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Use of extended laser plasma for generation of high-order harmonics of picosecond duration

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Abstract. **We report the results of experimental investigations on the generation of picosecond radiation harmonics in extended laser plasma produced on the surface of different metal targets. The effect of plasma length, heating pulse duration and delay between the heating and transformable pulses on the efficiency of conversion to higher harmonics is studied. The λ = 1064 nm radiation conversion to a short-wavelength (down to 50 nm, 21st harmonic) range in extended plasma of several metals is demonstrated.**

Keywords: *high-order harmonic generation, laser-produced plasma, picosecond laser radiation.*

1. Introduction

High-order harmonic generation (HOHG) of laser radiation in gases [1, 2] and plasma plumes [3, 4] is a conventional technique of making coherent short-wavelength radiation sources. Research in the area of HOHG of laser radiation in gases has been especially intensive during the two past decades. A technical boost for the execution of this research has been provided by the development of high-power femtosecond laser radiation sources. The high radiation intensity achievable for a relatively small size and design simplicity of these laser sources has made it possible to radically increase the highest harmonic order and make a start on the practical application of coherent short-wavelength radiation in plasma diagnostics, biology, microscopy, photolithography, etc. [5, 6].

The use of plasma opens far broader possibilities for varying the parameters of a nonlinear medium due to a wide diversity of plasma media in comparison with the gaseous ones. However, the initial experiments did not permit raising the limiting harmonic energy and the HOHG efficiency in laser plasmas produced in the course of ablation of solid targets in vacuum [7, 8]. Further research revealed the expedience of using a weakly excited low-temperature plasma as the nonlinear medium for HOHG in the vacuum ultraviolet and soft X-ray ranges [9, 10]. Providing optimal conditions permitted raising both the highest generated harmonic order (up to the 101st) and the HOHG efficiency under close-to-resonance conditions (up to $\sim 10^{-4}$ in certain spectral regions) [11].

Despite a certain progress in HOHG, the efficiency of energy conversion to higher harmonics remains at a level suited only to diagnostic purposes. In this connection, permanently under way are attempts to develop new techniques and to search for new media for the purpose of improving the efficiency and raising the higher harmonic energy. For the same HOHG efficiency, with the use of picosecond pulses the harmonic energy density will evidently be higher than with the use of femtosecond laser radiation due to higher pulse energy. Furthermore, a relatively low cost of picosecond facilities facilitates the practical application of these coherent radiation sources in the vacuum ultraviolet and soft X-ray ranges.

It is noteworthy that the majority of HOHG studies in plasmas were performed with the use of narrow plasma plumes measuring no more than 0.5 mm [12]. The use of long plasma plumes with dimensions larger by an order of magnitude than those of the previously used plasma media [13], along with the use of picosecond pulses to suppress several adverse effects, will permit improving the efficiency of short-wavelength photon production in the course of HOHG in plasma plumes.

In this paper we outline the results of experimental investigation into the generation of picosecond radiation harmonics in an extended laser plasma produced at the surfaces of different metal targets. Investigations are made of the effect of the plasma length, the heating pulse energy and the delay between the heating and transformable pulses on the conversion efficiency to the highest harmonics in a long plasma medium. In our experiments we demonstrated the λ = 1064 nm radiation conversion to the short-wavelength (down to 50 nm, 21st harmonic) range in extended plasmas of several metals using pulses of relatively low intensity (1.5×10^{13} W cm^{-2}).

2. Experimental setup for HOHG in extended laser plasma

HOHG was investigated in an extended laser plasma produced with cylindrical focusing of heating radiation on the surfaces of several metal targets. In the course of experiments we analysed the characteristics of the higher harmonics generated in 5-mm long plasma plumes and made a comparison with the case of relatively short (0.5 mm) plasma.

The radiation source was a passively mode-locked Nd:YAG laser (λ = 1064 nm), which produced a train of 38-ps long pulses at a pulse repetition rate of 1.5 Hz. Upon extraction of a single pulse and its amplification in two ampli-

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fiers, the radiation was split into two parts. The first part of the radiation (a pulse energy $E_p \approx 5$ mJ) was used as a heating pulse (Fig. 1a) and was directed via a cylindrical lens to the target surface in a vacuum chamber. The peak intensity of the heating pulse at the target surface was equal to 2×10^{10} W cm^{-2}; the plasma plume measured 5 mm$\times$70 μm. Figure 1b shows the image of the extended plasma produced on the surface of an aluminium target. After a certain time delay, the second part of the laser pulse (a probing pulse with an energy of 28 mJ) was focused parallel to the target surface into a lengthy plasma plume. The peak intensity of the probing pulse was equal to 1.5×10^{13} W cm^{-2}. We investigated the HOHG efficiency to determine the optimal time delay between the pulses in the 5–150 ns range.

The radiation of the probing pulse and higher harmonics was analysed in a VMR-2 vacuum monochromator. The 50–300 nm harmonic radiation was detected using a scintillator (sodium salicylate) and a photomultiplier (PM).

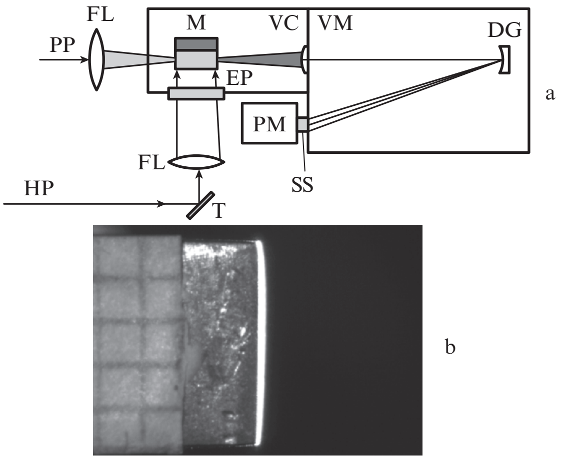

Figure 1. (a) Schematic of the experimental facility [(PP) picosecond probing pulse; (HP) picosecond heating pulse; (M) mirror; (FL) focusing lenses; (VC) vacuum chamber; (T) target; (EP) extended plasma; (VM) vacuum monochromator; (DG) diffraction grating; (SS) sodium salicylate; (PM) photomultiplier] and (b) photograph of the extended plasma at the surface of an aluminium target.

The targets were platelets of different metals (Al, Ag, Sn, Cu and Pb) measuring 5×5×2 mm. A three-axis manipulator was employed to vary the target position and, accordingly, the domain of plasma interaction with the probing radiation relative to the target plane.

3. Experimental results

The harmonics were studied both in the ultraviolet and vacuum ultraviolet spectral domains. Shown below are the results of investigation of 5th harmonic generation using plasma plumes of different length. Excess of the plasma length over the 5th harmonic coherence length must result in a decrease in the efficiency of conversion to this harmonic. If it is assumed that the amplitude of the harmonic radiation field is defined only by microscopic processes, and the role of macroscopic processes is not too important, one would expect a quadratic dependence of the 5th harmonic intensity on the length of the nonlinear medium ($I_{5h} \propto d^2$ [14]) as long as the length does not exceed the coherence length for this harmonic. The data for aluminium, tin and copper plasmas (Fig. 2) show that there occurs a moderate saturation in extended plasma, as evidenced by the slope of the $I_{5h}(d)$ dependence, which is close to two.

The qth harmonic coherence length is defined by the free-electron density N_e ($L_{coh} \sim 1.4\times10^{18}(qN_e)^{-1}$ (mm) [15, 16]), which was calculated for the aluminium target plasma with the use of a 38-ps long heating pulse and an energy density of 0.8 J cm^{-2}. These calculations were made employing the ITAP IMD code [17]. The calculated free-electron density in the aluminium plasma at a distance of 100 μm from the target surface was equal to 2.7×10^{16} cm^{-3} 30 ns after the onset of ablation. Accordingly, the coherence length for the 5th harmonic was equal to 10 mm, i.e. exceeded the plasma length (5 mm). Therefore, the phase mismatch due to the effect of free electrons should not play an important role for this harmonic. Similar results were obtained for the plasmas produced at the surface of tin and copper. It is noteworthy that the saturation and unstable generation for the 15th harmonic were observed for plasma lengths exceeding 3 mm, which is consistent with the calculated data given above.

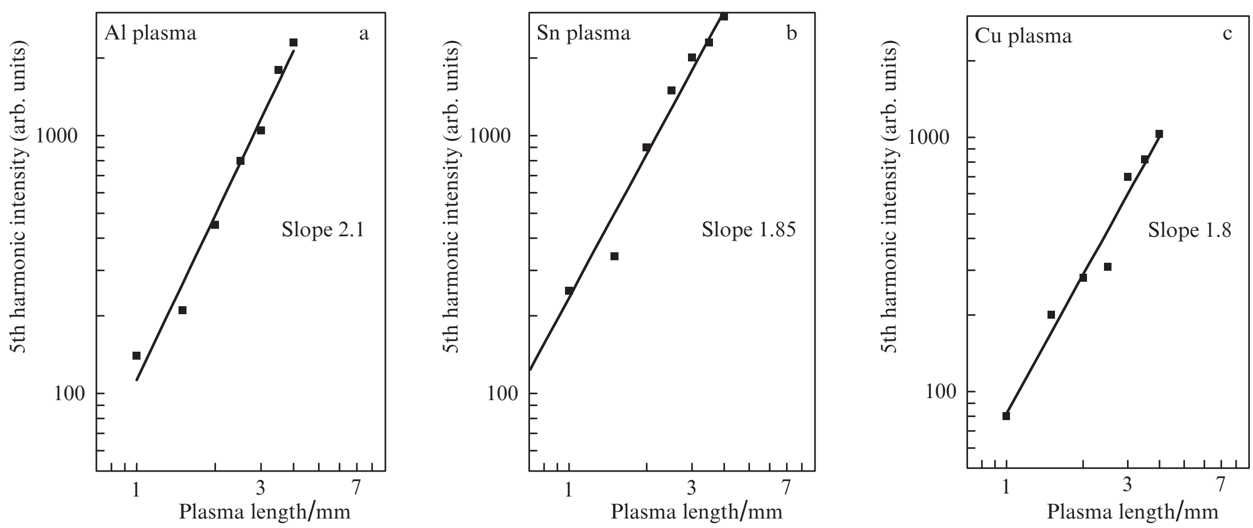

Figure 2. Dependences of 5th harmonic intensity on the lengths of plasma plumes produced at the surfaces of Al, Sn and Cu targets.

Variation of the optimal time delay between the heating and transformable pulses in the case of plasma harmonics is due to the difference in the initial velocities of particles of different elements. In accordance with the hydrodynamic plasma evaporation model subject to gas-dynamic boundary conditions [18], the initial particle mass is defined by their mass, binding energy in a solid, Debye frequency and intensity of absorbed laser radiation. A common feature of the resultant $I_h(E_p)$ dependences was a sharp decrease in harmonic intensity when the surface was irradiated by a heating pulse of energy above a certain level. The reason for this decrease is overexcitation of the plasma, which results in the emergence of free electrons and multiply charged ions and in the corresponding phase mismatch between the probing radiation and harmonic waves. This effect is more significant in the case of low-order harmonic generation. It is pertinent to note the lowering of laser frequency conversion efficiency in a highly excited plasma was also observed for the harmonics of much higher order earlier [19].

We analysed the dependence of the 9th and 11th harmonic efficiencies on the delay Δt between the heating and transformable pulses using several targets for the same extended-plasma dimensions. A typical dependence of 11th harmonic intensity on the time delay between the pulses is shown in Fig. 3 for the copper plasma. In this case, the peak of the $I_h(\Delta t)$ dependence was observed for E_p=3 mJ. Similar dependences were observed for Al, Ag, Pb and Sn targets.

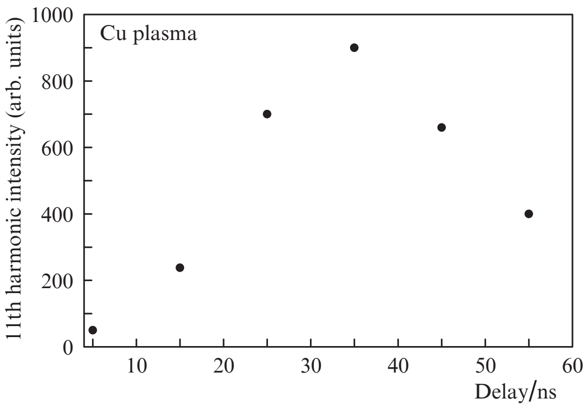

Figure 3. Dependence of the 11th harmonic intensity on the time delay between the heating and probing laser pulses in the case of extended copper plasma.

Below we analyse the mechanism of plasma formation on the target surface. The occurrence of a plasma medium above its surface is not based on the simple target surface heating and stationary processes of melting, evaporation and particle propagation with velocities defined by thermodynamic laws. For a temperature of 1000 K, the atomic velocities lie in a range of $(2-7)\times10^2$ m s^{-1}, depending on the target atomic number. During the first nanoseconds (~10 ns) after the onset of ablation, atoms and ions fly away a short distance (7 μm) from the target surface. Considering that the plasma is produced by short laser pulses, no harmonics would be observed in this experimental configuration. In particular, the laser ablation of silver produces a highly efficient 'nonlinear' plasma, which permits generating the highest harmonics, while the femtosecond pulse travels at a distance of 100 μm from the target surface. If we consider conventional particle evaporation, the velocities of the ions and neutral atoms of silver must be equal to about 5.5×10^2 m s^{-1}. During the first 30 ns they would fly away a distance of only 16 μm away from the target surface, which is insufficient to reach the axis of transformable beam propagation. This reasoning applies to the generation both of the highest and lowest harmonics.

This contradiction may be explained by another model of particle cloud production, namely by the explosive target ablation mechanism. The dynamics of plasma front propagation during laser ablation was investigated in several papers (see, for instance, [20] and references therein). A numerical analysis of this plasma development in the case of a single pulse–target surface interaction was made in Ref. [21]. The plasma formation and propagation dynamics may be analysed from plasma shadowgrams.

The features of B and Mo plasma front propagation were earlier analysed in Ref. [22]. For heavy targets (Mo), the plasma front propagated with a velocity of 6×10^4 m s^{-1}. In this case, the plasma travelled a distance of 130 μm away from the target within several nanoseconds (and not hundreds of nanoseconds if the previous estimate is adopted). It is evident that the 'optimal' plasma formation is not limited to the emergence of the plasma front in the region of transformable pulse propagation. In this case, it is necessary to produce the conditions whereby the particle density is high enough for efficient harmonic generation, while the free-electron density remains below the level at which the processes that limit the efficiency of the nonlinear conversion of laser frequency play only an insignificant role. This assumption, which was borne out by numerous experiments in high-order harmonic generation, and the experimental results outlined above suggest that optimising the generation of a low-order harmonic permits determining the conditions of plasma plume formation for the efficient conversion to higher-order harmonics.

While the efficiency of HOHG with picosecond pulses still remains low (due to the low intensity of laser radiation and lower thresholds of multiphoton ionisation of plasma particles), research along this line is being continued by the example of low-order harmonics [23].

Important information about the parameters of the extended plasma employed for HOHG may be acquired from its emission spectra. Below we present an analysis of the integral emission spectra of the plasmas of different metals employed for HOHG in the propagation of picosecond probing radiation through the plasma plume. These investigations were aimed at determining the optimal conditions for producing extended plasmas suited to efficient HOHG. The atomic and ionic emission spectra of laser plasmas were analysed in the visible and ultraviolet regions (300–700 nm). An HR4000 (Ocean Optics) fibre spectrometer was employed for recording the plasma emission spectra.

Figure 4a shows the plasma emission spectrum obtained in the ablation of silver surface. This spectrum corresponds to 'optimal plasma' conditions formed to maximise the HOHG efficiency. To this end, we maximised the efficiencies of harmonic generation in the 80–200 nm range and determined the spectral characteristics of the laser plasma produced under these conditions. One can see in Fig. 4 that the recorded spectral lines arise primarily from transitions between the states of neutral atoms and singly charged ions.

In the irradiation of metals, increasing the intensity of the heating pulses above 10^{11} W cm^{-2} resulted in an increase in the emission intensity of neutral atoms and singly charged ions, in spectral broadening and in the emergence of new spectral

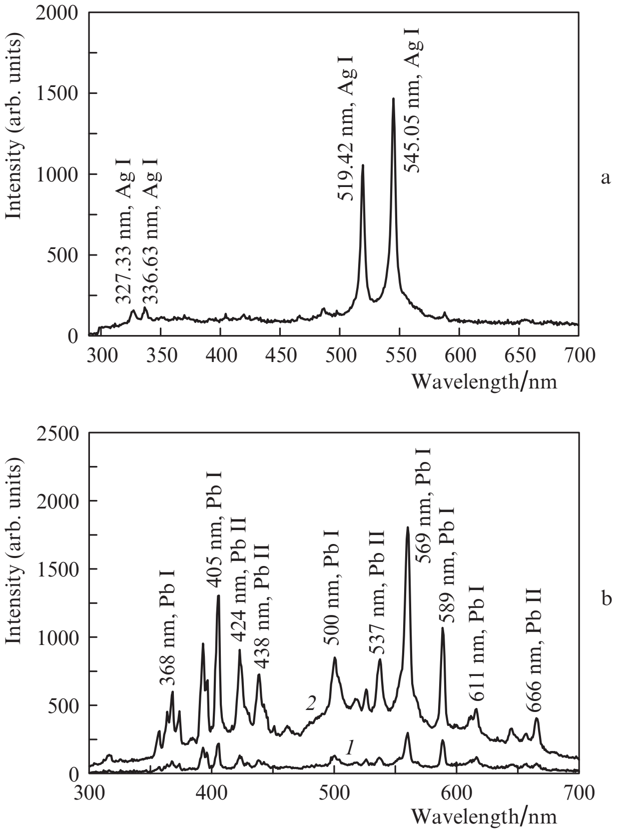

Figure 4. Emission spectra of (a) silver and (b) lead plasmas. In the case of lead plasma, the difference between the spectra is caused by the diference in heating pulse energies [(1) 3 mJ, (2) 6 mJ].

lines inherent in ions of higher charge. These changes in composition and degree of excitation of the plasma led to a significant lowering in HOHG efficiency in almost all plasma objects investigated. Figure 4b shows the lead plasma emission spectra for high and low energies of the heating pulse. One can see changes in the spectrum structure and plasma emission intensity under variation in the degree of target excitation.

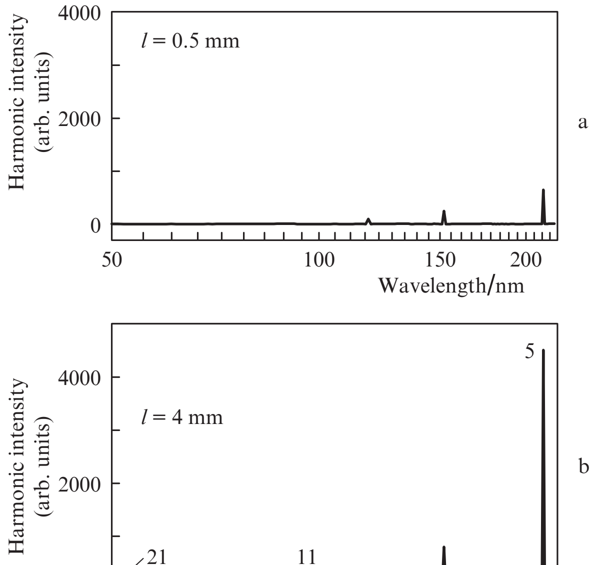

Figure 5. Harmonic spectra obtained in (a) 0.5- and (b) 4-mm long aluminium plasmas (the figures in Fig. 5b indicate harmonic orders).

The harmonic radiation in the 50–300 nm range was detected using a scintillator and a PM. The shortest wavelength corresponded to the 21st harmonic of the $\lambda = 1064$ nm radiation. Figure 5 shows the spectral harmonic distribution for a plasma produced at the surface of aluminium. Shown are the harmonic distributions generated in short and extended plasmas. In the course of experiments we demonstrated an improvement in low-order harmonic generation efficiency and an extension of the domain of plateau harmonic distribution for a 4-mm long plasma (Fig. 5b) in comparison with a 0.5-mm long plasma (Fig. 5a). A similar behaviour was observed for other plasma plumes. Our investigations suggest that the use of extended laser plasma plumes offers several advantages for harmonic generation.

4. Discussion

The recent interest in HOHG in plasma plumes, which has been displayed in different laboratories in the world [24–33], is due not only to the possibility of using this method to obtain coherent short-wavelength photons, but also to the more general problem of investigating the characteristics of substances by spectroscopic techniques invoking an analysis of higher-order nonlinear-optical properties of these substances [4, 34].

In this connection, the harmonic generation in plasma plumes should be compared in the first place with the similar process in gas jets. Three HOHG methods are known: from surfaces irradiated by extremely high fields with a high main pulse/prepulse contrast ratio, HOHG in gases, and HOHG in plasmas produced under irradiation by a heating laser pulse. The two last-mentioned mechanisms lend themselves to a similar physical description (the so-called three-stage model) and are much different from the first HOHG technique.

A comparison of HOHG in gases and in a laser plasma performed in several papers [30, 35] earlier with the use of very short pulses (3.5 and 8 fs, respectively) demonstrated the advantage of the latter medium (its part was played by a graphite plasma, while the comparison was made with argon, which is most frequently employed for HOHG) in the case of 'the lowest' (approximately from the 9th to the 19th) high-order harmonics. Quite recently, we note, for these low-order harmonics it has been possible to obtain a high conversion efficiency of 5×10^{-5} [36] in a zinc plasma, which exceeds the conversion efficiency in a graphite plasma. The methods employed for determining the absolute harmonic energies in our work and in the works mentioned above will be discussed below. So far, neither the surface- nor the gas-produced harmonics have reached this efficiency in the range between the 10th and 20th orders. Only the resonance conditions for the 13th harmonic in an indium plasma permitted approaching an efficiency of 10^{-4}. Without setting ourselves ambitious goals, we focused on the investigation of a specific problem entered in the title of our paper. Furthermore, the intensities of transformable pulses in use did not give hope for obtaining record HOHG efficiencies.

According to our investigations, when relatively long pulses are employed the effect of plasma electrons is limited to that group of free carriers which is produced in the course of laser ablation. The other group of electrons, which is produced in the course of multiphoton ionisation, does not significantly increase the phase mismatch between the interacting waves. By contrast, in the case of femtosecond pulses the laser field intensities (4×10^{14} W cm^{-2} and over) exceed the intensity required for the tunnel ionisation of singly charged ions, which leads to a growth of the density of this group of electrons. This seemingly minor factor may substantially increase the nonlinear-optical response of an extended medium at higher picosecond pulse intensities (unfortunately, unattainable with the laser facility in use).

In our case, we are dealing with determination of the optimal heating radiation flux at the target surface to maximise the harmonic yield. As the heating pulse energy increases, two processes affect the plasma dynamics. These are a growth of the density of harmonic emitters, which increases the number of converted photons due to the participation of additional individual elements of the medium, and the rise in electron density, which lowers the HOHG efficiency due to collective processes. They compete with each other and permit determining an optimum in every specific case, which was realised in these investigations. The problem may be formulated as follows: what is the cause of saturation (and the subsequent decrease) of conversion efficiency to high-order harmonics in the case of extended plasma and relatively long pulses? It is not merely that free electrons play a greater role (like in the case of short plasma) but, to a greater degree, that the coherence length (beginning with certain harmonic orders) becomes shorter than the length of the medium. Furthermore, the self-phase modulation of the interacting waves and the self-focusing of the laser pulse may be the additional effects expected in this case. These adverse effects are most likely to take place in the case of short (femtosecond) laser pulses, while the use of longer (picosecond) pulses will permit obviating involvement of these effects in HOHG limitation (under the appropriate experimental conditions). This consideration also argues in favour of the use of picosecond pulses for generating harmonics, especially low-order ones.

The absolute conversion efficiency was measured using the following technique. At the first stage, using the 'monochromator + sodium salicylate + PM' recording system we measured the signal produced by the 4th harmonic of laser radiation with a wavelength of 266-nm and an energy determined with a calorimeter. This permitted calibrating the monochromator in this spectral region. Since the quantum efficiency of sodium salicylate (25%) is constant over a broad range (between 30 and 350 nm) in the far ultraviolet, the system calibration at 266 nm permitted calculating the conversion efficiency for higher-order harmonics. The monochromator permitted recording short-wavelength radiation down to a wavelength of 50 nm. With the use of this technique we measured the conversion efficiencies for all materials. In particular, the conversion efficiency in the plateau region (15th–21st harmonics) was equal to 4×10^{-6} for aluminium plasma. Although it falls far short of the values obtained to date with the use of femtosecond pulses in plasmas, we do not regard it as being so very low, considering the low intensity of transformable radiation. We attribute this 'achievement' to the use of relatively long pulses, which limited the manifestation of adverse effects in extended plasmas. The absolute values of conversion efficiencies for the plasmas produced at the surfaces of other materials under investigation (zinc, silver, manganese, copper and tin) were somewhat lower than for the aluminium plasma. The uncertainty of efficiency measurements was estimated at 30%. A detailed description of these measurements is set forth in Ref. [37].

Investigations of extended plasmas, which were initially assumed to show no promise due to the growing adverse effect of collective phase-mismatch processes, were earlier carried out only in Ref. [25]. It was not until recently, with the discovery of quasi-phase matching in a modulated extended plasma [38, 39], that papers began to emerge in which such plasma systems are investigated with the use of femtosecond pulses (for instance, Ref. [13]).

5. Conclusions

In this work we set out the results of experimental investigations into the generation of harmonics of picosecond radiation in an extended laser plasma produced at the surfaces of targets of different metals. The effects of plasma length, heating pulse energy and time delay between the heating and transformable pulses on the efficiency of conversion to high-order harmonics were investigated. We demonstrated the conversion of $\lambda = 1064$ nm radiation to the short-wavelength region (down to 50 nm, the 21st harmonic) in the extended plasmas of several metals.

In the course of research we demonstrated an increase in harmonic intensity in an extended plasma in comparison with the short one and analysed the saturation and suppression of harmonic generation under a strong target excitation. These investigations may become the first step in the subsequent study of phase quasi-matching of coherent short-wavelength radiation with the use of relatively long laser pulses.

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