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Polarisation characteristics of the harmonics and reflected radiation upon the interaction of 100-GW picosecond pulses with aluminium films

R A Ganeev, J A Chakera, M Raghuramaiah, A K Sharma, P A Naik, P D Gupta

Abstract. The generation of harmonics from a solid surface irradiated by 27-ps pulses ($I \leqslant 1.5 \times 10^{15}$ W cm^{-2}) from a neodymium laser is studied. The conversion efficiency of p-polarised laser radiation was more than ten times greater than that of the s-polarised radiation for the second harmonic generation and more than one hundred times greater for the third harmonic generation. The optical rotation of the second harmonic generated by s- and p-polarised laser radiation is studied. The efficiencies of the second, third, and fourth harmonic generation were 2×10^{-8}, 2×10^{-10}, and 5×10^{-12}, respectively. The intensities of the second, third, and fourth harmonics exhibit a power dependence on the laser radiation intensity with exponents equal to 1.5, 1.8, and 3.8, respectively.

1. Introduction

Generation of harmonics from surfaces of solids irradiated by high-power femto- and picosecond laser pulses has lately been extensively studied both theoretically and experimentally [1 – 10]. The main features of this process had the same character in the entire range of laser pulse durations. The conversion efficiency of the p-polarised pump into harmonics was significantly greater than that of the s-polarised pump. The conversion efficiency smoothly decreased with increasing ordinal number of the harmonic.

The divergence of the harmonic beams was close to that of the laser pump for intensities of up to 10^{16} W cm^{-2}. For the parameter $I\lambda^2 > 10^{18}$ W μm^2 cm^{-2} (where I is the laser radiation intensity and λ is the radiation wavelength), the harmonics propagated isotropically. This result was obtained with the contrast $\sim 10^{-6}$, which was probably insufficient to create a steep electron-density gradient at the intensities used.

At the same time, many results are known on polarisation dependence of the conversion efficiency, the red and blue shift of the converted radiation, and the transition from the beam propagation regime of harmonics to the isotropic one. These results were different for laser pulses with different durations.

J A Chakera, M Raghuramaiah, A K Sharma, P A Naik, P D Gupta Centre for Advanced Technology, 452013 Indore, India;
R A Ganeev 'Akadempribor' Scientific and Production Enterprise, Academy of Sciences of Uzbekistan, 700143 Tashkent, Akademgorodok, Uzbekistan; e-mail: ganeev@acpr.silk.org

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The latest results on generation of harmonics from solid surfaces demonstrate that laser sources with different pulse durations produce harmonics with different spatial characteristics, different dependence of the conversion efficiency on the polarisation properties of the laser radiation, different frequency shifts, etc. The most important parameters in these experiments are the temporal characteristics of the converted radiation. In earlier experiments, laser pulses were used whose duration varied from tens of femtoseconds [11] to 2.5 ps [12]. In this work, we extend the temporal range of laser sources and study the surface generation of harmonics using 27-ps neodymium laser pulses. The main purpose of this work is to study the influence of various polarisation effects on the generation of harmonics (up to the fourth one). The results obtained are compared to the results of earlier experiments on conversion of shorter laser pulses.

2. Experimental setup

To study the generation of harmonics from the surface, we used a 100-GW neodymium-phosphate-glass laser. The laser consisted of a Nd:YLF-crystal generator with active-passive mode locking, an electrooptical selector of individual pulses, two two-pass and two one-pass neodymium-glass amplifiers, three vacuum spatial filters, a Faraday isolator, which blocked reflected radiation, and a Faraday polarisation rotator, which produced the required output polarisation.

The laser system produced one 27-ps, 2.5-J pulse every three minutes. The divergence of the laser beam was 175 μrad; the beam had a width of 33 mm (at the e^{-2} level). The ratio of the noise energy to the energy of the laser pulse was better than 10^{-5}, ensuring that no plasma was created until the arrival of the main pulse.

The laser beam illuminated a 100-μm-thick aluminium target deposited on an optically polished glass plate. After each laser shot, the target was turned about its axis so that each subsequent laser pulse interacted with an intact area of the aluminium film. The target was put in a vacuum chamber evacuated to 10^{-4} Pa.

Fig. 1 shows the scheme of the experiments. The laser beam, focused by a lens with a focal length of 750 mm, was incident on the target at an angle of 67.5°. The focused beam formed an oval spot with principal axes of 47 and 120 μm on the target surface. The maximum radiation intensity at the target surface was 1.5×10^{15} W cm^{-2}; the depolarisation degree of the radiation was less than 0.01. The Faraday polarisation rotator transformed the p-polarised radiation into the s-polarised radiation. The reflected pump and the harmonics were collimated by a quartz lens and directed into a CVI, DK480 monochromator.

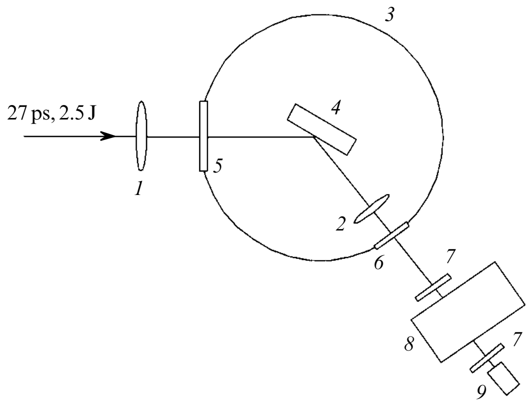

Figure 1. Scheme of the experiments: (*1*) focusing lens, (*2*) quartz collimator lens, (*3*) vacuum chamber, (*4*) target, (*5, 6*) input and output quartz windows, (*7*) filters, (*8*) monochromator, and (*9*) photomultiplier.

The second, third, and fourth harmonics were detected by three photomultipliers (Phillips 2232 at 526.5 nm, RCA 4840 at 351 nm, and Hamamatsu R166 at 263 nm) with the 3-ns time resolution. The photomultipliers were calibrated using broad-band and interference filters with known transmission at the relevant wavelengths. Digital Tektronix TDS-360 and Lecroy 9350A oscillographs detected the photomultiplier outputs with the time resolution that allowed us to separate the harmonic and laser plasma signals.

We took special precautions to avoid saturation of the photomultipliers. To measure the absolute conversion efficiencies, we employed two methods. In the first one, the number of harmonic photons incident on the photomultiplier was calculated from the photomultiplier signal, its gain, and efficiency.

Using the parameters of the calibrated filters and the monochromator transmission, we calculated the number and energy of the harmonic photons produced on the surface. The conversion efficiency was defined as the ratio of the calculated harmonic energy to the laser pump energy, which was measured by a calorimeter.

In the second method, the laser radiation was converted into the second, third, and fourth harmonics in KDP crystals; after passing through the vacuum chamber, the harmonics were detected by a calorimeter and then attenuated by the calibrated filters. Knowing the monochromator transmittivity, we determined the calibration factor between the energy of each harmonic and the measured photomultiplier signal. Using these two methods, we could find absolute harmonic conversion efficiencies differing by a factor of 2.

3. Experimental results and discussion

In the first series of the experiments, we measured the dependence of the intensities of the second and third harmonics on the target position with respect to the pump beam waist. The ratio of the second harmonic intensity to the third harmonic intensity was 100 in the region of the maximally efficient conversion.

Fig. 2 shows the dependence of the second harmonic intensity on the laser intensity for p- and s-polarised radiation. The maximum efficiency of the second harmonic conversion was 2×10^{-8} for p-polarised radiation and was 10 times less for s-polarised radiation. The experimental data can be described by power functions $I_{2\omega} \propto I_{\omega}^{1.5}$ in the case of p polarisation and $I_{2\omega} \propto I_{\omega}^{1.7}$ in the case of s polarisation.

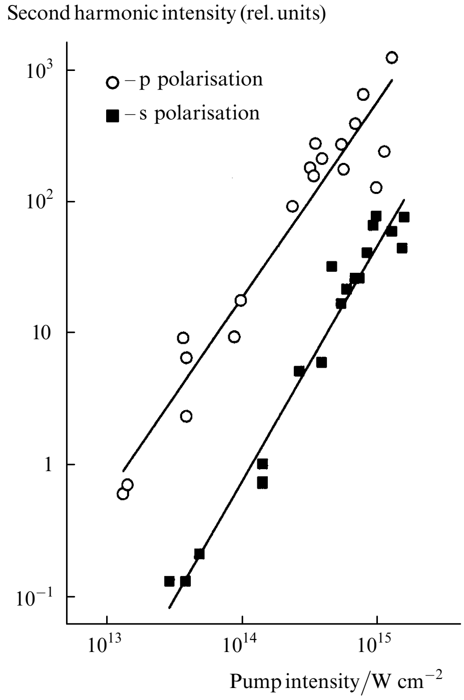

Figure 2. Dependences of the second harmonic intensity on the pump intensity in the case of s and p polarisation.

These results are comparable to the data of Refs [7, 8] (picosecond range), where the ratio between the second harmonic intensities generated by p- and s-polarised pumps was 2 – 10. In the case of shorter pulses (femtosecond range), this ratio was much greater ($\sim 10^3$, see, for example, Ref. [13]).

This difference can be explained by the Faraday rotation of longer pulses of the s-polarised pump. The rotation is caused by spontaneous megagauss magnetic fields produced in the laser plasma. Our technique of measuring the optical rotation angle was similar to the one earlier used in Ref. [14] to measure megagauss magnetic fields. To analyse the polarisation of the laser radiation reflected from the target, we utilised a Glan prism. Each of the orthogonal components was measured by calibrated calorimeters.

Fig. 3 shows the dependence of the turn angle of the pump polarisation on the pump intensity. In the case of p-polarised pump, we did not detect any noticeable optical rotation. In the case of the s-polarised pump with an intensity of $I = 10^{15}$ W cm^{-2}, the polarisation plane rotated by 9°. The high energy of the orthogonally polarised component observed in the latter case can be explained by both the optical rotation and the depolarisation caused by the interaction with the laser plasma.

To prove that this effect is caused exactly by the optical rotation, we measured the ratio of the energies detected by two calorimeters when the Glan prism was rotated about the axis of the reflected beam. This allowed us to measure

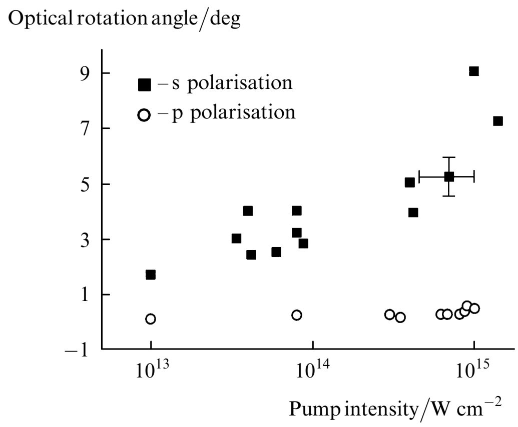

Figure 3. Dependences of the rotation angle of the pump polarisation on the intensity of the s- and p-polarised pump.

the exact angle of the optical rotation ignoring the depolarisation effects. We observed the minimum angle of the optical rotation when the Glan prism was rotated by 10°. This angle coincided with the angle of the optical rotation detected at the maximum intensity (9°, Fig. 3).

Thus, the depolarisation of radiation in the laser plasma was not important in our case. Large angles of optical rotation (22.5°) were earlier observed in Ref. [14] at similar intensities (10^{15} W cm^{-2}) and pulse durations of 100 ps. Absorption of the p-polarised radiation resulted in the creation of a magnetic field whose vector lied in the polarisation plane of the radiation. This, in turn, lowered the optical rotation angle of the p-polarised pump as compared to the case of the s-polarised pump.

The authors of Ref. [8] observed only a twofold increase in the intensity of the harmonic generated by the p-polarised pump with respect to the intensity of the harmonic generated by the s-polarised pump. They also explained this fact by the influence of optical rotation in spontaneous magnetic fields generated in the plasma. Their estimates of the optical rotation angle were based on the results of Ref. [14], where 100-ps pulses were used.

In Ref. [8] it was found that the estimates of the optical rotation angle for s-polarised radiation coincided with the measured ratio between the intensities of the harmonics generated by the two orthogonally polarised pumps even when the 2.2-ps pulses were used. In our experiments, we had longer pulses and greater interaction lengths. Thus, in our case, the Faraday rotation of the s-polarised radiation should have a greater effect on the generation of harmonics.

The increase in the optical rotation angle with increasing laser intensity can be explained by the fact that a greater fraction of the laser intensity is absorbed at greater intensities. Under our conditions, the initial target reflection of 50 % observed at low intensities ($\sim 10^{13}$ W cm^{-2}), decreased to 15 % at intensities exceeding 10^{15} W cm^{-2}. Accordingly, the strength of the spontaneous magnetic fields generated upon absorption of laser radiation reached the megagauss level, which resulted in the corresponding increase in the optical rotation angle of the reflected radiation.

We performed the experiments on the second harmonic generation from the target surface for the two orthogonal polarisations. Fig. 4 shows the dependence of the second harmonic intensity on the polarisation angle φ of the laser pump. The angles $\varphi = 0$ and $\varphi = 90°$ correspond to the p- and s-polarised pumps. The two experimental points (● and ■) at $\varphi = 90°$ show the average relative intensity of the second harmonic measured for two intensities of the s-polarised pump, 3×10^{13} and 10^{15} W cm^{-2}. One can see that the relative intensity of the second harmonic generated by the s-polarised pump increases with increasing intensity and, correspondingly, increasing absorption.

For comparison, we show in Fig. 4 the data of Ref. [5] ($I = 10^8$ W cm^{-2}, $t_p = 100$ fs, and $\lambda = 800$ nm), which correspond to the dependence $I_{2\omega} \propto \cos^4\varphi$. This dependence was obtained for pump energies significantly lower than the plasma generation threshold. The experiments [5] have shown that the generation of harmonics depends exclusively on processes that take place on the surface-vacuum interface. Comparing these results, we see that the second harmonic can be generated from the s-polarised pump only if the laser plasma is produced. In this case, a fraction of the s-polarised wave changes its polarisation, satisfying the conditions for the harmonic generation.

The earlier studies of the polarisation properties of the harmonics demonstrated that the harmonics have the same polarisation as the pump reflected from the target [1, 2, 5, 15]. Similar results are predicted by theoretical calculations. According to the moving-mirror model, the polarisation of the pump and the harmonic should be the

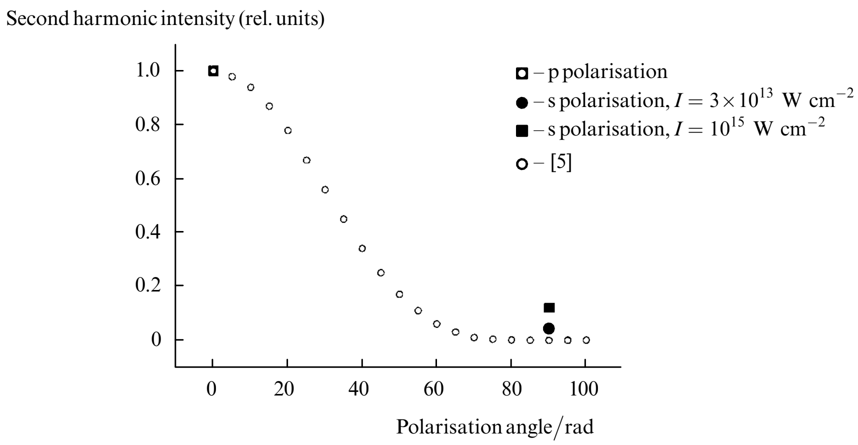

Figure 4. Dependences of the second harmonic intensity on the pump polarisation angle measured in Ref. [5] and this work.

same. Some of the rules for the polarisation dependences, which were formulated in Ref. [16] and apply to the case of a sharp plasma-vacuum interface predict that the p-polarised laser radiation should produce p-polarised odd and even harmonics.

At the same time, if the pump is s-polarised, only odd harmonics will have the same polarisation. Even harmonics will be p-polarised in this case. However, these properties are characteristic of a sharp plasma-vacuum interface, when $L/\lambda < 0.1$. In this case, the laser radiation creates a small gradient of the electron density ($L/\lambda \sim (10^7$ cm s$^{-1} \times$ 27 ps/1 μm ~ 2.7).

Far from interpreting our results within the model of the sharp plasma-vacuum interface, we would still like to note some coincidences in the behaviour of the polarisation characteristics in the two cases. We have measured the fraction of the orthogonal polarisation in the second harmonic for the s- and p-polarised pump by placing a Glan prism behind the monochromator. In the case of the p-polarised pump, virtually all radiation of the second harmonic (97 %) had the same polarisation, and this picture remained unchanged with increasing laser intensity.

At the same time, more than 30 % of the second-harmonic radiation (at the maximum intensity) had the opposite polarisation in the case of the s-polarised pump. As noted above, this effect was caused by the rotation of the pump polarisation. Even small optical rotations of the s-polarised pump increased the SHG efficiency. As a result, the curve of the SHG efficiency versus the pump intensity had a steeper slope in the case of the s-polarised pump than in the case of the p-polarised pump (see Fig. 2).

The difference between the energies of the second harmonics generated by the s- and p-polarised pumps decreases with increasing pump intensity. The creation of strong magnetic fields and the consequent optical rotations are not the only causes of this effect. Earlier, the authors of Ref. [12] have not observed any variation in the harmonic conversion efficiencies due to different pump polarisations at extreme intensities ($\sim 10^{19}$ W cm^{-2}). They argued that the radiation of such intensities disrupts the homogeneous interface between the target and vacuum.

These polarisation effects are particularly important in the case of relatively long pulses. The authors of Refs [7, 8] stressed the importance of the optical rotation observed for 0.8- and 2.2-ps pulses. Our results and their comparison with the results of earlier studies suggest that, in the case of long pulses, the optical rotation can change the course of some physical processes that take place at the surface – vacuum interface.

The efficiencies of converting the s- and p-polarised pumps into the third harmonic differed much more than the second harmonic efficiencies. The ratio of the intensities of the third harmonic generated by the p- and s-polarised pump was above 100. This agrees qualitatively with the theoretical calculations of Ref. [17], which predicted this ratio to be between ten and one hundred. In the case of the p-polarisation, the intensities of the third harmonic and the pump were related by the power function $I_{3\omega} \propto I_{\omega}^{1.8}$.

We have measured the fourth harmonic intensity as a function of the pump intensity only for the p-polarised radiation. It also obeyed a power law with an exponent of 3.8. The signal of the fourth harmonic that was generated by the s-polarised pump was below the background noise and the sensitivity threshold of our experimental setup.

Note again that our experimental conditions (small gradient of the electron density) differed substantially from the conditions of a sharp plasma-vacuum interface. The fact that some polarisation properties predicted by the existing models (electron motion induced by the optical field at the sharp plasma-vacuum interface [4, 11, 18, 19] and the $\mathbf{v} \times \mathbf{B}$ mechanism [17, 20]) coincide with our results suggests that these two regimes of the radiation-matter interaction have some common features.

4. Conclusions

Thus, we have studied the polarisation properties of the surface harmonic generation using 27-ps laser pulses of the intensity up to 1.5×10^{15} W cm^{-2}. The efficiencies of converting into the second, third, and fourth harmonics were 2×10^{-8}, 10^{-10}, and 5×10^{-12}, respectively. The dependence of the intensities of the second, third, and fourth harmonics on the pump intensity obeyed power laws with exponents 1.5, 1.8, and 3.8, respectively. The divergence of the second harmonic coincided with that of the pump.

The efficiencies of converting the p-polarised pump into the second and third harmonics were respectively 10 and 100 times greater than those in the case of the s-polarised pump. We have studied the optical rotation that was caused by the Faraday effect in strong magnetic fields created in the laser plasma. The polarisation characteristics of the reflected pump and the second harmonic have been studied in detail.

Our experiments have shown that the generation of harmonics from solid surfaces under the action of 27-ps pulses has a number of polarisation features that are similar to those observed upon pumping by femtosecond pulses. At the same time, there are a number of significant differences between them.

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