Third-Harmonic Generation with Ultrahigh-Intensity Laser Pulses

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When an intense, plane-polarized, laser pulse interacts with a plasma, the relativistic nonlinearities induce a third-harmonic polarization. A phase-locked growth of a third-harmonic wave can take place, but the difference between the nonlinear dispersion of the pump and driven waves leads to a rapid unlocking, resulting in a saturation. What become third-harmonic amplitude oscillations are identified here, and the nonlinear phase velocity and the renormalized electron mass due to plasmon screening are calculated. A simple phase-matching scheme, based on a resonant density modulation, is then proposed and analyzed.

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Recent advances in laser pulse compression [1] now make possible the exploration of laser-plasma interactions at fluxes above 10^{18} W/cm^2. Harmonic generation is among the important new processes that take place at such high laser intensities, and has been recently investigated [2] and identified as a promising candidate for a coherent light source at very short wavelength.

The nonlinear orbit of an electron in an intense, plane-polarized, laser pulse can be the current source of two different processes. (i) Spontaneous harmonic Compton scattering [3]: This process is incoherent, so that the emitted power scales as the density, i.e., as the square of the plasma frequency ω_p^2. (ii) Collective forward harmonic Compton scattering: In a cold plasma the phases of the currents are fixed by the pump, so that a coherent harmonic wave, in phase with the nonlinear currents, can grow or decay. The efficiency of this phase-locked coherent harmonic generation was recently studied [2], and was shown to scale as ω_p^8.

The main issue of harmonic generation in condensed and gaseous media is the phase-velocity mismatch between the pump and the harmonic waves [4]. This is also the case for harmonic generation in a plasma, and the efficiency of the conversion of power to high harmonics is dramatically sensitive to this mismatch. What happens then, we demonstrate, is that the harmonic wave does not really grow at all; rather, there are amplitude oscillations at a saturated level, scaling with ω_p^2. Also, we demonstrate that, by modulating the density, linear growth can be accomplished with an efficiency scaling as ω_p^4 or as $\omega_p^{8/3}$.

Consider an intense pulse, such that the plasma period ω_p^{-1} is shorter than the pulse duration $\delta\omega^{-1}$. In this regime, each electron is displaced in the direction of the pulse as the pulse passes it by. Then, after a transient response, a nonlinear oscillation, driven by the wave, and modulated by the plasma collective effects, is set up. To analyze the nonlinear response, we use a Lagrangian description of the plasma, rather than a Eulerian one. This method has proven to be powerful in studying the generation of beat waves [5] and plasma wakes [6].

The nonlinearity parameter of an intense electromagnetic wave, with vector potential A, is eA/mc, where c is the velocity of light, and $-e$ and m are, respectively, the electron charge and mass. For ultraintense waves, $eA/mc > 1$, and the electron quiver velocity becomes relativistic, so that the polarization currents saturate at the value $\varepsilon_0\omega_p^2 mc/e$. On the other hand, the displacement currents increase with A, as $\varepsilon_0\omega^2 A$, where $\omega/2\pi$ is the wave frequency. Because of the saturation of the polarization currents, the wave dynamics is dominated by nonlinearity, and the normalized density can be used as an expansion parameter, with all orders in eA/mc kept.

The large value of the pump field causes electrons to respond with an effective mass [7] M, so that a density expansion scheme is valid only if $\omega_p^2/\omega^2 M \approx \omega_p^2/\omega^2 \times (eA/mc)$ is a small parameter. This parameter is small at high power even for frequencies below the linear cutoff, i.e., $\omega_p = \omega$; this is a very favorable circumstance, because efficient harmonic generation requires a dense plasma, and the smallness of this parameter at high power assures both the wave penetration and the validity of our analysis.

Intermediate, but important, calculations here include the nonlinear phase velocity of an intense wave and the electron renormalized effective mass, due to plasmon inertia.

Finally, to overcome the problem that we identify, we propose and analyze two phase-matching schemes, based on a resonant density modulation. In the following, except in the final part, we will use $e = m = c = \omega = 1$.

Consider an intense, plane-polarized, laser wave propagating along the z axis:

$$\mathbf{A}(z,t) = A(z,t)\cos[t - z + \phi(t)]\mathbf{e}_x, \qquad (1)$$

where $\phi(t)$ is a slowly varying phase $[d\phi/dt = O(\omega_p^2/A)]$, which accounts for the nonlinear dispersion of the phase velocity, and where $A(z,t)$ is a slowly varying envelope, whose dynamics is insignificant to the problem, provided that $\partial A/\partial z < A$, $\partial A/\partial t < A$, and $\delta\omega < \omega_p$.

Under such conditions, when an electron enters the pulse, it behaves essentially as in an infinite wave. The power transfer from the pump to the harmonic wave is diminished primarily by the phase-velocity mismatch, and, to a much lesser extent, by the group-velocity mismatch. This latter mismatch accounts for the imperfect overlapping of the two pulses, it takes place on a far longer time scale, and will be evaluated at the end of this Letter.

Each electron is described by its unperturbed position z_0, and follows a Lagrangian orbit, $h(z_0,t)=z(t)-z_0$, $x(z_0,t)$, about its rest position. To lowest order in ω_p^2/A, when collective plasma effects are neglected, the electrons perform the well-known "figure-8" motion [8]:

$$x=\frac{A}{M}\sin[M\tau(t,z_0)+\phi],$$
$$h=\frac{A^2}{8M^2}\sin[2M\tau(t,z_0)+2\phi], \quad (2)$$
$$t=z_0+M\tau+\frac{A^2}{8M^2}\sin[2M\tau+2\phi],$$

where we used the effective mass of the electron in an intense wave [7] $M=(1+A^2/2)^{1/2}$, and where $\tau(z_0,t)$ is the proper time. The sum of all the Lagrangian currents, $-(dx/dt)\delta[z-z_0-h(t,z_0)]$, gives the Eulerian current, which is the source term of the Maxwell equations. With the Lorentz gauge we obtain

$$\frac{\partial^2\mathbf{A}}{\partial z^2}-\frac{\partial^2\mathbf{A}}{\partial t^2}=\omega_p^2\int dz_0\,\delta(z-z_0-h(t,z_0))\frac{\mathbf{A}(z,t)}{\gamma(z_0,t)}=\omega_p^2\frac{\mathbf{A}}{\gamma(1+\partial h/\partial z_0)}, \quad (3)$$

where we have introduced the relativistic energy, $\gamma^2=1+(dh/d\tau)^2+(dx/d\tau)^2$, and used the conservation of the transverse canonical momentum, $\gamma dx/dt=A$. To evaluate $\gamma(1+\partial h/\partial z_0)$ as a function of the longitudinal momentum, $p=dh/d\tau$, we consider $h=\int^\tau p(u)du$, to obtain $\partial h/\partial z_0=p\,\partial\tau/\partial z_0$. Then, we note that all the electrons have the same orbit but translated in space and time in order to accommodate the delay between their excitation; thus, to calculate the global plasma response, we write the time variable as $t=z_0/V+\int^\tau\gamma(u)du$, where V is the slope of this space-time translation. At the front of the pulse, we have $V=1$, while, in the bulk, we take $V=1$ to lowest order, and $V=V^*$, the phase velocity, to higher order. This expression for t is then differentiated with respect to z_0, so that we obtain $\gamma(1+\partial h/\partial z_0)=\gamma-p/V$. To lowest order in the plasma effects, Eqs. (2) give $\gamma-p=M$, with the result that, although the microscopic Lagrangian currents contain the various harmonics of ω, the Eulerian current, to this order, contains only the fundamental.

The relativistic nonlinearity does manifest itself through an effective plasma frequency, ω_p^2/M. The nonlinear dispersion, described by the slowly varying phase ϕ, is then easily calculated with Eqs. (1)-(3) to get $d\phi/dt=\omega_p^2/2M$. The nonlinear phase velocity of the pump, V^*, can be written as $V^*=1+\omega_p^2/2M$.

Because of the cancellation between the relativistic velocity anharmonicity and the relativistic density oscillations, $\gamma(1+\partial h/\partial z_0)=M$, harmonic generation occurs only at the order ω_p^4/A^2, i.e., at the order ω_p^2/A for the Lagrangian orbits. In this order, the Coulomb interactions, responsible for the plasma collective effects, enter the Lorentz equations. The transverse x dynamics remains unaltered, but, on applying the Gauss theorem to the perturbed density, one finds an additional restoring force [9], proportional to the density, ω_p^2, and the displacement h. In addition to this force we have to take into account the first-order nonlinear dispersion which introduces a $d\phi/dt$ term. We have

$$\frac{dh}{d\tau}=p,\quad \frac{dp}{d\tau}=-\frac{A^2}{2}\sin[2(t-z)+2\phi]-\omega_p^2\gamma h, \quad (4a)$$

$$\frac{dt}{d\tau}=\gamma,\quad \frac{d\gamma}{d\tau}=-\frac{A^2}{2}\sin[2(t-z)+2\phi]\left[1+\frac{\omega_p^2}{2M}\right]-\omega_p^2 ph. \quad (4b)$$

These equations describe a perturbed nonlinear oscillator. To implement an ω_p^2/A expansion scheme, we must be careful to avoid secular terms [10]. On the basis of the unperturbed solution, Eqs. (2), we seek a first-order solution of the form

$$h\approx\frac{A^2}{8M^{*2}}\sin[2M^*\tau+2\phi]+O(\omega_p^2/A),$$
$$t\approx M^*\tau+\frac{A^2}{8M^{*2}}\sin[2M^*\tau+2\phi]+O(\omega_p^2/A). \quad (5)$$

The plasma effects add up higher-order harmonic terms, $O(\omega_p^2/A)$, and renormalize the nonlinear fundamental frequency M, to give a new effective mass, $M^*=M[1+O(\omega_p^2/A)]$, dressed by plasmons. Solving Eqs. (4) with Eqs. (5) leads to

$$\gamma-p=M^*\left[1+\omega_p^2\frac{A^2}{16M^{*3}}\cos(2M^*\tau+2\phi)\right],$$
$$t-z=M^*\tau+\omega_p^2\frac{A^2}{32M^{*3}}\sin[2M^*\tau+2\phi]. \quad (6)$$

The dressed effective mass is then obtained by demanding that there be no secular drift along the z direction. After some algebra, we obtain a relation between M and M^*, and to first order

$$M^*=M\left[1-\omega_p^2\frac{A^4}{64M^5}\right]. \quad (7)$$

This last result is not specific to the problem of harmonic generation, and is, in fact, quite general. Equation

(7) is the effective mass of an electron in an intense wave, when Coulomb collective interactions are taken into account. The plasma collective effects decrease the bare effective mass M, because the collective forces are restoring forces, $-\omega_p^2/h$, which oppose the driving fast oscillations, and screen A.

On the basis of Eqs. (3) and (6), we see that the transverse Eulerian polarization current has a third-harmonic component, of order ω_p^4/A^2, which is able to excite a third-harmonic wave,

$$\mathbf{a}(z,t)=a(t)\cos[3(t-z)+\varphi(t)]\mathbf{e}_x\,, \tag{8}$$

with the amplitude a and phase φ evolving on the slow time scale of the problem. Provided that $a<A$, the coupled Maxwell equations are

$$\frac{\partial^2\mathbf{A}}{\partial z^2}-\frac{\partial^2\mathbf{A}}{\partial t^2}=\frac{\omega_p^2}{M^*}\mathbf{A}-\frac{\omega_p^4 3A^2}{32M^{*4}}\mathbf{A}\,,$$
$$\frac{\partial^2\mathbf{a}}{\partial z^2}-\frac{\partial^2\mathbf{a}}{\partial t^2}=\frac{\omega_p^2}{M^*}\mathbf{a}-\frac{\omega_p^4 3A^2}{32M^{*4}}A\cos[3(t-z)+3\phi(t)]\mathbf{e}_x\,. \tag{9}$$

The first term on the right-hand side of the second equation describes the reactive dispersion due to the polarization currents, but with dressed electrons, whose inertia is given by Eq. (7). The second term drives the harmonic generation. On the basis of the first equation, one can calculate the nonlinear dispersion of A, $2d\phi/dt=\omega_p^2/M^*-\omega_p^4 3A^2/32M^{*4}$; thus, to this order, the nonlinear phase velocity of the pump is

$$V^*=1+\frac{\omega_p^2}{2M}-\frac{\omega_p^4 3A^2}{64M^4}+\frac{\omega_p^4A^4}{128M^6}\,. \tag{10}$$

The equations for the slowly varying amplitude and phase, a and φ, share some similarity with the Rosenbluth-Liu [5] equations for beat-wave generation, but here the detuning term is independent of the amplitude. Rather than using the variable $\varphi(t)$, where $d\varphi/dt=\omega_p^2/6M-\omega_p^4A^3\cos(\theta)/64M^4a$, it is more convenient to use the variable $\theta(t)=\varphi(t)-3\phi(t)$, so that we get

$$\frac{da}{dt}=-\omega_p^4\frac{A^3}{64M^4}\sin(\theta)\,, \tag{11a}$$

$$\frac{d\theta}{dt}=-\frac{4\omega_p^2}{3M}-\omega_p^4\frac{A^3}{64M^4}\frac{\cos(\theta)}{a}\,. \tag{11b}$$

The tendency to phase lock at large a, due to the second term on the right-hand side of Eq. (11b), is canceled by the phase-velocity mismatch, described by the first term. No phase locking occurs, so that instead of growing linearly with time, the amplitude oscillates. The orbits are arranged around the elliptic points ($a=\pm\omega_p^2 3A^3/256M^3$, $\theta=n\pi$). These equations are a Pfaffian system, and the first integral is $I=a^2+\omega_p^2(3A^3/128M^3)a\cos(\theta)$. Two classes of orbits are easily identified: circulating orbits ($I>0$, for large $|a|$) and trapped orbits ($I<0$, for small $|a|$). The equation describing either orbit is $[Ut]=-\arcsin(2UI-U^2a^2+2V^2)/2V(V^2+2UI)^{1/2}$, where $U=-4\omega_p^2/3M$ and $V=-\omega_p^4A^3/64M^4$. If the amplitude is large enough, the $I>0$ orbits can be approximated by $a=[-V/U]\cos(Ut)$.

It is interesting to note that Eqs. (11) can be mapped onto a Hamiltonian system. Let us define $P=a\sin(\theta)$, $Q=a\cos(\theta)$, and the time $s=-Ut/2$. Equations (11a) and (11b) can be obtained from the Hamiltonian $H=P^2+Q^2+2VQ/U$, describing an offset linear oscillator whose orbits are circles in P-Q phase space.

The length over which the harmonic generation is detuned is $l\approx(\omega_p/\omega)^3c/\omega_p$, which is too hard to set up in a plasma. In addition, in the regime $A\approx 1$, the conversion is only $P_3/P_1\approx 10^{-3}(\omega_p/\omega)^4$. We now proceed to show a method that overcomes the serious problem of the short detuning length, and moreover, does it with higher conversion efficiencies. A resonant density modulation is to be used in order to detrap the $I=0$ orbit.

Imagine a one-dimensional plasma media, with alternating, along z, high- and low-density sections. The laser pulse will induce harmonic generation in the active high-density sections, but the interaction with the low-density plasma will result only in a reactive phase shift between the pump and the harmonic. Choosing the width of the reactive low-density sections to compensate the phase mismatch due to the active one now restores linear growth.

After s steps, a reaches $s\omega_p^2 3A^3/128M^3$. Thus, the power conversion efficiency scales as $P_3/P_1\approx 10^{-3}s^2\times(eA/mc)^\alpha(\omega_p/\omega)^4$, where the α exponent is 4, if $A<1$, 0, if $A\approx 1$, and -2, if $A>1$. Strong density modulations might be set up by, for example, the laser ablation of a multiple-layered media, or a nonlinear plasma wave.

However, such a strong resonant modulation may not be necessary, and it would be more convenient to use a low-frequency, small-amplitude density wave in an homogeneous plasma, such as, for example, a long-wavelength ion-acoustic wave. Actually, even a weak modulation has a dramatic effect which can be studied within the framework of the previous model. Because the density is modulated, $\delta n/n=\varepsilon\sin(\Omega t)$, with $\varepsilon<1$ and $\Omega<\delta\omega<\omega_p<\omega$, we substitute $\omega_p^2[1+\varepsilon\sin(\Omega t)]$ for ω_p^2 in Eqs. (11). As soon as the amplitude becomes larger than $\omega_p^2/256$, the second term on the right-hand side of the phase equation is negligible compared to the first one, and the amplitude equation becomes

$$\frac{da}{dt}=-\frac{\omega_p^4A^3}{64M^4}[1+2\varepsilon\sin(\Omega t)]\sum_N J_N(-4\varepsilon\omega_p^2/3M\Omega)\sin\left[\frac{4\omega_p^2}{3M}t+N\Omega t+N\frac{\pi}{2}\right]. \tag{12}$$

The amplitude is thus driven by a sum of oscillating terms, and, if one of these oscillations is resonant, it induces a secular linear growth. When N is odd the resonance condition is

$$N\Omega = +4\omega_p^2/3M = 0\,. \tag{13}$$

The associated resonant term quickly dominates the other bounded components, and we can average out the oscillating part of a, to study the secular part of the third-harmonic amplitude, $\langle a\rangle$. Taking the asymptotic expansion of $J_N(N\varepsilon)$ in the resulting equation, $d\langle a\rangle/dt = J_N(N\varepsilon)\omega_p^4 A^3/64M^4$, we obtain

$$\langle a\rangle = (18\times 10^{-3})A^3 M^{-11/3}\omega_p^{10/3}\varepsilon^{-1/3}\Omega^{1/3}\mathrm{Ai}[1.52M^{-2/3}\omega_p^{4/3}\varepsilon^{-1/3}\Omega^{-2/3}(1-\varepsilon)]t\,, \tag{14}$$

where Ai is the Airy function, and one can take the typical value $\varepsilon^{-1/3}\mathrm{Ai}\approx 0.1$. The efficiency scaling becomes $P_3/P_1 \approx 10^{-5}(eA/mc)^{\alpha}(\omega_p/\omega)^{20/3}(\Omega/\omega)^{2/3}(\omega t)^2$. The exponent α is now $-10/3$ if $A > 1$.

So far, we have solved a nonlinear initial value problem; i.e., an infinite plane wave $A(t)$ is turned on adiabatically in an infinite plasma, and we have studied the associated response $a(t)$. This solution is relevant to the corresponding initial boundary value problem, i.e., the study of the $a(z,t)$ response to an $A(z,t)$ pump, provided that the wave-packet overlapping problem, due to group-velocity mismatch, takes place on a long time scale. A departure from overlapping appears after a time $c\delta\omega^{-1}/2\omega(\partial v_g/\partial\omega)\approx\delta\omega^{-1}\omega^2/\omega_p^2$. This time is to be compared with the time needed to complete one generation cycle, $\approx\omega/\omega_p^2$. Since $\omega\gg\delta\omega$ for a wave packet, the overlapping mismatch comes into play for a time scale far longer than all the other processes. Nevertheless, this overlapping does limit the maximum number of steps in the strong modulation scheme, as well as the time in Eq. (14) ($t_{\max}=\delta\omega^{-1}\omega^2/\omega_p^2$).

To assess more carefully the potential of phase matching with a small amplitude wave, consider an ion-acoustic wave. The quasineutral, low-frequency, long-wavelength dispersion relation for a wave with $\delta n/n = \varepsilon\sin(\Omega_s t - K_s z)$ is $\Omega_s = K_s(T/m_i)^{1/2}$, where T is the electron temperature and m_i is the ion mass. Under typical laboratory conditions, $\Omega_s < \omega_{pi} < \delta\omega < \omega_{pe} < \omega$, the pulse length is smaller than the ion-acoustic wavelength, so that, Eq. (12) applies, provided that we use the effective modulation frequency, $\Omega = \Omega_s v_g/(T/m_i)^{1/2}$, seen by the pulse. We do not need to know the exact expression of v_g, the nonlinear group velocity of the pulse, because $v_g = 1 + O((\omega_p/\omega)^2)$ and we are working to the lowest relevant order in ω_p/ω. Thus the equivalent frequency to be used in Eq. (12) is $\Omega = \Omega_s/(T/m_i)^{1/2}$. Taking the group velocity mismatch as the ultimate limitation, and considering the regime $A\approx 1$, we obtain $P_3/P_1 \approx 10^{-2}[T/(1\text{ eV})]^{1/3}(\omega_p/\omega)^{8/3}(\Omega_s/\omega)^{2/3}(\omega/\delta\omega)^2$.

To summarize, we have set up, discussed, and solved, the equations for relativistic third-harmonic generation in a plasma. The important problem of phase mismatch has been identified and addressed. The nonlinear phase velocity, the renormalized electron mass, and the conversion efficiency have been calculated. Two simple modulation schemes, to overcome saturation, have been proposed and analyzed. Using plasma for third-harmonic generation has advantages over using other nonlinear condensed or gaseous media; plasmas do not suffer material breakdown at high intensities, and can convert radiation over a very broad range of frequencies.

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