Single Particle Analysis of Second and Third Harmonic Generation on Solid Surfaces

S. Varró[1] and K. Gál[2]

[1]*Research Institute for Solid State Physics and Optics, H-1525 Budapest, P.O.Box 49, Hungary*
[2]*Department of Experimental Physics, University of Szeged, Dóm tér 9, H-6720 Szeged, Hungary*

Abstract

In the last few years it has been demonstrated [1] that second and third harmonics generated in laser plasmas produced on solid surfaces preserve the polarization of the incident laser beam. Theoretical works published so far [1,2,3,4] predict that even-harmonics should be p-polarized for p- and s-polarized laser beams as well. In the present analysis of harmonic generation at surfaces having large density gradients a possible explanation is given for the experimentally observed polarization dependence of the harmonics on that of the fundamental beam.

Introduction

A widely used method for generating high-order harmonics is based on the interaction of high power ($I>10^{16}$W/cm^2) lasers with solid targets. Recent experiments demonstrated, that ultrashort laser pulses produce even and odd harmonics for both laser polarisations [1]. Some theories [1,2,3,4] were developed to explain the polarisation dependence of harmonic generation. These theories predict that only odd harmonics will be s-polarised for s-polarised incoming laser beam. The even harmonics will be p-polarised no matter what the incident laser polarisation is. These theories contradict the experiments in case of even harmonics produced by s-polarised incoming beams. In the model to be presented below we employ the theory of reflection and refraction to understand how the electric and magnetic fields behave in overdense plasmas. It is stressed that the magnetic term in the Lorentz force can be of importance in such a medium. We have solved exactly the nonrelativistic equation of motion of the electrons in the plasma by taking into account the complete Lorentz force due to the penetrating electromagnetic field. On the basis of these solutions high harmonic components of the electron current were obtained which are responsible for the appearance of s-polarised secondary fields for s-polarized incoming radiation.

Electromagnetic field configuration at the vacuum-plasma boundary

Let us consider an obliquely incident monochromatic electromagnetic wave on a sharp vacuum-plasma interface.

$$\vec{E}_i = \vec{E}_0 e^{i(\vec{k}\vec{r}-\omega t)} \tag{1}$$

The Fresnel equations [5] split automatically to two separate sets of equation for s- respectively for p-polarised monochromatic light [see Fig.1]. We concentrate our attention to the s-polarised laser light, for which the contradiction between the earlier theories and the experiments appears.

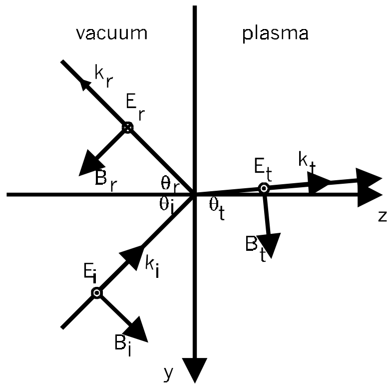

Figure 1. The geometry of reflection and refraction

On [Fig.1] k denotes the wave vector and θ the angle. E is the electric field vector and B is the magnetic induction vector. In both cases the index i refers to the quantities that characterise the incident, the index r the reflected and the index t the transmitted laser light respectively.

Our model starts with the study of the refraction from a sharp vacuum-plasma interface (Fresnel formulae). In case of an s polarised incident laser light in complex notation the electric and magnetic fields have the following form for a plasma whose dielectric function is negative.

$$\vec{E}_t = \sqrt{\frac{4\cos^2\vartheta_i}{1-\varepsilon}}E_0 e^{i\left(ArcTan\left(-\frac{\sqrt{\sin^2\theta_i-\varepsilon}}{\cos\theta_i}\right)-\omega t\right)}e^{-k_i\sqrt{-\varepsilon}z}\hat{x} \tag{2}$$

$$\vec{B}_t = \sqrt{\frac{-4\varepsilon\cos^2\vartheta_i}{1-\varepsilon}}E_0 e^{i\left(ArcTan\left(-\frac{\sqrt{\sin^2\theta_i-\varepsilon}}{\cos\theta_i}\right)-\omega t+\frac{\pi}{2}\right)}e^{-k_i\sqrt{-\varepsilon}z}\hat{y} \tag{3}$$

For simplicity we assume a linear density dependence on the steepness s and put this density n parametrically into the Fresnel formulae $n = n_{cr}\left(1 - s\,{}^{z}\!/_{\lambda}\right)$ [Fig.2]. The dielectric function of laser produced plasmas is determined by the density profile $\varepsilon = 1-\frac{n}{n_{cr}}$, where n_{cr} represents the critical density, the density where the plasma frequency ω_p gets the value of the fundamental laser frequency ω.

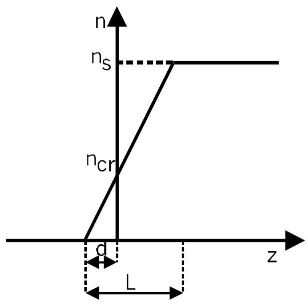

Figure 2. The density profile

Of course, we are well aware of the fact that the Fresnel formulae we are using are only a crude approximation for the transmission and reflection coefficient for a continuously varying dielectric function. This more complex treatment will be the subject of our future work.

To determine the steepness of the density profile we must define the following quantities. The plasma scale length is defined as the scale length of the inhomogeneity: $L = n\left(\frac{\partial n}{\partial z}\right)^{-1}$. The scale length of the underdense region is $d = n_{cr}\left(\frac{\partial n}{\partial t}\right)^{-1}$. The skin depth is the depth where the field quantities will decay to e-th part of their maximum value and it can be determined $d_{skin} = \left(\frac{3}{4\pi}\right)^{2/3}\frac{1}{s^3}\lambda$. We denoted with n_s the solid state density. The steepness of the density profile is calculated from the plasma scale length multiplied with the ratio between the solid-state density and the critical density. We have chosen a very steep density profile, so the underdense region will be so thin, that we can neglect all the processes which take place here if d_{skin}>>d. The electron's amplitude is a few per cent of the wavelength and the skin depth must be larger than this displacement. Considering the ratio between the solid-state density and the critical density of order of 40 we get for the plasma scale length (L) the following conditions: $0.1\lambda<L<<10\lambda$. Further on we assume a plasma scale length of 0.2λ. This way the dielectric function becomes: $\varepsilon=-s\ z/\lambda$. The index of refraction becomes complex in the region where the basic excitation processes take place ($z/\lambda>0$) and its absolute value gets very high. If the density profile is almost step like profile, then the angle of refraction is very small and for simplicity we take it 0.

The source of high harmonics

In the overdense region the phase difference between the magnetic and electric fields is $\pi/2$ and their amplitudes will differ. In [Fig.3] we can see the ratio between the two amplitudes B_{ta}/E_{ta}.

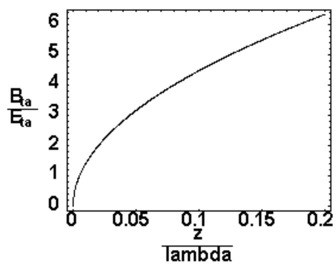

Figure 3 . The ratio between the magnetic and electric fields amplitudes

Even in the non-relativistic case (v/c<<1) the smallness of the term $(v/c)\times B_t$ is compensated by the smallness of the E_t. By solving the Newton equation we get high harmonic components of the velocity. The electron will oscillate in the x-z plane and it will have two velocity components. Both components will be proportional with the fundamental term $\frac{e}{m}\frac{1}{\omega}E_{ta}$ and the amplitudes of different high harmonic components will contain higher order Bessel functions, whose magnitude is governed by the ratio $\omega_L/\omega = (B_{ta}/E_0)\mu$, where μ is the well known intensity parameter: $\mu = 10^{-9}\lambda\sqrt{I}\left[\mu m \cdot W/cm^2\right]$ and ω_L denotes the local Larmor frequency. This components will contain high harmonic components for $I>10^{16} W/cm^2$. In this case the amplitudes of the different components will be comparable. This way the velocity component parallel to the wavenumber vector gives rise to an electron-plasma wave. The other component being parallel to the electric vector, gives source term for the reflected light, with high harmonic components. The wave equation of the harmonics in this case is:

$$\Delta\vec{E}_s - \frac{1}{c^2}\frac{\partial \vec{E}_s}{\partial t^2} - \nabla\left(\nabla\cdot\vec{E}_s\right) = \frac{4\pi}{c^2}\frac{\partial j_x}{\partial t}\hat{x} \quad (4)$$

where E_s represents the electromagnetic field vector of the reflected laser light. This secondary field vector will be parallel with the v_x component of the electron's velocity, so the reflected secondary field vector will be s polarised for s polarised incident laser beam.

Conclusions

In the present work we studied the refraction of the s polarised electromagnetic field. The main source of harmonic generation is considered to be the electron's motion in the overdense plasma due to the penetrating electromagnetic field. In such a field the electron's oscillations are governed by the *whole* Lorentz force, even in the nonrelativistic regime. The nonlinearities in the overdense region is getting considerable if the intensity exceeds a well defined threshold, which scales with the intensity parameter μ. The index of refraction contains implicitly the collective response of the plasma. In case of a 248nm KrF laser the intensity threshold is of the order of 10^{16}W/cm^2. Making some approximation we get for I=10^{16}W/cm^2 a secondary field intensity of 10^{10}W/cm^2 for the second harmonic which will decrease with the order.

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