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On the influence of a strong pump-field ellipticity on the third harmonic generation

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Abstract**. The third harmonic generation in plasma ionised by a high-power pump field is theoretically studied. The dependences of the harmonic generation efficiency on the electric pump-field strength and polarisation are found. It is assumed that the observation of the maximum of generation efficiency at the nonzero pump-field ellipticity is the universal property of the harmonic generation. This effect is nonlinear and has a threshold.**

Keywords: *harmonic generation, inverse bremsstrahlung, ionisation, generation efficiency, polarisation.*

1. The study of dependences of the generation efficiency of laser-radiation harmonics on the pump-field parameters is important both for determining the conditions providing the maximum conversion efficiency of the laser pump field to harmonics and for a deeper understanding of the physical mechanism of this effect. One of the important dependences characterising the harmonic generation process is the dependence of the generation efficiency on the polarisation of the pump field.

It was found experimentally [1 – 3] that the efficiency of generation of harmonics in an atomic medium decreased monotonically with increasing the modulus $|A|$ of the degree of circular polarisation of the pump field and harmonics were not generated when the pump field was completely circularly polarised ($|A| = 1$). This corresponds to the theoretical model [4], according to which an electron knocked out of an atom to generate harmonics returns to the atomic nucleus. However, the maximum generation efficiency of some harmonics was observed experimentally [3] not for $A = 0$, i.e., not for a linear polarisation of the pump field. The authors of Ref. [3] posed the problem of finding a mechanism responsible for such a property of harmonic generation.

One of the simplest mechanisms of harmonic generation is nonlinear bremsstrahlung. The simplest medium from the theoretical point of view where this mechanism can be observed is a stripped plasma. It cannot be said that the generation of harmonics in this medium has been studied in detail. However, it was found in Ref. [5] devoted to the theory of harmonic generation by the elliptically polarised pump field in a preliminary prepared stripped plasma with electrons having the Maxwell velocity distribution that the maximum of generation efficiency is shifted from the value $A = 0$ (as in experiments [3]). It was also shown that this shift has a threshold.

Harmonics are often generated in plasmas ionised by the pump field. In this case, the velocity distribution of ionised electrons can substantially differ from the usual Maxwell distribution. In this respect, the mutual influence of the ellipticity of the pump polarisation and the velocity distribution offers a broad field for studies. In this paper, we will analyse the dependence of the efficiency of third harmonic generation on the pump-field intensity and the degree of its circular polarisation. However, plasma, considered below as a medium in which harmonics are generated, differs qualitatively from the Maxwell plasma studied in Ref. [5]. We consider the plasma of hydrogen-like ions, which is ionised by a strong electric field providing the fulfilment of the Bethe condition [6]

$$E > \frac{I_Z^2}{4Z|e|^3}, \tag{1}$$

where Z is the charge of an atomic nucleus;

$$I_Z = \frac{Z^2 m_e e^4}{2n^2\hbar^2} \tag{2}$$

is the ionisation potential of a hydrogen-like atom; n is the principal quantum number; m_e and e are the electron mass and charge. When condition (1) for the electric field strength $\boldsymbol{E}$ is fulfilled, there is no bound electrons in atoms. In other words, a gas is transformed to a plasma.

Along with (1), we also assume that a laser pulse very rapidly rises in time, so that its rise time is shorter than the time

$$t = \frac{n^2\hbar^2}{m_e e^4} = 2.5 \times 10^{-17} n^2, \tag{3}$$

which is conventionally called the time (cycle) of the electron rotation over its orbit (measured in seconds). This time corresponds to the approach to the attosecond time scale [7, 8].

Consider hydrogen-like atoms with electrons in the ground state with the quantum numbers $n = 1$, $l = 0$, and $m = 0$. The wave functions for such electrons in the momentum ($\boldsymbol{p}$) representation has the form

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$$a(p,\theta_p,\phi_p)=\frac{2^{3/2}}{\pi v_Z^{3/2}}\frac{1}{\left[1+(v/v_Z)^2\right]^2}. \tag{4}$$

Here, $v=p/m_e$ is the electron velocity and $v_Z=Ze^2/\hbar=Zv_a$ is the Coulomb velocity [9].

2. The pump field $\boldsymbol{E}$ at the frequency ω has the form

$$\begin{aligned}E_x&=E\varepsilon_x\cos(\omega t-\varphi_x),\\E_y&=E\varepsilon_y\sin(\omega t-\varphi_x),\end{aligned} \tag{5}$$

where the components of the polarisation vector ε_x and ε_y satisfy the conditions

$$\varepsilon_x^2+\varepsilon_y^2=1,\ \ \varepsilon_x\geqslant\varepsilon_y\geqslant 0. \tag{6}$$

These components determine, in particular, the degree A of circular polarisation and the degree ρ of maximum linear polarisation

$$A=-2\varepsilon_x\varepsilon_y, \tag{7}$$

$$\rho^2=\varepsilon_x^2-\varepsilon_y^2=\left(1-A^2\right)^{1/2}. \tag{8}$$

An electron oscillates in the pump field (5) at the rate $\boldsymbol{u}_E(t)$:

$$\begin{aligned}u_{Ex}&=\varepsilon_x v_E\sin(\omega t-\varphi_x),\\u_{Ey}&=-\varepsilon_y v_E\cos(\omega t-\varphi_x),\end{aligned} \tag{9}$$

where

$$v_E=\frac{|e|E}{m_e\omega} \tag{10}$$

is the amplitude of the oscillation rate.

Having been, according to (3), knocked out of atoms virtually instantly, the ionised electrons retain the memory about their distribution in atoms. As a result, the distribution function of electrons in the coordinate system oscillating at the rate $\boldsymbol{u}_E(t)$ is determined by expression (4) and has the form

$$f_{n=1}(\boldsymbol{v})=\frac{8N_e}{\pi^2 v_Z^3}\frac{1}{\left[1+(v/v_Z)^2\right]^4}. \tag{11}$$

Here, the distribution function is normalised to the electron density N_e per unit volume.

3. We will describe the nonlinear action of the pump field (5) on plasma electrons by the kinetic Boltzmann equation, which gives in the dipole approximation

$$\frac{\partial f}{\partial t}+\frac{e}{m_e}\boldsymbol{E}\frac{\partial f}{\partial\boldsymbol{v}}=J_{ee}+J_{ei}[f(\boldsymbol{v},t)], \tag{12}$$

where $f(\boldsymbol{v},t)$ is the distribution function of electrons. The form of the electron–electron collision integral J_{ee} is not important for our calculations, while the electron–ion collision integral J_{ei} will be used in the approximation

$$J_{ei}(f)=\frac{2\pi Z_{eff}e^4N_e\Lambda}{m_e^2}\frac{\partial}{\partial v_r}\frac{v^2\delta_{rs}-v_rv_s}{v^3}\frac{\partial f}{\partial v_s}, \tag{13}$$

where

$$Z_{eff}=\frac{\sum_i e_i^2N_i}{e^2N_e}; \tag{14}$$

Λ is the Coulomb logarithm; N_i is the density of the ith ions; and e_i is the charge of the ith ion. The summation in (14) is performed over the types of ions of hydrogen-like atoms.

The current of the fundamental harmonic of the pump filed is obtained in a usual way assuming that the electron collision frequency is small compared to the pump frequency (cf. [10]). For the electron-current density $\delta\boldsymbol{j}=\{\delta j_x,\delta j_y,0\}$ of higher harmonics, we obtain from (12)

$$\begin{aligned}\delta j_x&=\sum_{N=1}^{\infty}\frac{e^2N_e}{m_e\omega^2}\,\nu_{xx}^{(2N+1)}\varepsilon_xE\cos[(2N+1)(\omega t-\varphi_x)],\\\delta j_y&=-\sum_{N=1}^{\infty}\frac{e^2N_e}{m_e\omega^2}\,\nu_{yy}^{(2N+1)}\varepsilon_yE\sin[(2N+1)(\omega t-\varphi_x)],\end{aligned} \tag{15}$$

where, in particular, the effective collision frequencies $\nu_{xx}^{(3)}$ and $\nu_{yy}^{(3)}$, corresponding to the third harmonic generation, are determined by expressions

$$\nu_{xx}^{(3)}=\frac{16e^4Z_{eff}N_e\Lambda}{m_e^2\rho^3v_E^3}D\alpha_+^{(3)}, \tag{16}$$

$$\nu_{yy}^{(3)}=\frac{16e^4Z_{eff}N_e\Lambda}{m_e^2\rho^3v_E^3}D\alpha_-^{(3)}, \tag{17}$$

where

$$D=1-\frac{d}{db}+\frac{1}{3}\frac{d^2}{db^2} \tag{18}$$

is the differential operator (with the subsequent substitution $b=1$);

$$\begin{aligned}\alpha_+^{(3)}=&\frac{\sqrt{2}\rho}{(1+\rho^2)^{1/2}}\left[\left(-\frac{1}{3}+\frac{4}{3\rho^2}\right)\right.\\&\times\mathscr{F}\left(\arctan\frac{(1+\rho^2)^{1/2}}{\sqrt{2}\alpha},\frac{\sqrt{2}\rho}{(1+\rho^2)^{1/2}}\right)-\left(1+\frac{4}{3\rho^2}\right)\\&\left.\times\mathscr{E}\left(\arctan\frac{(1+\rho^2)^{1/2}}{\sqrt{2}\alpha},\frac{\sqrt{2}\rho}{(1+\rho^2)^{1/2}}\right)\right]\\&+\frac{8\alpha}{3\rho}\left[1-\left(\frac{1+2\alpha^2-\rho^2}{1+2\alpha^2+\rho^2}\right)^{1/2}\right];\end{aligned} \tag{19}$$

$$\begin{aligned}\alpha_-^{(3)}=&\frac{\sqrt{2}\rho}{(1+\rho^2)^{1/2}}\left\{\left(\frac{1}{3}+\frac{4}{3\rho^2}\right)\right.\\&\times\mathscr{F}\left(\arctan\frac{(1+\rho^2)^{1/2}}{\sqrt{2}\alpha},\frac{\sqrt{2}\rho}{(1+\rho^2)^{1/2}}\right)-\left(-1+\frac{4}{3\rho^2}\right)\\&\times\frac{1+\rho^2}{1-\rho^2}\left[\mathscr{E}\left(\arctan\frac{(1+\rho^2)^{1/2}}{\sqrt{2}\alpha},\frac{\sqrt{2}\rho}{(1+\rho^2)^{1/2}}\right)\right.\\&\left.\left.-\frac{2\sqrt{2}\alpha}{(1+\rho^2)^{1/2}}\frac{\rho^2}{\left[(1+2\alpha^2)^2-\rho^4\right]^{1/2}}\right]\right\}\\&+\frac{8\alpha}{3\rho}\left[1-\left(\frac{1+2\alpha^2+\rho^2}{1+2\alpha^2-\rho^2}\right)^{1/2}\right];\end{aligned} \tag{20}$$

$\alpha = b(\upsilon_Z/\upsilon_E)$; and $\mathscr{F}(\varphi, s)$ and $\mathscr{E}(\varphi, s)$ are the elliptic integrals of the first and second kinds, respectively.

4. Let us define the efficiency $\eta^{(3)}$ of third harmonic generation as the ratio of the time-averaged squares of the electric fields of the third harmonic and pump in the plane-wave geometry, when $\varphi_x = kz$ (where k is the wave number):

$$\eta^{(3)} = \frac{\langle (\boldsymbol{E}^{(3)})^2 \rangle}{\langle \boldsymbol{E}^2 \rangle}. \quad (21)$$

Then, according to Maxwell's equations, we obtain

$$\eta^{(3)} = \left(\frac{3}{8}\right)^2 \frac{\varepsilon_x^2 (v_{xx}^{(3)})^2 + \varepsilon_y^2 (v_{yy}^{(3)})^2}{\omega^2}. \quad (22)$$

We assume here that the dispersion condition $\omega^2 = \omega_{\rm Le}^2 + c^2k^2$, which is usual for plasma, is satisfied, where $\omega_{\rm Le}$ is the Langmuir electron frequency and c is the speed of light. By using expressions (7), (8), (16), (17), (19), and (20), we can represent (22) in the form

$$\eta^{(3)} = \left(6\,\frac{e^4 Z_{\rm eff} N_{\rm e} \Lambda}{m_{\rm e}^2 \upsilon_Z^3 \omega}\right)^2 F^{(3)}\left(\frac{\upsilon_E}{\upsilon_Z}, \rho\right), \quad (23)$$

where

$$F^{(3)}\left(\frac{1}{\alpha}, \rho\right) = \frac{\alpha^6}{2\rho^6}\Big\{(1+\rho^2)\big[(D\alpha_+^{(3)})|_{b=1}\big]^2 + (1-\rho^2)\big[(D\alpha_-^{(3)})|_{b=1}\big]^2\Big\}. \quad (24)$$

The coefficient at the function $F^{(3)}$ in (23) is equal to $5\times10^{-3}[(\Lambda/10)(N_{\rm e}/N_{\rm ec})\lambda^{-1}Z_{\rm eff}/Z^3]^2$, where $N_{\rm ec}$ is the critical electron density and λ is the pump wavelength in μm. This relation gives a quantitative estimate of the third harmonic generation efficiency.

Figures 1–3 show the dependences of the function $F^{(3)}$, determining, according to (23), the third harmonic generation efficiency, on its two arguments. Figure 1 illustrates most clearly the dependence of $F^{(3)}$ on the relative strength of the pump field υ_E/υ_Z: the generation efficiency increases at small values of υ_E/υ_Z, achieves a maximum and then decreases as $\sim \upsilon_E^{-6}$. Figure 1 shows that the dependence of the generation efficiency on the degree of circular polarisation of the pump field is nonmonotonic. Figure 2 demonstrates the behaviour of the third harmonic generation efficiency normalised to its value at $A = 0$, i.e., to the value corresponding to the linearly polarised pump. We see that the dependence of the generation efficiency on the degree of circular polarisation is nonmonotonic and has a maximum equal to 1.53 at $\upsilon_E/\upsilon_Z = 5$.

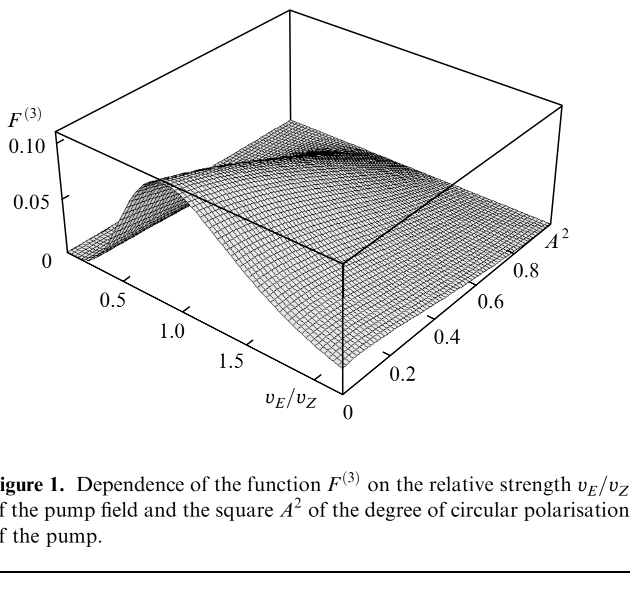

Figure 1. Dependence of the function $F^{(3)}$ on the relative strength υ_E/υ_Z of the pump field and the square A^2 of the degree of circular polarisation of the pump.

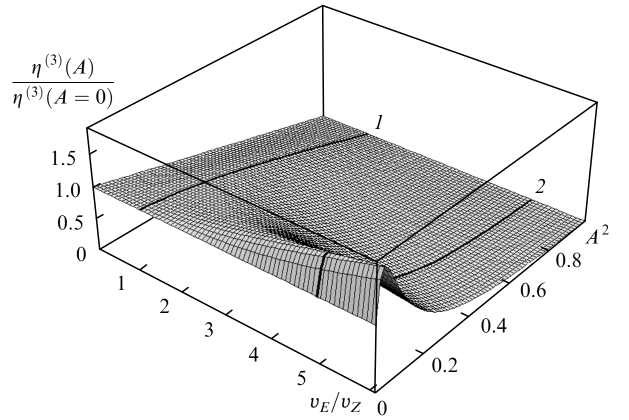

Figure 2. Dependence of the relative efficiency of third harmonic generation $\eta^{(3)}(A)/\eta^{(3)}(A=0)$ on the relative strength υ_E/υ_Z of the pump field and the square A^2 of the degree of circular polarisation of the pump. Curves (*1*) and (*2*) correspond to curves (*1*) and (*2*) in Fig. 3.

Finally, Fig. 3 illustrates the nonlinear property of the nonmonotonic dependence of the third harmonic generation efficiency on the ellipticity of the varying pump. The two curves in Fig. 3 correspond to two thick curves in three-dimensional Fig. 2. One can see that for $\upsilon_E/\upsilon_Z = 1$ [curve (*1*)], the generation efficiency decreases monotonically with increasing the degree of circular polarisation of the pump field. On the contrary, curve (*2*) in Fig. 3, corresponding to $\upsilon_E/\upsilon_Z = 5$, demonstrates the nonmonotonic dependence with a maximum achieving approximately 1.5. These two curves show that the nonmonotonic dependence $\eta^{(3)}(A)/\eta^{(3)}(A=0)$ in Fig. 3 is not only a nonlinear effect but also has a threshold.

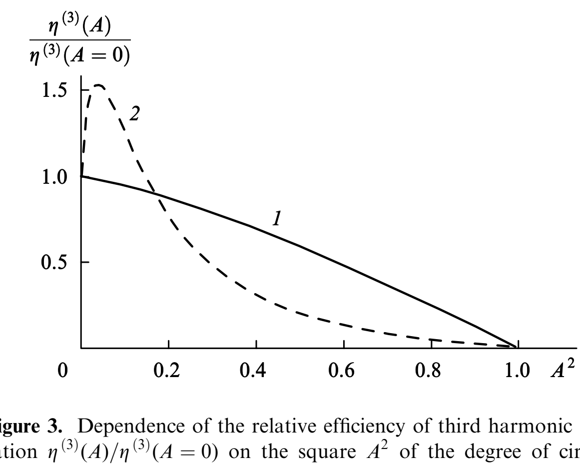

Figure 3. Dependence of the relative efficiency of third harmonic generation $\eta^{(3)}(A)/\eta^{(3)}(A=0)$ on the square A^2 of the degree of circular polarisation of the pump for $\upsilon_E/\upsilon_Z = 1$ (*1*) and 5 (*2*).

The latter is demonstrated in Fig. 4 where the absolute value of the maximum degree of circular polarisation $|A_{\max}|$, at which the maximum third harmonic generation efficiency is achieved, is shown as a function of the relative strength v_E/v_Z of the pump filed. One can see from Fig. 4 that, below the threshold value $(v_E/v_Z)_{\text{th}} = 1.46$, the generation efficiency is maximum for the linearly polarised pump. Above this threshold, the elliptic polarisation of the pump becomes preferable for achieving the maximum generation efficiency. In this case, the value $|A_{\max}| = 0.262$, corresponding to the maximum generation efficiency, is achieved at $v_E/v_Z = 2.49$.

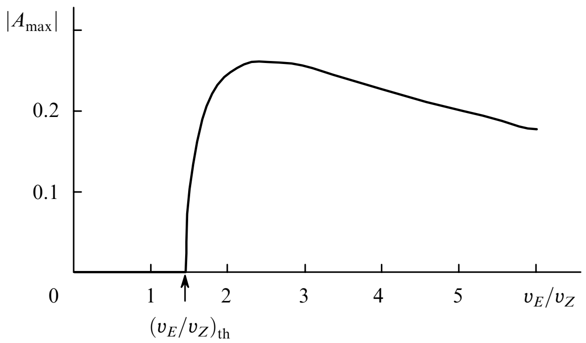

Figure 4. Absolute value $|A_{\max}|$ of the maximum degree of circular polarisation of the pump, at which the third harmonic generation efficiency achieves a maximum, as a function of the relative strength v_E/v_Z of the pump field.

5. In conclusion, we can assume that the property of harmonic generation observed in Ref. [3], which consists in the fact that the maximum generation efficiency can be achieved when the pump field is elliptically rather than linearly polarised, is a common property of the bremsstrahlung mechanism of harmonic generation. This property is essentially nonlinear and is realised above a pump-intensity threshold.

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