Nonlinear Analysis of Relativistic Harmonic Generation by Intense Lasers in Plasmas

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Abstract— A linearly polarized, ultra-intense laser field induces transverse plasma currents which are highly relativistic and nonlinear, resulting in the generation of coherent harmonic radiation in the forward direction (i.e., co-propagating with the incident laser field). A nonlinear cold fluid model, valid for ultrahigh intensities, is formulated and used to analyze relativistic harmonic generation. The plasma density response is included self-consistently and is shown to significantly reduce the current driving the harmonic radiation. Phase detuning severely limits the growth of the harmonic radiation. The effects of diffraction are considered in the mildly relativistic limit. No third harmonic signal will emerge from a uniform plasma of near infinite extent. A finite third harmonic signal requires the use of a semi-infinite or finite slab plasma. For an initially uniform plasma, no second harmonic radiation is generated. Generation of even harmonics requires transverse gradients in the initial plasma density profile.

I. INTRODUCTION

THE interaction of ultra-intense laser pulses in plasmas is rich in a variety of phenomena [1]-[22]. These phenomena have gained particular relevance due to the development of compact lasers which produce ultrashort pulses (≤ 1 ps) at ultrahigh powers (≥ 1 TW) and intensities ($\geq 10^{18}$ W/cm^2) [2], [23]. For example, the production of harmonic radiation may occur by several mechanisms. At modest intensities, lasers interacting with neutral gases have been observed to produce coherent harmonic radiation at well past the 53st harmonic due to atomic effects [13]. At ultrahigh intensities, a gas is readily ionized and the effects of the free plasma electrons become exceedingly important. The ionization process itself results in electron currents which can produce harmonic radiation [14], [15]. In a fully ionized plasma, harmonics can be produced by i) relativistic harmonic generation in the forward direction (the propagation direction of the incident laser) [1], [8], [17]-[20], ii) stimulated backscattered harmonic generation [1], [21], and iii) incoherently, by nonlinear Thomson scattering [1], [22]. Recent experiments on these processes have also been performed [2], [15]-[18]. This paper discusses the details of process i), relativistic harmonic generation in fully ionized plasmas. The growth, dephasing, and saturation of the harmonics will be analyzed.
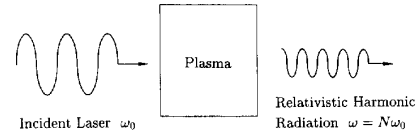
Fig. 1. Schematic of an intense laser interacting with a plasma to produce coherent relativistic harmonic radiation in the forward direction.

An intense laser field interacting with a plasma induces transverse currents associated with the quiver motion of the electrons. For ultrahigh intensities and linear polarizations, the induced plasma current becomes highly relativistic and nonlinear, resulting in the generation of coherent harmonic radiation in the forward direction (see Fig. 1). The transverse quiver momentum, $\mathbf{p}_q$, of an electron in a 1-D laser field is given by $\mathbf{p}_q = m_0 c \mathbf{a}_0$, where $\mathbf{a}_0 = e\mathbf{A}_0/m_0 c^2$ is the normalized vector potential of the incident laser field. The quiver velocity, $\mathbf{v}_q$, is given by $\mathbf{v}_q = c\mathbf{a}_0/\gamma_\perp$, where $\gamma_\perp \simeq (1 + a_0^2)^{1/2}$ is the relativistic factor associated with the transverse electron motion. Consider a linearly polarized incident laser field of the form $\mathbf{a}_0(z,t) = \hat{a}_0 \cos\theta_0 \mathbf{e}_x$, where $\theta_0 = k_0 z - \omega_0 t$ and k_0 and ω_0 are the wavenumber and frequency of the incident laser field, respectively. In the mildly relativistic limit, $\hat{a}_0^2 \ll 1$, the quiver velocity is sinusoidal. At ultrahigh intensities, $\hat{a}_0^2 \geq 1$, the quiver velocity contains a spectrum of harmonic components. This nonlinear electron quiver motion leads to the generation of relativistic harmonic radiation [1], [8], [17]-[20]. The laser strength parameter, $\hat{a}_0$, is related to the intensity, I_0, of the incident laser field by

$$\hat{a}_0 \simeq 8.5 \times 10^{-10} \lambda_0[\mu m] I_0^{1/2} [\text{W/cm}^2] \qquad (1)$$

and to the laser power, P_0, by $P_0[\text{GW}] \simeq 21.5(\hat{a}_0 r_0/\lambda_0)^2$, where r_0 is the spot size of the Gaussian transverse profile and $\lambda_0 = 2\pi/k_0$ is the incident laser wavelength. For $\lambda_0 \simeq 1\ \mu$m, ultrahigh intensities $I_0 \gtrsim 10^{18}$ W/cm^2 imply $\hat{a}_0^2 \gtrsim 1$ and, hence, highly nonlinear and relativistic electron motion. Such intensities are currently available from compact laser systems based on the method of chirped-pulse amplification [2], [23].

Relativistic harmonic generation was first described and analyzed by Sprangle, *et al.* [8]. In [8], the independent variables $\zeta = z - ct$ and $\tau = t$ were used along with a quasistatic plasma response. Expressions were derived for the growth of the harmonic radiation in the linear regime in which the harmonic field amplitude is proportional to the laser–plasma interaction distance, L. These expressions are qualitatively valid for interaction distances less than the phase

detuning length, $c\tau_d$. Use of the variables ζ, τ along with the quasistatic approximation is not adequate to accurately describe saturation of the relativistic harmonics by phase detuning. More recently, by using the variables $\xi = z - v_{ph}t$ and $\tau = t$, where v_{ph} is the phase velocity of the incident laser field, along with the quasistatic plasma response, the authors accurately analyzed the saturation of the relativistic harmonics by phase detuning [1]. Independent analyses of saturation of the third harmonic by phase detuning have also been recently performed [19], [20].

This paper is organized as follows. The remainder of the introduction describes the basic physics of third harmonic generation using a simplified 1-D model in the mildly relativistic limit, $\hat{a}_0^2 \ll 1$. The importance of collective, space-charge, and detuning effects are discussed. It is shown that the self-consistent plasma density response significantly reduces the source current driving the harmonic radiation. Phase detuning places a severe limit on the growth of the harmonic radiation. In Section II, a 1-D nonlinear model valid for ultrahigh intensities, $\hat{a}_0^2 \geq 1$, is formulated and used to study the generation of coherent radiation at odd harmonics. A general expression for the nonlinear index of refraction and the dispersion relation for a laser field in the limit $\hat{a}_0^2 \geq 1$ are presented. The collective plasma response is included self-consistently and the saturation amplitude of the harmonics by phase detuning is calculated. This is done in the long pulse limit, $c\tau_L \gg \lambda_p$, where τ_L is the incident laser pulse duration and $\lambda_p = 2\pi c/\omega_p$, where ω_p is the electron plasma frequency. For a $\tau_L = 1$ psec laser pulse, $c\tau_L \gg \lambda_p$ implies $n_0 \gg 10^{16}$ cm^{-3}, where n_0 is the ambient plasma density. In Section III, the effects of diffractive spreading of the radiation fields are determined using a 3-D model in the mildly relativistic limit, $\hat{a}_0^2 \ll 1$. Third harmonic generation from semi-infinite and finite slab plasmas are analyzed. The effects of transverse gradients in the initial plasma density are discussed, which lead to the generation of radiation at even harmonics. A conclusion is presented in Section IV.

A. Quiver Model, Collective Effects and Detuning

The process by which relativistic effects produce coherent harmonic radiation may be understood by considering a simplified "quiver" model, which includes only the effects of the relativistic electron quiver motion. Other effects, such as the plasma density response, will be discussed below. A linearly polarized laser field will be assumed, $a_0(z,t) = \hat{a}_0 \cos\theta_0$. In the quiver model, the transverse plasma current is $J_q = -en_0 v_q$, where v_q is the relativistic electron quiver velocity (discussed above) and n_0 is the ambient plasma density. The quiver current J_q acts as the source term in the wave equation which drives the harmonic radiation, $(\nabla^2 - \partial^2/c^2\partial t^2)a = S_q$, where

$$S_q \simeq k_p^2 \hat{a}_0 \cos\theta_0 \left(1 + \hat{a}_0^2 \cos^2\theta_0\right)^{-1/2} \qquad (2)$$

and $\omega_p = ck_p = (4\pi e^2 n_0/m)^{1/2}$ is the plasma frequency. In the limit $\hat{a}_0^2 \ll 1$, the denominator in (2) may be expanded and the component driving the Nth odd harmonic ($\omega_N = N\omega_0$) may be determined. For example, the ratio

of the third harmonic power to the fundamental is $P_3/P_0 \simeq (\hat{a}_0^2 k_p^2 L/16k_0)^2 \equiv R_q$, where L is the laser–plasma interaction length and $L < L_d$ has been assumed, where L_d is the detuning distance (discussed below). Hence, in the quiver limit, $P_3 \sim L^2 n_0^2 I_0^3$.

The quiver model assumed that the plasma response is dominated by the electron quiver motion. This is an oversimplification and collective plasma effects, i.e., the plasma density response, cannot be neglected. Including the density response, $\delta n(z,t)$, in the transverse current gives $J_\perp = -e(n_0 + \delta n)v_\perp$. Letting $v_\perp = v_q$ gives $S = k_p^2 a_0(1 + \delta n/n_0 - a_0^2/2)$, where the term $a_0^2/2$ arises from expanding the relativistic factor, assuming $a_0^2 \ll 1$. Using 1-D cold fluid theory, the density response δn may be calculated, giving

$$S \simeq k_p^2 \hat{a}_0 \cos\theta_0 \left(1 - \frac{\hat{a}_0^2}{4} - \frac{3}{16}\frac{\omega_p^2}{\omega_0^2}\hat{a}_0^2 \cos 2\theta_0\right). \qquad (3)$$

Hence, the effect of including the density response is to reduce [7], [8] the source term for the third harmonic, $\sim \exp(\pm 3i\theta_0)$, by the factor $3\omega_p^2/4\omega_0^2 \ll 1$ as compared to the quiver model. The power in the third harmonic will be reduced by the square of this factor, $P_3/P_0 = R_q(3\omega_p^2/4\omega_0^2)^2$. Hence, in the 1-D limit, $P_3 \sim L^2 n_0^4 I_0^3$. Physically, this reduction arises from the longitudinal ponderomotive force, $F_z \sim -\nabla_z a_0^2 \sim k_0 \hat{a}_0^2 \sin 2\theta_0$. This modulates the density, $\delta n/n_0 \sim \exp(\pm 2i\theta_0)$, in such a way that it nearly cancels (to order ω_p^2/ω_0^2) the contribution from the relativistic factor, $a_0^2/2$. In the absence of the space charge potential, $\omega_p^2 \to 0$, this cancellation is exact and no third harmonics are generated in the 1-D limit.

The harmonic radiation will reach a maximum amplitude after a detuning distance [1], [19], [20], $L = L_d$. The phase velocity v_p of an electromagnetic wave of frequency ω in a plasma is given by $v_p/c \simeq 1 + \omega_p^2/2\omega^2$, where $\omega_p^2/\omega^2 \ll 1$ and $a_0^2 \ll 1$ have been assumed. The phase velocity of the incident laser, $\omega = \omega_0$, is greater than that of the third harmonic, $\omega = 3\omega_0$. Hence, the third harmonic, which is being driven by the incident laser, eventually becomes out of phase with the incident laser. The maximum amplitude of the third harmonic occurs after the detuning distance defined by $L_d \Delta v_p/c = \lambda_3/2$, where $\Delta v_p = 4c\omega_p^2/9\omega_0^2$ is the difference in the phase velocities of the incident and third harmonic fields and $\lambda_3 \simeq \lambda_0/3$ is the wavelength of the third harmonic. Hence, $L_d \simeq 3\lambda_p^2/8\lambda_0$, where $\lambda_p = 2\pi/k_p$ is the plasma wavelength. The maximum amplitude of the third harmonic power at saturation may be estimated by setting $L = L_d$, giving $P_3/P_0 \simeq (\hat{a}_0\lambda_0/\lambda_p)^4\%$. As an example, consider a $\lambda_0 = 1$ μm laser with $I_0 = 5\times10^{17}$ W/cm^2($\hat{a}_0 \simeq 0.6$)and a plasma of density $n_0 = 10^{19}$cm^{-3}($\lambda_p \simeq 11$ μm). The third harmonic power is given by $P_3/P_0 \simeq 9 \times 10^{-8}$. The detuning length is prohibitively short, $L_d \simeq 45$ μm. The third harmonic pulse length is approximately equal to that of the incident laser pulse.

II. NONLINEAR FORMULATION

The 1-D fields associated with the laser–plasma interaction can be described by the normalized transverse vector potential,

$a(z,t) = eA_\perp(z,t)/m_0c^2$, and the normalized scaler potential, $\phi(z,t) = e\Phi(z,t)/m_0c^2$. Coulomb gauge will be used, $\nabla \cdot A = 0$, which implies $A_z = 0$. It is convenient to introduce the independent variables $\xi = z - c\beta_t t$ and $\tau = t$, where β_t is the normalized transform velocity which will be specified below. Using the ξ, τ variables, the normalized potentials a and ϕ satisfy the 1-D wave equation and Poisson's equation, which are given by

$$\left(\frac{1}{\gamma_t^2} \frac{\partial^2}{\partial \xi^2} + \frac{2\beta_t}{c} \frac{\partial^2}{\partial \xi \partial \tau} - \frac{1}{c^2} \frac{\partial^2}{\partial \tau^2} \right) a = k_p^2 \frac{n}{\gamma n_0} a \qquad (4)$$

$$\frac{\partial^2}{\partial \xi^2} \phi = k_p^2 \left(\frac{n}{n_0} - 1 \right) \qquad (5)$$

where $\gamma_t^{-2} = 1 - \beta_t^2$, $\omega_p = ck_p = (4\pi e^2 n_0/m_0)^{1/2}$ is the ambient plasma frequency, $n(\xi, \tau)$ is the plasma electron density, n_0 is the ambient density, $\gamma(\xi, \tau) = (1 - \beta^2)^{-1/2}$ is the relativistic factor associated with the plasma electrons and $\beta(\xi, \tau) = v/c$ is the normalized electron fluid velocity. In deriving the right side of (4), use was made of the fact that the transverse canonical momentum is invariant and that prior to the laser interaction the plasma is assumed to be stationary, i.e., $\beta_\perp = a/\gamma$. Also, the ions are assumed to be stationary.

The electron fluid quantities n, β_z and γ are assumed to satisfy the cold, relativistic fluid equations. Using the ξ, τ variables, the continuity equation and the axial momentum equation may be written as

$$\frac{\partial}{\partial \xi} [n(\beta_t - \beta_z)] = \frac{1}{c} \frac{\partial}{\partial \tau} n \qquad (6)$$

$$\frac{\partial}{\partial \xi} [\gamma(1 - \beta_t\beta_z) - \phi] = -\frac{1}{c} \frac{\partial}{\partial \tau} (\gamma\beta_z) \qquad (7)$$

respectively. Equations (4)–(7), together with $\gamma = (1 + a^2)^{1/2}/(1 - \beta_z^2)^{1/2}$, form a complete set of fully nonlinear, relativistic cold fluid equations which describe the 1-D laser–plasma interaction. The 1-D assumption is valid provided that the radiation spot size r_s is large compared to the plasma wavelength $\lambda_p = 2\pi/k_p$, i.e., $r_s \gg \lambda_p$. The cold fluid assumption is valid provided that i) the electron quiver velocity is much greater than the transverse thermal velocity and ii) the phase velocity of the driven plasma oscillations is much greater than the axial thermal velocity, as is the case in the following.

The primary focus of this section is to determine the coherent harmonic radiation excited by a long pulse incident laser field, $c\tau_L \gg \lambda_p$. To study coherent harmonic radiation, it is convenient to set the transform velocity, β_t, equal to the phase velocity, β_{ph}, of the incident laser field, $\beta_t = \beta_{ph}$. The incident laser field is of the form $a_0 = \hat{a}_0 \cos k_0 \xi$, where $k_0\xi = k_0(z - c\beta_{ph}t)$, $\beta_{ph} = \omega_0/ck_0$ is the normalized phase velocity, ω_0 is the frequency, and k_0 is the wavenumber of the pump laser field. The pump laser amplitude is slowly varying, $|\partial \hat{a}_0/\partial \xi| \sim \hat{a}_0/c\tau_L$, and is assumed to be independent of τ, i.e., pump depletion effects are neglected. The effects of diffraction are consider in Section III. In determining the plasma response to an incident laser field of the form $a_0 = a_0(\xi)$, the τ derivatives are dropped in (6)–(7). Strictly

speaking, for a general laser field of the form $a_0 = a_0(\xi, \tau)$, the τ derivatives may be neglected provided that the transit time of the electrons through the laser pulse, τ_L (= laser pulse duration), is small compared to the evolution time, τ_E, of the laser envelope, $\tau_L \ll \tau_E$, i.e., the quasistatic approximation [8]. Assuming a quasistatic plasma response, (6)–(7) imply the existence of two constants of the motion,

$$n(\beta_t - \beta_z) = \beta_t n_0, \qquad (8a)$$

$$\gamma(1 - \beta_t\beta_z) - \phi = 1. \qquad (8b)$$

Equations (8a)–(b) may be solved to give expressions for the fluid quantities n, β_z and γ in terms of the fields a and ϕ,

$$n/n_0 = \gamma_t^2 \beta_t \left[(1 - 1/\psi^2\gamma_t^2)^{-1/2} - \beta_t \right], \qquad (9a)$$

$$\beta_z = \frac{\beta_t - (1 - 1/\psi^2\gamma_t^2)^{1/2}}{1 - \beta_t(1 - 1/\psi^2\gamma_t^2)^{1/2}}, \qquad (9b)$$

$$\gamma = \gamma_t^2(1 + \phi)\left[1 - \beta_t(1 - 1/\psi^2\gamma_t^2)^{1/2} \right], \qquad (9c)$$

$$n/\gamma n_0 = \beta_t(1 + \phi)^{-1}(1 - 1/\psi^2\gamma_t^2)^{-1/2} \qquad (9d)$$

where $\psi^2 = (1 + \phi)^2/(1 + a^2)$. Notice that for the case $\beta_t = \beta_{ph}$, $\gamma_t^{-2} = \gamma_{ph}^{-2} < 0$, since $\beta_{ph}^2 > 1$ for a radiation field in a plasma.

Using the above expressions for the fluid quantities, expressions for the normalized transverse plasma current, $k_p^2 na/n_0\gamma$, and the normalized charge perturbation, $k_p^2(n/n_0 - 1)$, can be found and inserted into the wave equation and Poisson's equation (4)–(5). This results in two coupled nonlinear equations for the potentials a and ϕ,

$$\left(\frac{1}{\gamma_t^2} \frac{\partial^2}{\partial \xi^2} + \frac{2\beta_t}{c} \frac{\partial^2}{\partial \xi \partial \tau} - \frac{1}{c^2} \frac{\partial^2}{\partial \tau^2} \right) a$$
$$= \frac{k_p^2}{(1 + \phi)} \left(1 + \frac{1}{k_p^2 \gamma_t^2} \frac{\partial^2 \phi}{\partial \xi^2} \right) a \qquad (10)$$

$$\frac{\partial^2}{\partial \xi^2} \phi = k_p^2 \gamma_t^2 \left[\beta_t(1 - 1/\psi^2\gamma_t^2)^{-1/2} - 1 \right]. \qquad (11)$$

Equations (10)–(11) completely describe the nonlinear generation of relativistic harmonic radiation in 1-D within the quasistatic approximation, i.e., $\tau_L \ll \tau_E$. For the case where the transform velocity, β_t, is set equal to the phase velocity, β_{ph}, of the incident laser, (10)–(11) may be used to analyze relativistic harmonic generation, as is done in detail in the following. For the case where β_t is set equal to the group velocity, β_g, of a laser pulse with a pulse length $\simeq \lambda_p$, (11) may be used to analyze laser wakefield generation [5]–[9]. The limiting case of $\beta_t = 1$ has been analyzed in detail in [8].

It is of interest to define the nonlinear index of refraction n_R by setting the right side of the wave equation (10) equal to $(\omega^2/c^2)(1 - n_R^2)a$, which gives

$$n_R^2 = 1 - \frac{\omega_p^2}{\omega^2(1 + \phi)} \left(1 + \frac{1}{k_p^2 \gamma_t^2} \frac{\partial^2 \phi}{\partial \xi^2} \right). \qquad (12)$$

In particular, the slow part of n_R determines the dispersion relation for the radiation field a, whereas the fast part of

n_R determines the generation of harmonic radiation, as is discussed below.

A. Dispersion Relation

To describe the generation of relativistic harmonics, the electrostatic potential ϕ is separated into slow and fast components, $\phi = \phi_s + \phi_f$, where ϕ_s is approximately constant, $|\partial \phi_s/\partial \xi| \ll |k_0 \phi|$, and $|\partial \phi_f/\partial \xi| \sim |k_0 \phi|$. The fast component ϕ_f contains the harmonic content and is important in determining the source current which drives the Nth harmonic radiation. The slow component ϕ_s is important in determining the dispersive characteristics of the harmonic radiation as well as of the pump laser field. In the following, it is assumed that $\phi_f^2/(1 + \phi_s)^2 \ll 1$. Furthermore, it is assumed that the vector potential of the harmonic radiation $\mathbf{a}_N$, where N is the harmonic number, is much less than that of the pump laser, i.e., $|a_N|/|\hat{a}_0| \ll 1$.

The dispersion relation is determined by examining the slow part of the n_R^2, (12). Assuming $|(k_p \gamma_{ph})^{-2} \partial^2 \phi_s/\partial \xi^2| \ll 1$, the dispersion relation for the radiation field, a, to leading order (i.e., neglecting ϕ_f), is given by

$$\omega_N^2/c^2 = k_N^2 + k_p^2/(1 + \phi_s) \tag{13}$$

where $\mathbf{a} = \hat{a} \exp(ik_N z - i\omega_N t)$ has been assumed with $\hat{a}$ slowly varying compared to the phase factor $\exp(ik_N z - i\omega_N t)$. Equation (13) holds for the pump laser field ($\omega_N = \omega_0, k_N = k_0$) as well as for the various harmonics. Notice that the transform velocity is the phase velocity of the incident laser field, $\beta_{ph} = \omega_0/ck_0$, and, hence, $\gamma_{ph}^{-2} = 1 - \beta_{ph}^2 = -k_p^2/k_0^2(1 + \phi_s) < 0$. Furthermore, for a nonevolving, long incident laser pulse of the form $a_0 = \hat{a}_0 \exp(ik_0 z - i\omega_0 t)$, the quantity $(1 + \phi_s)$ is approximately constant, as is discussed in the following.

Consider a long pump laser pulse with a slowly varying envelope, $|\partial \hat{a}_0/\partial \xi| \sim |\hat{a}_0/c\tau_L|$, where τ_L is the laser pulse duration, propagating in a plasma with $c\tau_L \gg \lambda_p, \lambda_0$. The slow part of the potential ϕ_s may be determined from examining the slow part of Poisson's equation (11). In the long pulse limit, $\partial^2 \phi_s/\partial \xi^2$ may be neglected and (11) implies that, to leading order (i.e., neglecting ϕ_f) [8], [21],

$$(1 + \phi_s) = (1 + \hat{a}_0^2/2)^{1/2}. \tag{14}$$

In particular, notice that in the long pulse limit, $c\tau_L \gg \lambda_p, \lambda_0$, propagation (i.e., real k) requires $\omega_p^2/\omega^2(1 + \phi_s) < 1$. This implies that for intense pump laser fields with $\hat{a}_0 \geq 1$, propagation in an overdense plasma in which $\omega_p^2/\omega^2 \geq 1$ may be possible. Physically, $\gamma_s = 1 + \phi_s = (1 + \hat{a}_0^2/2)^{1/2}$, and the reduction in the effective plasma frequency, $\omega_p/\gamma_s^{1/2}$, is due to the relativistic quiver motion of the electrons.

It should be emphasized that (14) only applies to the long pulse limit $c\tau_L \gg \lambda_p, \lambda_0$. For $\tau_L = 1$ psec, $\tau_L \gg \lambda_p$ implies plasma densities $n_0 \gg 10^{16}$ cm^{-3}. In the short pulse limit, $\lambda_p \gg c\tau_L \gg \lambda_0$, it can be shown [8], [21] that $|\phi_s| \ll 1$ provided $c\tau_L \ll \lambda_p/(1 + \hat{a}_0^2/2)^{1/2}$. Physically, $|\phi_s| \ll 1$ holds for pulses sufficiently shorter than λ_p, since λ_p is the characteristic length scale (in the ξ frame) for collective electron motion (i.e., collective electron motion leading to

charge separation and significant values of ϕ_s does not occur on time scales sufficiently shorter than $1/\omega_p$).

B. Plasma Response

The source current driving the harmonic radiation is determined by the harmonic content of n_R. As indicated by (12), n_R may be specified entirely in terms of ϕ. Hence, it is necessary to determine the various harmonic components of ϕ. This is done by analyzing the plasma response to the pump laser field, $a_0 = \hat{a}_0 \cos k_0 \xi$, via Poisson's equation (11). The various harmonic components of $\phi_f = \sum_\ell \phi_{2\ell}$, where $\phi_{2\ell} \sim \cos(2\ell k_0 \xi)$ with $\ell = 1, 2, 3...$, may be determined analytically in the limit $\epsilon = k_p^2/k_0^2(1 + \phi_s) < 1$. In this limit $|\phi_{2\ell}/(1 + \phi_s)| \sim \epsilon^\ell$, and the various harmonic components of ϕ_f may be solved order by order in ϵ.

To solve for ϕ_f via (11), two expansions will be used. Assuming $|(\psi \gamma_{ph})^{-2}| \leq \epsilon$, gives

$$\left(1 - \frac{1}{\psi^2 \gamma_{ph}^2}\right)^{-1/2} = \sum_{\ell=0}^{\infty} \frac{(2\ell - 1)!!}{(2\ell)!!} \left(\frac{1}{\psi^2 \gamma_{ph}^2}\right)^\ell, \tag{15a}$$

where $(2\ell - 1)!! = 1 \cdot 3 \cdot \ldots \cdot (2\ell - 1)$ and $(2\ell)!! = 2 \cdot 4 \cdot \ldots \cdot (2\ell)$. Assuming $|\phi_f/(1 + \phi_s)| \leq \epsilon$, gives

$$\left(\frac{1}{\psi^2 \gamma_{ph}^2}\right)^\ell = \left[\frac{(1 + a^2)}{\gamma_{ph}^2 (1 + \phi_s)^2}\right]^\ell$$
$$\cdot \sum_{m=0}^{\infty} \frac{2\ell(2\ell + 1) \ldots (2\ell + m - 1)}{m!}$$
$$\cdot \left[\frac{-\phi_f}{(1 + \phi_s)}\right]^m. \tag{15b}$$

Letting $\phi_f = \sum_\ell \phi_{2\ell}$, where $\phi_{2\ell} \sim \epsilon^\ell \cos(2\ell k_0 \xi)$ for $\ell = 1, 2, 3 \ldots$, and using the expansions in (15a) and (15b), allows $\phi_{2\ell}$ to be solved order by order in ϵ. For example, ϕ_2 (order ϵ), ϕ_4 (order ϵ^2) and ϕ_6 (order ϵ^3) are given by

$$\frac{\partial^2}{\partial \xi^2} \phi_2 = \frac{k_p^2 (a_0^2)_2}{2(1 + \phi_s)}, \tag{16a}$$

$$\frac{\partial^2}{\partial \xi^2} \phi_4 = k_p^2 \left[-\frac{\phi_2 (a_0^2)_2}{(1 + \phi_s)^3} + \frac{3(a_0^4)_4}{8\gamma_{ph}^2 (1 + \phi_s)^4}\right], \tag{16b}$$

$$\frac{\partial^2}{\partial \xi^2} \phi_6 = \frac{-k_p^2 (a_0^2)_2}{(1 + \phi_s)^3} \left[\phi_4 - \frac{3\phi_2^2}{2(1 + \phi_s)}\right.$$
$$\left. + \frac{3(a_0^2)_2}{2\gamma_{ph}^2 (1 + \phi_s)^2} \left(\phi_2 - \frac{5(a_0^2)_2}{24\gamma_{ph}^2 (1 + \phi_s)^2}\right)\right] \tag{16c}$$

where the subscript $(Q)_\ell$ refers to the ℓth harmonic component of Q, i.e., $(Q)_\ell \sim \exp(\ell k_0 \xi)$. These equations can be solved iteratively to yield

$$\phi_2 = -\frac{k_p^2 \hat{a}_0^2 \cos 2k_0 \xi}{16 k_0^2 (1 + \phi_s)^2}, \tag{17a}$$

$$\phi_4 = \frac{k_p^4 \hat{a}_0^4 \cos 4k_0 \xi}{2^9 k_0^4 (1 + \phi_s)^5}, \tag{17b}$$

$$\phi_6 = -\frac{17 k_p^6 \hat{a}_0^6 \cos 6 k_0 \xi}{9 \cdot 2^{14} k_0^6 (1 + \phi_s)^8}. \tag{17c}$$

For order of magnitude scaling purposes, (17a)–(17c) imply that the $2\ell^{th}$ harmonic component of ϕ_f scales as

$$\phi_{2\ell} \sim (-1)^\ell \frac{(2\ell - 1)!!}{(2\ell)!!} \left(\frac{k_p}{2 k_0}\right)^{2\ell} \frac{\hat{a}_0^{2\ell} \cos 2\ell k_0 \xi}{2\ell^2 (1 + \phi_s)^{3\ell - 1}}. \tag{18}$$

Knowing the harmonic components of ϕ_f, it is possible to determine the harmonic components of the source current S which drives the harmonic radiation. The source current S is given by the right side of (10),

$$S = \frac{k_p^2 a}{(1 + \phi)} \left(1 + \frac{1}{k_p^2 \gamma_{ph}^2} \frac{\partial^2 \phi}{\partial \xi^2}\right). \tag{19}$$

Using the expansion

$$(1 + \phi)^{-1} = (1 + \phi_s)^{-1} \sum_{m=0}^{\infty} \left[-\phi_f (1 + \phi_s)^{-1}\right]^m \tag{20}$$

allows the harmonic components of S to be solved order by order in ϵ. For example, the third, S_3, fifth, S_5, and seventh, S_7, harmonic components are given by (21) at the bottom of this page. Using (17a)–(17c) gives

$$S_3 = -\frac{3 k_p^4 \hat{a}_0^3 \cos 3 k_0 \xi}{32 k_0^2 (1 + \phi_s)^4}, \tag{22a}$$

$$S_5 = \frac{3 k_p^6 \hat{a}_0^5 \cos 5 k_0 \xi}{2^8 k_0^4 (1 + \phi_s)^7}, \tag{22b}$$

$$S_7 = -\frac{5 k_p^8 \hat{a}_0^7 \cos 7 k_0 \xi}{3 \cdot 2^{10} k_0^6 (1 + \phi_s)^{10}}. \tag{22c}$$

For order of magnitude scaling purposes, (22a)–(22c) imply that the $(2\ell + 1)^{th}$ harmonic component of S scales as

$$S_{2\ell+1} \sim (-1)^\ell k_p^2 \frac{(2\ell - 1)!!}{(2\ell)!!} \left(\frac{k_p}{2 k_0}\right)^{2\ell} \frac{\hat{a}_0^{2\ell+1} \cos(2\ell + 1) k_0 \xi}{\ell (1 + \phi_s)^{3\ell + 1}}. \tag{23}$$

C. Harmonic Radiation

The above expressions for the harmonic components of the source current, $S_{2\ell+1}$, may be used in the wave equation (10), to determine the growth and saturation of the relativistic

harmonic radiation. It is convenient to represent the Nth harmonic radiation field, a_N, by the form

$$a_N = \hat{a}_N(\tau) \exp(i k_N \xi - i \Delta \omega_N \tau) \tag{24}$$

with $\Delta \omega_N = \omega_N - \beta_{ph} c k_N$ and $k_N \xi - \Delta \omega_N \tau = k_N z - \omega_N t$, where k_N and ω_N are the wavenumber and frequency of the Nth harmonic radiation field. Inserting this into (10) gives a reduced wave equation for the harmonic amplitude $\hat{a}_N$,

$$\left(\frac{\partial}{\partial \tau} - 2 i \omega_N\right) \frac{\partial}{\partial \tau} \hat{a}_N = -c^2 \hat{S}_N \exp(i \Delta \omega_N \tau) \tag{25}$$

where ω_N and k_N satisfy the dispersion relation, (13), with $k_N = N k_0$ and $N = 2\ell + 1$. The amplitude of the source current for the Nth harmonic, $\hat{S}_N$, is given by $S_N = \hat{S}_N \exp(i N k_0 \xi)$, where S_N is given in (22)–(23).

The reduced wave equation (25) has the solution

$$\hat{a}_N \simeq -c^2 \hat{S}_N \frac{[\exp(i \Delta \omega_N \tau) - 1]}{(\omega_N^2 - c^2 \beta_{ph}^2 k_N^2)} \tag{26}$$

where

$$\omega_N^2 - c^2 \beta_{ph}^2 k_N^2 = -\frac{c^2 k_p^2}{(1 + \phi_s)}(N^2 - 1). \tag{27}$$

For small times $|\Delta \omega_N \tau| \ll 1$, the harmonics grow linearly in time [8],

$$\hat{a}_N \simeq -i \hat{S}_N \frac{c \tau}{2 N k_0}. \tag{28}$$

The Nth harmonic reaches its maximum amplitude [1], [19], [20] after a dephasing length $c \tau_{d_N} = \pi c / |\Delta \omega_N|$, where $\Delta \omega_N = c k_N (\beta_N - \beta_{ph})$ and $\beta_N = \omega_N / c k_N$, i.e.,

$$\Delta \omega_N = N c k_0 \left[\left(1 + \frac{k_p^2}{N^2 k_0^2 (1 + \phi_s)}\right)^{1/2} - \left(1 + \frac{k_p^2}{k_0^2 (1 + \phi_s)}\right)^{1/2}\right]. \tag{29}$$

For $k_p^2 / k_0^2 (1 + \phi_s) \ll 1$, (29) may be expanded giving

$$c \tau_{d_N} \simeq \frac{N \lambda_p^2 (1 + \phi_s)}{(N^2 - 1) \lambda_0}. \tag{30}$$

$$S_3 = \frac{k_p^2 a}{(1 + \phi_s)} \left[-\frac{\phi_2}{(1 + \phi_s)} + \frac{1}{k_p^2 \gamma_{ph}^2} \frac{\partial^2 \phi_2}{\partial \xi^2}\right], \tag{21a}$$

$$S_5 = \frac{-k_p^2 a}{(1 + \phi_s)} \left[\frac{\phi_4}{(1 + \phi_s)} - \frac{\phi_2^2}{(1 + \phi_s)^2} - \frac{1}{k_p^2 \gamma_{ph}^2} \left(\frac{\partial^2 \phi_4}{\partial \xi^2} - \frac{\phi_2}{(1 + \phi_s)} \frac{\partial^2 \phi_2}{\partial \xi^2}\right)\right], \tag{21b}$$

$$S_7 = \frac{-k_p^2 a}{(1 + \phi_s)} \left\{\frac{\phi_6}{(1 + \phi_s)} + \frac{\phi_2^3}{(1 + \phi_s)^3} - \frac{2 \phi_2 \phi_4}{(1 + \phi_s)^2}\right.$$
$$\left. - \frac{1}{k_p^2 \gamma_{ph}^2} \left[\frac{\partial^2 \phi_6}{\partial \xi^2} - \frac{\phi_2}{(1 + \phi_s)} \frac{\partial^2 \phi_4}{\partial \xi^2} - \left(\frac{\phi_4}{(1 + \phi_s)} - \frac{\phi_2^2}{(1 + \phi_s)^2}\right) \frac{\partial^2 \phi_2}{\partial \xi^2}\right]\right\}. \tag{21c}$$
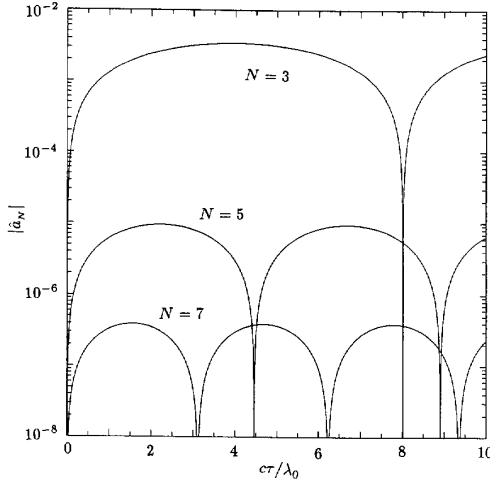
Fig. 2. The amplitude of the normalized vector potential of the harmonic radiation, $|\hat{a}_N|$, for the first three harmonics, $N = 3$, 5, and 7, versus the normalized propagation distance, $c\tau/\lambda_0$, for a $\lambda_0 = 1\mu$m wavelength laser of intensity $I_0 = 6.7 \times 10^{18}$ W/cm^2 ($\hat{a}_0 = 2.2$) interacting with a plasma of density $n_0 = 10^{20}$ cm^{-3} ($\lambda_p = 3.4\mu$m).

The maximum amplitude of the Nth harmonic at $c\tau = c\tau_{d_N}$ is given by

$$|\hat{a}_N|_{max} = \frac{2|\hat{S}_N|(1 + \phi_s)}{k_p^2(N^2 - 1)}. \tag{31}$$

In particular, for the third, fifth, and seventh harmonics,

$$|\hat{a}_3|_{max} = \frac{3k_p^2\hat{a}_0^3}{2^7 k_0^2(1 + \phi_s)^3}, \tag{32a}$$

$$|\hat{a}_5|_{max} = \frac{k_p^4\hat{a}_0^5}{2^{10} k_0^4(1 + \phi_s)^6}, \tag{32b}$$

$$|\hat{a}_7|_{max} = \frac{5k_p^6\hat{a}_0^7}{9 \cdot 2^{13} k_0^6(1 + \phi_s)^9}. \tag{32c}$$

An order of magnitude expression for the maximum amplitude of the $(2\ell + 1)$ harmonic is given by

$$|\hat{a}_{2\ell+1}|_{max} \sim \frac{2(2\ell - 1)!!(k_p/2k_0)^{2\ell}\hat{a}_0^{2\ell+1}}{\ell(2\ell)!![(2\ell + 1)^2 - 1](1 + \phi_2)^{3\ell}}. \tag{33}$$

Equations (32)–(33) are valid for all $\hat{a}_0$, including $\hat{a}_0 \geq 1$. It is easily verified that $|\hat{a}_N|_{max} \ll \hat{a}_0$. A plot of the harmonic radiation amplitude $|\hat{a}_N|$ verses the normalized propagation distance $c\tau/\lambda_0$ is shown in Fig. 2 for the first three harmonics for the parameters $\lambda_0 = 1$ μm, $\lambda_p = 3.4$ μm ($n_0 = 10^{20}$ cm^{-3}) and $\hat{a}_0 = 2.2$ ($I_0 = 6.7 \times 10^{18}$ W/cm^2).

At saturation for the Nth harmonic, i.e., $c\tau = c\tau_{d_N}$, the ratio of the power in the Nth harmonic, P_N, to the power in the pump radiation field, P_0, is given by $P_N/P_0 = N^2|\hat{a}_N|^2/\hat{a}_0^2$. Using (32)–(33), this can be written as

$$\frac{P_N}{P_0} \simeq C_N\left(\frac{\lambda_0\hat{a}_0}{\lambda_p}\right)^{2(N-1)}\left(1 + \frac{\hat{a}_0^2}{2}\right)^{-3(N-1)/2} \tag{34}$$

where C_N are constants which decrease rapidly with increasing harmonic number, i.e., $C_3 = 4.9 \times 10^{-3}$, $C_5 = 2.4 \times$

10^{-5} and $C_7 = 2.3 \times 10^{-7}$. Furthermore, driven relativistic harmonic generation is a nonresonant interaction; hence, the process is not sensitive to thermal plasma effects.

As an example, consider an incident laser with $P_0 = 10$ TW, $\lambda_0 = 1$ μm and spot size $r_0 = 10$ μm ($\hat{a}_0 = 2.2$), interacting with a plasma of density $n_0 = 10^{20}$ cm^{-3} ($\lambda_p = 3.4$ μm). For the third (and fifth) harmonic, $P_N/P_0 = 2.2 \times 10^{-5}$ (4.6×10^{-10}) and $L_d = 8.0$ μm (4.3 μm). Hence, 220 MW (4.6 kW) should be observed at a wavelength of $\lambda = 3300$ Å (2000 Å). Clearly, the limitations due to phase detuning are restrictive. If a scheme for phase matching could be conceived, the interaction distance, L, and thus the harmonic power, $P_N \sim L^2$, could be increased.

III. DIFFRACTIVE EFFECTS

The above nonlinear theory indicates that the Nth harmonic saturates (i.e., reaches maximum amplitude) after a detuning distance $c\tau_{d_N} = \pi c/|\Delta\omega_N|$, where $\Delta\omega_N = ck_N(\beta_N - \beta_{ph})$. In particular, for the third harmonic, $c\tau_{d_3} \simeq 3\lambda_p^2(1+\phi_s)/8\lambda_0$. Physically, saturation by detuning arises due to the fact that the phase velocity of the fundamental pump radiation is greater that than the phase velocity of the harmonic radiation, i.e., $\beta_{ph} > \beta_N$. In a realistic 3-D configuration, however, diffractive effects will limit the effective radiation–plasma interaction length to a few vacuum Rayleigh lengths, Z_R, where $Z_R = k_0r_0^2/2$ and r_0 is the minimum spot size of the radiation field (assumed to be Gaussian). For interaction distances $L \ll \pi Z_R$, diffraction effects are unimportant and the 1-D theory is an adequate description. In the limit $\pi Z_R \lesssim L$, however, and in particular $\pi Z_R \lesssim c\tau_{d_N}$, diffractive effects are important and must be included in the analysis.

In the following, the generation of second and third harmonic radiation is analyzed using 3-D relativistic fluid equations in the limit $a_0^2 \ll 1$. Higher order harmonic generation can be consider by solving the fluid equations to higher order in a_0^2. Furthermore, the radiation fields are assumed to undergo vacuum diffraction. Notice that in the regime in which 3-D effects are important, the condition $\pi Z_R < c\tau_{d_3}$ implies $\delta = 2k_p^2r_0^2/3 < 1$. Hence, the effects of relativistic optical guiding [3]–[10], which become important when $P/P_c = k_p^2r_0^2\hat{a}_0^2/32 \gtrsim 1$, may be neglected in the 3-D regime when $\hat{a}_0^2 \ll 1$. Assuming vacuum diffraction, a long pulse, Gaussian, incident radiation field evolves according to $a(r, z, t) = a_v \exp i\theta_0$, where $\theta_0 = k_0z - \omega_0t$ and the radiation envelope a_v is given by [24]

$$a_v(r, z) = \frac{\hat{a}_0r_0}{r_s}\exp\left[-(1 - i\alpha)\frac{r^2}{r_s^2} - i\tan^{-1}\alpha\right] \tag{35}$$

where $r_s = r_0(1 + \alpha^2)^{1/2}$ is the radiation spot size, $\alpha = z/Z_R$ and $z = 0$ is the location of the laser focus at which $r_s = r_0$.

A. 3-D Formulation

The 3-D pump laser interaction with the plasma electrons will be modeled using the cold, relativistic fluid equations. In particular, the momentum equation and the continuity equation may be written as

$$\frac{1}{c}\frac{d}{dt}\mathbf{u} = \nabla\phi + \frac{1}{c}\frac{\partial}{\partial t}\mathbf{a} - \frac{1}{\gamma}\mathbf{u} \times (\nabla \times \mathbf{a}) \tag{36}$$

$$\frac{1}{c}\frac{\partial}{\partial t}(\rho\gamma) + \nabla \cdot (\rho\mathbf{u}) = 0 \qquad (37)$$

respectively, where $\mathbf{u} = \mathbf{p}/mc$ is the normalized momentum, $\gamma = (1+u^2)^{1/2}$ is the relativistic factor, and $\rho = n/(n_0\gamma)$. The evolution of the normalized potentials are determined from the wave equation and Poisson's equation, which may be written as

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{a} = k_p^2\rho\mathbf{u} + \frac{1}{c}\frac{\partial}{\partial t}\nabla\phi \qquad (38)$$

$$\nabla^2\phi = k_p^2(\rho\gamma - 1) \qquad (39)$$

respectively, where Coulomb gauge, $\nabla \cdot \mathbf{a} = 0$, has been assumed.

To study the generation of the third harmonic, the above equations will be solved perturbatively, i.e., order by order in the pump laser amplitude $\hat{a}_0$, assuming $\hat{a}_0^2 \ll 1$. The various plasma quantities, denoted by Q, will be represented by an expansion $Q = Q_0 + Q_1 + Q_2 + \ldots$, where $Q_n \sim a_0^n$. The pump laser vector potential is assumed to be known and given by $\mathbf{a}_{0\perp} = a_v \exp i\theta_0\mathbf{e}_x$, where $a_v(r, z)$ is the vacuum solution given by (35). The effects of pump depletion and of various laser-plasma instabilities will be neglected. The zeroth order plasma quantities (in the absence of the pump laser) are given by $\mathbf{u}_0 = 0$, $\gamma_0 = 1$, $\rho_0 = 1$ and $\phi_0 = 0$, i.e., the plasma is assumed to be initially uniform, stationary, and neutral. To first order in the normalized vector potential, (36) implies that $\mathbf{u}_1 = \mathbf{a}_0$, which is simply the quiver motion of the electrons. Furthermore, (36)–(39) imply $\rho_1 = \gamma_1 = \phi_1 = 0$. The first order form of the wave equation (38) implies that the pump laser field obeys the dispersion relation $\omega_0^2/c^2 = k_0^2 + k_p^2$.

To second order, (36)–(39) imply that quantities ϕ_2, $\mathbf{u}_2$ and ρ_2 are related to a_0^2 by

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} + k_p^2\right)\phi_2 = k_p^2\frac{a_0^2}{2}, \qquad (40a)$$

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} + k_p^2\right)\mathbf{u}_2 = -\frac{1}{c}\frac{\partial}{\partial t}\nabla\frac{a_0^2}{2}, \qquad (40b)$$

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} + k_p^2\right)\rho_2 = \left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2} - k_p^2\right)\frac{a_0^2}{2}. \qquad (40c)$$

Furthermore, (40a) and (40c) imply that the second-order wave equation describing the generation of the second harmonic may be written as

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2} - k_p^2\right)\mathbf{a}_2 = 0. \qquad (41)$$

Hence, no second harmonic radiation will be generated.

To third order, (36)–(39) imply that quantities ϕ_3 and $\mathbf{u}_3$ are related to a_0^3 by

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} + k_p^2\right)\nabla^2\phi_3 = k_p^2\left[\nabla^2(\mathbf{a}_0 \cdot \mathbf{u}_2) - \frac{1}{c}\frac{\partial}{\partial t}\nabla \cdot (\mathbf{a}_0\rho_2)\right], \qquad (42a)$$

$$\frac{1}{c}\frac{\partial}{\partial t}\mathbf{u}_3 = \nabla\phi_3 - \nabla(\mathbf{a}_0 \cdot \mathbf{u}_2) \qquad (42b)$$

Furthermore, the third-order transverse wave equation, describing the generation of third harmonic radiation, is given by

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2} - k_p^2\right)\mathbf{a}_{3\perp}$$
$$= \left(k_p^2\rho_2\mathbf{a}_{0\perp} + k_p^2\mathbf{u}_{3\perp} + \frac{1}{c}\frac{\partial}{\partial t}\nabla_\perp\phi_3\right)_3. \qquad (43)$$

where $(\ldots)_n$ signifies the nth harmonic component.

In particular, to determine the source term driving the third harmonic radiation, it is necessary to determine the fast (i.e., third harmonic) components of ϕ_3 and $\mathbf{u}_3$. Assuming $\phi_3, \mathbf{u}_3 \sim \exp 3i\theta_0$, (42a) and (42b) imply

$$\phi_3 \simeq -\frac{\omega_p^2}{9\omega_0^2}(\mathbf{a}_0 \cdot \mathbf{u}_2)_3, \qquad (44a)$$

$$\mathbf{u}_3 \simeq -\frac{ic}{3\omega_0}\nabla(\mathbf{a}_0 \cdot \mathbf{u}_2)_3 \qquad (44b)$$

where terms of order $k_p^2/k_0^2 \ll 1$ and $1/r_0^2k_0^2 \ll 1$ have been neglected. It can be shown that the leading order contributions of each of the three terms on the right side of (43) are of the same order. However, to leading order, $k_p^2\mathbf{u}_{3\perp} + \nabla_\perp\partial\phi_3/\partial ct = 0$, and, hence, the wave equation describing third harmonic generation is given by

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2} - k_p^2\right)\mathbf{a}_{3\perp} = k_p^2(\rho_2\mathbf{a}_{0\perp})_3 \qquad (45)$$

where higher order terms (of order k_p^2/k_0^2 and $1/r_0^2k_0^2$) have been neglected. The transverse wave equation (45) along with the second harmonic component of ρ_2,

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} + k_p^2\right)\rho_2 = \left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2} - k_p^2\right)\frac{(a_0^2)_2}{2} \qquad (46)$$

completely determine the generation of third harmonic radiation in 3-D in the limit $a_0^2 \ll 1$.

B. Third Harmonic Generation

The pump laser is assumed to be a long pulse, Gaussian laser beam which is diffracting according to the relation $a_0(r, z, t) = a_v \exp i\theta_0$, where $a_v(r, z)$ is the envelope given by (35) and $\omega_0^2/c^2 = k_p^2 + k_0^2$. Using this form in (46) gives

$$\rho_2 \simeq -\frac{1}{2r_0^2k_0^2}\left[\frac{3}{8}r_0^2k_p^2 + \frac{1}{(1+i\alpha)}\right]a_v^2 \exp 2i\theta_0. \qquad (47)$$

Hence, the source term driving the third harmonic radiation is given by

$$k_p^2(\rho_2a_0)_3 \simeq -\frac{k_p^2}{4r_0^2k_0^2}\left[\frac{3}{8}r_0^2k_p^2 + \frac{1}{(1+i\alpha)}\right]a_v^3 \exp 3i\theta_0. \qquad (48)$$

It is convenient to denote the third harmonic radiation field, $a_3(r, z, t)$, by the following form,

$$a_3(r, z, t) = f(z)a_{v3}(r, z) \exp i\theta_3, \qquad (49a)$$

$$a_{v3}(z, r) = \frac{r_3}{r_{s3}}\exp\left[-(1 - i\alpha_3)\frac{r^2}{r_{s3}^2} - i\tan^{-1}\alpha_3\right] \qquad (49b)$$

where $\theta_3 = k_3 z - \omega_3 t$, ω_3 and k_3 are the third harmonic frequency and wavenumber, $r_{s3} = r_3(1 + \alpha_3^2)^{1/2}$ is the third harmonic spot size, $\alpha_3 = z/Z_{R3}$, $Z_{R3} = k_3 r_3/2$ is the Rayleigh length associated with the third harmonic, and $\omega_3^2/c^2 = k_3^2 + k_p^2$. Inserting this into the wave equation (45) gives

$$2ik_3 a_{v3} \exp(i\theta_3) \frac{\partial}{\partial z} f \simeq k_p^2 (\rho_2 a_0)_3 \qquad (50)$$

where the right side is given by (48). Letting $\omega_3 = 3\omega_0$ and $r_3^2 = r_0^2/3$ implies $Z_{R3} \simeq Z_R$ and

$$\frac{\partial}{\partial z} f \simeq \frac{ik_p^2 \hat{a}_0^3}{24 r_0^2 k_0^3} \left[\frac{3}{8} r_0^2 k_p^2 + \frac{1}{(1 + i\alpha)} \right] \frac{\exp(i\Delta k z)}{(1 + i\alpha)^2} \qquad (51)$$

where $\Delta k = 3k_0 - k_3 = -4k_p^2/3k_0$.

For a pump laser pulse which interacts with a uniform plasma extending from $z_{min} < z < z_{max}$, (51) can be integrated giving

$$f = |f_{1-D}|[h(\delta, \alpha_{max}) - h(\delta, \alpha_{min})], \qquad (52a)$$

$$h(\delta, \alpha) = -\frac{4}{9} \left\{ \left(\frac{\delta}{8} + \frac{1}{(1 + i\alpha)} \right) \frac{e^{-i\delta\alpha}}{(1 + i\alpha)} \right.$$
$$\left. - \frac{\delta^2}{8} e^\delta E_1[\delta(1 + i\alpha)] \right\} \qquad (52b)$$

where $|f_{1-D}| = |3\hat{a}_0^3 k_p^2/2^7 k_0^2|$ is the maximum amplitude of the third harmonic in the 1-D limit, $\delta = |\Delta k| Z_R = 2k_p^2 r_0^2/3$ ($\delta = \pi Z_R/c\tau_{d_3}$), $\alpha_{min} = z_{min}/Z_R$, $\alpha_{max} = z_{max}/Z_R$ and E_1 is the exponential integral.

In particular, consider a finite slab plasma centered about the focal point of the pump laser (chosen to be $z = 0$) which extends from $z_{min} = -z_0$ to $z_{max} = z_0$. The amplitude function f of the third harmonic signal emerging from the slab plasma at $z = z_0$ is given by

$$f = 2|f_{1-D}|\text{Im}[h(\delta, \alpha_0)] \qquad (53)$$

where $\text{Im}[h(\delta, \alpha_0)]$ is the imaginary part of $h(\delta, \alpha_0)$ and $\alpha_0 = z_0/Z_R$. The ratio of the third harmonic power to the pump laser power is given by

$$P_3/P_0 = \omega_3^2 r_3^2 |f|^2 / \omega_0^2 r_0^2 \hat{a}_0^2 = 3|f|^2/\hat{a}_0^2. \qquad (54)$$

Hence, for a slab plasma centered about the laser focus, $P_3 \sim 4|\text{Im}[h(\delta, \alpha_0)]|^2$. A plot of the function $H(\delta, \alpha_0) = 4|\text{Im}[h(\delta, \alpha_0)]|^2$ verses the normalized plasma slab width, α_0, is shown in Fig. 3 for $\delta = 0.1$, 1.0, and 10.0. Notice that for $\delta = 10.0$ ($c\tau_{d_3} \ll \pi Z_R$) and $\alpha_0 \ll 1$, the first peak, which occurs when the slab width is equal to the detuning length, $2z_0 = c\tau_{d_3}$, is close to the 1-D value of $H(\delta, \alpha_0) = 1$. For $\delta = 0.1$ ($c\tau_{d_3} \gg \pi Z_R$), the maximum amplitude of the third harmonic occurs for a plasma length of $\simeq Z_R$, at which $H(\delta, \alpha_0) \simeq 1/3$. In the limit of a slab plasma with a spatial extent large compared to Z_R, i.e., $z_0 \gg Z_R$ ($\alpha_0 \gg 1$), $f \to f_I$, where $f_I = |f_{1-D}|\alpha_0^{-2} \sin(\delta\alpha_0)$. Hence, $f_I \to 0$ as $\alpha_0 \to \infty$, i.e., the third harmonic radiation emerging from a plasma centered about the laser focus vanishes for plasma dimensions large compared to Z_R. The conclusion that no third harmonic is generated by an infinite medium is also
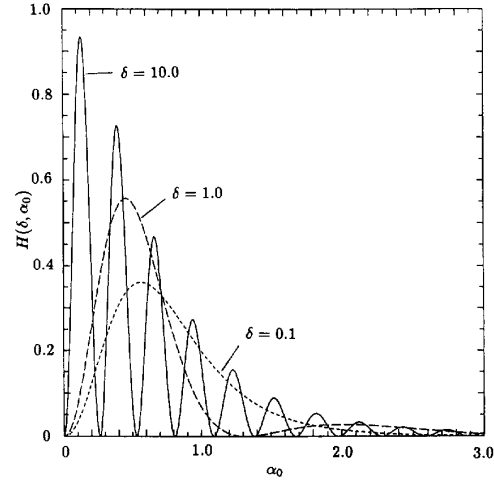

Fig. 3. The function $H(\delta, \alpha_0)$, proportional to the third harmonic power emerging from a plasma of width $2z_0$ centered about the laser focus, verses $\alpha_0 = z_0/Z_R$, for $\delta = 2k_p^2 r_0^2/3 = 10.0$ (solid curve), 1.0 (dashed curve) and 0.1 (dotted curve), where $Z_R = k_0 r_0^2/2$ is the Rayleigh length.

the case for third harmonic generation due to the nonlinear susceptibility associated with bound electrons [25].

A finite third harmonic signal, however, may be detected by focusing the pump laser on the trailing edge ($z = 0$) of a plasma slab extending from $z_{min} \leq z \leq z_{max} = 0$, with dimensions large compared to Z_R, i.e., $z_{min} \ll -Z_R$. In practice, this may be achieved by focusing the pump laser pulse on the trailing edge of a pre-ionized gas get. Approximating $\alpha_{min} = -\infty$ and $\alpha_{max} = 0$ gives $f = f_S$, where

$$f_S = -\frac{k_p^2 \hat{a}_0^3}{96 k_0^2}[1 + g(\delta)], \qquad (55a)$$

$$g(\delta) = (\delta/8)[1 - \delta e^\delta E_1(\delta)]. \qquad (55b)$$

Furthermore, it can be shown that $0 \leq 1 - \delta e^\delta E_1(\delta) < (1 + \delta)^{-1}$. Hence, $g^2 \ll 1$ and may be neglected. The amplitude of the third harmonic emerging from the plasma is given by $|a_3| = |f_S||a_{v3}|$. At the laser focus, this is a factor of $4/9$ smaller than that obtained from the 1-D theory in the limit $\hat{a}_0^2 \ll 1$. Hence, for a semi-infinite plasma including the effects of diffraction in the limit $\hat{a}_0^2 \ll 1$, the ratio of third harmonic power to the pump laser power, $P_3/P_0 = 3|f_S^2|/\hat{a}_0^2$, is given by

$$P_3/P_0 \simeq 3 \times 10^{-4} (\lambda_0 \hat{a}_0/\lambda_p)^4 \qquad (56)$$

which is a factor of $(4/9)^2/3 \simeq 1/15$ smaller than the corresponding 1-D expression for the maximum power.

C. Second Harmonic Generation

The above results indicate that for an initially uniform plasma density, no second harmonics are excited. This is true even when the effects of a finite incident laser spot size, r_0, and the transverse gradients associated with self-consistent 3-D density perturbation are included, i.e., (41). Generation of even harmonics requires that the plasma have initial transverse

density gradients prior to the arrival of the incident laser, i.e., $\nabla_\perp n_0 \neq 0$, where $n_0(r)$ is the initial plasma density profile. Electrons undergoing quiver motion in the presence of a density gradient produce density oscillations. For a pump laser of the form $\mathbf{a}_0 = \hat{a}_0 \cos\theta_0 \mathbf{e}_x$, the continuity equation, to first order in $\hat{a}_0 \ll 1$, implies a density oscillation δn_1 of the form $\delta n_1 \simeq (\hat{a}_0 \sin\theta_0)\nabla_x n_0/k_0$. This produces a source current $S_2 \simeq k_p^2(\delta n_1/n_0)\hat{a}_0 \cos\theta_0 \sim \sin 2\theta_0$ which drives the second harmonic radiation. The amplitude of the second harmonic radiation may be estimated by approximating the density gradient by $\nabla_x n_0 \simeq -n_0/r_p$, assuming $r_p^2 \gg r_0^2$ and neglecting the effects of diffraction. At saturation, i.e., after a detuning length $L_{d_2} = 2\lambda_p^2/3\lambda_0$, the amplitude of the second harmonic is given by $|\hat{a}_2| \simeq \hat{a}_0^2/3k_0 r_p$. The ratio of second harmonic power to pump laser power is $P_2/P_0 \simeq (\hat{a}_0\lambda_0/r_p)^2\%$ and, hence, $P_2 \sim I_0^2$. Not only do initial density gradients lead to the generation of even harmonics for linearly polarized incident lasers, it also implies that circularly polarized incident lasers will generate harmonics, both even and odd.

Transverse plasma density gradients can occur when a Gaussian laser pulse produces intensity-dependent ionization of a neutral gas. For laser pulses with peak intensities $I_0 > I_S$ (where I_S is the saturation intensity for which the gas is fully ionized) ionizing a uniform gas, density gradients will exist in the "halo" region about the laser focus. This halo region is the portion of the interaction region in which the gas is not fully ionized and corresponds approximately to focal regions in which the laser intensity lies within the band $I_{min} < I < I_S$ (where I_{min} is the minimum intensity required to produce ionization). As the peak intensity, I_0, increases, one can show that the volume of this halo region increases as $I_0^{3/2}$, where a Gaussian laser pulse of the form given by (35) has been assumed (i.e., a spherical lens and a double cone focal geometry) [26]. Second harmonics will be produced from the halo region. The intensity scaling of the second harmonic power with peak intensity will tend to be dominated by effects associated with the increasing volume of the halo region. Hence, for $I_0 > I_S$, the volume effect implies a second harmonic power scaling of $P_2 \sim I_0^{3/2}$. Furthermore, in the partially ionized halo region, the production of harmonic radiation can be significantly enhanced and/or dominated by atomic [13] and ionization [14] processes [17], [18]. In long-pulse laser-plasma experiments, filamentation may be a dominant mechanism in second harmonic generation [15], [27].

IV. Conclusion

A nonlinear cold fluid model valid in the regime $\hat{a}_0^2 \geq 1$, given by (10) and (11), has been formulated and used to analyze relativistic harmonic generation. The self-consistent collective plasma response is included and shown to significantly reduce the source current driving the harmonic radiation. A general expression for the nonlinear index of refraction was derived (12), and is a function of only the electrostatic (space-charge) potential of the plasma, ϕ. The slow part of the index of refraction determined the dispersion

relation (13) and the fast part determined the source current for the harmonics. Saturation of the Nth harmonic occurs after a detuning length, $L_{d_N} = c\tau_{d_N}$, given by (30). The harmonic amplitude is maximum when the laser-plasma interaction length, L, is an integer multiple of L_{d_N}. The ratio of saturated power in the Nth harmonic to that in the incident laser is given by (34). This expression is valid for long lasers pulses, $c\tau_L \gg \lambda_p$ ($n_0 \gg 10^{16}$ cm^{-3} for $\tau_L \simeq 1$ psec), and for interaction distances short compared to the diffraction length, $L \ll \pi Z_R$. Relativistic harmonic generation favors the use of high densities and intense lasers, $\hat{a}_0 \sim 1$. The saturated power given by (34), in the limit $\hat{a}_0^2 \ll 1$, is a factor of $b_N(\lambda_0/\lambda_p)^{2(N-1)}$ smaller, where b_N are constants, than that predicted by the simplified quiver model, which neglects the self-consistent plasma response. This reduction in the harmonic power by collective plasma effects is supported by recent experiments on harmonic generation in pre-ionized plasmas [2], [17], [18].

The effects of a diffracting incident laser field with a finite spot size r_0 have been analyzed in the limit $\hat{a}_0^2 \ll 1$. Diffraction is important for interaction lengths $L \gtrsim \pi Z_R$ ($L_{d_N} \geq \pi Z_R$ implies $\delta = 2k_p^2 r_0^2/3 \leq 1$ for the third harmonic). It is shown that no third harmonic signal emerges from a plasma of near infinite extent. A finite third harmonic signal requires the use of a semi-infinite or finite slab plasma. The third harmonic power emerging from the edge of a semi-infinite plasma, which corresponds to the focal point of the incident laser, is given by (55) and is a factor of 15 smaller than the corresponding 1-D saturation power. No second harmonics are generated from an initially uniform plasma. The generation of even harmonics requires the existence of transverse gradients in the initial plasma density. Circularly polarized light will also generate both even and odd harmonics when initial transverse density gradients are present.

The most severe constraint on the production of coherent relativistic harmonic radiation is that of phase detuning. In a dense plasma with $n_0 = 10^{20}$ cm^{-3} ($\lambda_p = 3.4~\mu$m) and a $\lambda_0 = 1~\mu$m laser with $I_0 = 6.7 \times 10^{18}$ W/cm^2 ($\hat{a}_0 = 2.2$), the saturation efficiencies for the third and fifth harmonics are $P_3/P_0 = 2.2 \times 10^{-5}$ and $P_5/P_0 = 4.6 \times 10^{-10}$, respectively. At such a high density the detuning lengths are extremely short, $L_{d_3} = 8.0~\mu$m for the third harmonic and $L_{d_5} = 4.3~\mu$m for the fifth harmonic. If a scheme for phase matching could be conceived, the interaction distance, L, and, hence, the harmonic power, $P_N \sim L^2$, could be dramatically increased.

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