Second-harmonic generation by intense lasers in inhomogeneous plasmas

H. S. Brandi, P. A. Maia Neto, and E. S. Guerra
Instituto de Física, Universidade Federal do Rio de Janeiro, Caixa Postal 68.528, 21945-970, Rio de Janeiro, Brazil
(Received 15 December 1995)

We study the second-harmonic generation by laser interaction with a cold inhomogeneous underdense plasma, using a perturbative approach to solve the coupled set of Maxwell fluid, and momentum equations. The laser field is assumed to be Gaussian with diffractive effects included. The solution of the second-order inhomogeneous wave equation is obtained from the Green's function formalism in the paraxial approximation. We show that the total power generated is proportional to the square of the laser intensity and independent of the plasma density in the low-density limit. Depending on the laser beam waist and on the plasma density profile, the second-harmonic generation can be highly efficient. [S1063-651X(96)02407-5]

PACS number(s): 52.40.Db, 42.25.−p, 42.65.Jx, 52.35.Mw

Until the end of the past decade, the intensities of the available focused lasers were limited to the range of $10^{15}-10^{16}$ W/cm^2. The new laser technology based on chirped pulse amplification in solid state amplifiers followed by temporal compression at the picosecond or subpicosecond duration [1] made possible the production of compact, intense, terawatt lasers. After focusing, they may reach intensities above 10^{18} W/cm^2, which represents laser field strengths larger than one atomic field unit. When the laser pulse reaches intensities of the order of 10^{15} W/cm^2, all atoms irradiated by the laser are ionized, a plasma is formed with densities in the range $10^{10}-10^{20}$ cm^{-3}, the laser propagates in these plasmas, and the interaction is then dominated by electron-photon coupling. For high-intensity short pulses, the plasma can be considered as a mixture of a cold electron fluid and a fixed-ion background ensuring the electroneutrality of the plasma. The motion of the electrons in the presence of such fields is weakly relativistic, and connected to this behavior, various effects such as relativistic self-focusing, wake field generation, production of strong magnetic fields, electron-positron pair production, harmonic generation, have been theoretically predicted. This last effect is treated in the present work, where we consider the second-harmonic generation by interaction of an intense laser with a preformed cold underdense inhomogenous plasma, with plasma frequency $\omega_p = ck_p = (4\pi e^2 N_c/m_0)^{1/2}$, where m_0 is the electron rest mass and N_c the initial plasma density at the center of the laser beam, as discussed below. As previously shown [2,3], no second harmonic is generated for an initially uniform plasma density; generation of even harmonics requires that the laser must interact with plasmas having initially density gradients, which occurs when a laser pulse ionizes neutral gas. When an intense laser pulse is focused in a gas, it produces a plasma through multiphoton or tunnel ionization. The plasma density builds up when the laser intensity exceeds the gas ionization threshold. Since the multiphoton or tunnel ionization has a strong nonlinear dependence on the laser intensity, a Gaussian beam radial intensity profile produces a plasma with strong radial density gradients, the maximum being on the beam axis. This effect has been observed in connection with defocusing of laser beams by plasmas formed from noble gases [4]. Due to the complexity of the harmonic-generation problem, most of the theoretical approaches consider the one-dimensional (1D) approximation (where all physical quantities are functions of a single variable $\zeta = z - vt$). In this case the existence of constants of motion simplifies the problem for the harmonic generation by intense laser-plasma interaction. Of course, a 3D scheme is required to treat the situation where the plasma is radially inhomogeneous. Recently, Brandi *et al.* [3] have studied the third-harmonic generation by an homogeneous plasma, using the Green's function approach to solve the 3D Helmholtz equation. We closely follow this work adopting a perturbative scheme, i.e., all the physical quantities are expanded as powers of the incident laser field. This corresponds to a situation where relativistic corrections are small. Furthermore, as in Refs. [3,5], we introduce the expansion parameters $\alpha = \lambda_0/2\pi r_0 \ll 1$, where λ_0 and r_0 are the laser beam wavelength and waist, and $\delta = \omega_p/\omega_0 \ll 1$, with $\omega_0 = 2\pi c/\lambda_0$ being the laser frequency. The first assumption corresponds to the paraxial approximation and the second to the requirement that the plasma is very underdense, $N_c \ll 10^{20}$ cm^{-3}. We have also defined the parameter $\alpha_S = \lambda_0/2\pi L_S \ll 1$, where L_S is the typical electron density gradient length. The paraxial approximation is valid near the propagation axis, which justifies the modeling of the initial electron plasma density by taking the parabolic approximation

$$N_0 = N_c(1 + \sigma\rho^2/L_S^2) = N_c f(\rho),$$

where $\sigma = -1(+1)$ denotes the electronic plasma density as a maximum (minimum) on the laser axis. Here we have supposed that the laser beam propagating along the z direction has maximum intensity at $\rho = \sqrt{x^2+y^2} = 0$.

The electric field $\mathbf{E}$ and the magnetic field $\mathbf{B}$ are written in terms of the scalar potential Φ and the vector potential $\mathbf{A}$, i.e., $\mathbf{E} = -\nabla\Phi - 1/c\,\partial\mathbf{A}/\partial t$, and $\mathbf{B} = \nabla\times\mathbf{A}$. The relativistic cold hydrodynamic equations for the electron fluid consider the electron density N and the electron momentum $\mathbf{P} = m_0\gamma\mathbf{V}$ [$\mathbf{V}$ is the fluid velocity and $\gamma = (1+P^2)^{1/2}$ is the Lorentz factor]. These equations are coupled to the wave equation and Poisson's equation through the charge density $-e(N-N_0)$ and the current density $\mathbf{j} = -eN\mathbf{V}$. It is convenient to normalize the quantities appearing in these equations as

$$\mathbf{a}=e\mathbf{A}/m_0c^2, \quad \phi=e\Phi/m_0c^2,$$

$$\mathbf{p}=\mathbf{P}/m_0c=\gamma\mathbf{V}/c, \quad n=N/N_c.$$

We then have the set of coupled nonlinear equations

$$\frac{1}{c}\frac{\partial}{\partial t}(\mathbf{p}-\mathbf{a})=\nabla(\phi-\gamma)+\frac{\mathbf{p}}{\gamma}\times\nabla\times(\mathbf{p}-\mathbf{a}), \tag{1}$$

$$\frac{1}{c}\frac{\partial n}{\partial t}+\nabla\cdot\left(\frac{n}{\gamma}\mathbf{p}\right)=0, \tag{2}$$

$$\nabla^2\phi=k_p^2[n-f(\rho)], \tag{3}$$

$$\left(\nabla^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{a}_\perp=k_p^2\left(\frac{n}{\gamma}\mathbf{p}\right)+\frac{1}{c}\frac{\partial\nabla\phi}{\partial t}=k_p^2\left(\frac{n}{\gamma}\mathbf{p}\right)_\perp, \tag{4}$$

where the Coulomb gauge $\nabla\cdot\mathbf{a}=0$ has been assumed and $\mathbf{F}_\perp$ and $\mathbf{F}_L$ are the transverse and longitudinal components of a vector field $\mathbf{F}$.

To study the harmonic generation, we develop a perturbative expansion of all plasma quantities in terms of powers of the known laser vector potential amplitude a_0. This amplitude is related to the laser intensity I_0 by

$$a_0\simeq 4.25\times 10^{-10}[\lambda_0(\mu\text{m})][I_0(\text{W/cm}^2)]^{1/2}. \tag{5}$$

For $\lambda_0\simeq 1$ μm, the perturbative treatment breaks for laser intensities of the order of $I_0\simeq 10^{18}$ W/cm^2, which imply $a_0\simeq 1$.

If the plasma is stationary and neutral, in zero order of the laser field, from Eqs. (1)–(4), the plasma quantities are $p_\mathbf{0}=0$, $n_0=f(\rho)$, $\phi_0=0$, and $\gamma_0=1$. To first order, the equations are

$$\frac{1}{c}\frac{\partial}{\partial t}(\mathbf{p}_1-\mathbf{a}_0)=\nabla\phi_1, \tag{6}$$

$$\frac{1}{c}\frac{\partial n_1}{\partial t}+\nabla\cdot[f(\rho)\mathbf{p}_1]=0, \tag{7}$$

$$\nabla^2\phi_1=k_p^2 n_1, \tag{8}$$

$$\left(\nabla^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{a}_1=k_p^2 f(\rho)\mathbf{p}_1+\frac{1}{c}\frac{\partial}{\partial t}\nabla\phi_1. \tag{9}$$

In order to calculate the second-harmonic field, it is necessary to obtain only the first-order correction in the electronic density. From Eqs. (6)–(8), we find up to second order in δ

$$n_1=\frac{i}{k_0}\nabla f\cdot\mathbf{a}_0, \tag{10}$$

where $k_0=2\pi/\lambda_0$. Using Eq. (10) and the second-order expansion of Eq. (4), we obtain to lowest order in δ

$$[\boldsymbol{\nabla}^2+k_2(\rho)^2]\mathbf{a}_2=k_p^2 n_1(\mathbf{p}_{1\perp}+\mathbf{p}_{1L})+k_p^2 n_0\mathbf{p}_{2L}+\frac{1}{c}\frac{\partial\nabla\phi_2}{\partial t}, \tag{11}$$

where we have defined $k_2(\rho)=\sqrt{4\omega_0^2/c^2-k_p^2 f(\rho)}$. Following the same procedure as in Ref. [3], one obtains $k_p^2(n_1\mathbf{p}_{1L}+n_0\mathbf{p}_{2L})+1/c\,\partial\nabla\phi_2/\partial t=O(\alpha_s\delta^2)$ and therefore we have

$$[\nabla^2+k_2(\rho)^2]\mathbf{a}_2=i\delta^2 k_0(\nabla f\cdot\mathbf{a}_0)\mathbf{a}_0. \tag{12}$$

The right-hand side of Eq. (12) corresponds to the source for the second-harmonic field. It is a transverse field (to lowest order in δ) associated with the gradient of the electronic density.

To solve Eq. (12) we assume that the pump laser is Gaussian with diffractive effects included. In this case the vector potential is given by

$$\mathbf{a}_0=\mathbf{e}_x a_s(\rho,z)\exp(i\Psi_0), \tag{13}$$

where

$$a_s(\rho,z)=-\frac{iz_R}{z-iz_R}\tilde{a}_0\exp\left[ik_0\frac{\rho^2}{2(z-iz_R)}\right], \tag{14}$$

$\Psi_0=k_0z-\omega_0t$, and $z_R=k_0r_0^2/2$ is the Rayleigh length. The approximate solution of Eq. (12) may be obtained neglecting to order δ^2 the spatial dependence in $k_2(\rho)^2$, which is reinforced by the assumption of the paraxial approximation justifying the parabolic modelling of the electronic density. From the Green's function solution of the Helmholtz equation

$$(\nabla^2+k_2^2)g(\mathbf{r}-\mathbf{r}')=-4\pi\delta(\mathbf{r}-\mathbf{r}'), \tag{15}$$

with

$$g(\mathbf{r}-\mathbf{r}')=\frac{\exp ik_2|\mathbf{r}-\mathbf{r}'|}{|\mathbf{r}-\mathbf{r}'|} \tag{16}$$

and $k_2=\sqrt{4\omega_0^2/c^2-k_p^2 f(\rho)}\simeq 2k_0[1+3/8\,\delta^2 f(\rho)]$, we integrate Eq. (12) to find

$$\mathbf{a}_2=-\frac{i}{2\pi}\frac{\sigma\delta^2k_0}{L_S^2}\tilde{a}_0^2 I\mathbf{e}_x, \tag{17}$$

with

$$I=\int_{-Z_0}^{Z_M}dz'\int_{-\infty}^{\infty}x'dx'\int_{-\infty}^{\infty}dy'\left(\frac{iz_R}{z-iz_R}\right)^2 \times\exp\left[ik_0\frac{x'^2+y'^2}{(z'-iz_R)}\right]\frac{\exp ik_2|\mathbf{r}-\mathbf{r}'|}{|\mathbf{r}-\mathbf{r}'|}. \tag{18}$$

The primed quantities in Eq. (18) are to be integrated over the corresponding integration variable and $\mathbf{r}$ is the observer's position. The plasma is modeled by a finite slab extending from $-Z_0$ to Z_M.

In the paraxial approximation the radiation zone is defined by the relation $|z-z'|\gg|x-x'|,|y-y'|$ and therefore one has

$$I=\exp ik_2z\int_{-Z_0}^{Z_M}\frac{dz'}{(z-z')}\left(\frac{iz_R}{z'-iz_R}\right)^2\times\exp i\Delta kz'\int_{-\infty}^{\infty}x'dx'\int_{-\infty}^{\infty}dy'\exp\left[ik_0\frac{x'^2+y'^2}{(z'-iz_R)}\right]\times\exp\left[i\frac{k_2}{2}\frac{(x-x')^2+(y-y')^2}{(z-z')}\right],\qquad(19)$$

where we have defined $\Delta k=2k_0-k_2\simeq-3/4\,k_0\delta^2$. We solve Eq. (19) in the limit $\Delta kz_R\gg1$, which is equivalent to the condition $k_pr_0\gg1$. In the high-intensity chirped pulse Nd:YAG laser experiments (where YAG denotes yttrium aluminum garnet) the spot size r_0 is typically of the order of 10 μm [7]. Thus our approximation is valid for electronic plasma densities larger than 5×10^{17} cm^{-3}. We obtain

$$\mathbf{a}_2=\mathcal{A}(L)\frac{\sigma\tilde{a}_0^2}{k_0L_S^2}x\frac{\exp(ik_2z)}{(1+iz/z_R)^2}\exp\left[ik_0\frac{x^2+y^2}{z-iz_R}\right]\mathbf{e}_x,\qquad(20)$$

where the factor $\mathcal{A}(L)$ depends on the length of the plasma $L=(Z_M+Z_0)$. For $L\ll z_R$, the diffraction of the laser beam is negligible over the extension of the plasma and then we find

$$\mathcal{A}(L)=-\frac{4}{3}i\exp[i\Delta k(Z_M-Z_0)/2]\sin(\Delta kL/2).\qquad(21)$$

On the other hand, for a very long plasma $L\gg z_R$, the amplitude of the second-harmonic field approaches the constant value $\mathcal{A}(L)=2/3$.

The second-harmonic field found in Eq. (20) corresponds to the transverse Hermite-Gaussian mode $\mathrm{TEM}_{1,0}$. According to Eq. (20), the second-harmonic field is an odd function of x. For $\sigma=1(-1)$, and at a point in the half-space corresponding to $x>0$, when $\mathbf{a}_0$ is along the positive-x direction, it points towards the higher (lower) plasma density region of space, whereas the negative-x direction corresponds to lower (higher) densities. Such asymmetry is at the origin of the second-harmonic generation. In the half-space $x<0$, however, the asymmetry occurs in the opposite sense, thus explaning the change of sign of the second-harmonic field. This results in the generation of a beam characterized by a profile corresponding to a zero intensity on the beam axis.

The power radiated by the second harmonic is calculated from the flux of the Poynting vector through the surface defined by a plane perpendicular to the z axis. Since

$$\mathbf{S}=\mathrm{Re}\left[\frac{c}{8\pi}\mathbf{E}\times\mathbf{H}^*\right]\simeq\frac{ck_2^2}{8\pi}|\tilde{a}_2(L)|^2\mathbf{e}_z,\qquad(22)$$

we find from Eq. (20) the following expression for the power in the second harmonic P_2:

$$P_2=\frac{1}{64}c|\mathcal{A}|^2\left(\frac{\alpha_S}{\alpha}\right)^4\tilde{a}_0^4.\qquad(23)$$

The power conversion rate is defined as the ratio of the power in the second harmonic P_2 to the power in the pump laser field $P_1=\omega_0k_0r_0^2/16\tilde{a}_0^2$. Its maximum value occurs for odd multiples of $L=\pi/\Delta k\ll z_R$:

$$\max\left(\frac{P_2}{P_1}\right)=\frac{4}{9}\frac{\alpha_S^4}{\alpha^2}\tilde{a}_0^2.\qquad(24)$$

On the other hand, for $L\gg z_R$ we have

$$\frac{P_2}{P_1}=\frac{1}{9}\frac{\alpha_S^4}{\alpha^2}\tilde{a}_0^2.\qquad(25)$$

The power conversion rate varies as the intensity of the laser power I_0. Moreover, it is independent of the electronic plasma density, which may be explained by the exact compensation between the increase of the source for the second-harmonic field with δ^2 and the phase mismatch represented by Δk, which also varies as δ^2. A qualitative discussion by Esarey *et al.* [2] predicts that volume effects due to the ''halo'' region (the portion of the interaction region where the gas is not fully ionized) of the laser pulse should change the I_0^2 dependence of the second-harmonic total power generated to $I_0^{3/2}$, as is the case of atomic harmonic generation [6]. Since the emitted power is independent of the electronic plasma density, no modifications in this intensity dependence should occur due to the ''halo'' region effects. We should notice that although $\alpha_S,\alpha\ll1$, the experimental conditions for a chirped pulse laser may be such that $\alpha_S/\alpha>1$, which may lead to a highly efficient mechanism of second-harmonic generation.

Summarizing, the emission of second-harmonic radiation by a cold, inhomogeneous, underdense plasma has been studied using a Green's function formulation to calculate the total power generated by the second-harmonic radiation. All the physical quantities have been expanded in terms of the parameters $\alpha=\lambda_0/2\pi r_0\ll1$, $\delta=\omega_p/\omega_0\ll1$, and $\alpha_S=\lambda_0/2\pi L_S\ll1$. Furthermore, our calculations are restricted to the condition $k_pr_0\gg1$, which implies $\delta\gg\alpha$. Typically, those approximations correspond to the range of electronic plasma densities from 5×10^{17} to 10^{20} cm^{-3}. We have considered a perturbative expansion in terms of powers of the pump laser vector potential. For a chirped pulse laser with $\lambda_0\approx1$ μm, our perturbative approach is valid for laser intensities smaller than 10^{18} W/cm^2. Nonperturbative results are at the moment restricted to 1D models [2], which are clearly inadequate to treat the inhomogeneous case discussed in this paper. To study the harmonic generation by very intense fields such as those expected in the relativistic self-focusing regime [5], new 3D theoretical schemes must be developed.

The results show that the emision occurs in a transverse Hermite-Gaussian mode, that the total power is proportional to I_0^2, and that the power conversion rate is independent of the electronic density within the range considered in this paper. Under certain conditions on the preformed plasmas, the interaction of intense lasers with inhomogeneous plasmas may be an efficient mechanism of second-harmonic generation.

The authors acknowledge the partial support from the Brazilian Agency Conselho Nacional de Desenvolvimento Científico e Tecnológico.

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