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Tim James Morrow Boyd, Ricardo Ondarza-Rovira. Modulation of harmonic emission spectra from intense laser-plasma interaction. 2004. <hal-00003279>

**HAL Id: hal-00003279**

**<https://hal.science/hal-00003279v1>**

Preprint submitted on 15 Nov 2004

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# Modulation of Harmonic Emission Spectra from Intense Laser-Plasma Interactions

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**Abstract.** We report results from a series of PIC simulations carried out to investigate the interaction of intense laser light with overdense plasma and in particular, aspects of the harmonic emission spectrum generated in these interactions. Recent observations of emission in this range by Watts *et al.* [1] show emission above the 20th harmonic extending to the 37th with conversion efficiencies of order  $10^{-6}$  with this spectrum showing distinct, if variable, modulation of between two and four harmonics. This spectral feature proved highly sensitive to the incident intensity, with more pronounced modulation at higher intensities. These authors also reported results from a set of model PIC simulations which showed a modulated emission spectrum across the entire harmonic range, but with a wider modulation bandwidth.

In the results to be presented we report on features of the emission spectrum across a wide parameter range and its dependence on the detailed structure of the plasma density profile. While we do see evidence of a modulated spectrum, this appears to be sensitive not only to the incident intensity but to the plasma density profile. In addition plasma emission present further complicates the harmonic emission spectrum. Our results lead us to conclude that attempts to interpret modulation in emission spectra in terms of one particular theoretical model and from this to infer the dynamics of the critical surface may be misguided.

## I. INTRODUCTION

The interaction of high power laser pulses with ultradense plasma has proved a rich source of new plasma phenomena as varied as channel formation and the generation of intense-localized magnetic fields. One of the earliest laser-plasma interactions to be studied in detail - and one of enduring interest - is the generation of multiple harmonics of the incident light. Different mechanisms for harmonic generation apply across differing intensity ranges and target characteristics but perhaps most attention now attaches to harmonic production by short intense laser pulses incident on solid targets. Short pulses without significant pre-pulse create target plasmas free from extensive underdense coronal regions.

Harmonic generation is not only of intrinsic interest but holds promise as a diagnostic of plasma conditions in the surface interaction region and, potentially of other physical processes localized there. As well as targets of dense slab plasmas the other determinant for efficient harmonic production is the brightness of the source. For light of intensity  $I$  and wavelength  $\lambda_L$ , values of  $I \lambda_L^2$  in the range  $10^{18} - 10^{19} \text{ W cm}^{-2} \mu\text{m}^2$  typically generate a harmonic range of 50 harmonics and Norreys *et al.* [2] detected harmonics up to the 75th (with conversion efficiency of  $10^{-6}$ ) for  $1.053 \mu\text{m}$  wavelength light interacting with a solid target. Such a harmonic range holds potential as a source of coherent XUV radiation. The harmonics in the spectrum of reflected light are generated by electrons accelerated across the vacuum-solid interface by the electric field of the pulse.

The spectral characteristics of the reflected light are broadly understood. Conditions favouring optimal harmonic generation include intensities in the range  $10^{18} - 10^{19} \text{ W cm}^{-2} \mu\text{m}^2$  with plasma densities of a few tens  $n_c$ , where  $n_c$  denotes the critical density. Typically this combination of intensity and target plasma density gives rise to a spectrum of 50 or more harmonics with a conversion efficiency of  $10^{-6}$ . An important transition takes place at an intensity threshold below which plasma expansion occurs thus creating a region of underdense plasma and above which channel boring and ponderomotive steepening are dominant.

Particle-in-Cell (PIC) simulations have been invaluable in understanding and characterizing this harmonic emission. Gibbon [3] used this approach to good effect in identifying the physical mechanism underlying harmonic generation, attributing it to those electrons that make vacuum excursions and then free-stream back into the target plasma. Gibbon's work showed that the current sources generating the harmonics are localized just within the overdense plasma boundary. These simulations led to an empirical expression for the efficiency of high-order harmonic generation  $\eta_m \sim 9 \times 10^{-5} (I_{18} \lambda_L^2)^2 (10/m)^{-5}$  where  $I_{18}$  denotes light intensity in units of  $10^{18} \text{ W cm}^{-2}$ .

In the work presented here we use a PIC code to further interpret aspects of the reflected light spectrum and to extend these simulations over a wider range of parameters. Boyd and Ondarza [4,5] examined in detail the effects of plasma line emission on the harmonic spectrum. Strong plasma emission was recorded, with harmonics up to the fifth detected in both the reflected and transmitted spectra. A robust feature of the reflected spectra appeared on the blue side of the plasma line between  $(1.2-1.6) \omega_p$ , where  $\omega_p$  is the plasma frequency. In this work jets of electrons of relativistic energy penetrating the ultra-dense plasma generated strongly nonthermal Langmuir waves. The spectrum of Langmuir waves was localized at the front edge of the plasma slab. This localization is important for the subsequent coupling of the plasma waves to the radiation field by means of the steep density gradient in the (perturbed) peak density region. Fig. 1 reproduces the spectrum obtained by Boyd and Ondarza [5] showing the reflected harmonic spectrum and the plasma line for  $n_e/n_c = 30$  and  $a_0 = 0.5$ . In this case the feature between  $\omega_p$  and  $2\omega_p$  shows up as enhanced harmonic lines between  $8\omega_L$  and  $12\omega_L$ , in contrast to the broad less well-resolved feature recorded for other choices of parameters.

We were prompted to re-examine the plasma line effects on the spectrum by results reported

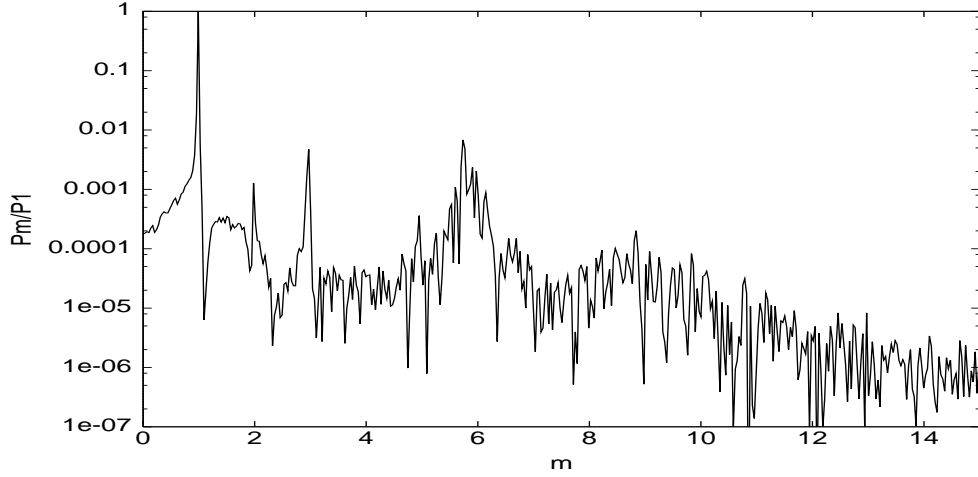


Figure 1: *Reflected harmonic spectrum for  $n_e/n_c = 30$  and  $a_0 = 0.5$ .*

by Watts *et al.* [1]. In this work using light intensities up to  $1.0 \times 10^{19} \text{ W cm}^{-2}$  incident on a solid surface, harmonics up to the 37th were recorded with a conversion efficiency  $\geq 10^{-7}$ . The striking feature of the spectra at the highest intensity is a distinct if irregular modulation with a frequency between 2 and 4 times that of the incident light (Fig. 2). Watts *et al.* found that although modulations peak at different harmonics they were observable across the entire spectral range although their published spectra show only a part of this range, and that at relatively low conversion efficiencies.

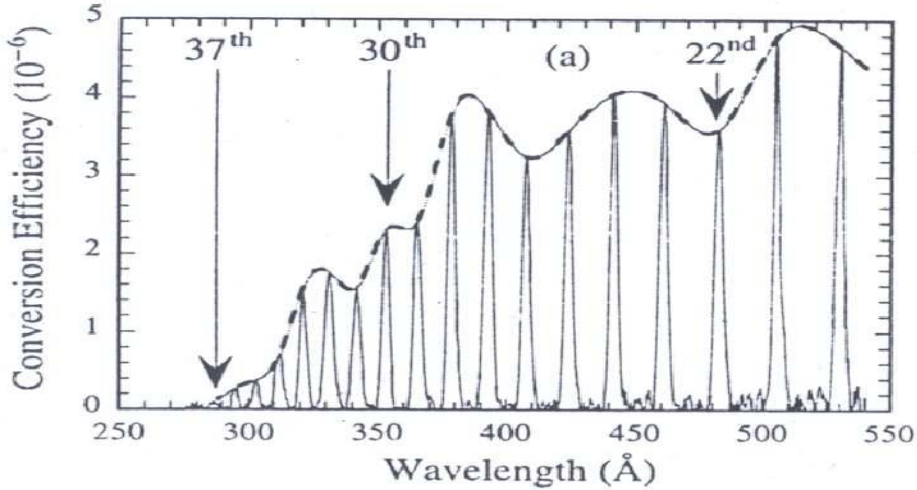


Figure 2: *Modulation of harmonic spectra, for a laser-dense plasma interaction,  $I \lambda_L^2 \sim 10^{19} \text{ W cm}^{-2} \mu\text{m}^2$ , from Watts *et al.* [1].*

The corresponding spectrum at  $I \lambda_L^2 \sim 10^{18} \text{ W cm}^{-2} \mu\text{m}^2$  in contrast shows only weak modulation. In conjunction with these observations they simulated the emission spectrum for conditions approximating those in the experiment apart from their choice of thickness of the simulated plasma which was only one wavelength across. In addition they used a pulse with a sine-squared temporal profile. The simulated spectra showed a clear modulation with an

amplitude that increased with laser intensity. In contrast to previous work using the same code where modulations were attributed to internal reflections of transmitted light within the plasma box (Lichters *et al.* [6]), Watts *et al.* suggest that the modulation structure is intrinsic to harmonic generation. For a plasma one wavelength thick and with density  $n_e = 13 n_c$  they record modulations at between 4 and 5 harmonics for  $I\lambda_L^2 \sim 10^{19} \text{ W cm}^{-2} \mu\text{m}^2$  across a spectral range of  $m = 40$  whereas for  $I\lambda_L^2 \sim 10^{18} \text{ W cm}^{-2} \mu\text{m}^2$  this range contracts to about  $m = 7$ , too limited to discern modulation even if it were present. These authors undertook a correlation of the modulated spectra with an examination of the dynamics of the critical surface, showing that surface modes excited by nonlinearities could have relatively large amplitudes. For the higher intensity studied, it appeared that harmonics at  $m = 4, 5$  were dominant. From this they concluded that the oscillating-mirror model of harmonic generation proposed by Bulanov *et al.* [7] was capable of interpreting the modulation observed.

The spectra reported by Watts *et al.* and the inferences drawn by them prompted us to look again at spectra from our earlier simulations. The PIC code we use, like the Pfund-Lichters code used by Watts *et al.*, embeds the Bourdier technique to allow for oblique incidence but in contrast to theirs, our simulation plasma can be up to ten wavelengths in extent, with the plasma density ramped at the front surface. 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. The initial electron temperature was chosen to be 1 keV and we used a gaussian pulse of variable length. The normalized quiver velocity  $a_0 \sim 0.85(I_{18}\lambda_L^2)^{1/2}$  lay in the range 0.5-2.0 with slab densities between  $4 n_c$  and  $64 n_c$ . The ions formed a neutralizing background.

Our attention focused on the effect of the plasma density on the modulated spectra. Given the wealth of detail in the spectra presented in terms of the dependence of the modulation on laser light intensity and pulse length as well as on the density and the density profile, it is perhaps worth highlighting at the start the principal finding from our work.

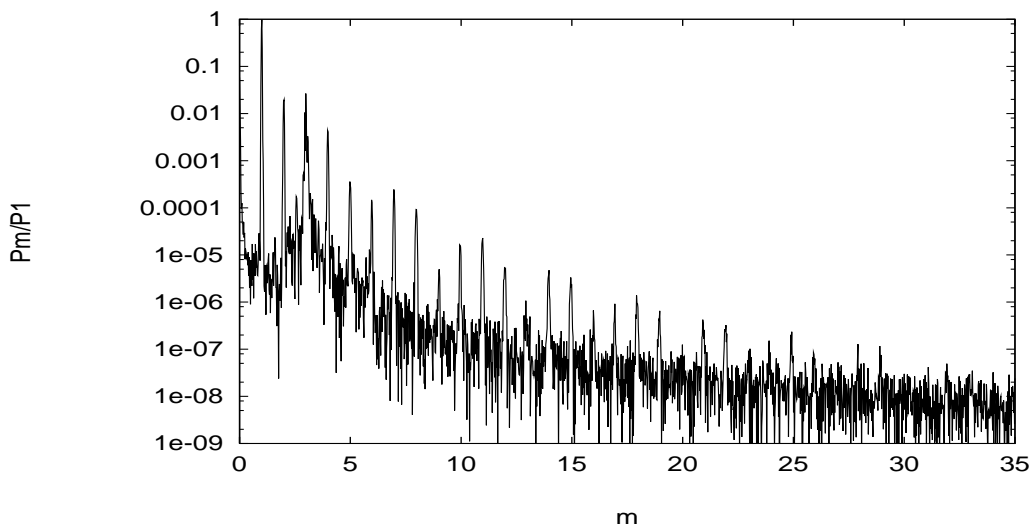


Figure 3: Harmonic modulation for  $a_0 = 0.5, n_e/n_c = 10$  and  $\tau_p = 17 \text{ fs}$ .

This is evident from Fig. 3 in which  $a_0 = 0.5, n_e/n_c = 10$  and the pulse length  $\tau_p = 17 \text{ fs}$ .

The spectrum shows clear and uniform modulation with an average modulation frequency of  $3.3 \omega_L$  for a plasma with  $\omega_p = 3.2 \omega_L$ . We contend that the spectrum of reflected light shows harmonic structure modulated at the Langmuir frequency.

To support this interpretation we carried out simulations across a range of intensities and pulse lengths. In all of these we used a plasma slab 4 wavelengths thick without any density ramping. Fig. 4 shows spectra for runs with  $a_0 = 0.6$  and  $n_e = 9 n_c$  and  $19 n_c$  respectively.

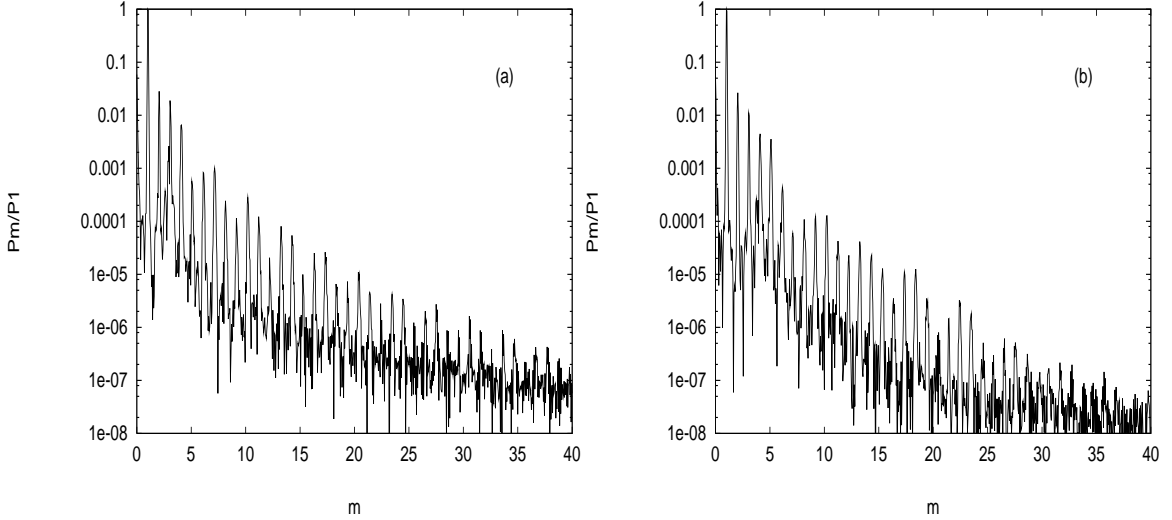


Figure 4: *Harmonic modulation for  $a_0 = 0.6$ ,  $\tau_p = 17$  fs, (a)  $n_e/n_c = 9$  and (b)  $n_e/n_c = 19$ .*

These spectra are modulated at frequencies of  $3.3 \omega_L$  and  $4.4 \omega_L$ , consistent with the Langmuir frequencies for these densities. Note in passing that for this combination of parameters the level of plasma line emission is low enough for the harmonic spectrum to be relatively unperturbed. With increasing density the harmonic range narrows correspondingly until a point where just one modulation cycle persists as in Fig. 5 for  $a_0 = 0.5$ ,  $n_e/n_c = 64$  with a pulse length of 17 fs. For this density the plasma line is centred on  $m = 8$  and we see the effects of the Langmuir wave spectrum on neighbouring harmonic lines, notably on the blue wing. Apart from the strength of the plasma line, this spectrum shows somewhat similar structure to that in Fig. 1. It now appears that the detail in the spectrum identified by Boyd and Ondarza [5] as the “combination line” is nothing other than the amplification of individual harmonic lines in the spectrum by the nonthermal Langmuir waves excited in the supra-dense plasma.

If we wish to reproduce spectra showing several cycles of modulation at higher densities we need to generate emission spectra at higher  $a_0$ . However the combination of higher intensity and increased plasma density results in the Langmuir wave being more strongly driven and at the same time significantly broadened. Simple linear estimates of the bandwidth of Langmuir waves  $\Delta\omega$ , excited by a beam of fast electrons give  $\Delta\omega \sim (\Delta V_b/v_b) (n_e/n_c)^{1/2} \omega_L$  for a beam velocity  $v_b$  with velocity spread  $\Delta V_b$ . Our PIC simulations show strongly driven Langmuir waves at the front edge of the plasma slab and this localization is crucial for coupling to the radiation field by the steep gradients in the perturbed peak density zone (cf. Boyd and

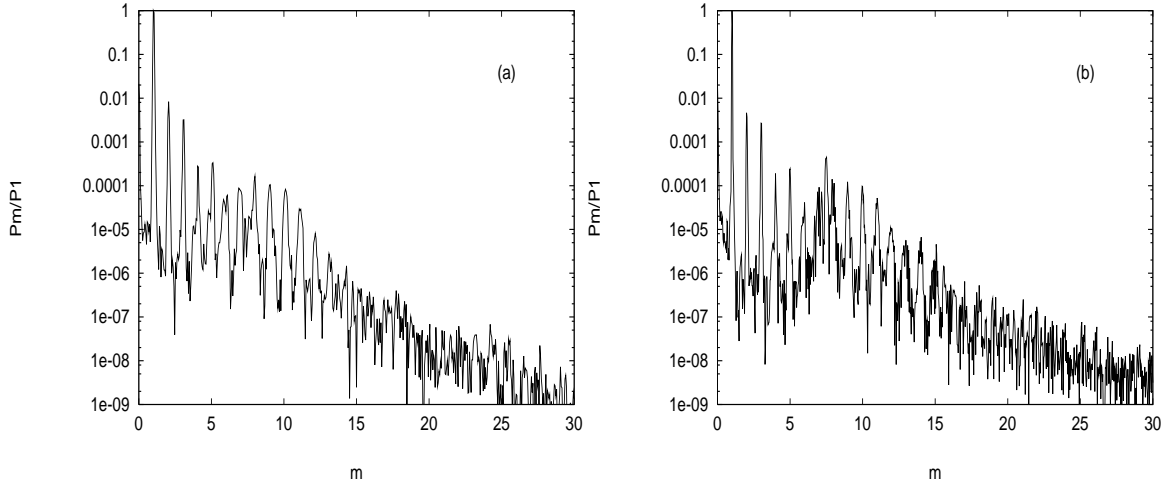


Figure 5: *Harmonic modulation for  $a_0 = 0.5, n_e/n_c = 64$ , (a)  $\tau_p = 17$  fs and (b)  $\tau_p = 25$  fs.*

Ondarza [5]). As the plasma emission line becomes more strongly driven it modifies the harmonic spectrum in the region of the plasma frequency (cf. Fig. 1). This in turn means that one has to look for modulation cycles at frequencies well above  $\omega_p$ . As we have remarked already the number of harmonic lines (above a conversion efficiency of  $10^{-6}$ , say) reduces with increasing density so that the spectral window available is squeezed from both ends.

Fig. 6 shows a more strongly driven result ( $a_0 = 1.0$ ) for densities  $n_e = 25 n_c$  and  $n_e = 50 n_c$ . The plasma line is now evident and affects neighbouring harmonic lines so that in Fig. 6a with  $\omega_p/\omega_L = 5$  the first full modulation cycle appears at  $m = 8$ . Three cycles with an average modulational frequency of  $5.3 \omega_L$  can be resolved above a conversion level of  $10^{-6}$ . For  $n_e = 50 n_c$ ,  $\omega_p/\omega_L \simeq 7.1$  and there are now just two cycles above this level with an average modulational frequency of  $8 \omega_L$ .

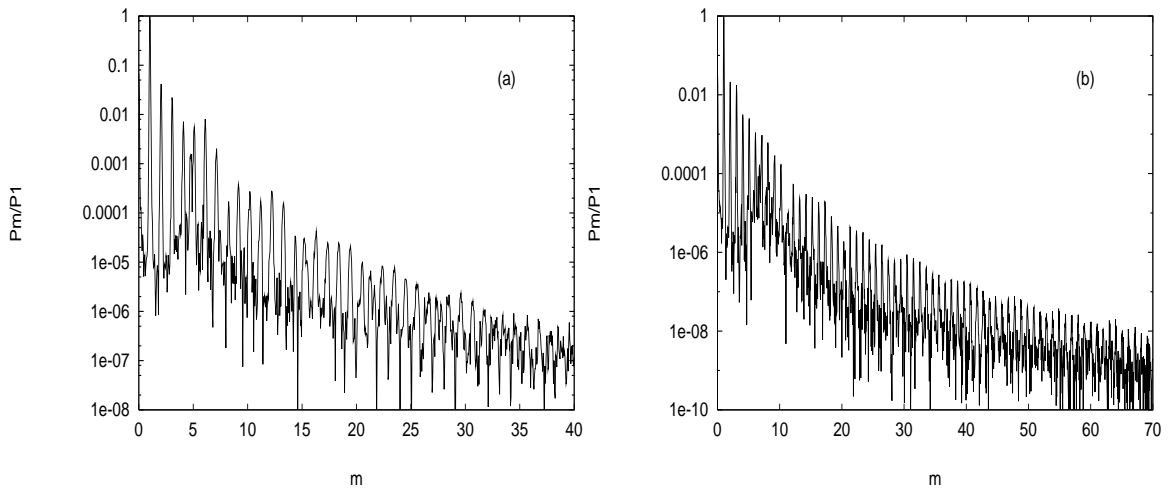


Figure 6: *Harmonic modulation for  $a_0 = 1.0, \tau_p = 17$  fs, (a)  $n_e/n_c = 25$  and (b)  $n_e/n_c = 50$ .*

At yet higher intensities ( $a_0 = 2.0$ ) and densities  $n_e = 64 n_c$ , plasma line emission is broader

again than in Fig. 6. The harmonic spectrum above a conversion level  $10^{-6}$  now extends to approximately  $m = 60$  (Fig. 7). However at this intensity not only is in the plasma line a strong feature but its second harmonic is strong enough to perturb the line spectrum. Consequently one can resolve only 3 modulation cycles with an average frequency of  $8.7 \omega_L$ .

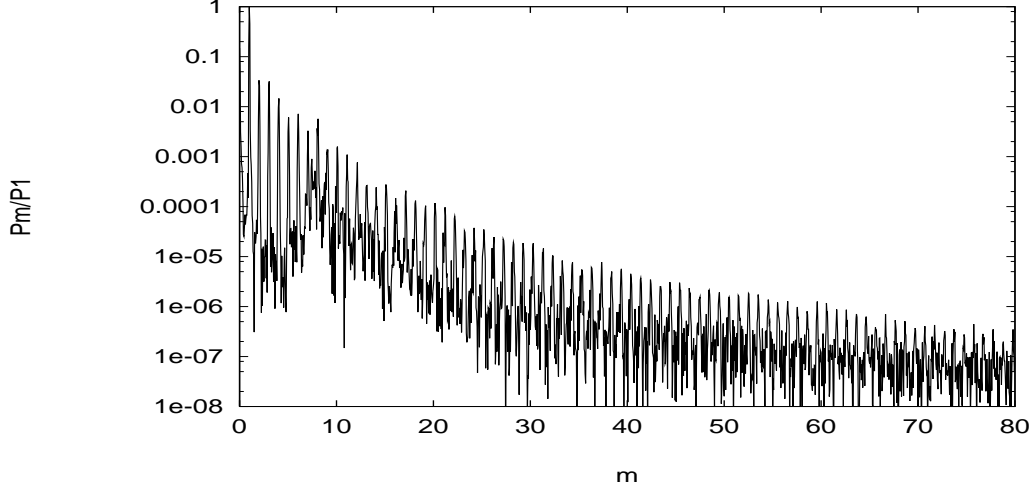


Figure 7: *Harmonic modulation for  $a_0 = 2.0$ ,  $\tau_p = 17$  fs, and  $n_e/n_c = 64$ .*

In all the cases discussed thus far, the PIC simulation plasma has been in the form of a slab. We have also examined the effect on the modulation of a ramped density profile to check the claim by Watts *et al.* that density ramping served to suppress the effect. We too found a comparable decay in modulation as the thickness of the ramp is increased. Fig. 8 shows our result for  $a_0 = 0.5$ ,  $n_e/n_c = 10$  for different thickness of ramp  $\Delta$ ; (a)  $\Delta = 0.2 \lambda_L$  and (b)  $\Delta = 0.4 \lambda_L$ . From this it is evident that although some modulation persists when a ramp of thickness  $0.2 \lambda_L$  is present, the effect disappeared completely in Fig. 8b.

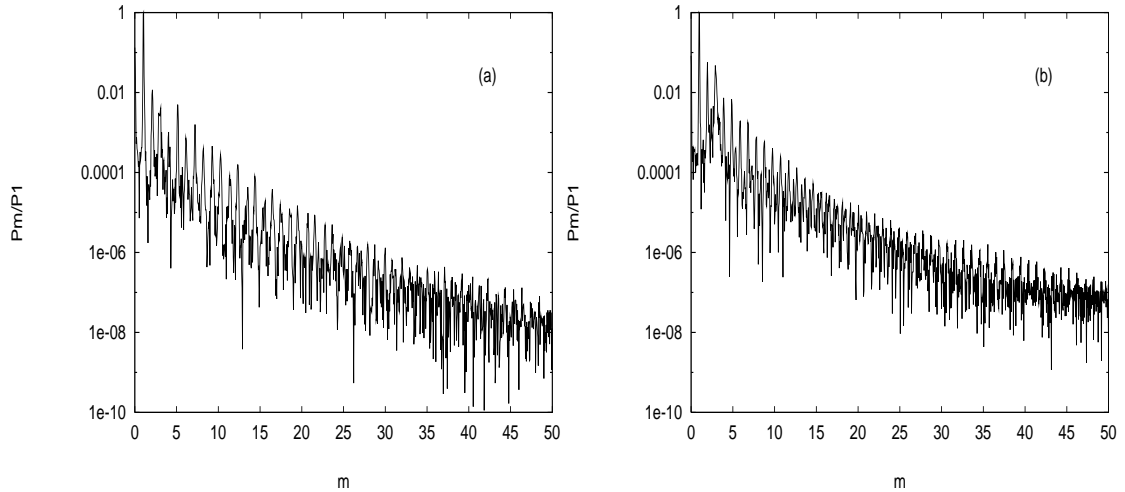


Figure 8: *Harmonic modulation for  $a_0 = 0.5$ ,  $\tau_p = 17$  fs,  $n_e/n_c = 10$ , (a)  $\Delta = 0.2 \lambda_L$  and (b)  $\Delta = 0.4 \lambda_L$ .*



## II. CONCLUSIONS

Our simulations lead us to conclude that the modulations we see in the harmonic spectrum stem from the effect of Langmuir 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. The modulation is evident for choices of intensity and pulse length for which plasma line emission does not distort the spectrum to any marked degree. On this interpretation the modulation frequency affords a measure of plasma density as distinct from the claim by Watts *et al.* of its potential as a diagnostic of the dynamics of the critical surface. The presence of a density ramp a fraction of a wavelength thick appears to suppress the modulation as Watts *et al.* found. We see this as consistent with our interpretation in that the presence of a ramp ensures that there is no longer a unique plasma density at the surface.

## Acknowledgments

One of us (ROR) acknowledges support from CONACyT under Contract No. 43621-F.

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