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## Third-harmonic generation in the skin layer of a hot dense plasma

G. Ferrante and M. Zarcone\*

Istituto Nazionale per la Fisica della Materia and Dipartimento di Fisica e Tecnologie Relative, Viale delle Scienze, edificio 18, 90128 Palermo, Italy

## S. A. Uryupin

P. N. Lebedev Physical Institute, Leninsky Prospekt 53, 119991, Moscow, Russia (Received 11 June 2003; published 21 July 2004)

The third-harmonic generation of a pump wave, resulting from the electron-ion collision frequency dependence on the electric field in the skin layer of a hot dense plasma is investigated. The relation of the current third harmonic with the high-frequency field in the skin layer is established for arbitrary ratios of the electron-ion collision frequency to the field frequency. For arbitrary ratios of these two frequencies, the field structure inside the skin layer is determined, and the field of the wave irradiated by the plasma at tripled frequency, too, is calculated. It has permitted us to find the explicit dependencies of the third-harmonic generation efficiency on the plasma and pump field parameters.

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#### I. INTRODUCTION

Harmonic generation (HG) occurring as a result of electron-ion collisions in the presence of a strong highfrequency electromagnetic field has been attracting attention for almost 40 years [1-5]. As a rule, excluding numerical calculations of papers [4], theoretical papers addressing this subject restrict their analysis to conditions when the frequency of the generating field is much larger than the electron-ion collision frequency. The consideration of such conditions is of interest and is appropriate when the highfrequency field interacts with a sufficiently hot plasma of relatively low density. At the same time, according to theoretical papers [1–3,5], the HG efficiency in a low-density plasma grows proportionally to the square of the electron (or the ion) density. We also recall that in the experiments [6], due to the spatial coherence effect, the dependence of HG efficiency on density may be not quadratic. In any case, in experiments, too, the efficiency in a low-density plasma is found to be higher the more dense the plasma. These findings are recalled here to underline that both theory and experiments for rare plasmas suggest that the increase of HG efficiency in electron-ion collisions requires the desirable increase of the plasma density. In turn, the increase of plasma density causes the increase of electron-ion collision frequency  $\nu$ , which may become comparable to the frequency  $\omega$ of the generating field. So, to extend the investigation of the HG efficiency to the interesting domain of sufficiently dense plasmas, one needs to work out a theory, appropriate to deal with the process of HG in the conditions, when the frequency  $\nu$  is not small as compared with  $\omega$ . Such a theory is required also in another important context of laser-plasma interaction; namely, in that concerned with experiments in which intense ultrashort laser pulses interact with solid targets. In such experiments, on the surface of a solid target, a hot dense

The paper is organized as follows: In Sec. II we obtain an appropriate solution of the kinetic equation for the electron distribution function in the presence of a given periodic electric field. The field strength is assumed to be relatively small, allowing us to describe its influence on the electron motion on the basis of perturbation theory. Within such an approach we find the linear, quadratic, and cubic corrections to the initially Maxwellian electron distribution function. The corrections are obtained for arbitrary ratios of the electron-ion collision frequency to the field frequency. In Sec. III, we determine the current density in the plasma at the frequency  $3\omega$  and investigate how it depends on the parameter  $\Omega$  $=\nu/\omega$ . It is shown, that for small  $\Omega$  values, the current density grows proportionally to  $\Omega$ , reaches its maximum value at approximately  $\Omega \approx 10$ , and then monotonically decreases according to  $\sim \Omega^{-2}$ . In Sec. IV, we describe the absorption and reflection of an electromagnetic wave normally impinging on the surface of a dense hot plasma with a sharp boundary. The relation is established between the strength of the field generating the current third harmonic in the plasma skin layer and the strength of the field impinging in the plasma. In Sec. V we report the solution of the equation for the field, containing the current source at the frequency  $3\omega$ . The relation is established between the field inside the plasma and the field of the wave emitted by the plasma at  $3\omega$ . An analytical expression for the third HG efficiency in the skin layer of a dense hot plasma is obtained and analyzed. In Sec. VI, finally, the conditions are indicated, allowing us to observe the characteristics of the third HG established in the present investigation.

plasma with a sufficiently sharp boundary is formed. The latter fact has been used in the theoretical treatment given in papers [7–11]). Besides, the conditions occur when the electron plasma frequency is much larger than both the fundamental frequency of the laser pulse and the electron-ion collision frequency. The ratio between  $\nu$  and  $\omega$  may be arbitrary. The present paper takes a step towards a theory of the third HG in just such conditions.

<sup>\*</sup>Electronic address: zarcone@unipa.it

# II. APPROXIMATE SOLUTION OF THE KINETIC EQUATION

Let us consider a fully ionized plasma, in which the multiplicity Z of ionized ions is much larger than unity,  $Z \gg 1$ . This assumption will allow us to neglect the influence of electron-electron collisions in describing the kinetics of the electron bulk. We note that the condition  $Z \gg 1$ , in its essence, does not add limitations to the domain of validity of the present theoretical treatment. As a matter of fact, the necessary condition required below, that the particles' coulomb interaction energy be small as compared to their kinetic energy for a plasma with almost solid-state density, is fulfilled only when the ion and electron temperatures are larger than hundreds of electron volts. At such temperatures, for all the media, except those formed by the most light atoms, the degree of atom ionization is much larger than unity. Let us consider that in the plasma is present an electric field of the form

$$\vec{E}(t) = \vec{E}\cos(\omega t - \delta),\tag{1}$$

where  $\vec{E} = (0,0,E)$ , while the strength E and the phase shift  $\delta$  weakly change over the distance covered by thermal electrons in the field period  $2\pi/\omega$ . In these conditions, to describe the electron response to the electric field, Eq. (1), we may use the spatially uniform kinetic equation

$$\frac{\partial}{\partial t}f + \frac{e}{m}\vec{E}(t) \cdot \frac{\partial f}{\partial \vec{v}} = St(f), \tag{2}$$

where the electron-ion collision integral is naturally taken in the Fokker-Planck form

$$St(f) = \frac{1}{2}\nu(v)\frac{\partial}{\partial v_i}(v^2\delta_{ij} - v_iv_j)\frac{\partial f}{\partial v_i}.$$
 (3)

The electron-ion collision frequency  $\nu(v)$  entering to Eq. (3) is given by

$$\nu(v) = \frac{4\pi Z e^4 n\Lambda}{m^2 v^3},\tag{4}$$

where e, m, and n are, respectively, the electron charge, mass, and density, while  $\Lambda$  is the Coulomb logarithm. Let us confine our consideration to the action on the plasma of a relatively weak electric field, in which the velocity of the electron directed motion is small compared to the electron thermal velocity. It allows us to look for a solution to Eq. (2) as a series of field strength powers. Accordingly, in the linear approximation, from Eq. (2) we have

$$\frac{\partial}{\partial t} \delta f_1 - St(\delta f_1) = -\frac{e}{m} \vec{E}(t) \cdot \frac{\partial f_m}{\partial \vec{v}}, \tag{5}$$

where  $f_m$  is the equilibrium electron Maxwellian distribution,

$$f_m = \frac{n}{(2\pi)^{3/2} v_T^3} \exp\left[-\frac{v^2}{2v_T^2}\right],\tag{6}$$

where  $v_T$  is the electron thermal velocity. Equation (5) has the solution

$$\delta f_1 = -\frac{\omega}{\omega^2 + \nu^2(v)} \left[ \omega \sin \psi + \nu(v) \cos \psi \right] \left( \vec{v}_E \cdot \frac{\partial f_m}{\partial \vec{v}} \right), \quad (7)$$

where  $\psi = \omega t - \delta$ ,  $\vec{v}_E = e\vec{E}/m\omega$ .

In the quadratic approximation, from Eq. (2) we have

$$\frac{\partial}{\partial t} \delta f_2 - St(\delta f_2) = -\frac{e}{m} \left( \vec{E}(t) \cdot \frac{\partial}{\partial \vec{v}} \right) \delta f_1. \tag{8}$$

Taking into account the solution to Eq. (7), we write the right-hand side (r.h.s.) of Eq. (8) as

$$-\frac{e}{m}\left(\vec{E}(t)\cdot\frac{\partial}{\partial\vec{v}}\right)\delta f_{1} = \frac{1}{2}\omega^{2}\left(\vec{v}_{E}\cdot\frac{\partial}{\partial\vec{v}}\right)\left\{\nu(v) + \left[\nu(v)\cos 2\psi\right] + \omega\sin 2\psi\right\}\right\} \frac{1}{\nu^{2}(v) + \omega^{2}}\left(\vec{v}_{E}\cdot\frac{\partial f_{m}}{\partial\vec{v}}\right). \tag{9}$$

According to Eq. (9), the solution to Eq. (8) may be looked for as the sum of two independent terms

$$\delta f_2 = \delta f_{20} + \delta f_{22}. \tag{10}$$

The function  $\delta f_{20}$  does not contain the periodic time dependency and it describes the quadratic in the field strength correction to the initial Maxwell electron distribution function. As we are considering a weak field case,  $\delta f_{20}$  gives negligibly small corrections to the third HG efficiency. Accordingly, there is no need to write down the solution to the equation for  $\delta f_{20}$ . At the contrary,  $\delta f_{22}$  changes with the frequency  $2\omega$  and is crucial for the following analysis. The function  $\delta f_{22}$  is found from Eq. (8), leaving in its r.h.s. [see Eq. (9)] only the terms changing with frequency  $2\omega$ . The solution to the corresponding equation has the form

$$\delta f_{22} = \frac{1}{4} \left( \frac{v_E^2}{3v^2} \frac{\partial}{\partial v} \right) \frac{\omega v^2}{v^2(v) + \omega^2} [\nu(v) \sin 2\psi - \omega \cos 2\psi] \frac{\partial f_m}{\partial v}$$

$$+ \frac{1}{2} \left[ (\vec{v}_E \cdot \vec{v})^2 - \frac{1}{3} v_E^2 v^2 \right] \frac{\omega^2}{9 v^2(v) + 4\omega^2}$$

$$\times \left\{ [3 \nu(v) \sin 2\psi - 2\omega \cos 2\psi] \times \left( \frac{1}{v} \frac{\partial}{\partial v} \right) \frac{\omega}{v^2(v) + \omega^2} \right.$$

$$\times \left( \frac{1}{v} \frac{\partial f_m}{\partial v} \right) + [3 \nu(v) \cos 2\psi + 2\omega \sin 2\psi]$$

$$\times \left( \frac{1}{v} \frac{\partial}{\partial v} \right) \frac{\nu(v)}{v^2(v) + \omega^2} \left( \frac{1}{v} \frac{\partial f_m}{\partial v} \right) \right\}.$$

$$(11)$$

Finally, taking into account the role of the function  $\delta f_{20}$ , to determine the cubic correction to electrons distribution, it is sufficient to consider the equation

$$\frac{\partial}{\partial t} \delta f_3 - St(\delta f_3) = -\omega \left( \vec{v}_E \cdot \frac{\partial}{\partial \vec{v}} \right) \delta f_{22} \cos \psi. \tag{12}$$

From Eqs. (11) and (12) it is seen that in the r.h.s. of Eq. (12) are present terms changing with frequencies  $\omega$  and  $3\omega$ . Accordingly to the linearity of Eq. (12), in its r.h.s. we leave only the terms with the frequency  $3\omega$  necessary to describe the generation of the third harmonic of the fundamental fre-

quency. Besides, in writing down the solution to the truncated Eq. (12), we write only the terms of the function  $\delta f_3$  which give nonzero contributions to the current density at  $3\omega$ . It allows, in particular, to omit that part of the function

 $\delta f_3$ , which is proportional to the combination  $(\vec{v}_E \cdot \vec{v})^3 - 3v_E^2 v^2 (\vec{v}_E \cdot \vec{v})/5$ . On the basis of the above considerations, for  $\delta f_3$  needed in what follows we have:

$$\begin{split} \delta f_{3} &= \frac{1}{3v} (\vec{v}_{E} \cdot \vec{v}) v_{E}^{2} \omega^{3} \frac{1}{\nu^{2}(v) + 9\omega^{2}} [\nu(v)\cos 3\psi + 3\omega \sin 3\psi] \\ &\times \left\{ \frac{1}{8} \frac{\partial}{\partial v} \left( \frac{1}{v^{2}} \frac{\partial}{\partial v} \right) v^{3} + \frac{2}{5} \left( \frac{1}{v^{3}} \frac{\partial}{\partial v} \right) \frac{\omega^{2}}{9\nu^{2}(v) + 4\omega^{2}} \left( v^{4} \frac{\partial}{\partial v} \right) - \frac{3}{5} \left( \frac{1}{v^{3}} \frac{\partial}{\partial v} \right) \frac{\nu(v)}{9\nu^{2}(v) + 4\omega^{2}} \left( v^{4} \frac{\partial}{\partial v} \right) \nu(v) \right\} \frac{1}{\nu^{2}(v) + \omega^{2}} \left( \frac{1}{v} \frac{\partial f_{m}}{\partial v} \right) \\ &+ \frac{1}{3v} (\vec{v}_{E} \vec{v}) v_{E}^{2} \omega^{3} \frac{1}{\nu^{2}(v) + 9\omega^{2}} [3\omega \cos 3\psi - \nu(v)\sin 3\psi] \left\{ \frac{1}{8} \frac{\partial}{\partial v} \left( \frac{1}{v^{2}} \frac{\partial}{\partial v} \right) v^{3} \frac{\nu(v)}{\omega} + \frac{3}{5} \left( \frac{1}{v^{3}} \frac{\partial}{\partial v} \right) \frac{\nu(v)\omega}{9\nu^{2}(v) + 4\omega^{2}} \left( v^{4} \frac{\partial}{\partial v} \right) \right. \\ &+ \frac{2}{5} \left( \frac{1}{v^{3}} \frac{\partial}{\partial v} \right) \frac{\omega}{9\nu^{2}(v) + 4\omega^{2}} \left( v^{4} \frac{\partial}{\partial v} \right) \nu(v) \right\} \frac{1}{\nu^{2}(v) + \omega^{2}} \left( \frac{1}{v} \frac{\partial f_{m}}{\partial v} \right). \end{split} \tag{13}$$

In Eq. (13), the differential operators over the velocity act on all the functions staying on the right side.

#### III. CURRENT THIRD HARMONIC

Let us calculate the current density  $\delta \vec{j}_3$  at the frequency  $3\omega$ . By definition, the current density is given by

$$\delta \vec{j}_3 = e \int d\vec{v} \, \vec{v} \, \delta f_3. \tag{14}$$

Using the relation (13), after averaging over the velocity vector angles from Eq. (14), we find

$$\delta \vec{j}_3 = 10^{-4} J_3(\Omega) \frac{\omega_L^2}{\omega} \frac{v_E^2}{v_T^2} \vec{E} \cos[3\psi - \Delta_3(\Omega)],$$
 (15)

where  $\omega_L = \sqrt{4\pi e^2 n/m}$  is the electron plasma frequency,  $\nu = \nu(v_T)$ , the effective frequency of thermal electron collisions with ions. The functions  $J_3(\Omega)$  and  $\Delta_3(\Omega)$ , which determine the amplitude and the phase shift of the current third harmonic, depend only on the parameter  $\Omega$  and are given by the expressions

$$J_3(\Omega) = \{ [j_{c1}(\Omega) + j_{c2}(\Omega)]^2 + [j_{s1}(\Omega) + j_{s2}(\Omega)]^2 \}^{1/2},$$
(16)

$$\cos[\Delta_3(\Omega)] = \frac{[j_{c1}(\Omega) + j_{c2}(\Omega)]}{J_3(\Omega)},$$
(17)

$$\sin[\Delta_3(\Omega)] = \frac{[j_{s1}(\Omega) + j_{s2}(\Omega)]}{J_3(\Omega)},$$
(18)

in which use is made of the notations

$$j_{c1(s1)} = -\frac{10^4}{18\pi\sqrt{2\pi}} \int_0^\infty dt \, I_{c1(s1)} \frac{t^6}{\Omega^2 + 9t^6}$$

$$\times \left\{ \frac{1}{8} \frac{\partial}{\partial t} \left( \frac{1}{t^2} \frac{\partial}{\partial t} \right) t^3 + \left( \frac{2}{5t^3} \frac{\partial}{\partial t} \right) \frac{t^6}{9\Omega^2 + 4t^6} \left( t^4 \frac{\partial}{\partial t} \right) \right.$$

$$\left. - \left( \frac{3}{5t^3} \frac{\partial}{\partial t} \right) \frac{\Omega^2}{9\Omega^2 + 4t^6} \left( t^7 \frac{\partial}{\partial t} \right) \frac{1}{t^3} \right\} \frac{t^6}{\Omega^2 + t^6}$$

$$\times \exp\left( -\frac{t^2}{2} \right), \tag{19}$$

$$j_{c2(s2)} = -\frac{10^4}{18\pi\sqrt{2\pi}}\Omega\int_0^\infty dt \, I_{c2(s2)} \frac{t^6}{\Omega^2 + 9t^6}$$

$$\times \left\{ \frac{1}{8} \frac{\partial}{\partial t} \left( \frac{1}{t^2} \frac{\partial}{\partial t} \right) + \left( \frac{3}{5t^3} \frac{\partial}{\partial t} \right) \frac{1}{9\Omega^2 + 4t^6} \left( t^7 \frac{\partial}{\partial t} \right) \right.$$

$$\left. + \left( \frac{2}{5t^3} \frac{\partial}{\partial t} \right) \frac{t^6}{9\Omega^2 + 4t^6} \left( t^4 \frac{\partial}{\partial t} \right) \frac{1}{t^3} \right\} \frac{t^6}{\Omega^2 + t^6}$$

$$\times \exp\left( -\frac{t^2}{2} \right), \tag{20}$$

$$I_{c1} = -I_{s2} = \Omega, \quad I_{s1} = I_{c2} = 3t^3.$$
 (21)

The functions  $J_3(\Omega)$  and  $\Delta_3(\Omega)$  take a simple form in the limits of small and large  $\Omega$ . In the case of a high-frequency field, when

$$\omega \gg \nu$$
, (22)

from Eqs. (16)–(21), up to terms linear in  $\Omega = \nu/\omega \ll 1$ , we find

$$J_3(\Omega) \simeq \frac{10^3}{24\pi\sqrt{2\pi}}\Omega, \quad \Omega \ll 1,$$
 (23)

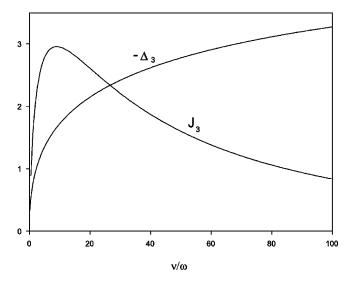


FIG. 1. Phase shift  $-\Delta_3(\nu/\omega)$  and current third-harmonic amplitude  $J_3(\nu/\omega)$ , in relative units, versus the ratio of the electron-ion collision frequency  $\nu$  to the field frequency  $\omega$ .

$$\Delta_3(\Omega) \ll 1, \quad \Omega \ll 1.$$
 (24)

Obviously, the results (23) and (24) also follow from the theory of HG based on the mechanism of electron-ion collisions (see Refs. [1] and [5]), where from the beginning it is assumed that the electron-ion effective collision frequency is much smaller than the field frequency. In the opposite limiting case, when

$$\nu \gg \omega$$
, (25)

from Eqs. (16)-(21) we approximately have

$$J_3(\Omega) \simeq \frac{105}{16\pi} \frac{10^4}{\Omega^2}, \quad \Omega > 100,$$
 (26)

$$\Delta_3(\Omega) \simeq -\frac{3\pi}{2}, \quad \Omega > 100.$$
(27)

The results of the numerical calculations of  $J_3(\Omega)$  and  $\Delta_3(\Omega)$ are shown in Fig. 1. Comparing the curves obtained numerically with the simple analytic dependencies Eqs. (23), (24), (26), and (27), one can see that the domains of validity of the latter are restricted to very small and to very large values of the parameter  $\Omega$ , respectively. In the range of  $\Omega$  values that are important for applications, where the efficiency of current third HG is the most large, from Fig. 1 it is seen that the function  $J_3(\Omega)$  exhibits a useful quantitative property. Namely, according to Fig. 1, the function  $J_3(\Omega)$  changes by no more than a factor of 3 in the wide and most interesting region of the  $\Omega$  values, when  $1 \le \Omega \le 100$ . We note also that the current third HG efficiency has its maximum at  $\Omega \simeq 10$ with the value  $\max[J_3(\Omega)] \approx 3$ . The phase shift in the maximum, as seen from Fig. 1, is close to  $-\pi/2$ . From Fig. 1 it is also seen that by increasing  $\Omega$ , the phase shift is monotonically decreasing from zero to  $-3\pi/2$ . This last value of the phase shift  $\Delta_3(\Omega)$  takes place at rather high  $\Omega$  values, and corresponds to the situation when the current is generated in opposition of phase with the field. In concluding this section we observe that the nonmonotonic dependence of the function  $J_3(\Omega)$  on  $\Omega$  established here is similar to the dependence on plasma density of the current third harmonic found in Ref. [4] as a result of numerical solution of a quantum kinetic equation.

#### IV. THE FIELD IN THE PLASMA

In this section we study the field in the plasma in the conditions when the ratio of the electron-ion effective collision frequency to the field frequency is assumed neither small nor large, as it is usually done in the theory of high-frequency  $(\omega \gg \nu)$  or normal  $(\nu \gg \omega)$  skin effect. With this aim, we first find the current density at the field frequency. From Eq. (7), by definition, we have

$$\delta \vec{j}_1 = e \int d\vec{v} \, \vec{v} \, \delta f_1 = J_1(\Omega) e n \vec{v}_E \cos[\psi - \Delta_1(\Omega)], \quad (28)$$

where the functions  $J_1(\Omega)$  and  $\Delta_1(\Omega)$  are given by

$$J_1(\Omega) = [j_c^2(\Omega) + j_s^2(\Omega)]^{1/2}, \tag{29}$$

$$\Delta_1(\Omega) = \arctan \left[ \frac{j_s(\Omega)}{j_c(\Omega)} \right]. \tag{30}$$

In writing down the relations (29) and (30), the following notations are used:

$$j_{c(s)} = \frac{2}{3\sqrt{2\pi}} \int_0^\infty dt \frac{t^7}{\Omega^2 + t^6} I_{c(s)} \exp\left(-\frac{t^2}{2}\right), \tag{31}$$

where  $I_c = \Omega$ ,  $I_s = t^3$ . For small and large  $\Omega$  values, the functions  $J_1(\Omega)$  (29) and  $\Delta_1(\Omega)$  (30) yield known asymptotic expressions

$$J_1(\Omega) \simeq 1, \ \Delta_1(\Omega) \simeq \frac{\pi}{2} - \frac{2\Omega}{3\sqrt{2\pi}}, \ \Omega \ll 1,$$
 (32)

$$J_1 \simeq \frac{32}{\sqrt{2\pi\Omega}}, \quad \Delta_1(\Omega) \simeq \frac{315\sqrt{2\pi}}{32\Omega} \ll 1, \quad \Omega \gg 10. \quad (33)$$

In Fig. 2,  $J_1(\Omega)$  and  $\Delta_1(\Omega)$  are plotted for intermediate  $\Omega$  values. According to Fig. 2, both the current density phase shift and amplitude decrease monotonically, increasing the ratio of the collision frequency  $\nu$  to the field frequency  $\omega$ .

Let us use the current density (28)–(31) to determine the field in the plasma, which we represent as [see Eq. (1)]

$$\vec{E}\cos(\omega t - \delta) = \frac{1}{2}\vec{E}(x)\exp(-i\omega t) + \text{c.c.},$$
 (34)

where  $\vec{E}(x) = (0, 0, E(x))$ . Then, for the function E(x) from the Maxwell equations and Eq. (28), we have the equation

$$\frac{d^2}{dx^2}E(x) + \frac{\omega^2}{c^2}E(x) = -iJ_1(\Omega)\frac{\omega_L^2}{c^2}E(x)\exp[i\Delta_1(\Omega)].$$
(35)

The expression (28) for the current density, determining the r.h.s. of Eq. (35), has been obtained for arbitrary values of

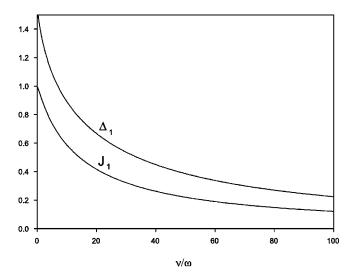


FIG. 2. Phase shift  $\Delta_1(\nu/\omega)$  and current amplitude  $J_1(\nu/\omega)$  of the fundamental frequency vs  $\nu/\omega$ .

the ratio  $\nu/\omega$ . At the same time, in deriving (28), use has been made of a kinetic equation suited for weak interaction of electrons with ions and themselves, when  $\nu \ll \omega_L$ . It means that in the case when  $\nu \gg \omega$  is implied, the fulfillment of the inequality  $\omega \ll \omega_L$  is also implied. Thus, the field frequency is assumed to be smaller than the electron plasma frequency. A field with such a frequency penetrates inside the plasma only to the skin-layer depth. In other words, here we can speak only about the current third HG in the skin layer. Keeping this in mind, let us assume that also in the case in which  $\nu \ll \omega$  takes place, the inequality  $\omega \ll \omega_L$  also occurs. As a result, we arrive at the conclusion that it is necessary to consider the current third HG in the skin-effect conditions. Further, we assume that

$$\omega \leqslant \omega_L \sqrt{J_1(\Omega)}$$
. (36)

When  $\nu \ll \omega$ , this last inequality is equivalent to  $\omega \ll \omega_L$ , while when  $\nu \gg \omega$  from (36), we have  $\nu \omega \ll \omega_L^2$ . The inequality (36) allows us to neglect the second term proportional to the squared-field frequency in the left-hand side of the equation for the field (35). Furthermore, considering that the plasma fills the half space x>0, from Eq. (35) we have approximately the following solution going to zero at  $x\to\infty$ :

$$E(x) = E(0)\exp(-i\kappa x), \tag{37}$$

$$\kappa = \frac{\omega_L}{c} \sqrt{J_1(\Omega)} \exp\left[\frac{i}{2} \Delta_1(\Omega) - i\frac{\pi}{4}\right]. \tag{38}$$

We now use the field distribution in the plasma given by Eqs. (37) and (38) to solve the problem of pump wave reflection and absorption.

Let us assume that on the plasma filling the half space x>0, normally to its surface impinges a linearly polarized electromagnetic wave of the form

$$\frac{1}{2}E_o \exp(-i\omega t + ikx) + \text{c.c.}.$$
 (39)

This wave penetrates inside the plasma to the skin-layer depth and is reflected by it. The reflected wave field is written as

$$\frac{1}{2}RE_o \exp(-i\omega t - ikx) + \text{c.c.}, \tag{40}$$

where R is the complex reflection coefficient. In the conditions we are considering, the magnetic field  $\vec{B}(x,t) = (0, B(x,t), 0)$  both in vacuum and inside the plasma, is connected to the electric one by

$$\frac{\partial}{\partial x}E(x,t) = \frac{1}{c}\frac{\partial}{\partial t}B(x,t). \tag{41}$$

From Eqs. (37)–(41) and the continuity requirements on the magnetic and electric fields on the plasma surface we have

$$E_o + RE_o = E(0), \tag{42}$$

$$-E_o + RE_o = -i\frac{c}{\omega}\kappa E(0). \tag{43}$$

From these relations, we find the complex reflection coefficient

$$R = \frac{\omega - ic\kappa}{\omega + ic\kappa} \simeq -1 + \frac{2\omega}{\omega_L \sqrt{J_1(\Omega)}} \exp\left[-\frac{i}{2}\Delta_1(\Omega) - i\frac{\pi}{4}\right]$$
(44)

and the relation connecting the field inside the plasma to the electric field of the impinging wave

$$E(0) = \frac{2E_o}{1 + i\kappa c\omega} \simeq \frac{2\omega}{\omega_L \sqrt{J_1(\Omega)}} E_o \exp\left[-\frac{i}{2}\Delta_1(\Omega) - i\frac{\pi}{4}\right]. \tag{45}$$

From Eq. (44), a relatively simple expression follows for the absorption coefficient due to electron-ion collisions:

$$A = 1 - |R|^2 \simeq \frac{4\omega}{\omega_L \sqrt{J_1(\Omega)}} \cos\left[\frac{1}{2}\Delta_1(\Omega) + \frac{\pi}{4}\right]. \tag{46}$$

Using the relations (32) and (33) in two limiting cases from Eq. (46) we have

$$A \simeq \frac{2\nu_{ei}}{\omega_L}, \quad \nu_{ei} \leqslant \omega_L, \tag{47}$$

$$A \simeq \sqrt{\frac{3\pi}{4}} \frac{\sqrt{\nu_{ei}\omega}}{\omega_L}, \quad \nu_{ei} \gg \omega_L, \tag{48}$$

where use has been made of the notation  $\nu_{ei} = \sqrt{2} \nu/3 \sqrt{\pi}$  familiar in the theory of high-frequency field absorption. For the general case, the dependence of the absorption coefficient A, given by formula (46), on the ratio  $\nu/\omega$  is reported in Fig. 3.

We conclude the description of the field inside the plasma by reporting the relations that connect the strength and the

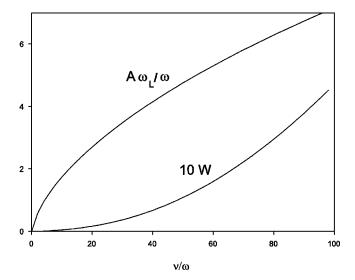


FIG. 3. Fundamental wave absorption coefficient A and third-harmonic generation efficiency W, Eq. (63), in relative units vs  $\nu/\omega$ .

phase shift of the generating field (1) and (34) to the electric field in vacuum. From Eqs. (34), (38), and (45), we have approximately

$$E(x) \simeq \frac{2\omega e^{i\delta}}{\omega_L \sqrt{J_1(\Omega)}} E_o \times \exp\left\{-x\frac{\omega_L}{c} \sqrt{J_1(\Omega)} \cos\left[\frac{1}{2}\Delta_1(\Omega) - \frac{\pi}{4}\right]\right\},$$
(49)

$$\delta \simeq -\frac{\pi}{4} - \frac{1}{2}\Delta_1(\Omega) - x\frac{\omega_L}{c}\sqrt{J_1(\Omega)}\sin\left[\frac{1}{2}\Delta_1(\Omega) - \frac{\pi}{4}\right]. \tag{50}$$

According to these relations, the field strength and the phase shift depend on the space coordinate. As mentioned above (Sec. II), we neglect this dependence in dealing with the electron kinetics in the skin layer. It is allowed, if the distance covered by thermal electrons in the field period is much smaller than the effective depth of the skin layer, which, according to Eq. (49), is  $c/\omega_L\sqrt{J_1(\Omega)}$ .

#### V. THIRD-HARMONIC EMISSION

Let us consider now the emission of the third harmonic by the plasma. The field inside the plasma at the frequency  $3\omega$  is written as

$$\frac{1}{2}\vec{E}_3(x)\exp(-3i\omega t) + \text{c.c.} \equiv \vec{E}_3\cos(3\omega t - \delta_3), \quad (51)$$

where  $\vec{E}_3(x) = [0, 0, E_3(x)]$  and  $\delta_3$  is the corresponding phase shift. In accordance with the relation (28), the field (51) yields the current at  $3\omega$  with the density

$$\delta \vec{j}_{3\omega} = \frac{e^2 n}{3\omega m} \vec{E}_3 J_1 \left(\frac{\Omega}{3}\right) \cos \left[3\omega t - \delta_3 - \Delta_1 \left(\frac{\Omega}{3}\right)\right]. \quad (52)$$

Taking into account the relation (52), similar to Eq. (35), to determine the field  $E_3(x)$  from the Maxwell equations we find

$$\frac{d^2}{dx}E_3(x) + \frac{9\omega^2}{c^2}E_3(x) - \kappa_3^2 E_3(x) = F \exp(-3\kappa x), \quad (53)$$

where F is given below by Eq. (55) and the notation

$$\kappa_3 = \frac{\omega_L}{c} \sqrt{J_1(\frac{\Omega}{3})} \exp\left[\frac{i}{2}\Delta_1(\frac{\Omega}{3}) - i\frac{\pi}{4}\right]$$
 (54)

is used. A substantial difference of this equation as compared to Eq. (35) is that its r.h.s. contains a field source at frequency  $3\omega$ , due to nonlinear dependence of the electron-ion collision frequency on the field. According to relations (15), (49), and (50), the current density value of the source F is proportional to the third power of the field strength and has the form

$$F = -i12\pi 10^{-4} J_3(\Omega) \left( \frac{e}{m\omega} \frac{\omega_L}{v_T} \frac{E_o}{c} \right)^2 \left[ \frac{2\omega}{\omega_L \sqrt{J_1(\Omega)}} \right]^3 E_o$$

$$\times \exp \left[ -i \frac{3\pi}{4} - i \frac{3}{2} \Delta_1(\Omega) + i \Delta_3(\Omega) \right]. \tag{55}$$

Furthermore, together with the condition (36), assumed in analyzing the field at  $\omega$ , we will consider that a similar condition takes place for the field at  $3\omega$  as well. Namely, we assume that

$$3\omega \ll \omega_L \sqrt{J_1\left(\frac{\Omega}{3}\right)}.$$
 (56)

In this case, the solution to the nonhomogeneous Eq. (53), going to zero inside the plasma, has the form

$$E_3(x) = E(0)\exp(-\kappa_3 x) + \frac{F}{\kappa_2^2 - 9\kappa^2} [\exp(-\kappa_3 x) - \exp(-3\kappa x)].$$
 (57)

Next, in accordance with Eq. (41), the electric field (57) univocally determines the magnetic field in the plasma

$$B_3(x) = \frac{i}{3k} \frac{d}{dx} E_3(x). {(58)}$$

At the plasma boundary x=0, the electromagnetic fields (57) and (58) transform into the field of the wave irradiated by the plasma at  $3\omega$ . According to Maxwell equations, the field of the irradiated wave has the form

$$\frac{1}{2}\vec{E}_r \exp(-3i\omega t - 3ikx) + \text{c.c.}, \tag{59}$$

where  $\vec{E}_r = (0,0,E_r)$ . Besides, according to Eq. (41)  $\vec{B}_r = (0,E_r,0)$ . Furthermore, using the field continuity conditions at x=0, from Eqs. (57)–(59), we find the strength of electric and magnetic fields of the irradiated wave:

$$B_r = E_r = -i \frac{F}{(3k + i\kappa_3)(\kappa_3 + 3\kappa)}.$$
 (60)

From here we find the energy flux density irradiated by the plasma at the frequency  $3\omega$ 

$$\vec{S} = \frac{c}{4\pi T_r} \int_0^{T_r} dt [\vec{E}(x,t)\vec{B}(x,t)] = -\vec{i}I_r,$$
 (61)

where  $I_r = c|E_r|^2/8\pi$  and  $T_r = 2\pi/3\omega$  the corresponding period. As the energy flux density of the impinging on the

plasma wave is  $I_o = cE_o^2/8\pi$ , using the inequality (56) from Eqs. (55), (60), and (61) for the third HG efficiency in the skin layer of a dense plasma, we have the following result:

$$\eta = \frac{I_r}{I_o} = \left(\frac{eE_o}{m\omega v_T}\right)^4 \frac{\omega^6}{\omega_I^6} W\left(\frac{\nu}{\omega}\right),\tag{62}$$

where the function  $W(\Omega)$  has the form

$$W(\Omega) = \left(\frac{6\pi}{625}\right)^{2} \frac{J_{3}^{2}(\Omega)}{J_{1}^{3}(\Omega)J_{1}(\Omega/3)} \left\{ \frac{1}{J_{1}(\Omega/3) + 9J_{1}(\Omega) + 6\sqrt{J_{1}(\Omega)J_{1}(\Omega/3)}\cos\left[\frac{\Delta_{1}(\Omega) - \Delta_{1}(\Omega/3)}{2}\right]} \right\}.$$
(63)

In the limit of small and large values of the ratio  $\nu/\omega$  from Eq. (63), we have the simple asymptotic dependencies

$$W(\Omega) = \frac{5}{\pi} 10^{-3} \Omega^2, \quad \Omega \ll 1, \tag{64}$$

$$W(\Omega) \simeq \frac{\sqrt{2\pi}}{2+\sqrt{3}} \left(\frac{105\pi}{2048}\right)^2 \Omega, \quad \Omega \gg 10.$$
 (65)

In Fig. 3 we report the curve of the function  $W(\Omega)$ . From Fig. 3 it is seen that increasing  $\Omega = \nu/\omega$  one has a monotonical increase of the third HG generation efficiency.

## VI. CONCLUSIONS

Let us recall the basic points of our work. First of all, we note that, in investigations on harmonic generation due to electron-ion collisions, an analytical treatment is given in which the effective electron-ion collision frequency is comparable to, or greater than, the radiation frequency. These conditions have great relevance for present-day experiments concerning ultrashort laser pulses interacting with dense plasmas generated on the solid-state target surface. Thus, the domain of validity of the theory is extended to the conditions required by present-day and future experiments. Second, an essential merit of third-harmonic generation treatment given here is that we establish the explicit dependencies of the radiation flux density at  $3\omega$  measured experimentally on the plasma and fundamental wave parameters. It is at variance with previous treatments (see, for instance [1-5]), where only the current density or the harmonics field inside the plasma were determined. The theory of the third-harmonic generation presented here is easily extended to higher odd harmonics. Here we are confined to the most important third harmonic, because due to the strong weakening of the fundamental wave electric field in the skin layer, the generation of higher-order harmonics is significantly smaller. Harmonics generation is an important nonlinear effect, interesting in its own right. At the same time, the prediction of the nonlinear theory of harmonics generation may be used for application purposes as well. In particular, the simultaneous measurement of the plasma radiation at  $3\omega$  and of the reflected signal at the fundamental frequency makes it possible to find the plasma density and temperature on the solid-state surface, using the analytical dependencies established above. We recall that the idea of using high-order harmonics for plasma diagnostics is not new and has been verified in Ref. [12].

Let us now discuss in some detail the domain of validity of our treatment. It is valid in the conditions of normal and high-frequency skin effects, which, as it is known [7–11], takes place in broad ranges of radiation frequencies, and plasma temperatures and densities. We refrained from extending the treatment to the anomalous skin-effect conditions, which are not easy to realize, because in such a case, the influence of electron-ion collisions is severely reduced, which is the physical mechanism responsible for harmonics generation.

As far as the radiation intensity is concerned, the domain of validity of our theory extends from vanishingly small flux densities (having in mind radiation interaction with prepared plasma) up to very high values, at which the electron-directed motion velocity in the skin layer, where the fundamental wave electric field is strongly reduced, becomes comparable to the electron thermal velocity. For instance, in the visible frequency range, taking into account the effective electron heating in the skin layer of a solid-state plasma, the above condition of velocity comparability allows us to use the present theory up to flux densities of the order  $10^{18}$ – $10^{17}$  W/cm², when the anomalous skin-effect domain is approached.

Finally, concerning the limitations stemming from the laser pulse duration, they are very weak. On the side of very long pulse durations, a limitation comes from the possible plasma hydrodynamic flying away, which yields the plasma density decrease and the need to describe harmonics generation in a rarefied plasma with a density smaller than the critical one. Usually, the plasma flying away time is larger than picosecond order times. We note that the basic pulse may be very short. As a matter of fact, the characteristic time of high-order harmonics generation is smaller than the period of the fundamental wave. It means that the dependencies established above for the third-harmonic generation hold true if the field intensity of the pump wave and the plasma state change weakly in a field period. In the visible frequency range, such a time is of the femtosecond order.

As a consequence of plasma heating and flying away, its temperature and density may change, with the ensuing consequence that the harmonic generation efficiency, too, may change. In the physical conditions that are realized more frequently, the characteristic times of variation of plasma density and temperature are larger than the field period of the fundamental wave. So in discussing the harmonic generation efficiency, it is enough to substitute, in the above derived formulas, the results concerning plasma heating and flying away, available in the literature.

The regularities established above concerning the third HG in the skin layer of a dense hot plasma allow us to understand at which plasma and laser parameter the generation of radiation at  $3\omega$  is most effective. According to relations (62) and (63) and Fig. 3, the third HG efficiency is higher the greater the electron-ion collision frequency. At the same time, the expression for  $\eta$  (62) has been obtained in the framework of the ideal plasma theory, when  $\nu < \omega_L$ . The dependence of  $\eta$  on the radiation frequency  $\omega$  is such that for  $\omega < \nu$  the function  $\nu$  increases proportionally to  $\omega$  and does not depend upon  $\omega$  for  $\omega > \nu$ . The presence of such scalings on  $\nu$  and  $\omega$  allows us to formulate the hypothesis, that the third HG efficiency in the skin layer takes its largest values when all the characteristic frequencies are the same order of magnitude  $\omega \leq \nu \leq \omega_L$ , though their numerical values may differ by several times.

Another parameter, on which the function  $\eta$  depends in an essential way, is the ratio  $eE_o/m\omega v_T$ , characterizing the strength of the pump wave electric field. According to (62),  $\eta$  increases proportionally to  $(eE_o/m\omega v_T)^4$ . Such a dependence is derived in the assumption that in the skin layer, the electron-directed motion velocity in the field E is much smaller than the electron thermal velocity. For  $\omega > \nu$ , this assumption amounts to  $v_E/v_T \ll 1$ , while for  $v > \omega$  to  $|eE/mv| \ll v_T$ . The limitation on the field strength value for  $v > \omega$  makes it possible to neglect the runaway electrons, at least before the electron temperature doubling. It is just the electrons run away effect that may be responsible of the relative third HG efficiency weakening in the most favorable conditions when  $\omega \ll v \ll \omega_L$  and  $v_E$  is comparable to  $v_T$ .

In choosing the best conditions to generate the third harmonic, it is wise to take into account two other important processes. One of them is the electron heating due to inverse bremsstrahlung in the skin layer. We should remark that the theory given above is applicable only when the time of electron temperature doubling is much greater than the period of the fundamental wave. As a result of the electron temperature increase, the generation efficiency  $\eta$  (62) decreases:  $\eta$ 

 $\sim T^{-7/2}$  for  $\nu > \omega$ , and  $\eta \sim T^{-5}$  for  $\nu < \omega$ , where  $\nu \sim T^{-3/2}$ . The other one is the plasma matter flying away, yielding its density decrease and the destruction of the sharp plasma-vacuum boundary. The relations (62)–(65) indicate that for  $\omega < \omega_L$ , the generation efficiency  $\eta$  must increase with the density decrease. However, it must be noted that this conclusion is derived considering a plasma with a sharp boundary, and evidently a different analysis is required if the plasma boundary loses its sharpness because of the plasma flying away. We note also that the plasma matter flying away is irrelevant if the third HG is studied for times smaller than the skin-layer depth  $c/\omega_L\sqrt{J_1(\Omega)}$  divided by the acoustic velocity  $v_s$  in the plasma.

Let us give an example illustrating the possibility of third HG. For an estimate we assume that a laser radiation with frequency  $\omega \simeq 2 \times 10^{15} \text{ s}^{-1}$  and flux density  $I_o = 4$  $\times 10^{16} \, \text{W/cm}^2$  interacts with a fully ionized beryllium plasma with electron density  $n=5\times10^{23}$  cm<sup>-3</sup>, ionization multiplicity Z=4, and temperature T=500 eV. In such conditions  $v_T \approx 9.4 \times 10^8 \text{ cm/s}, \ \nu \approx 5.4 \times 10^{15} \text{ s}^{-1}, \ \omega_L \approx 4$  $\times 10^{16} \, s^{-1}$ ,  $\Omega = \nu/\omega \simeq 2.7$ ,  $2\omega/\omega_L \sqrt{J_1(\Omega)} \simeq 0.11$ , while for the ratio  $v_E/v_T$  in the skin layer of the Be plasma, we have  $v_F/v_T \approx 0.5$ . In these estimates we do not take into account the small corrections to the distribution function related to the Langdon effect. The resulting third HG efficiency is  $\eta \simeq 3 \times 10^{-6} \text{ W}(\Omega) \simeq 2 \times 10^{-9}$ , which corresponds to the radiation flux density irradiated by the plasma at  $3\omega$  equal to  $I_r \approx 10^8 \text{ W/cm}^2$ . In the same conditions, the time of the electron temperature doubling  $\simeq 3v_T^2 [2\omega v_F^2 J_1(\Omega)\cos\Delta_1(\Omega)]^{-1} \simeq 20$  fs, while the time of plasma boundary loss of sharpness is  $\tau_{exp} \simeq c/\omega_L v_s \sqrt{J_1(\Omega)}$  $\approx$  50 fs. In other words, for about 20 fs the flux density of the radiation emitted by the plasma at  $3\omega$  is rather high, while in the subsequent time instants it rapidly decreases basically due to electron heating. The reported estimates show that it is possible to carry out a relatively simple experiment aimed at observing the third HG regularities established above and those of higher odd harmonics as well. In connection to this possibility we remark that the radiation at  $3\omega$  is concentrated in the opposite direction to that of the impinging fundamental wave, and has a narrow spectral width, which should allow us to single it out from the background plasma thermal radiation. The latter may be intense enough, but is distributed over all the frequencies and angles of the wave vector.

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