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Generation of laser-pulse-field harmonics in a gas upon impact ionisation of atoms

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***Abstract*. The generation of harmonics of a high-power-laser-pulse field in a gas during impact ionisation of atoms by oscillating electrons is studied theoretically. Fields are considered under conditions when the oscillation energy of electrons in the radiation field, remaining nonrelativistic, considerably exceeds the ionisation potential of an atom. In addition, the radiation field was assumed weak compared to the atomic field ($E_a = 5.1 \times 10^9$ V cm^{-1}), which allowed us to neglect the field ionisation of atoms, taking into account only the impact ionisation of atoms by oscillating electrons. Under such conditions, along with the elastic scattering of electrons, the inelastic scattering of oscillating electrons accompanied by ionisation of gas atoms can make a significant contribution to a nonlinear current induced in the plasma.**

Keywords: *generation of harmonics, strong high-frequency field, elastic collisions, inelastic collisions.*

1. Introduction: Formulation of the problem

The generation of harmonics of a high-frequency radiation field in a plasma caused by elastic scattering of oscillating electrons from ions was first considered by Silin [1]. It was assumed that the plasma was completely ionised and only elastic collisions of electrons with ions were taken into account. In this paper, we consider a plasma with an arbitrary degree of ionisation, taking into account, along with elastic collisions, also inelastic (ionising) collisions of electrons with neutral atoms. It is known (see Chapters 17 and 18 in [2]) that at large, but still non-relativistic energies, the cross sections for ionising collisions of electrons with gas atoms decrease with decreasing energy slower than cross sections for elastic collisions. Already for energies exceeding the ionisation energy, ionising collisions of electrons with atoms become dominating.

We will describe the dynamics of electrons in a weakly ionised gas in a strong high-frequency electromagnetic field of frequency ω_0

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$$\boldsymbol{E} = \boldsymbol{E}_0 \cos(\omega_0 t), \tag{1}$$

by using the kinetic equation for the electron distribution function f:

$$\frac{\partial f}{\partial t} + \frac{e\boldsymbol{E}}{m}\frac{\partial f}{\partial \boldsymbol{v}} = J_{\rm inel}(f) + J_{\rm el}(f). \tag{2}$$

Here, e and m are the electron charge and mass, and $J_{\rm inel}$ and $J_{\rm el}$ are the integrals of inelastic (ionising) and elastic collisions, respectively. We neglected in (2) the magnetic component of the Lorentz force and the gradient term in the left-hand side, which is justified for nonrelativistic oscillation velocities of electrons in field (1), i.e.

$$v_{\rm e} \equiv eE_0/(m\omega_0) \ll c. \tag{3}$$

At the same time, the field is assumed strong, so that the electron oscillation energy $\varepsilon_{\rm E}$ greatly exceeds the ionisation potential I of an atom, i.e.

$$\varepsilon_{\rm E} = \frac{1}{2}mv_{\rm e}^2 \gg I. \tag{4}$$

The representation of the filed in form (1) does not exclude its spatial inhomogeneity. In particular, this can be the field of a diverging or converging wave or of a plane wave with the nonzero wave vector. We will consider the coordinate dependence of the field below by performing spatial differentiation.

Inequality (4) allows us to neglect the chaotic motion of electrons after ionisation and to write the inelastic (ionising) collision integral in (2) in the form [3]

$$J_{\rm inel} = \delta(\boldsymbol{v})\int \nu_{\rm inel}(|\boldsymbol{v}'|) f(\boldsymbol{v}')\mathrm{d}\boldsymbol{v}'. \tag{5}$$

Here, $\nu_{\rm inel}(|\boldsymbol{v}|) = n_0 v\sigma(v)$ is the frequency of ionising electron collisions depending on v; $\sigma(v)$ is the ionisation cross section of an atom by an electron; $v = |\boldsymbol{v}|$; n_0 is the atomic density in the gas. The cross section $\sigma(v)$ can be described with good accuracy by the Born approximation [2]

$$\sigma(v) = \frac{\alpha}{v^2}\eta(v - v_{\rm i})\ln\frac{v}{v_{\rm i}}, \tag{6}$$

where

$$\eta(x) = \begin{cases} 1 & \text{for } x > 0, \\ 0 & \text{for } x < 0 \end{cases}$$

is the Heaviside formula, and $v_i = (2I/m)^{1/2}$ and $\alpha = 2\pi Z e^4/(mI)$ (Z is the charge of the atom nucleus) depend on the gas (for hydrogen, $I = 13.6$ eV and $\alpha = 16.3$ cm^4 s^{-2}).

Except restrictions (3) and (4), it is necessary to take into account another very important restriction inherent in the model under study, namely, the neglect of the tunnel ionisation of atoms in a strong electromagnetic filed compared to the impact ionisation. This is possible only in relatively weak fields, weaker than the atomic field: $E < E_a = 5.1 \times 10^9$ V cm^{-1}. For gas (hydrogen) pressures of the order of atmospheric and the field frequency $\omega_0 = 2 \times 10^{15}$ s^{-1} (the wavelength $\lambda = 1$ μm), this neglect is valid for electromagnetic field power densities $P < 10^{17}$ W cm^{-2}, and according to (3), the inequality $P > 10^{14}$ W cm^{-2} should be fulfilled. Note that the electron oscillation energy in the high-frequency field can achieve a few kiloelectronvolts.

As for the elastic collision integral, the scattering of electrons by atoms under condition (4) does not differ from the Coulomb scattering of electrons by nuclei, and therefore $J_{el}(f)$ can be written in the form of the Landau electron–ion collision integral [4]

$$J_{el}(f) = \frac{2\pi e^2 e_i^2 n_i L_0}{m^2} \frac{\partial}{\partial v_k} \left(\frac{v^2 \delta_{kj} - v_k v_j}{v^3} \frac{\partial f}{\partial v_j} \right), \qquad (7)$$

where e_i is the ion charge; $L_0 = 10 - 20$ is the Coulomb logarithm; and n_i is the total ion density in the gas, i.e. the ion density of both ionised and unionised atoms; and δ_{kj} is the Kronecker delta.

2. Frequencies of ionisation and elastic electron collisions

Before proceeding to the problem of generation of harmonics of field (1) in a gas under conditions being considered, we investigate the ionisation of the gas by using the above-presented equations and obtain the expression describing the rise of the electron density $n_e(t)$. It follows from Eqn (2), taking into account (5) and (7) that the electron density rises exponentially in time:

$$\frac{\partial n_e}{\partial t} = \gamma(E_0) n_e = n_0 \int \nu_{inel}(|\boldsymbol{v}'|) f(\boldsymbol{v}') d\boldsymbol{v}'. \qquad (8)$$

To determine the rise increment $\gamma(E_0)$ (the ionisation collision frequency of electrons), it is necessary to calculate $f(\boldsymbol{v})$. Let us assume that the inequalities

$$\omega_0 \gg \gamma(E_0), \nu_{eff}(E_0), \omega_p \qquad (9)$$

are fulfilled, where $\nu_{eff}(E_0)$ is the effective inelastic collision frequency, which is determined below, and $\omega_p = (4\pi e^2 n_e/m)^{1/2}$ is the Langmuir electron frequency. Under these assumptions, the right-hand side of Eqn (2) can be neglected in the first approximation and the solution can be represented in the form of the function of the characteristic $\boldsymbol{v} - \boldsymbol{v}_e \sin\omega_0 t = \text{const}$ [3]

$$f(t, \boldsymbol{v}) = n_e f_0(t, \boldsymbol{v}), f_0 = \delta(\boldsymbol{v}_\perp)\delta\left(v_\parallel - v\sin\omega_0 t + v_e \sin\varphi\right). \qquad (10)$$

Here, $\boldsymbol{v}_\perp$ и $v_\parallel$ are the transverse and longitudinal (with respect to the electric field $\boldsymbol{E}$) components of the electron velocity; φ is the field phase at the instant of the electron creation due to ionisation; and the function f_0 is normalised to unity. Taking into account inequalities (9), function (10) should be averaged over phases φ to obtain the known function

$$\langle f_0 \rangle = \frac{\pi^{-1}\delta(\boldsymbol{v}_\perp)}{\left[v_e^2 - (v_\parallel - v_e \sin\omega_0 t)^2\right]^{1/2}} \qquad (11)$$

describing the uniform distribution over phases [5]. By substituting (11) into (8), we obtain the expression

$$\gamma(E_0) \approx \frac{2\alpha n_0}{\pi v_i} \ln^2 \frac{v_e}{v_i} \qquad (12)$$

for the avalanche ionisation constant [6].

In conclusion of the section, we calculate the effective frequency of elastic collisions of electrons oscillating in a strong high-frequency field with gas atoms. For this purpose, we will use distribution (11) and the Coulomb cross section for scattering of electrons by ions. This approximation is justified under condition (4). In this case, the Coulomb scattering cross section should be written in the form

$$\sigma_K = \frac{2e^2 e_i^2 L_0}{m^2 v^4} \eta(v - v_i). \qquad (13)$$

Taking distribution (11) into account, we obtain

$$\nu_{eff}(E_0) = n_i \int_{v_i}^{v_e} dv \frac{2e^2 e_i^2 L_0}{\pi m^2 v^3} \frac{1}{\left(v_e^2 - v^2\right)^{1/2}} \approx \frac{e^2 e_i^2 L_0 n_i}{\pi m^2 v_i^2 v_e}. \qquad (14)$$

By comparing (14) and (12), we see that the ionisation frequency exceeds the elastic collision frequency under conditions $v_e > v_i$ and $n_e > n_i v_i / v_e$.

3. Generation of high-frequency field harmonics

Consider now the generation of harmonics of a strong high-frequency field upon gas ionisation. The generation mechanism is emission by oscillating electrons during ionisation of atoms. Here, the electron distribution function averaged over phases (11) can no longer be used, and Eqn (2) should be solved by the method of successive approximations and the correction f_1 to unaveraged distribution (10) should be found. According to (2), we have

$$\frac{\partial f_1}{\partial t} + \frac{e\boldsymbol{E}}{m} \frac{\partial f_1}{\partial \boldsymbol{v}} = J_{inel}(f_0) + J_{el}(f_0), \qquad (15)$$

where $J_{inel}(f_0)$ and $J_{el}(f_0)$ are described by expressions (5) and (7) taking (6) into account. Equation (15) differs from the equation studied in [1] by the presence of the first term taking into account inelastic electron collisions*. But because the solution of Eqn (15) is additive with respect to the right-hand side, we find first its solution taking into account only inelastic collisions and then – taking into account only elastic collisions. In the case of only inelastic collisions, the solution of Eqn (15) has the form

$$f_1(\boldsymbol{v}) = n_0 \int_{-\infty}^{t} dt' J_{inel}\left[f_0(\boldsymbol{v} - \boldsymbol{v}_e \sin\omega_0 t + \boldsymbol{v}_e \sin\varphi)\right]. \qquad (16)$$

*In addition, as mentioned above, the ion density in the term containing the elastic scattering integral is the density of all neutral and ionised atoms, which is justified by condition (4).

Because the electron density changes slowly, we find

$$\frac{\partial \boldsymbol{j}_1}{\partial t} = \frac{\partial}{\partial t}\int \boldsymbol{v} f_1 \mathrm{d}\boldsymbol{v}$$

$$= 2en_0 n_e \alpha \boldsymbol{i}_{\|} \int_{v_i}^{\infty} \mathrm{d}v \ln\frac{v}{v_i}\,\delta[v - v_e(\sin\omega_0 t - \sin\varphi)], \quad (17)$$

where $\boldsymbol{i}_{\|}$ is the unit vector along the high-frequency field direction. Expression (17) was obtained by using cross section (6) and assuming that n_e slowly varies in time with the rise increment (12). Because of the ionisation of atoms, the density of neutral atoms in the plasma also slowly varies in time and $n_e + n_0 = n_i = \mathrm{const}$.

Further calculations of the right-hand side are similar to the calculation of the Landau collision integral performed in [1]. Let us represent the delta function in the integral form, expand the integrand in the harmonics of the fundamental frequency ω_0, average over φ, assuming that the inequality $v_e \gg v_i$ is fulfilled, and carry out $\ln(v_e/v_i)$ from the integrand in the form $\ln(v_e/v_i) = L$. After simple calculations, we obtain

$$\frac{\partial \boldsymbol{j}_1}{\partial t} = \frac{4}{\pi} en_0 n_e \alpha L \boldsymbol{i}_{\|} \sum_{n\geqslant 1} \sin(n\omega_0 t) F(n), \quad (18)$$

where

$$F(n) \equiv \left[1 - (-1)^n\right]\int_0^{\infty} \frac{\mathrm{d}x}{x} J_0(x) J_n(x) = \frac{2}{\pi n^2}\sin\frac{\pi}{2}n, \quad (19)$$

and J_0 and J_n are the Bessel functions.

One can see from (18) that the expansion of the current $\boldsymbol{j}_1$ in harmonics contains only odd harmonics of the laser field, and therefore only these harmonics will be produced by this current. Then, the ratio of the amplitudes of harmonics of the electric field to the fundamental-harmonic amplitude is described by the expression

$$\frac{E_n^{\mathrm{inel}}}{E_0} \approx \frac{8n_0\alpha}{\pi^2\omega_0 v_e}\ln\frac{v_e}{v_i}\,\frac{1}{n^3(n+1)}\sin\frac{\pi}{2}n. \quad (20)$$

We derived this expression by using Maxwell's equations with current (18)

$$\mathrm{rot}\,\mathrm{rot}\,\boldsymbol{E} + \frac{1}{c^2}\frac{\partial^2\boldsymbol{E}}{\partial t^2} = -\frac{4\pi}{c^2}\frac{\partial \boldsymbol{j}_1}{\partial t}, \quad (21)$$

and assuming that the laser-pulse field (1) is a plane wave.

Relation (20), as all expressions presented above, was obtained by taking into account in (2) and (15) only inelastic (ionising) electron collisions and neglecting elastic collisions.

Elastic collisions can be simply taken into account. Thus, when only elastic collisions are taken into account in Eqn (2), the correction to the plasma current can be found in the same way as (18) and is given by the expression [cf. (18) and (19)]

$$\frac{\partial \boldsymbol{j}_1}{\partial t} = \frac{8e^2 e_i^2 e n_e L_0}{m^2 v_e v_i n(n+1)}\boldsymbol{i}_{\|}\sum_{n\geqslant 1}\sin(n\omega_0 t)\sin\frac{\pi}{2}n. \quad (22)$$

One can see from here that the current induced in the plasma is purely ohmic in this case as well and contains only odd harmonics of the high-frequency field. Taking into account only elastic collisions, the ratio of the nth harmonic amplitude to the fundamental-harmonic amplitude is described by the expression

$$\frac{E_n^{\mathrm{el}}}{E_0} \approx \frac{8e^2 e_i^2 n_i L_0}{m^2\omega_0 v_i v_e^2}\,\frac{1}{n(n+1)}\sin\frac{\pi}{2}n. \quad (23)$$

By comparing expressions (20) and (23), we see that although the amplitudes of harmonics in the case of only elastic electron collisions decrease with increasing n slower than upon inelastic collisions, they are smaller by a factor of v_i/v_e in the high-frequency field strength and therefore dominate at small n^2.

4. Discussion of results

It is interesting to compare the results obtained above with the results of paper [1] in which a completely ionised plasma in an external high-frequency field with the Maxwell distribution function with oscillating electrons was considered. Let us present the expression

$$\boldsymbol{j}_1 = \boldsymbol{E}_0\sum_{n\geqslant 0}\cos[(2n+1)\omega_0 t]\,\frac{e^2 n_e}{m\omega_0^2}\,\frac{16 n_i e_i^2\omega_0^3}{eE_0^3}\,L_0\ln\frac{eE_0}{m\omega_0 v_T} \quad (24)$$

for the current induced in the plasma and the ratio of the nth harmonic amplitude of the high-frequency field to the fundamental-harmonic amplitude

$$\frac{E_n}{E_0} \approx \frac{2n+1}{n(n+1)}\,\frac{n_i e^2 e_i^2 L_0}{m^2\omega_0 v_e^3}\ln\frac{eE_0}{m\omega_0 v_T}, \quad (25)$$

obtained in [1]. Here, v_T is the thermal velocity of plasma electrons. It follows from a comparison of expressions (23) and (25) (we can compare the results only for elastic collisions) that under the condition

$$\frac{v_i^2}{v_e^2}n^2 < 1 \quad (26)$$

the emission of field harmonics during ionisation of the gas by the field dominates, whereas in the opposite limit the field harmonics in a preliminarily prepared completely ionised plasma can be generated more intensely. Note, however, that inequality (26) determines the condition of applicability of the results obtained in [1], and this means that the most intense field harmonics will be always generated during gas ionisation by the field itself.

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