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Third harmonic generation by an obliquely incident laser on a vacuum – plasma interface

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Abstract A high power laser obliquely incident on a plasma produces a third harmonic component in the reflected component The process is sensitive to the plasma frequency and the angle of incidence. At higher plasma densities, the efficiency peaks at smaller angles of incidence and as the plasma becomes rarer, the efficiency peaks at higher angles of incidence. The efficiency also drops with plasma frequency In gaseous plasma, the efficiency is higher by more than an order as compared with solid-state plasmas.

Keywords : Third harmonic generation, laser plasma interaction

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1. Introduction

Harmonic generation of intense laser radiation in plasmas is an important nonlinear process[1-4]. In a uniform plasma, one observes the generation of odd harmonics[5]. The nonlineartity arises *via* ponderomotive force and relativistic mass effect. The laser of frequency ω_1 induces an oscillatory velocity v_1 of electrons at ω_1 frequency and exerts a ponderomotive force $F_{2\omega_1}$ at $2\omega_1$. $F_{2\omega_1}$ induces density oscillations at $2\omega_1$ which couple with the oscillatory velocity v_1 to produce the third harmonic current and the third harmonic electromagnetic radiation. The electron mass also contains a component at $2\omega_1$ and results in the generation of a third harmonic velocity leading to the third harmonic generation. In inhomogeneous plasma, one may generate second harmonic and other even harmonics as the electron density may have an oscillatory component at ω_1. This process could become quite efficient near the critical layer where laser may generate a Langmuir wave *via* linear mode conversion or *via* decay instability producing a pair of Langmuir and ion acoustic waves. The Langmuir wave thus generated, beats with the laser to produce second harmonic generation.

In recent years, there has been considerable interest on harmonic emissions from laser irradiated overdense plasma or metallic targets. Watts *et al* [6] have reported generation of harmonics using a 0.7-1.0ps, 1.053μm, 10^{19}W/cm^2 p-polarized laser over a fused silica slab set at 45° angle of incidence with the laser beam. Modulation of the harmonic emission spectrum with a periodicity of 2 to 4 harmonics is observed at higher laser intensities. The dynamics of the critical surface can be inferred from the shape of the harmonic spectrum. Teubner *et al* [7] have reported the harmonic emission from thin solid carbon and aluminum foils, irradiated by 150fs long, Ti:Sapphire laser pulses at λ = 395nm and peak intensities of a few 10^{18}W/cm^2. In addition to the harmonics emitted from the front side in the specular direction, they observed harmonics up to 10th order, including the fundamental from the rear side in the direction of the incident beam, while the foil is still strongly over dense. Von-der-Linde and Rzázewski[8] have given a theoretical formulation for high order optical harmonic generation from solid surface assuming the sharp plasma - vacuum boundary as an oscillating mirror. It was shown that the generation of reflected harmonics can be interpreted as a phase modulation experienced by the light, upon reflection from the oscillating boundary. The modulation sidebands of the reflected frequency spectrum correspond to odd and even harmonics of the laser frequency. Norreys *et al* [9] have observed harmonics up to 75th order using 2.5ps, 10^{19}W/cm^2 laser over CH plastic-coated aluminum targets. The harmonic generation was independent of s or p polarization.

In this paper, we study third harmonic generation by an obliquely incident laser on a step boundary between vacuum and uniform plasma. The plasma density could be either underdense ($n_0^0 < n_{cr}$, n_{cr} , ncr is the critical density) or overdense ($n_0^0 > n_{cr}$). In the underdense region, laser gets partly transmitted into the plasma and gives rise to nonlinear interaction. However, at angles of incidence greater than the critical or at supercritical densities, the laser gets totally reflected back into free space, nevertheless, the evanescent wave near the surface possesses large enough amplitude to produce harmonics. The laser induces an oscillatory velocity $\boldsymbol{v}_\omega$ to the plasma electrons, which couples with the transmitted magnetic field of the laser $\boldsymbol{B}_T$ to produce a second harmonic ponderomotive force $\boldsymbol{F}_{p2}$. The plasma density perturbation n_2 n_2 due to $\boldsymbol{F}_{p2}$ couples with the electron velocity $\boldsymbol{v}_\omega$ to produce a third harmonic current $\boldsymbol{J}_3$ which gives rise to third harmonic electromagnetic radiation in the reflected component. In Section 2, we obtain the third harmonic current and solve the wave equation to obtain the third harmonic field. In Section 3, we obtain the efficiency of the process and discuss our results.

2. Third harmonic field

Consider a vacuum-plasma interface at $x = 0, x < 0$ being vacuum and $x > 0$ being a uniform plasma of density n_0^0 . A semiconductor or a metal could replace the plasma. A laser is incident on the interface at an angle θ (c.f. Figure 1),

$$\boldsymbol{E}_i = E(\hat{z} - \hat{z}\tan\theta)e^{-i\left(\omega t - \frac{\omega}{c}\cos\theta\, x - \frac{\omega}{c}\cos\theta\, z\right)}. \tag{1}$$

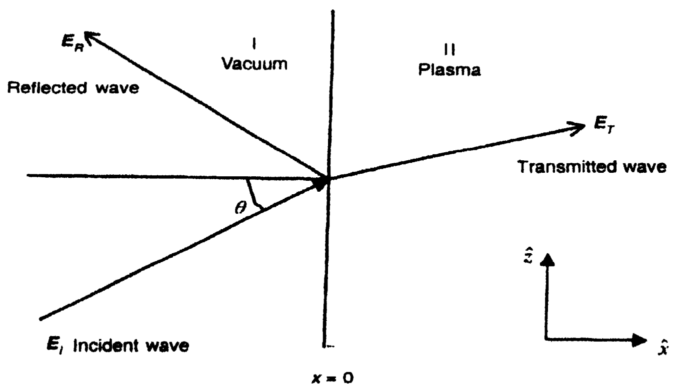

Figure 1. Geometry of the process.

The reflected and transmitted waves can be written as

$$\boldsymbol{E}_R = RE(\hat{z} + \hat{x}\tan\theta)e^{-i\left(\omega t - \frac{\omega}{c}\cos\theta\, x - \frac{\omega}{c}\cos\theta\, z\right)}, \tag{2}$$

and

$$\boldsymbol{E}_R = TE(\hat{z} + \hat{x}\frac{i\sin\theta}{\alpha c/\omega})e^{-i\left(\omega t - i\alpha x - \frac{\omega}{c}\sin\theta\, z\right)} \tag{3}$$

respectively. Here, R and T are amplitude reflection and transmission coefficients respectively. $\alpha = \left[k_z^2 - \left(\omega^2 - \omega_p^2\right)/c^2\right]^{1/2}$, $k_z = (\omega/c)\sin\theta$, $\omega_p = \left(4\pi n_0 e^2/m\right)^{1/2}$, $-e$ and m are the electronic charge and mass, respectively.

On demanding the continuity of E_z and εE_x at x = 0 where $\varepsilon = \varepsilon_r - \omega_p^2/\omega^2$, ε_r being lattice permittivity in the case of a metal / semiconductor and for plasmas $\varepsilon_r = 1$, we obtain

$$R = \frac{\tan\theta\alpha c/\omega - \varepsilon^2\sin^2\theta}{\tan\theta\alpha c/\omega - i\varepsilon\sin\theta}, \tag{4}$$

and

$$T = \frac{2\tan\theta\alpha c/\omega}{\tan\theta\alpha c/\omega - i\varepsilon\sin\theta}. \tag{5}$$

Using the Maxwell's equation $\nabla\times\boldsymbol{E} = -(1/c)(\partial\boldsymbol{B}/\partial t)$, the transmitted magnetic field $\boldsymbol{B}_T$ can be written as

$$\boldsymbol{B}_T = \hat{y}iET\left(\frac{\omega}{\alpha c}\sin^2\theta - \frac{\alpha c}{\omega}\right)e^{-i\left(\omega t - i\alpha x - \frac{\omega}{c}\sin\theta\, z\right)}. \tag{6}$$

On solving the equation of motion $m(dv/dt) = -eE - (e/c)v\times\boldsymbol{B}$, using eq.(3), we obtain the electron velocity at $(\omega,\boldsymbol{k})$ as

$$\boldsymbol{v}_\omega = \frac{e\boldsymbol{E}_T}{mi\omega}. \tag{7}$$

$\boldsymbol{v}_\omega$ beats with $\boldsymbol{B}_T$ to produce the ponderomotive force $\boldsymbol{F}_{p2}$ at $(2\omega, 2\boldsymbol{k})$:

$$\begin{aligned}\boldsymbol{F}_{p2} &= -\frac{e}{2c}v_\omega\times\boldsymbol{B}_T \\ &= -\frac{e^2E^2T^2}{2m\alpha c}\left(\frac{\omega}{\alpha c}\sin^2\theta - \frac{\alpha c}{\omega}\right)\times \\ &\quad\left(-\hat{x} + \hat{z}\frac{i\sin\theta}{\alpha c/\omega}\right)e^{-2i\left(\omega t - i\alpha x - \frac{\omega}{c}\sin\theta z\right)}.\end{aligned} \tag{8}$$

On solving eq.(8), we obtain the electron velocity $v_{2\omega}$ at $(2\omega,2\boldsymbol{k})$ as

$$\boldsymbol{v}_{2\omega} = -\frac{\boldsymbol{F}_{p2}}{2mi\omega}. \tag{9}$$

Using eq.(9) in the continuity equation $\partial n_2/\partial t + \nabla.\left(n_0^0 \boldsymbol{v}_{2\omega}\right) = 0$, we obtain the perturbed electron density n_2 as

$$n_2 = \frac{n_0^0 (\nabla.\boldsymbol{v}_{2\omega})}{2i\omega}. \tag{10}$$

The third harmonic nonlinear current density $\boldsymbol{J}_3^{NL}$ is given by

$$\boldsymbol{J}_3^{NL} = -\frac{1}{2} n_2 e \boldsymbol{v}_{\omega}. \tag{11}$$

Using eq.(11) in the wave equation for the third harmonic field $\boldsymbol{E}_3$, we get

$$\nabla^2 \boldsymbol{E}_3 + \frac{9\omega^2}{c^2}\varepsilon_3 \boldsymbol{E}_3 = F\alpha^2\left(z + \frac{i\sin\theta}{\alpha c/\omega}\hat{x}\right)e^{-3i\left(\omega t - i\alpha x - \frac{\omega}{c}\sin\theta z\right)}, \tag{12}$$

where $\varepsilon_3 = \varepsilon_r - \omega_p^2/9\omega^2$,

$$F = \frac{3}{8}\frac{\omega_p^2}{\omega^2}\frac{v_1^2}{c^2} ET\left[\frac{\omega^2}{\alpha^2 c^2}\sin^2\theta - 1\right]^2, \text{ and } v_1 = \frac{eET}{m\omega}.$$

On solving eq.(12) for x-component, we obtain

$$E_{3x} = \left[A_1 e^{ik_{3x}x} + Qe^{3ik_x x}\right]e^{-i(3\omega t - 3k_z z)}, \quad x > 0;$$

$$= A_2 e^{-ik'_{3x}x} e^{-i(3\omega t - 3k_z z)}, \quad x < 0; \tag{13}$$

where $k'^2_{3x} = 9\omega^2/c^2 - 9k_z^2$, $k_{3x}^2 = \left(9\omega^2/c^2\right)\varepsilon_3 - 9k_x^2$, $k_x = i\alpha$, and $Q = \dfrac{F\,\omega i\sin\theta}{\alpha\, c}$.

Using eq.(13) in the Gauss law $\nabla.\boldsymbol{E}_3 = 0$ and solving, we obtain the z-component of $\boldsymbol{E}_3$ as

$$E_{3z} = \left[\frac{k_{3x}}{3k_z}A_1 e^{ik_{3x}x} + \frac{k_x}{k_z}Qe^{3ik_x x}\right]e^{-i(3\omega t - 3k_z z)}, \quad x > 0;$$

$$= -\frac{k'_{3x}}{3k_z}A_2 e^{-ik'_{3x}x} e^{-i(3\omega t - 3k_z z)}, \quad x < 0. \tag{14}$$

Using eqs.(13) and (14) in the Maxwell's equation $\nabla \times \boldsymbol{E} = -(1/c)(\partial \boldsymbol{B}/\partial t)$ and on solving, we obtain the y-component of third harmonic magnetic field $\boldsymbol{B}_3$ as

$$B_{3y} = -\frac{ck_z}{\omega}\left\{\left(\frac{k_{3x}^2}{9k_z^2} - 1\right)A_1 e^{ik_{3x}x} + \left(\frac{k_x^2}{k_z^2} - 1\right)Qe^{3ik_x x}\right\}e^{-i(3\omega t - 3k_z z)},$$

$$x > 0;$$

$$= -\frac{ck_z}{\omega}\left(\frac{k'^2_{3x}}{9k_z^2} - 1\right)A_2 e^{ik'_{3x}x} e^{-i(3\omega t - 3k_z z)}, \quad x < 0. \tag{15}$$

Applying the boundary conditions $E_{3z}\big|_I = E_{3z}\big|_{II}$ and $B_{3y}\big|_I = B_{3y}\big|_{II}$, at $x = 0$, we get the third harmonic field amplitude A_2 in the reflected component:

$$A_2 = \frac{3Q\left[-3k_x^2 k_{3x} + 3k_{3x}k_z^2 + k_x\left(k_{3x}^2 - 9k_z^2\right)\right]}{\left(k_{3x} + k'_{3x}\right)\left(k_{3x}k'_{3x} - 9k_z^2\right)}. \tag{16}$$

3. Efficiency and discussion

The ratio of the third harmonic reflected wave power $P_3 = c|A_2|^2/8\pi\cos^2\theta$ to the incident wave power $P_0 = cE^2/8\pi$ is

$$\frac{P_3}{P_0} = \left|\frac{81}{64}\frac{\omega^2}{\alpha^2 c^2}\frac{\omega_p^4}{\omega^4}\frac{v_1^4}{c^4}\left(\frac{\omega^2\sin^2\theta}{\alpha^2 c^2} - 1\right)^4 \right.$$

$$\left.\frac{3k_{3x}k_z^2 + k_x\left(k_{3x}^2 - 9k_z^2\right) - 3k_x^2 k_{3x}}{\left(k_{3x} + k'_{3x}\right)\left(k_{3x}k'_{3x} - 9k_z^2\right)}T^2\tan^2\theta\right|. \tag{17}$$

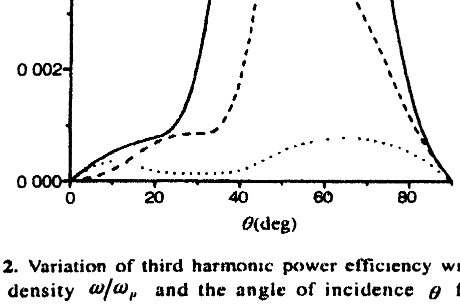

Figure 2. Variation of third harmonic power efficiency with normalized plasma density ω/ω_p and the angle of incidence θ for $\varepsilon_r = 1$ and $v_1/c = 0.01$.

In Figures (2) and (3), we have shown the variation of third harmonic power with the normalized plasma frequency ω/ω_p and the angle of incidence θ for ε_r =1 and 10, respectively for v/c = 0.1. The process is sensitive to the plasma frequency ω_p

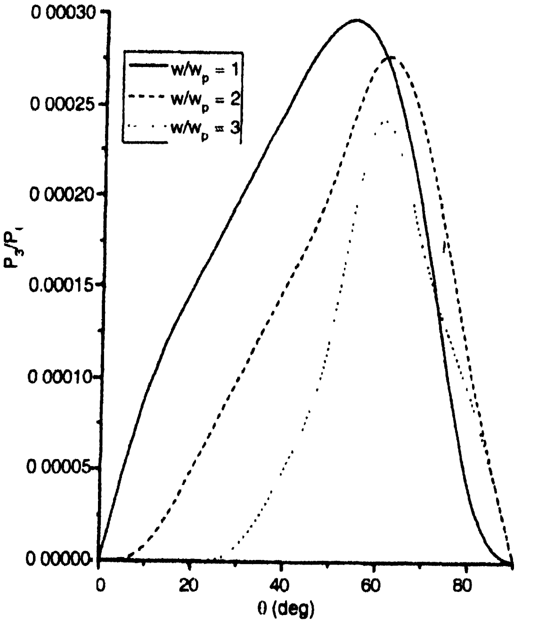

Figure 3. Variation of third harmonic power efficiency with normalized plasma density ω/ω_p and the angle of incidence θ for ε_r =10 and v_t/c = 0.01.

and the angle of incidence. At higher plasma densities, the efficiency peaks at smaller angles of incidence and as the plasma becomes rarer, the efficiency peaks at higher angles of incidence. The efficiency also drops with plasma density. In gaseous plasma, the efficiency is higher by more than an order as compared with solid-state plasmas. The frequency factor ω/ω_p signifies the resistance of the medium to perturbations of the electronic charge density. It appears that a slightly overdense plasma is preferable in higher order harmonic generation. One of the limitations of the proposed model is that in typical experimental situations, it is difficult to maintain a step like density discontinuity during the interaction with the laser pulse. More likely, one is dealing with a plasma-vacuum boundary having experienced some broadening during the interaction. One would expect that the simple model discussed here, ceases to be a good picture of harmonic generation when the plasma - vacuum boundary is spread out over a distance comparable with the electron excursion. The proposed work has applications in diagnostics and wavelength conversion.

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