Superstrong Light Fields of the International Laser Center, M V Lomonosov Moscow State University, during recent years. The aim of our work is to study the ways for controlling processes proceeding in the FLP. We have shown that, by controlling plasma processes, we can produce FLP with the required temperature and electron and ion densities, and X-ray spectrum, or obtain a plasma with new combinations of these basic parameters.

A promising method for increasing absorption and 'effective' nonlinear optical susceptibility of the FLP is the excitation of surface electromagnetic waves (SEW) on targets with a periodical surface relief considered in the second section of this paper. The third section is devoted to our experiments with FLP in ultrathin freely suspended carbon films, when the thermal flow inside the target proves to be suppressed, resulting in FLP overheating. In the fourth section, we present studies of the formation and properties of a plasma in porous nanostructured substances.

Experiments were performed using the femtosecond excimer laser system, which produced a superstrong light field and has been described in a number of papers [6 – 9]. We devised a diagnostic system for studies of the FLP in our laboratory, which provided the following measurements: control of the accuracy of focusing to a spot of diameter 3 μm with the help of the second-harmonic signal in reflection and by measuring the total energy of X-ray radiation from the plasma in the energy range $E > 3$ keV using a photomultiplier with a NaJ(Tl) scintillator; detection in the second identical channel equipped with changeable filters of quanta with energies $E > 5 - 50$ keV; and detection of soft X-ray radiation by p-i-n diodes with filters ($0.05 < E < 1$ keV). The velocity of plasma expansion was measured with a time-of-flight detector based on an electron multiplier equipped with a microchannel plate.

2. Excitation of surface electromagnetic waves

Because the modulus of the wave vector of the SEW in the FLP exceeds that of the wave vector of the incident radiation, we compensated for this mismatching by using a periodically modulated surface [9 – 11]. We demonstrated for the first time the resonance generation of the second harmonic upon excitation of the SEW in the FLP and showed that SEWs modify parameters of the FLP by increasing the local-field effect, and the process of the second-harmonic generation is a sensitive indicator of the SEW excitation.

3. 'Overheating' of the femtosecond laser plasma in freely suspended thin-film targets

We carried out a series of studies [9, 12 – 14] devoted to the efficient generation of X-rays in the region of the water window[1] upon irradiation of freely suspended carbon films by femtosecond pulses of 10^{15} W cm^{-2} intensity. We showed that the optimum thickness of the film was 20 – 30 nm. For a thinner film, the conversion efficiency decreases due to the rapid hydrodynamic disintegration of the film over a time shorter than the laser pulse duration. These experimental data were confirmed by a numerical experiment, which showed

[1] The water window is the transparency region of water between 2.3 and 4.4 nm, which is optimal for microscopy of biological objects.

that the electron temperature of the plasma increased from 200 to 700 – 800 eV. Note that in the case of a 'thick' target, such a temperature can be obtained only for a laser radiation intensity exceeding 10^{17} W cm^{-2}.

Our numerical experiments with freely suspended metal-film targets (Al, Ni, Bi) showed that these targets are promising for the enhancement of the efficiency of generation of X-rays with energies above 4 keV [9]. These results were confirmed experimentally [15].

4. Formation of the FLP in porous structures

The most interesting and promising type of target are nanostructured porous materials (NPMs).They offer a number of new possibilities for controlling the parameters of the plasma being produced, as well as various new applications. NPMs consist of individual clusters several nanometers in size and have an average density which is 2 – 100 times lower than the density of solids, but exceeds the critical density for the given wavelength . Nanoclusters can form both a random fractal structure (highly porous semiconductors) and regular networks (weakly porous semiconductors, metal brushes, etc.).

The interaction of the ultrashort laser pulse with NPMs is characterized by a number of special features, which determine the possibility of controlling the plasma parameters by changing the NPM parameters: the degree of porosity P (the ratio of densities of common and porous materials) and the average size D of an individual cluster [9, 14, 16 – 19].

The reflection coefficient of a porous target is lower than that of a smooth target, resulting in strong heating of porous materials. In addition, in a porous target under the condition $\varepsilon \approx -2$ (ε is the dielectric constant of a plasma), the local field can increase, resulting in its additional heating. This condition determines the average electron density and, respectively, porosity $P \sim 65$ for which the effect of the local field enhancement is maximal (in the approximation of spherical clusters).

The second important effect, which substantially affects limiting values and the dynamics of plasma parameters, is the limitation of the thermal flow inside the target, similarly to the case of freely suspended thin films. Estimates [18] show that the relation between the thermal conductivities of porous materials and common solids can be represented in the form $\zeta/\zeta_0 \sim P^{-0.5}$, $P \sim 2$. For $P = 8$, this leads to an increase in the absorbed energy per atom by approximately a factor of three.

The cluster structure of NPMs substantially affects the processes proceeding in a plasma. The main mechanisms of generation of hot electrons in a plasma are related to electron oscillations at the plasma-vacuum interface (vacuum heating) or to the resonance absorption of laser radiation in the region of critical density. The efficiency of these mechanisms directly depends on the area of the plasma surface, which in NPMs is several hundreds times larger than the area of the laser focal spot (for $P = 6$, the ration of these areas equals 600 [17]). Therefore, one should expect an enhancement of the generation efficiency of hot electrons and hard X-ray radiation, which was indeed observed in our experiments with porous silicon [16 – 19]. We detected a rapid enhancement of the conversion efficiency to hard X-ray radiation above the porosity threshold $P \sim 5$, the enhancement rate increasing with the energy of X-rays. The hard X-ray radiation was only observed for laser radiation intensity $I > (5 \pm 1) \times 10^{15}$ W cm^{-2}. We estimated from the spectra of hard X-ray radiation ($I \sim 10^{16}$ W cm^{-2}) the temperature of hot electrons as 2.5, 5, and 7 keV for $P = 1$ (a silicon crystal), 6, and 7, respectively [17]. Note that the value 2.5 keV is in good agreement with theoretical calculations and estimates [20]. Recent experiments with porous silicon samples [19] showed that the dependence of the temperature of hot electrons on the laser radiation intensity had the form $T_h \sim I^{0.6 \pm 0.1}$ for pure silicon and $T_h \sim I^{1.0}$ for $P = 7$.

In NPMs, the role of hydrodynamic ablation substantially changes. The ablation becomes explosive, and along with the mechanism of ambipolar acceleration of ions by electrons, the Coulomb repulsion of charged particles inside a cluster becomes important. The instant charge separation inside a microplasma of an individual cluster becomes possible, because, on the one hand, the amplitude of electron oscillations in the light-wave field (~ 10 nm for $I \sim 10^{16}$ W cm^{-2}) exceeds the cluster size, and on the other hand, the laser wavelength substantially exceeds the cluster size, i.e., the microplasma is located in the uniform oscillating film and all plasma electrons are moving in phase. The time-of-flight measurements showed that the velocity of the plasma expansion for highly porous Si samples ($I \sim 3 \times 10^{16}$ W cm^{-2}) exceeded 10^8 cm s^{-1}, which corresponds to an energy 2 ± 1 MeV [19]. Under the same conditions, the expansion velocity for common silicon did not exceed 3×10^7 cm s^{-1}. In addition, we detected X-ray quanta with 50 – 100 keV energies for porous silicon, whose number did not correspond even to the electron temperature 5 – 10 keV, which also suggests the presence of additional mechanisms of generation of fast electrons.

A plasma in an NPM becomes homogeneous over approximately 500 fs and its density decreases. The formation of the homogeneous plasma is accompanied by substantial heating of the ion component due to transformation of the energy of translational motion of ions to thermal energy in collisions of 'jets' of different clusters [9]. The kinetic energy of ions in the expanding plasma can be estimated as $E \sim 30$ keV for $T_e \sim 1$ keV and its degree of ionization $Z \sim 12$. The collision mean free path λ of an ion is of the order of 30 nm and the collision frequency between ions is of the order of 2×10^{13} s^{-1}. Therefore, upon collision of two plasma jets, the kinetic energy of ions transfers to the thermal energy for 50 – 100 fs, thereby increasing it up to several tens of kiloelectronvolts. As a result, a plasma is produced which has a quite unusual, inverse relation between electron and ion temperatures. This plasma is of interest first of all for problems related to nuclear excitations in plasmas.

5. The outlook for nanostructured porous material applications

Surfaces of clusters in NPMs contain a huge number of disrupted bonds passivated by H and OH groups. If these groups are replaced by D and OD groups, the explosion of clusters in such porous silicon would induce the reaction $D + D \rightarrow \alpha + n$ (3.8 MeV) [21]. Therefore, a powerful neutron flux could be produced in the plasma when the intensity of the heating laser pulse is as low as 10^{16} W cm^{-2}.

In the high-density plasma, a great number of isomers can be excited from the low-lying ground states (the level energy is lower than 20 keV) for a time comparable or even shorter than the level lifetime [22]. Measurements of the

parameters of such low-lying isomers of stable and long-lived nuclei (lifetimes, spin, energy, decay channels, etc.) by conventional methods of nuclear spectroscopy present considerable problems.

The main excitation channels of low-lying isomers in the FLP — excitation by X-ray radiation of the plasma itself, electron-impact excitation, and reverse internal electron conversion — were discussed in papers [22 – 24]. The excitation energy of most stable isomers exceeds 5 keV [23]. Therefore, the increase in the temperature and concentration of hot electrons observed in the FLP should increase the probability of the experimental detection of excitation of the isomer nuclear level in the laser plasma.

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Experimental investigation of the influence of subterawatt femtosecond laser radiation on transparent insulators at axicon focusing

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(1) The extensive recent development of lasers generating powerful femtosecond pulses has made it possible to study the interaction of intense radiation (which can produce electromagnetic fields of the order of or even higher than intra-atomic fields) with matter. As a result, new classes of physical phenomena have been experimentally investigated, such as the generation of large-amplitude plasma waves [1, 2], the obtaining of picosecond bursts of soft X-ray radiation [3], the generation of high harmonics extending to the soft X-ray region [4], etc. In this paper, we describe a subterawatt Ti:sapphire laser setup developed in the Institute of Applied Physics, Russian Academy of Sciences, and report the results of experimental studies of the action of intense laser radiation focused with an axicon lens on transparent dielectric targets.

(2) Figure 1 shows a block diagram of the femtosecond laser setup. The driving oscillator was pumped by a CW argon laser and produced a train of 100-fs pulses of 3 nJ energy and a repetition rate of 110 MHz at the wavelength $\lambda_0 \approx 795$ nm. The pulses were amplified using the common scheme [5]. To avoid nonlinear distortions and breakdowns in optical elements of the amplifier, the femtosecond pulses generated by the driving oscillator were stretched in time to approximately 150 ps with the help of a pulse stretcher. Then, the stretched pulses were first amplified in a regenerative oscillator and then in the output amplifier. The amplified pulses with a repetition rate determined by the repetition rate of the pump Nd:YAG laser were compressed in the diffraction compressor. The operation of separate units of the setup was synchronized with an accuracy of 1 – 2 ns with the help of a specially devised synchronization scheme. The diagnostic setup included a spectrograph (for current monitoring the spectrum of the driving oscillator), an autocorrelator (for continuous monitoring of the pulse duration of the driving oscillator), and a single-pulse autocorrelator (to control the duration of amplified pulses). All the output signals of the diagnostic setup were fed into a computer and were processed using an L-305 data acquisition and processing card.

The output pulses of the laser setup had the following parameters: pulse duration $\tau \approx 120 - 140$ fs, repetition rate $f = 10$ Hz, pulse energy $W \leqslant 20$ mJ, corresponding to the radiation power $P \leqslant 0.15$ TW. The diameter of the output radiation beam was $d \approx 8$ mm.

The laser radiation was focused by a conic axicon lens with an angle $\beta = 20°$ at the base. It is known [6] that an axicon focuses the incident radiation beam into a filament directed along its axis. The maximum radiation intensity on the axicon axis in our experiments was $I \sim (1 - 5) \times 10^{14}$ W cm^{-2}. The focusing scheme is shown in Fig. 2. A target made of a transparent dielectric was located in the region of maximum intensity. In most experiments, it was mounted on a rotating table, which provided irradiation of a given spot on the target by only one laser pulse.