Measurements of relativistic self-phase-modulation in plasma

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We report the first systematic observations of relativistic self-phase-modulation (RSPM) due to the interaction of a high intensity laser pulse with plasma. The plasma was produced in front of a solid target by the prepulse of a 100 TW laser beam. RSPM was observed by monitoring the spectrum of the harmonics generated by the intense laser pulse during the interaction. The multipeaked broadened spectral structure produced by RSPM was studied in plasmas with different density scale lengths for laser interactions at intensities up to 3.0×10^{19} W cm^{-2} ($a=p_{osc}/m_e c=4.7$). The results are compared with calculated spectra and agreement is obtained.

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I. INTRODUCTION

The rapid development of modern laser systems has led to the generation of focussed laser light with intensities exceeding 10^{20} W cm^{-2}, and has resulted in the observation of many novel phenomena from the interaction of high intensity pulses with matter [1]. These observations include the production of relativistic electrons [2], high energy γ rays [3], energetic protons [4], laser induced nuclear reactions [5], and huge magnetic fields [6]. In addition, relativistic nonlinear optical effects in the plasma begin to become important in this regime [7] and phenomena such as harmonic generation [8], relativistic self-focusing and parametric instabilities in the strongly-coupled regime have been observed [9].

Self-phase-modulation (SPM) of laser light has been studied extensively in solids and gases [10] and is a result of propagation of light through a medium with an intensity-dependent index of refraction. In plasmas, SPM has been studied theoretically [7,11,12] and at high intensities is predicted to occur when the quiver velocity of an electron in the laser field becomes relativistic ($a=p_{osc}/m_e c>1$) such that the refractive index of the plasma is modified by the relativistic increase in electron mass.

In this paper, we present the first direct measurements of relativistic SPM of laser light in a plasma. Qualitative observations of SPM have been observed previously in Zhang *et al.* [13]. However, to date, no systematic experiments addressing this effect have been reported. In our experiments, a preformed plasma is produced in front of a solid target by the prepulse of a high power laser. SPM of the light occurs as it travels through the underdense preplasma and consequently this effect alters the spectral profile of the incident laser pulse on the target, which thus is evident in the spectrum of harmonics of the laser frequency generated during the interaction. The harmonics are produced at the relativistic critical density of the laser $n_e=\gamma n_c$, where n_e is the electron density, n_c is the critical density for the laser and γ is the relativistic factor. Since scattered light at the fundamental frequency is not at relativistic intensities it may not be able to leave the plasma. For this reason detailed observations of the third harmonic, which has a critical density nine times larger, were performed. A similarly broadened spectrum was observed for all harmonics up to the 30th at 35.1 nm. The spectral profile is compared for two preplasmas with different density scale length and the observed SPM agrees well with theoretical predictions. A spectral shift is also observed which is consistent with the motion of the critical surface due to hole boring [14,15].

II. EXPERIMENT

The experiment was performed using the VULCAN laser system at the Rutherford Appleton laboratory. This laser delivers 0.7–1.0 ps pulses at a wavelength of 1.053 μm with energies up to 80 J on target. After compression, the laser beam was incident onto a mirror that allowed 1% transmission of the laser energy for diagnostics. These consisted of a far-field monitor for measurement of the focal spot, an autocorrelator for the laser pulse duration and a spectrometer for the laser spectral profile. The beam had dimensions of 22×11 cm^2 and was focused onto the target by an off-axis parabolic mirror of focal length $f=60$ cm. The target consisted of an optically polished fused silica slab at 45° angle of incidence with the beam p polarized. A penumbral imaging camera monitored the soft ($h\nu\sim1$ keV) x-ray spot size and this, combined with the laser energy, yielded the focused intensity. An independent measure of the intensity was also obtained by using CR39 plastic nuclear track detectors to measure the maximum ion energies in the blowoff plasma [4,16]. The preplasma produced by the laser prepulse was monitored with a 0.527 μm transverse optical probe. The spectrum of the third harmonic, at 351 nm, was observed with a spectrometer viewing along the target normal. Other higher-order harmonics—up to the 30th order—were observed using a flat-field x-ray ultraviolet (XUV) spectrometer in the specular direction.

The intensity on target was up to $\sim3.0\times10^{19}$ W cm^{-2}. The diameter of the focal spot was ~10 μm. The bandwidth of the laser was transform limited with $\Delta\lambda=1.89\pm0.16$ nm. The laser prepulse has been measured previously

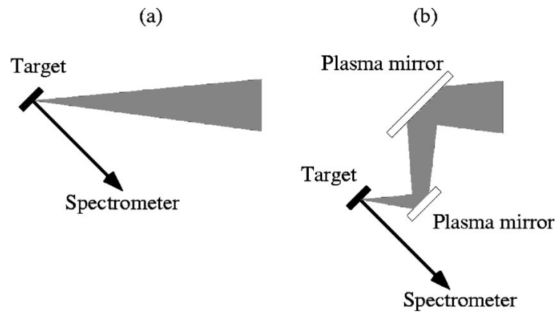

FIG. 1. The configuration of the plasma mirrors. (a) Without the plasma mirrors the laser prepulse typically produced $a \sim 10$ μm preplasma. (b) With the plasma mirrors the prepulse is reduced leading to a smaller scale length preplasma.

to be $\sim 10^{-6}$ of the peak intensity at 40 ps before the peak of the pulse [17]. With this prepulse the scalelength was found to be typically less than 10 μm [18].

In the experiments reported here a smaller scale length was produced by reducing the laser prepulse with a pair of plasma mirrors [19]. These consisted of two antireflection coated optical flats. The damage threshold of the coatings was $\sim 10^{13}$ W cm^{-2}. Incident light at intensities below this value passed through the glass and did not reach the target. The prepulse due to amplified spontaneous emission (ASE) in the system was reduced in this manner by a factor of $\sim 10^3$, this limit being due to the residual reflectivity of the glass flat. The setup is shown in Fig. 1. The laser energy reaching the target was measured with a calorimeter and found to be $\sim$46% of the incident laser energy. It should be noted that the effect of the plasma mirror in these experiments was not to eliminate preplasma completely since it was likely that some prepulse due to imperfect recompression of the laser beam remained after the plasma mirror.

Typically, a high intensity laser pulse like that produced by the VULCAN Nd:Glass laser system consists of three parts: (1) the high intensity main pulse, (2) a long duration ($\sim$1 nsec) component of amplified spontaneous emission (measured to be $\sim 10^{-6}$ of the main pulse intensity) (3) a lower intensity shoulder on the main pulse due to incomplete compression by the diffraction gratings (and which may only be tens of picoseconds long). While the plasma mirror is very effective in eliminating the ASE component it is less successful in reducing the shoulder of the pulse—and consequently even with the implementation of the plasma mirror it is difficult to completely eliminate the preplasma in our experiments.

Consequently, the effect of the plasma mirror in our experiments is not to increase the contrast ratio of the laser pulse—but rather to reduce the scalelength of the interaction—by reducing the duration of the prepulse.

If one simply considers the production of the preplasma by a self-similar expansion model of the plasma—the scale length of the plasma is given at any instant by $c_s\tau$ [where c_s is the sound speed which varies weakly with the prepulse intensity $\sim(I)^{0.2}$]. Therefore if the prepulse intensity (contrast ratio) remains the same—while the duration (τ) of the prepulse has been reduced this has the effect of a dramatic reduction in the plasma scale length. This is the principal effect of the plasma mirrors as implemented in our experiments.

The spectrum of the third harmonic was measured as a function of the laser intensity without and with the plasma mirrors, i.e., for large and small scale length preplasmas. Typical spectra are shown in Figs. 2(a) and 2(c). In both cases the spectrum is broadened and shifted and exhibits a multipeak structure. This is the characteristic of SPM. The broadening and shift are functions of the laser intensity, but the individual peaks remain constant with a width of 0.27 $\pm$0.04 nm (FWHM). The resolution of the spectrometer was 0.07 nm. A larger number of peaks is observed in the long scale length preplasma over a broader spectral width. The spectrum is redshifted, the shift scaling as $(I\lambda^2)^{0.5}$. This is in agreement with hole boring at the critical surface, as seen in the simulations by Wilks *et al.* [14], and measured by Zepf *et al.* [15]. In the smaller scale length preplasma a slight blue shift is observed as expected and is observed when the plasma pressure exceeds the laser ponderomotive pressure. The transition from a redshift to a blueshift depends on the incident laser intensity and the preplasma scale length, and is discussed in Kalashnikov *et al.* [20].

The full spectral width is plotted as a function of $I\lambda^2$ for large and small scale lengths in Fig. 3. Higher-order harmonics up to the 30th at 35.1 nm were observed using the XUV spectrometer and microchannel plate detector (see Fig. 4). The wavelength shift and broadening of the spectral profile was also evident for these harmonics—and was consistent with the measurements of the third harmonic. However, the resolution of this instrument was not high enough ($\sim$0.15 nm) to observe the associated multipeak structure.

In these experiments the laser intensity was varied systematically while the bandwidth of the generated harmonics was simultaneously examined under two separate conditions—one with high prepulse and one with low prepulse. The scale length of the preplasma in the interaction region is difficult to measure and it is difficult to make a precise claim as to the actual value of the scale length in either case with or without the plasma mirrors. It is clear, however, that decreasing the amount of prepulse has decreased the amount of preplasma and this is reflected in a reduction in the scale length. Indications of the scale length in the "large prepulse" case were obtained from transverse probing measurements of the plasma shadowgraphy [18]. While such measurements are taken of the lower density part of the plasma (up to 10^{20} cm^{-3}) they do give a fair indication of the plasma scale length at higher density.

III. RELATIVISTIC SELF-PHASE-MODULATION

Relativistic SPM can be calculated as follows. The angular frequency shift of the laser pulse is [10]

$$\delta\omega(t) = -\frac{d}{dt}[\phi(t)], \tag{1}$$

where $\phi(t)$ is the phase shift

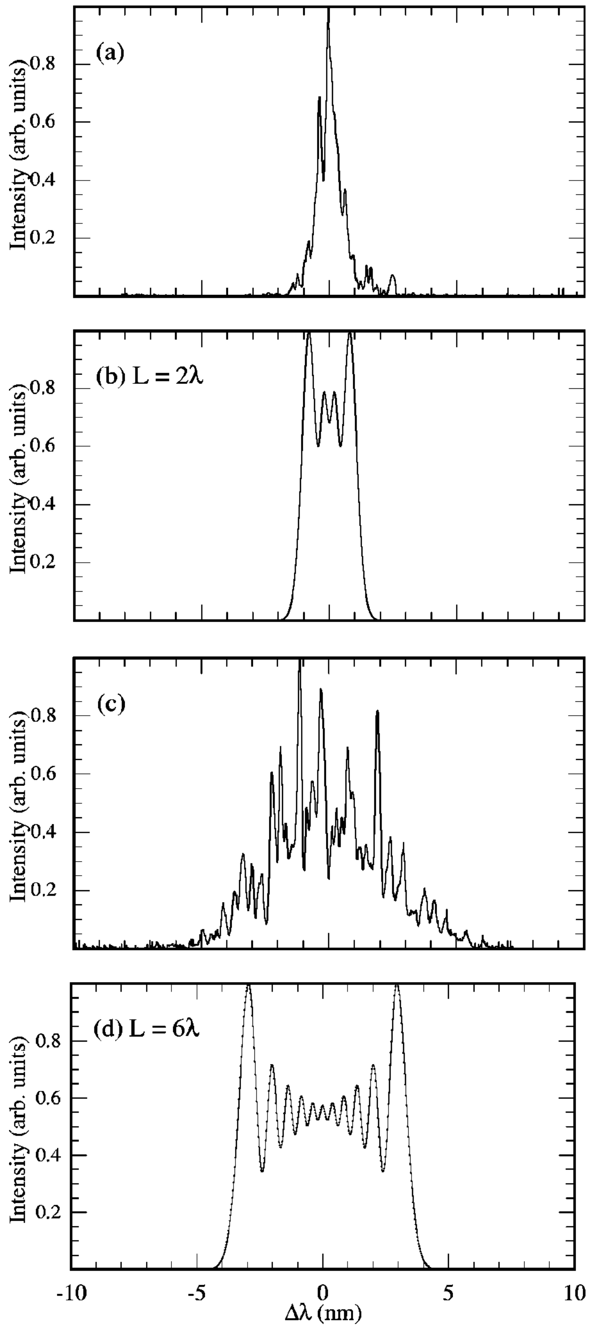

FIG. 2. Broadening of the third harmonic spectra at (a) $I\lambda^2 \sim 2\times10^{19}$ W cm^{-2} μm^2 with reduced prepulse and (c) at $I\lambda^2 \sim 5\times10^{18}$ W cm^{-2} μm^2 with prepulse. [Note that in (a) the central wavelength is slightly blueshifted with $\delta\lambda_3/\lambda_3 \approx 0.002$ and in (c) it is redshifted with $\delta\lambda_3/\lambda_3 \approx 0.01$. This has been corrected for in the above figures]. In (b) and (d) the calculated spectral profiles from the Fourier transform of the electric field are given for (b) L=2λ and (d) L=6λ.

$$\phi(t) = \frac{\omega}{c}\int_0^{x_c} \eta dx. \tag{2}$$

Here $\eta = [1 - n_e(x)/\gamma(t)n_c]^{1/2}$ is the refractive index and $\gamma(t) = [1 + a(t)^2/2]^{1/2}$ is the relativistic factor due to the transverse quiver motion of the plasma electrons in the focal region. The index of refraction only requires relativistic corrections to the motion of plasma electrons since the pulse duration is much shorter than typical ion motion time scales. The normalized momentum, $a = p_{osc}/m_e c$, is related to the laser intensity by $I\lambda^2 = a^2 \times 1.37\times10^{18}$ W cm^{-2} μm^2.

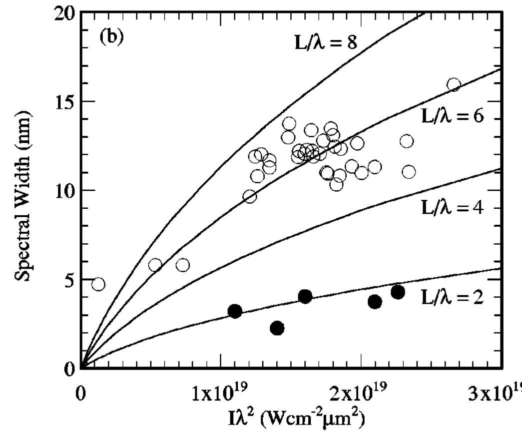

FIG. 3. The full spectral width of the third harmonic plotted as a function of $I\lambda^2$. The closed circles represent the data obtained with low prepulse. Open circles represent the data obtained for larger prepulse. The lines represent the calculated spectral width for relativistic SPM with varying linear ramp scale lengths.

For a linearly ramped density profile with $n_e(x) = n_c x/L$, Eqs. (1) and (2) give

$$\delta\omega(t) = -\frac{2}{3}\frac{\omega_o L}{c}\frac{d}{dt}[\gamma(t)], \tag{3}$$

where the integration has been up to γn_c.

The frequency change $\delta\omega(t)$ is to the red in the leading part of the pulse and to the blue in the rear. There are thus two times during the pulse having the same value of $\delta\omega(t)$. At these instants the pulse has the same frequency but will have different phase. The observed multipeak structure is a result of the constructive and destructive interference of these waves.

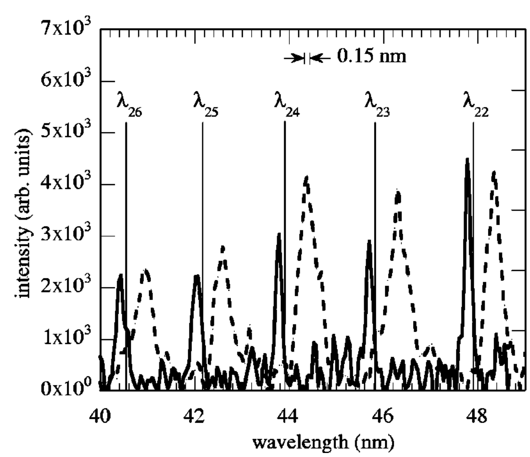

FIG. 4. Higher-order harmonic spectra at (a) $I\lambda^2 = 3.1\times10^{19}$ W cm^{-2} μm^2 with prepulse (dashed line) and (b) at $I\lambda^2 = 2.3\times10^{19}$ W cm^{-2} μm^2 with reduced prepulse (solid line).

The full spectral width, calculated from the peak to peak value of $\delta\omega(t)$ for preplasmas of different linear scale lengths, is also plotted in Fig. 3. A scale length of $L/\lambda \sim 6$ matches the experiment when the full laser prepulse was used. This agrees well with the scale length measured by transverse laser probing [18]. With the reduced prepulse obtained by use of the plasma mirrors a fit can be obtained using a scale length of $L/\lambda \sim 2$, but because of the complicated experimental geometry it was not possible to make probing measurements to confirm this scale length estimate during these experiments.

The number of peaks in the spectral profile is a linear function of the maximum phase change ϕ_{max} [10]. The number of peaks M is given by $\phi_{max} \simeq (M-1/2)\pi$. For scale lengths $L/\lambda = 2$ and $L/\lambda = 6$ calculating ϕ_{max} from Eq. (2) gives $M=4$ and $M=11$, respectively. These are in reasonable agreement with the spectral profiles shown in Fig. 2 and corroborates the scale lengths inferred from the data in Fig. 3.

The spectral profile can be calculated by taking the square of the Fourier transform of $E(t)\exp[i\phi(t)]$, where $E(t)$ is the laser electric field and $\phi(t)$ is from Eq. (2). This is shown in Figs. 2(b) and 2(d) for linear density distributions with $L/\lambda = 2$ and $L/\lambda = 6$, assuming a Gaussian pulse shape (0.5 ps $1/e$ intensity half width) with peak intensity $I_o = 1.2 \times 10^{19}\ \mathrm{W\,cm^{-2}}$. The width of the individual peaks is related to the assumed bandwidth, which for the third harmonic is 0.26 nm. The average measured width of the peaks in Fig. 2 is 0.27 ± 0.04 nm.

The calculated spectra in Fig. 2 have a minimum at the central wavelength whereas the experimental profiles have a maximum. The analysis of Bulanov *et al.* [11] found that the spectral profile is a minimum for a nonrelativistic pulse ($a < 1$) and a maximum for a relativistic pulse ($a > 1$) in the situation where a constant uniform plasma density was assumed. This is indeed found to be the case when the integration of Eq. (2) is carried out to the critical density n_c. However, when the integration is evaluated up to the relativistic cutoff γn_c the resulting profile is a minimum. An analysis of RSPM with an exponential density profile $n_e(x) = n_c \times \exp[-x/L]$ shows a similar behavior. For high intensity laser-plasma experiments the effect of ponderomotive steepening at the critical surface can result in extremely short scale length plasmas [21,22]. Therefore the difference in position of the relativistic and nonrelativistic critical densities is likely to be small and may explain the measured spectral profile.

IV. DISCUSSION

The observed spectral broadening in our experiments is primarily due to relativistic self-phase-modulation—a phenomenon which can have significant effects on interactions at high laser intensity. However, while the phenomenon of self-phase-modulation has been studied extensively in nonlinear optics (e.g., optical fibres), it has not previously been considered in detail in an experimental study of plasmas. This is an extremely important effect in the interaction of high power lasers with plasma especially as the peak intensity of lasers continues to rise.

There are four potential sources of self-phase-modulation in a laser produced plasma.

(1) that due to the nonlinear behavior of ion susceptibilities in the plasma—i.e., nonlinear susceptibility due to bound electrons of plasma ions. This effect has not previously been considered in the literature—but can be easily estimated. The nonlinear susceptibility of ionized atoms varies approximately like the inverse of the ionization potential to the third power and is very small in our experiments since the ionization stage of the atoms in the plasma is typically large —see Ref. [24]. The broadening caused by this effect should therefore be very small since the interaction distances are also quite short (a few microns).

(2) The second source of SPM can be that due to the nonlinear behavior of the temporally varying plasma index of refraction from electronic charge displacement in the plasma. It is likely that charge displacement self-phase-modulation (CDSPM) plays a role in the observed broadening but this effect can be easily calculated and is relatively small.

The peak charge displacement which is produced by an intense laser beam can be estimated by balancing the radial ponderomotive force of the laser with the space charge force created by electrons as they are forced radially out of the laser focal region. The total amount of charge that can be displaced by the laser ponderomotive force consequently only depends on the spot size and the peak intensity—it is not dependent on plasma density.

It is possible that laser self-focusing occurs in the plasma at higher density—which would cause a larger amount of charge displacement—however, in our experiments the hot electron temperature as measured through the use of γ ray spectroscopy indicates that the peak intensity is consistent with the vacuum intensity of the focused laser beam. In addition, the vacuum focal spot is consistent with penumbral images of the focal spot during the interaction.

Therefore if one equates the laser ponderomotive force with the space charge force via Gauss' law one calculates a peak dip in the plasma electron density at the center of the laser focal region. This is $\delta n_e \sim 5 \times 10^{17}\ \mathrm{cm^{-3}}$ for an intensity of $\sim 10^{19}\ \mathrm{W\,cm^{-2}}$ which is quite small compared to the background plasma density in our experiments and correspondingly produces little self-phase-modulation for the laser parameters in our experiments (i.e., a spectral broadening of much less than 1 nm for the third harmonic).

(3) A third source of SPM can be due to displacement of plasma (i.e., ion *and* electron motion in the focal region during the laser pulse). The experiments discussed here concern short pulse (~ 1 psec) high intensity laser plasma interactions. Ion motion during the time of the laser pulse is generally small—and the effect of the plasma pressure (temperature) balancing the ponderomotive pressure is only important for laser pulses with longer pulse durations. Indeed this effect (plasma displacement SPM) would only give rise to a redshift not a symmetric shift as we observe (see Fig. 2). The effect of plasma displacement SPM is even less than that for CDSPM for high intensity short pulse laser experiments and

only results in a small redshift—since the plasma density would always be decreasing throughout the duration of the laser pulse.

Previously observed channels generated in high intensity laser-plasma experiments are those created by the effect of the electric field of the displaced electrons on excess ions remaining in the focal region which consequently receive an impulse and are expelled radially. The time for these ions to move is about of the order of the laser pulse length in our experiments (i.e., greater than 1 ps). Therefore, the plasma channels created by the laser pulse in our experiments are more of a small dip in the background density rather than a vacuum hole bored in the high density plasma. The deep channels observed in some previous experiments are produced by the motion of ions after the passage of the intense laser pulse and would not affect the spectrum of the laser pulse itself.

(4) The the only effect which can successfully explain our measurements of broadening is relativistic self-modulation. It is theoretically expected to occur in experiments with lasers having similar intensities as VULCAN—and the effect agrees with the spectral broadening expected from interactions in plasma density profiles similar to those expected in our experiments. It also generally agrees with the modulated structure that we observe in the spectrum.

In these experiments there is also some broadening of the spectrum due to the hole-boring process (the laser pulse pushing against the critical surface). However, spectral broadening is also observable at lower intensities when no redshifting (hole-boring) or blue shifting can be observed. The fact that this occurs in situations which have large amounts of preplasma is indicative that the main source of the broadening is relativistic self-phase-modulation in the underdense plasma rather than hole boring. This is also evident in Fig. 4 —which shows the broadening effect on the higher-order harmonics. In addition redshifting is much less prominent in very short pulse experiments when hole boring is less important while the effects of self-phase-modulation remain observable [23].

The effect of ionization induced blue shifting [25] has always been observed as a blue shifted wing of the laser pulse—but only in underdense (gas target) interactions. This is to be expected since the trailing half of the laser pulse will pass through plasma which has already been ionized by the front of the pulse and should not be blue shifted.

This effect is not really observable in our experiments since the plasma would be ionized by the prepulse or by the very front of the main pulse (and also because the propagation distance in the plasma is small).

It should also be noted that the bandwidth of the harmonics ($\Delta\omega/\omega \sim \Delta\lambda/\lambda$) is the same for all of the different harmonics observed on a particular shot which suggests that this bandwidth is similar to the incident bandwidth of the fundamental laser pulse—so spectral structure of particular harmonics can be directly related to the spectral structure in the incident laser pulse.

To summarize, we have performed the first systematic experiments on self-phase-modulation in a plasma. The effect of preplasma is found to have a significant effect on the bandwidth of the harmonic radiation—principly due to the effect of relativistic self-phase modulation. Other sources of self-phase-modulation in plasma are estimated to be much less important including the contribution to the self-phase-modulation in the pulse from "charge displacement" as well as from nonlinearities due to partially ionized atoms. The broadening from relativistic self-phase-modulation affects not only the third harmonic—but actually the entire harmonic spectrum—and has important consequences for applications of high-order harmonic radiation. It is clear from these results that the reduction of preplasma also significantly enhances the conversion efficiency of the high-order harmonics.

This is clearly a very important topic for understanding the interaction of very high intensity laser-plasma interactions. There are many laser systems under construction around the world which expect to generate focussed intensities greater than $I = 10^{21}$ W/cm^2. At such intensities the effect of relativistic self-phase-modulation is likely to be extremely large. This consequently has very serious implications for energy deposition and absorption mechanisms during these interactions.

The suitability of the harmonics produced from intense laser-solid interactions as a source of coherent XUV radiation has been shown to be limited by preplasma formation [26]. This leads to a reduced source brightness at higher intensities due to spectral broadening, a broadened angular emission distribution and reduced conversion efficiencies. The results presented here demonstrate that the formation of preplasma is efficiently suppressed by the use of plasma mirrors. This may lead to enhanced brightness characteristics of the high-order harmonics produced in future experiments.

In conclusion, observations of the third harmonic spectral profile show varying degrees of shifting, broadening, and modulation with different levels of laser prepulse. These features are consistent with the effect of relativistic SPM of the fundamental laser light as it travels through the underdense preplasma. This is the first quantitative measurement of relativistic SPM in a plasma and demonstrates the importance of relativistic nonlinear plasma effects in the high intensity regime. As higher laser intensities are achieved the role of relativistic SPM in laser-plasma interaction experiments will become increasingly important. For instance, the spectral extent of the broadening given by Eq. (3) for $I_o \sim 10^{21}$ W cm^{-2} is such that $\Delta\omega/\omega \sim 1$ implying that the region of laser energy deposition will be greatly enlarged. This will clearly affect hot electron and x-ray production and may have important implications for some of the technological applications foreseen for these lasers. Consequently this confirmation of theory is also important for "fast-ignition" [27] and laser-plasma accelerator research in addition to fundamental studies of relativistic nonlinear optics.

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