Plasma Modulation of Harmonic Emission Spectra from Laser-Plasma Interactions

T. J. M. Boyd[1] and R. Ondarza-Rovira[2]

[1]Centre for Physics, University of Essex, Wivenhoe Park, Colchester CO4 3SQ, United Kingdom
[2]Instituto Nacional de Investigaciones Nucleares, A.P. 18-1027, México 11801, Distrito Federal, Mexico
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We report results from particle-in-cell simulations of the interaction of intense laser light with overdense plasma designed to examine the effects of plasma waves generated by pulses of fast electrons on high-order harmonic emission from the plasma. We show that the emission spectrum is modulated at the plasma frequency and identify combinations of parameters and circumstances favorable for modulation. In particular, the observed modulation is shown to depend not only on the chosen plasma electron density and intensity of the incident light but on the density profile and pulse shape.

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The generation of multiple harmonics of the incident light in laser-plasma interactions is a topic that has proved to be of enduring interest, not only because of its intrinsic importance but for its potential as a diagnostic of conditions at the target surface and as a means of generating intense coherent pulses at short wavelengths. Distinct mechanisms are responsible for harmonic generation across different intensity ranges and target characteristics. For gas-jet targets, harmonics up to several hundred are generated through the nonlinear response of atomic electrons. However, their intensities are limited by field ionization. No such constraint applies to harmonic generation when intense laser pulses are focused on the surface of solid targets. This was first observed in experiments with nanosecond pulses [1] and, subsequently, detected for picosecond pulse lengths [2]. Conditions in short-pulse experiments ensured two prerequisites essential for high-order harmonic generation, namely, an electron density much greater than the critical density n_c combined with high brightness of the source. Using light of intensity I and wavelength λ_L, at values of $I\lambda_L^2$ in the range 10^{18}–10^{19} W cm^{-2} μm^2 Norreys *et al.* [2] detected harmonics up to the 75th (with conversion efficiency of 10^{-6}) for 1.053 μm wavelength light interacting with a solid target. Subsequently, harmonic generation using pulse lengths of 100 fs has been observed [3]. Such short pulses with no significant prepulse create plasma with extremely steep density gradients at the plasma surface.

In the absence of any convincing theoretical model for high-order harmonic generation in this parameter regime, particle-in-cell (PIC) simulations have proved invaluable in understanding and characterizing the emission. Gibbon [4] used PIC simulations to identify the physical mechanism underlying harmonic generation, showing that the current sources generating the harmonics are localized just within the overdense plasma boundary. Gibbon showed empirically that the efficiency of high-order harmonic generation η_m scales as $\eta_m \sim 9 \times 10^{-5}(I_{18}\lambda_L^2)^2 \times (m/10)^{-5}$, where I_{18} denotes light intensity in units of 10^{18} W cm^{-2}, with λ_L in microns, and m is the harmonic number. Useful as this scaling is, it makes no allowance for any dependence of harmonic power levels on the plasma electron density which can affect levels of emission [5]. Alternative scaling laws have been proposed by Baeva *et al.* [6] on the basis of ultrarelativistic similarity theory, independent of details of the interaction. These authors report a scaling in which the spectrum decays with harmonic number as $m^{-8/3}$ with a cutoff at $m^* = \sqrt{8\alpha}\gamma_{\max}^3$, where α is of order 1 and γ is the maximum value of the relativistic factor associated with the motion of the plasma surface.

In contrast, the work described here deals with a regime that is only modestly relativistic but is concerned with ways in which the harmonic spectrum may reflect plasma effects. Boyd and Ondarza-Rovira [7] showed that the harmonic spectrum is affected by plasma line emission, seen in a number of experiments [8]. Moreover, they reported for the first time an unexpected feature in the spectrum, attributing it to the effect of the plasma wave on neighboring harmonic lines. This so-called "combination line" is in effect a footprint of the modulation subsequently observed experimentally [3,9]. In this Letter, we investigate these modulation effects using a model in which the source of the modulation is identified as plasma waves generated in the target plasma by pulses of reentrant electrons.

It is important at the outset to underline a limitation of our, or for that matter most other, PIC simulations of harmonic generation. We do not attempt to model the ionization of target material but treat instead the interaction of a laser pulse of length τ_p with a plasma slab of initially uniform density, apart from an interface at the front end. This vacuum-plasma interface is characterized by a density profile with scale length Δ that is some prescribed fraction of a wavelength across the ramp. The $1\frac{1}{2}$D PIC code we use embeds the Bourdier technique [10] to allow for oblique incidence. The simulation plasma can be up to ten wavelengths in extent. Vacuum gaps extend from both the front of the ramp and the planar rear surface of the plasma to the walls of the simulation box to allow for particle and wave propagation. The initial electron temperature was chosen to be 100 eV, and we used Gaussian pulses of variable length and profile. The normalized quiver momentum $a_0 \sim 0.85(I_{18}\lambda_L^2)^{1/2}$ lies in the range 0.5–2.0 with electron densities between $10n_c$ and $64n_c$. The reflected emission spectra showing the relative strength of the different oscillation modes is determined from the harmonic content of the reflected electric field at the vacuum gap, normalized to the fundamental.

Our earlier work [5,7] established that plasma line emission was present alongside the harmonics of the incident light. The source of the plasma emission was identified as plasma waves excited in the supradense plasma by jets of Brunel-accelerated electrons formed when p-polarized light is obliquely incident on the plasma surface [11]. The reentrant electrons bunch to generate plasma oscillations at the surface before penetrating the overdense plasma. Plasma oscillations at the target surface couple efficiently to the radiation field. In Fig. 1, we reproduce the effect for a plasma with slab electron density $n_e = 20n_c$. Figure 1(a) plots electron density contours across a $0.1\lambda_L$ section of the plasma target ($3\lambda_L$ wide in this simulation). The striations correspond to the pulses of fast electrons that excite plasma oscillations. A comparable density bunching effect and corresponding excitation of plasma oscillations have been reported by Quéré *et al.* [12] but in this case for a foil only $\lambda_L/15$ thick.

In Fig. 1(b), we present a Fourier analysis of the numbers of electrons at a point just inside the plasma for the parameters used in Fig. 1(a). Envelopes are sketched over the Fourier components of the density showing 3 cycles across a span of 14 harmonics, suggestive of modulation at the plasma frequency $\omega_p = 4.5\omega_L$, where ω_L is the frequency of the incident light. This modulation in turn is reflected in the power spectra.

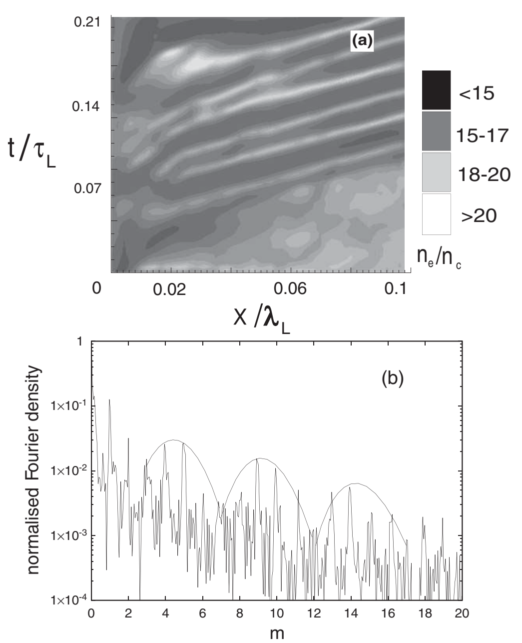

FIG. 1. (a) Plasma electron density contours for a target plasma density $n_e/n_c = 20$, $a_0 = 0.5$, with angle of incidence $\theta = 23°$. The time scale corresponds to 3 laser periods, τ_L, and the section of plasma shown extends over $0.1\lambda_L$. (b) Fourier decomposition of the electron density at a point $x = 0.1\lambda_L$ inside the plasma.

To explore the modulation of the emission spectrum, we carried out PIC simulations across a wide parameter range that included not only the intensity of the incident light and the density of the target plasma but both pulse shape and length and the steepness of the density gradient at the front edge of the plasma box. Insofar as the intensity dependence of the modulation is concerned, our conclusions are broadly in line with those of Watts *et al.* [9], namely, the higher the intensity, the more pronounced the modulation, albeit for different reasons. In our case, at values of a_0 significantly below the range of interest (0.5–2.0), the number of harmonics with intensities above threshold (in our case, 10^{-6}) is correspondingly reduced. This, in turn, is reflected in the number of modulation cycles observed or indeed by their disappearance altogether. For the model we have adopted, there is a more fundamental reason why modulation should disappear at lower laser intensity. Since the flux of electrons injected into the target plasma depends critically on intensity, a diminished flux, in turn, means that the level of plasma waves excited in the overdense plasma is correspondingly weaker.

A selection of spectra is shown in Fig. 2. Figures 2(a) and 2(b) use $a_0 = 0.5$, $n_e = 10n_c$ and $a_0 = 0.6$, $n_e = 19n_c$, respectively, where $\theta = 23°$ is the angle between the wave propagation vector and the normal to the target surface. For Fig. 2(c), $a_0 = 1.0$, $n_e = 40n_c$, and in the highest density run shown in Fig. 2(d), $a_0 = 0.5$, $n_e = 64n_c$. In all four cases, the pulse length used was 17 fs, chosen to minimize noise from plasma line emission. To highlight the effect, we show envelopes across the bands of modulation for $P_m/P_1 > 10^{-6}$ and for values of $m \gtrsim 1.5(\omega_p/\omega_L)$.

For the parameters used in Figs. 2(a)–2(c), the PIC results show 3 or 4 clear modulation cycles with average modulation frequencies $3.5\omega_L$, $4.3\omega_L$, and $6.6\omega_L$ respectively, values that correspond closely to the plasma frequencies for each of these densities. Figure 2(c) shows some fine structure in the modulation across the range $m = 10$ to $m = 18$. In this case, there is a contribution from second harmonic plasma emission which perturbs the emission spectra slightly in the neighborhood of $m = 12$.

The choice of parameters in Fig. 2(d) combines relatively high density and moderate light intensity, ensuring that the plasma wave is not too strongly driven. For $n_e/n_c = 64$, the plasma line is centered at $m = 8$ with the blue half of the modulation structure clearly defined (harmonic lines 9 to 12). The red wing is masked by the relative strength of the fourth and fifth harmonic lines.

Higher laser intensities are needed to extend the range of modulation at higher densities. This, in turn, results in the Langmuir waves being more strongly driven and significantly broadened. A linear estimate of the bandwidth of

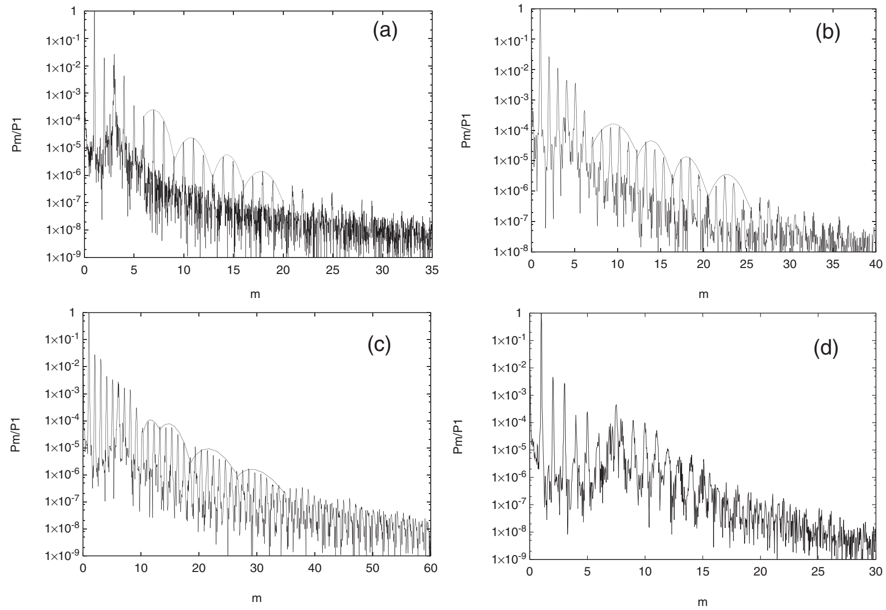

FIG. 2. Harmonic modulation for a laser pulse with $\tau_p = 17$ fs and $\lambda_L = 0.248$ μm: (a) $a_0 = 0.5$, $n_e/n_c = 10$, (b) $a_0 = 0.6$, $n_e/n_c = 19$, (c) $a_0 = 1.0$, $n_e/n_c = 40$, and (d) $a_0 = 0.5$, $n_e/n_c = 64$. Envelopes over the harmonics have been added to indicate the respective modulation frequencies.

Langmuir waves excited by a beam of fast electrons is $\Delta\omega \sim (\Delta V_b/v_b)(n_e/n_c)^{1/2}\omega_L$ for a beam velocity v_b with velocity spread ΔV_b. Broadened plasma line emission, in turn, affects neighboring harmonic lines.

Figure 3 illustrates the susceptibility of modulation to pulse shape and steepness of the density gradient at the plasma surface. Essentially, steepening the pulse over the $\sin^2$ profile used to obtain the data in Fig. 2 improves the definition of the modulation [Fig. 3(a)]. Varying the pulse length τ_p, on the other hand, has relatively little effect on the modulation as such, always provided the pulse is sufficiently longer than the growth time t_g of the electron-generated plasma waves. If we make use of the estimate given by Boyd and Ondarza-Rovira [7], this means $\tau_p > 10(n_c/n_b)(n_e/n_c)^{1/2}(\Delta V_b/v_b)^2\tau_L$, where n_b is the density of the electron pulse exciting the plasma waves. For the parameters in these simulations, this implies $\tau_p > 10$ fs.

Relaxing the steepness of the density gradient has the effect of weakening the modulation. This supports the conclusion by Watts *et al.* that a density ramp suppressed the modulation in their spectra. With a ramp present, electrons injected into the plasma excite plasma oscillations across the ramp. Thus, for example, for the parameters used in Fig. 3(b), $n_e = 16n_c$, $a_0 = 0.6$, the simulations now show an enhanced $m = 3$ line consistent with additional plasma excitation at $n_e = 9n_c$ in contrast to the sharp boundary case where only plasma oscillations with $\omega_p = 4\omega_L$ are present. A further distinction to be drawn is that the coupling of plasma waves to the radiation field is proportionately weaker in the case of an extensive ramp. Note that the disappearance of modulation is not the only difference between the two spectra. Whereas the maximum m over threshold ($P_m/P_1 \geq 10^{-6}$) in Fig. 2(b) is $m^* \sim 24$, the result in Fig. 3(b) for a ramp $0.6\lambda_L$ in width shows a range almost twice this value. Precisely why a ramp should have this effect is not clear.

The modulation first reported by Watts *et al.* was recorded over a range of about 15 harmonics. Their spectra showed a distinct if irregular modulation with a frequency of between 2 and 4 times that of the incident light. A less

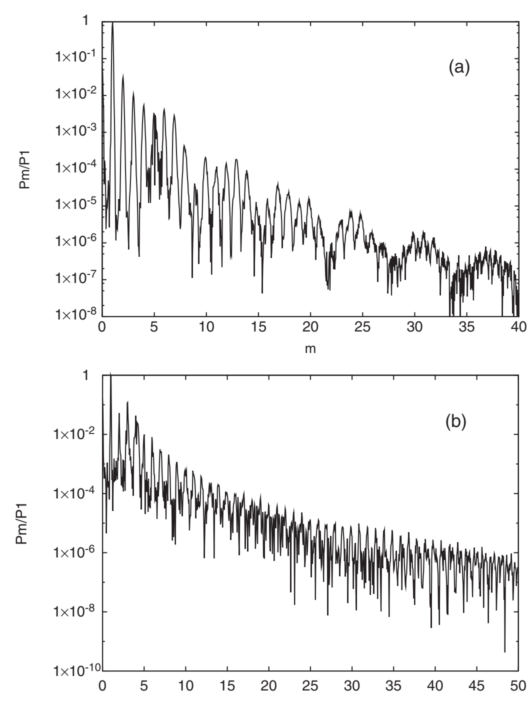

FIG. 3. Harmonic modulation for (a) a laser pulse with $a_0 = 1.0$, $\tau_p = 30$ fs, and $n_e/n_c = 30$, for a steepened pulse envelope; (b) effectiveness of a density ramp in suppressing modulation for a simulation with $a_0 = 0.6$, $\tau_p = 30$ fs, and $n_e/n_c = 16$ for a density ramp of width $\Delta = 0.6\lambda_L$.

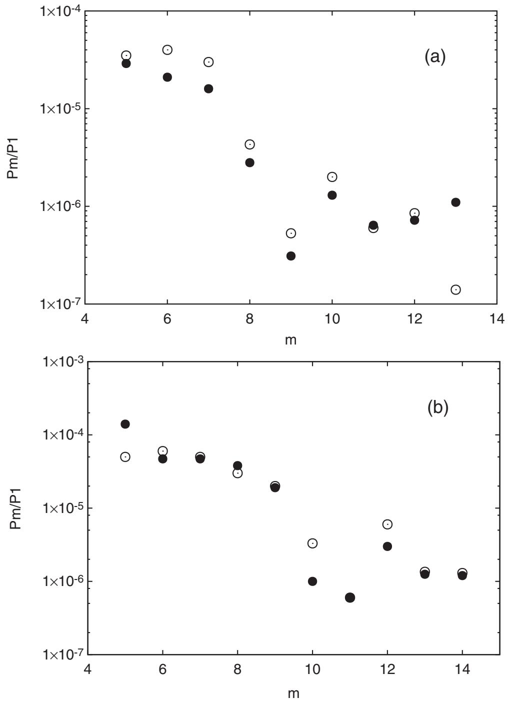

FIG. 4. Harmonic reflected power from PIC simulations and from experiment: (a) solid circles (●) correspond to $a_0 = 1.0$, $\lambda_L = 0.395$ μm, and $n_e/n_c = 30$; open circles (○) correspond to emission measured from a carbon target [3]; (b) ●: $a_0 = 1.0$, $\lambda_L = 0.395$ μm, $n_e/n_c = 45$, and $\tau_p = 17$ fs; ○: correspond to emission found from a glass target [3].

pronounced effect was subsequently found by Teubner *et al.* in experiments using pulses typically 100 fs in length. Over a harmonic range of about 10 lines, they noted anomalies in emission, though it is not possible to identify a modulation frequency over such a limited range. PIC simulations were carried out by both groups with a view to interpreting their data. The simulated spectrum in Ref. [9] showed an average modulation frequency of 5–6, as against the 2–4 observed. Both sets of authors interpreted the modulation in terms of the so-called "oscillating mirror" model proposed by Bulanov *et al.* [13].

The main stumbling block to a fuller understanding of these anomalies is the uncertainty over the ionization state of the target and, hence, the electron density. We illustrate in Fig. 4 data points from our simulated spectra alongside the harmonic efficiency measured in Ref. [3] for carbon and glass targets using frequency-doubled light from a Ti:sapphire laser (395 nm) at an intensity 2.4×10^{18} W cm^{-2}.

In Fig. 4(a), we show the results of a simulation in which $n_e/n_c = 30$ for which the plasma line appears at $\sim 5.5\omega_L$. From the pattern found in Fig. 2, we should expect to see the first minimum in the modulated spectrum at $m = 9$, and so it appears. We have added data points from the harmonic spectrum measured by Teubner *et al.* for a carbon target for comparison. In Fig. 4(b), we show results for a simulation with $n_e/n_c = 45$, which is characterized by a minimum at $m = 11$, along with the measured spectrum for a glass target. In each case, the harmonics, simulated and observed, show a broadly comparable dependence on the harmonic number. However, in making these comparisons, we stress that the electron densities corresponding to the measured spectra were not determined experimentally.

Our simulations lead us to conclude that the modulations we see in the harmonic spectrum stem from the effect of plasma waves excited in the plasma slab. We have shown that the modulation frequency varies with the plasma density as $n_e^{1/2}$ over a wide range. On this interpretation, the modulation frequency affords a measure of plasma density. The presence of a density ramp a fraction of a wavelength thick appears to suppress the modulation. This finding is consistent with our interpretation in that, in the presence of a ramp, there is no longer a unique surface plasma density. The question may properly be asked as to whether results obtained from a PIC code, limited in its dimensionality, can reliably describe the physics of this interaction. In particular, would one expect the modulation seen here to persist in three dimensions? While it would be incautious to press this claim too strongly, we are encouraged, not least by the comparisons drawn in Fig. 4, to believe that it might.

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