High-harmonic generation in plasmas from relativistic laser-electron scattering

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Results are presented on the generation of high harmonics through the scattering of relativistic electrons from high-intensity laser light. The characteristic signatures of this process are found to be the emission of even-order harmonics, linear dependence on the electron density, significant amount of harmonics with circular polarization, and small spatial extent of the source. The harmonics are emitted as a forward-directed beam with a divergence of 2°–3°. The measured spatial profile of the harmonics is in excellent agreement with calculations that assume that relativistic electrons play a significant part in the scattering process. © 2003 Optical Society of America

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1. INTRODUCTION

With the advent of high-power lasers it has become possible to study the interaction of free electrons in extremely high laser fields. Such lasers, based on the principle of chirp pulse amplification,[1] routinely produce multiterawatt pulses of subpicosecond duration, which can be focused to obtain peak intensities in excess of 10^{19} W cm^{-2}. At these ultrahigh intensities electrons quiver with velocities close to the velocity of light, the magnetic field associated with light becomes significant, and the motion of even free electrons becomes highly nonlinear. Under these conditions conventional electrodynamics has to be significantly modified. As was predicted many years ago, the nonlinear motion of relativistic electrons should lead to the emission of short-wavelength light in the form of harmonics, a process known as nonlinear Thomson scattering.[2–4] Over the years there have been several efforts to detect this process. One of the earliest was to directly scatter an electron beam off the laser.[5] However, the experimental evidence for such nonlinear scattering processes has been insufficient and of doubtful validity. With the development of tabletop high-power lasers it has become possible to easily access the intensities required for studying this process. Thus in the late 1990s the first signatures of nonlinear Thomson scattering were claimed to have been detected in experiments where a high-intensity laser pulse interacted with underdense plasma.[6] It was shown that the second and third harmonics emitted from the plasma had the characteristic angular distributions predicted by theory and scaled linearly with the number density as expected because the process is incoherent. Interest in this process has continued because it offers the possibility of studying some of the basic physics of the interaction of relativistic electrons with strong fields.

Harmonics produced from short-pulse laser-driven plasmas have several attractive characteristics. The fact that they are produced by short-pulse lasers means they are of femtosecond duration. The spatial extent is also extremely small (micrometer sized), and it is possible to produce these harmonics with compact setups. The conversion efficiencies of harmonics from bound electrons have been shown to be extremely large, and because of phase matching a beam of coherent extreme-UV radiation can be obtained.[7] Free-electron harmonics are even more promising in this respect since the plateau seen in atomic harmonic generation is not predicted to occur for this process and the radiation would extend into the kilo-electron-volt region of the spectrum. Combined with the fact that the calculated conversion efficiencies are significant it would be feasible to use this extreme-UV radiation for time-resolved and imaging experiments in chemistry and biology. There remain issues related to the source size and coherence of these harmonics. While these are not yet fully resolved, it can be expected that work in progress will provide information on this aspect.

This paper is organized as follows. In Section 2 we briefly describe the basic physics of the interaction of free electrons with light fields in the regime in which the motion becomes highly nonlinear. This is the well-known classical description that has been the centerpiece of theoretical understanding of this process for the last three decades. Modification of this for more realistic situations encountered in a plasma will be considered in later sections. A brief description is also given of the relevant processes that can generate high harmonics in underdense plasma, and the efficiencies of various processes are described with a brief review of previous results. The characteristic features expected if the harmonics are generated from free electrons are pointed out, which will serve as tests of nonlinear Thomson scattering. Section 3 provides relevant details of the experimental setup for studying high-order harmonic emission from the plasma. In Section 4 experimental results are presented, and consistency with relativistic Thomson scattering is verified. The consistency check is both qualitative and quantitative, and we seek to show that competing processes such as nonlinear mixing in the plasma can be ruled out. We then present experimental results and rigorous calcula-

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tions to show that the spatial profile of the harmonics can be explained if it is assumed that high-energy electrons play a significant role in the scattering process. Section 5 presents our conclusions and directions for future work.

2. NONLINEAR THOMSON SCATTERING

The interaction of free electrons with the electric field of light pulses has been studied for well over a century. In the limit of low fields the electron motion is linear and along the polarization of the light field, and the scattered radiation is at the frequency of the incident light field. This is the process of linear Thomson scattering,[8] which has been well known for well over a century. As the field strength increases, the electron starts to quiver at relativistic intensities in a nonlinear orbit. The equation of motion is given by

$$\frac{\mathrm{d}\mathbf{p}}{\mathrm{d}t} = e(\mathbf{E} + \mathbf{v} \times \mathbf{B}), \tag{1}$$

where $\mathbf{p} = m_0 \gamma \mathbf{v}$, $\mathbf{E}$ and $\mathbf{B}$ are the electric and magnetic fields associated with the laser light, $\mathbf{v}$ is the velocity of the electron, and γ is the relativistic correction factor given by $\gamma = (1 - v^2/c^2)^{-1/2}$.

Equation (1) can be recast into dimensionless form. It is easy to show that the intensity scale is set by a dimensionless parameter a_0, the value of which determines whether the electron motion is linear or nonlinear. This is related to whether the effect of the magnetic field on the motion of the electron needs to be considered. The parameter a_0 is given by

$$a_0 = \frac{eE}{m\omega c} \approx 10^{-9} \sqrt{I\,(\mathrm{W\ cm^{-2}})}\ \ \lambda\ \ (\mu\mathrm{m}), \tag{2}$$

where ω is the frequency of the incident laser light. For values of $a_0 \ll 1$ the electron motion is linear. The nonlinear regime is reached when a_0 approaches unity. It is easy to see from approximation (2) that this corresponds to intensities of approximately $10^{18}\ \mathrm{W\,cm^{-2}}$. The ultrarelativistic limit corresponds to $a_0 \gg 1$. In this paper we are primarily interested in the case $a_0 \approx 1$. One can solve for the momentum and position by solving the Hamilton–Jacobi equation to obtain exact analytic expressions in some simple, though important, cases.[3] For the case in which the vector potential of the electric field can be expressed in the form $\mathbf{A}(\eta) = f(\eta)\mathbf{a}(\eta)$, where $\mathbf{a}(\eta)$ is a periodic function of η, $f(\eta)$ is a suitable pulse-shape function, and $\eta = \omega t - \mathbf{k}\cdot\mathbf{r}$, it is easy to show that for an electron initially at rest the trajectory is given by

$$\mathbf{r}(\eta) = \mathbf{r}_0 + \frac{1}{k}\int_{\eta_0}^{\eta} \frac{e\mathbf{A}(\eta')}{m_0 c}\,\mathrm{d}\eta' + \frac{\mathbf{k}}{k^2}\int_{\eta_0}^{\eta}\left[\frac{e\mathbf{A}(\eta')}{m_0 c}\right]^2 \mathrm{d}\eta'. \tag{3}$$

This is the well-known figure-8 trajectory, with the linear term leading to the motion of the electron along the polarization vector of the light field and the quadratic term causing the electron to more along the propagation vector of the incident light. The velocity of the electron is then given by

$$\boldsymbol{\beta}(\eta) = \frac{[e\mathbf{A}(\eta)/mc] + (\hat{\mathbf{k}}/2)[e\mathbf{A}(\eta)/mc]^2}{1 + (1/2)[e\mathbf{A}(\eta)/mc]^2}, \tag{4}$$

where $\boldsymbol{\beta} = \mathbf{v}/c$ and the above equations describe motion in the laboratory frame. Knowing the time dependence of the electron coordinates and velocity, one can calculate the radiation pattern. The scattered power P per unit solid angle and unit frequency interval at an observation point defined by the unit vector n is then given by

$$\frac{\mathrm{d}^2 E}{\mathrm{d}\Omega\,\mathrm{d}\omega} = \frac{(e\omega)^2}{4\pi^2 c^3}\left|\int_{-\infty}^{\infty} \mathbf{n} \times [\mathbf{n} \times \mathbf{v}(t)] \times \exp\left[i\omega\left\{t - \frac{\mathbf{n}\cdot\mathbf{r}(t)}{c}\right\}\right]\mathrm{d}t\right|^2. \tag{5}$$

To simplify the calculations, one calculates the radiation pattern in the frame in which the electron is at rest and then transforms back to the laboratory frame. As a result of the figure-8 motion, the electron radiates at integral multiples of the fundamental frequency with a radiation pattern that has a characteristic angular distribution. The fundamental frequency is itself not well defined for large a_0, but for the case in which a_0 does not significantly exceed unity it is approximately equal to the laser frequency (see below). A closed-form solution for the radiation pattern can be obtained only for the case of circular polarization.

There are several characteristics of this process, which are of interest because they serve to provide signatures of nonlinear Thomson scattering. It is well known that in the case of harmonic generation from bound electrons in gases there are no even-order harmonics.[9] This arises from the fact that the isotropy of the medium leads to zero even-order susceptibilities. Similarly, harmonic generation from the atomic susceptibilities is forbidden when the incident light is circularly polarized.[10,11] Neither of these rules is obeyed when harmonics are generated by the interaction of free electrons with an intense light field. Thus one would expect to see even-order harmonics as well as substantial harmonic production when circularly polarized light is used if nonlinear Thomson scattering leads to the generation of harmonics. For the case of atomic harmonics, rescattering of the electrons from the ion core leads to the generation of light, and clearly this process should depend quadratically on the electron density.[12] In actual experiments the dependence may be less than two because of the loss of phase matching as a result of ionization in the medium. In the treatment of nonlinear Thomson scattering a single free electron is considered. The complete radiation pattern will be obtained by summing over all the electrons. Thus the strength of harmonics emitted will scale linearly with the density of electrons in the absence of any coherent effects. For an incoherent process the emitted radiation should therefore be a sum of the radiation of the individual electrons, and the strength of the harmonics should scale linearly with the density of the plasma.

Harmonic generation from gases because of bound electrons has been a well-studied process for a long time now. It has been shown to be a very efficient process for the generation of very UV radiation. It is well known that atomic harmonics are produced at much lower intensity ($\approx 10^{13}$ W cm^{-2}), and their yield scales as I^n till saturation is reached because of depletion of the focal volume. In any experiment on the generation of relativistic harmonics a strong contribution from atomic harmonics is to be expected.

Fortunately atomic harmonics have characteristics very different from free-electron harmonics, making it easy to distinguish between the two processes. Two differences have already been noted—namely, the dependence on the electron density and the production of harmonics with circularly polarized light. It is also important to note that the threshold for production of atomic harmonics is orders of magnitude smaller than that from free electrons. All experiments that achieve high intensities use tight focusing of the incident laser beam. Typically, for a focal spot of 10 μm, intensities that are in excess of the threshold for atomic harmonic production would cover a spatial extent ~3 orders of magnitude larger than that for relativistic harmonics when a peak intensity of 10^{18} W cm^{-2} is reached. Therefore for a tight-focusing geometry with Gaussian beams the atomic harmonics should be produced in a much larger spatial region than are the harmonics from free electrons, which would be emitted only from regions in which the highest intensities are reached. Moreover, the high harmonics from bound electrons are phase matched, and a large amount of ionization would tend to destroy the phase matching. Thus at high intensities, well above the threshold for producing multiply ionized atoms, there should be an enhancement of harmonics from free electrons and a depletion of harmonics from the bound electrons.

Early work on harmonics that are produced by relativistic Thomson scattering was on the low-order (second and third) emission in the UV–visible range. Definitive signatures for this process were obtained for both incoherent and coherent emission. In the former case the polarization dependence of the emission was observed at 90° to the incident laser direction.[6] It was found that the harmonics were dependent on the polarization and resulted in a multilobed structure exactly as predicted by theory. The scaling with number density was also established to be nearly linear, as is expected for incoherent process. Subsequent experiments were used to study the third harmonic in the forward direction.[13] It was found that the harmonics were distributed in a strong forward cone. The wavelength spectrum showed a broad emission with a narrow peak. Experiments carried out with two beams showed that the broad component of the spectrum disappeared when the plasma was preionized with the second harmonic of the laser leaving only the narrow component, the latter scaling linearly with density as in the incoherent case. The third harmonic yield was found to decrease at high densities because of ionization-induced defocusing of the beam. These two experiments are possibly the only known observations of nonlinear Thomson scattering. In addition, the fact that harmonics were produced when the incident laser beam was circularly polarized without any significant reduction in the yield was also cited as evidence for this process.

However, as is well known, in a plasma, harmonics, especially the low-order ones, can be produced by many different mechanisms. In fact other groups have observed low-order harmonics in the same regime; these harmonics were attributed to mechanisms other than relativistic Thomson scattering. The work of Krushelnick *et al.*[14] claimed to show that the second harmonic was produced by Raman processes in a plasma and scaled quadratically with the number density. Theoretically it has been predicted that relativistic Thomson scattering should be a very efficient process, especially for the generation of high-order harmonics. Thus it is imperative to look at much shorter wavelengths, first because it would serve to provide more definitive proof of relativistic scattering processes driven by intense light fields. In particular it has been found that there is substantial production of harmonics in the forward direction, in contradiction to what is predicted by theory. In fact the work of Sarachik and Schappert[3] shows that the harmonics should be identically zero along the laser propagation vector and should peak at some angle away from the laser vector, with the angle determined by the intensity of the incident laser light. A study of the higher-order harmonics, which would have negligible contribution from various other scattering processes, would serve to clarify this point. Second, the promise of an efficient source of extreme UV radiation for applications in imaging and real-time dynamics of chemical processes with high spatial resolution, one that does not suffer from the shortcomings of conventional atomic harmonic generation, has also spurred further research.

3. EXPERIMENTAL SETUP

The experiments were performed with a hybrid titanium:sapphire–neodymium:glass laser system that produced pulses of 400 fs in duration at 1.053 μm with a maximum output energy of 6 J. The peak power delivered at the experimental setup is 5 TW. The 50-mm-diameter beam with a flattop profile was focused onto the front of a supersonic gas jet with an *f*/3.3 gold-coated parabolic mirror. The focal spot under optimum conditions had a diameter of 10–12 μm (FWHM) and had a Gaussian-like profile, which contained ~60% of the total energy of the pulse. A large-diameter (≈ 100 μm) spot contains the remaining 40% of the pulse energy. The peak intensity available in our experiments is 5×10^{18} W cm^{-2}, which corresponds to a maximum $a_0 \approx 2$. Thus the electron motion would be well into the relativistic regime at the highest laser power available.

It is known that under the conditions of our experiment there is a strong self-channeling of the laser beam in the plasma because of relativistic self-focusing. Concomitantly, a high-energy electron beam is produced whose angular spread decreases as the laser power is increased.[15,16] At the highest power the electron beam may have an angular divergence of less than 1°. These well-established results are used as diagnostics in our experiment to ensure optimal coupling of the laser beam into the plasma. Specifically, the channeling is monitored by side imaging the Thomson scattered radiation from the plasma. The high-energy electron beam is measured by recording the fluorescence from a LANEX screen. At peak powers greater than 2 TW a plasma channel of 1 mm in length is obtained, which is the same as the length of the gas jet under conditions where the laser is focused on the front of the gas pulse (gas–vacuum interface). It is to be noted that the Raleigh range is $\approx$300 μm. The electron beam has an angular divergence of 1°–2° when the laser focus is at best position with respect to the gas jet. In our experiments it is found that there is an uncertainty of $\pm$50 μm in the position of the gas jet. This is consistent with the fact that the gas jet has a flow pattern such that the density increases from background level to maximum value over a distance of $\sim$100 μm.

High-harmonic emission from the plasma is measured with a Saye–Namioka spectrometer with a range of 30–200 nm. This covers the harmonic range of 6–30 for the fundamental wavelength of 1.053 μm. The spectrometer consists of a toroidal grating (1200 lines/mm) with a radius of curvature of 1 m. The resolution of the system is $\approx$0.3 nm. The spectrometer is configured so that it ideally acts as an imaging system with a magnification of 1:1. The imaging characteristic of the spectrometer implies wavelength resolution in the horizontal plane and spatial information in the vertical plane. Harmonics are detected with an imaging multichannel plate (MCP) (dual plate + phosphor screen) coupled to a high-sensitivity high-dynamic-range CCD camera. In the current experiment the laser beam is directed along the spectrometer axis. The gas jet and the MCP are located such that the former is on the object plane and the latter is on the image plane of the spectrometer. This is the so-called slit-free geometry, because in itself the small spatial extent of the plasma located at the object plane of the spectrometer acts as an entrance slit.

The spectrometer and the experimental chamber are differentially pumped by means of turbomolecular pumps. A slit of diameter 2 mm placed next to the gas jet allows the entire laser beam to enter the spectrometer and allows differential pumping of the system. Differential pumping is necessary because the vacuum requirements for the MCP are much more stringent than for other parts of the experiment. Typically the background pressure in the experimental chamber is in the range 10^{-4}–10^{-5} Torr, while the spectrometer chamber is maintained at a base pressure of 10^{-6} Torr. This increases to 10^{-2} Torr in the former and 10^{-5} Torr in the latter when the supersonic nozzle is operated at the highest backing pressures. The typical duration of the gas pulse is $\sim$10 ms. The maximum density reached in the interaction region is $\sim 10^{19}\,\mathrm{cm}^{-3}$. The gas pressure at which best results are obtained depends on the gas used. The use of a tight-focusing geometry leads to significant mismatch in the focal lengths of the spectrometer and the parabola. As a result the ideal magnification of 1:1 is not attained, although the imaging condition is still satisfied.

4. RESULTS AND DISCUSSION

Figure 1 shows the spectrum of high harmonics obtained from underdense helium plasma at an intensity of $2 \times 10^{18}\ \mathrm{W\,cm^{-2}}$ and for linearly polarized light. Because of the high resolution of the spectrometer and the limited size of the MCP, it is possible to obtain images of only a few harmonics for a given laser shot; in order to capture the complete spectrum, many laser shots are necessary to span the entire range of the grating. From the figure it is apparent that in addition to the seventh and ninth harmonic, which would be expected to arise from bound electrons long seen in atomic harmonic generation, there is a clear signal coming from the eighth harmonic. It was confirmed that this disappears at lower intensities or gas densities. The emission in the even order was weaker than that in the odd orders even at the highest intensities. For the case shown here the eighth order was eight times smaller than the seventh order and six times smaller than the ninth order. Data taken over the entire spectrometer range, which goes down to the 30th harmonic, reveals a similar trend–namely, that the even harmonics are present though weaker than the odd harmonics. The presence of the even harmonics is promising, and the fact that they are absent for intensities $< 10^{17}\ \mathrm{W\,cm^{-2}}$ ($a_0 \approx 0.2$) seems to be consistent with the fact that scattering from free electrons may be the major contributing factor. The presence of even-order harmonics, though promising, is in itself not sufficient or conclusive as far as a determination of the underlying mechanism is concerned. A series of further tests was carried out to gain insight into the physical process that led to the emission of even-order harmonics from the plasma.

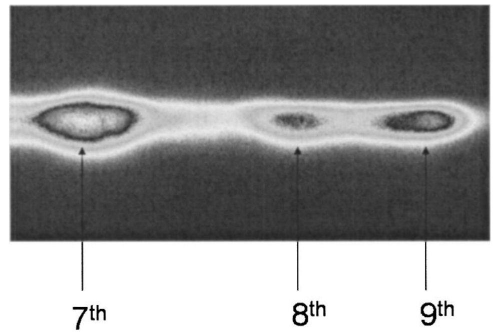

Fig. 1. Spectrum of high harmonics from helium at an intensity of $2 \times 10^{18}\ \mathrm{W\,cm^{-2}}$ and electron density of $10^{19}\,\mathrm{cm^{-3}}$. Shown are the seventh, eighth, and ninth harmonics.

As described previously, the very UV spectrometer used in these experiments is configured to operate as an imaging device. It has also been noted that the magnification of the system is not 1:1 on account of the focal-length mismatch. Hence a measurement of the magnification is required. Calibration of this was done with a helium–neon beam focused on the object plane and looking at the transmitted spot on the image plane by use of the zero order of the grating. The focused spot size on the object plane was changed by varying the input beam size, and the size of the image on the MCP plane was recorded on a screen with a CCD. It was found that the object size and the image size in the vertical direction were related in linear fashion as expected. On the basis of this calibration the magnification of the system could be obtained. Hence the actual spatial extent of the high-order harmonics could be calculated after a correction for the wavelength at which measurements are carried out.

Two different experiments were carried out to obtain data on the spatial region in which the harmonics were produced. As has been noted previously, the odd harmonics were stronger than the even-order harmonics. This is true for helium but not in other gases, a point that will be discussed below. One may take as a working hypothesis that the odd orders have contributions from both the bound and free electrons, while the even orders arise only from the free electrons. At low intensities only harmonics from bound electrons should be present, and one may expect that as a_0 approaches unity contributions from nonlinear Thomson scattering should modify the spectrum. Figure 2(a) shows the results obtained when linearly polarized light at low intensity and low density is used. As expected, only the odd order (seventh) is seen, and the spatial extent is large, since bound harmonics would be produced in the focal volume when the intensity exceeds $\approx 10^{13}$ W cm^{-2}. Shots taken for other harmonic orders show similar results, namely, the absence of all the even harmonics and large spatial extent of the region in which odd harmonics are produced (typically 100 μm). Next, data were obtained at high intensity to let us look at the contribution of free electrons to the harmonic generation process. To isolate the contribution from free electrons the bound electron signal was eliminated by use of circularly polarized light. The results are shown in Fig. 2(b). It is immediately obvious that both the seventh and the eighth harmonics are present and that their spatial extent is significantly smaller ($\approx$20 μm) compared with the atomic harmonics. Figures 3(a) and 3(b) show the corresponding lineout for these two cases. It is also obvious that when linear polarization is used the odd harmonics will always have a larger spatial extent than the even harmonics, since the focal volume in which the former is produced is larger than for the latter. This can be understood on the basis of the fact that the odd harmonics produced with linear polarization have contributions from both the bound and the free electrons. Figure 3(c) shows the spatial extent of the seventh and eighth harmonics at $I = 2 \times 10^{18}$ W cm^{-2} when linear polarization is used. The spatial extent of the seventh is greater than that of the eighth, though the difference is not so dramatic as when circular polarization is used, and a similar trend holds for higher-order harmonics, too. This may arise from the fact that at this high intensity the signal from atomic harmonics is no longer dominant because of a loss in efficiency of harmonic generation from bound electrons.

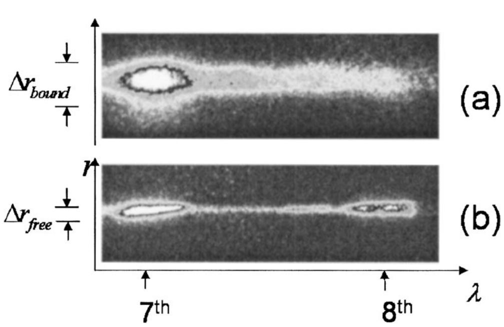

Fig. 2. Spatial profile of the high-order harmonics for (a) $I = 5 \times 10^{16}$ W cm^{-2}, $n = 10^{17}$ cm^{-3}, and linear polarization and (b) $I = 5 \times 10^{17}$ W cm^{-2}, $n = 10^{18}$ cm^{-3}, and circular polarization.

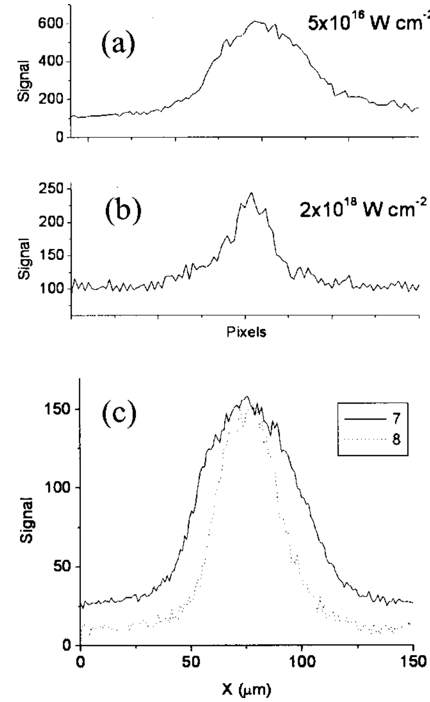

Fig. 3. (a), (b) Lineouts showing the transverse extent of the harmonics (see text); (c) data corresponding to Fig. 2(c) for $I = 2 \times 10^{18}$ W cm^{-2}, $n = 10^{19}$ cm^{-3}, and linear polarization.

Conventional theory predicts that the efficiency of harmonic generation should depend on $n_i n_e$, where n_i and n_e are the ion and electron densities, respectively. Thus they should scale as n^2, where n is the gas density. In contrast, if the free-electron scattering is considered to be incoherent then the dependence should be linear with gas density. Figure 4 shows the results obtained for the 11th and 12th harmonics in argon for various gas densities. In the former case there is initially a quadratic dependence, while the dependence is linear in the latter case. Both signals saturate and then decay, an effect that is well known to be due to ionization-induced defocusing[17] of the laser beam; that is, the peak intensity available at the focus decreases with increasing electron density. It should be noted that the dependence on the density for the even orders is not exactly linear—in fact in our experiments it ranged from 1.2 to 1.4 for the various harmonic orders from 3 through 30. While it is possible that there may be some collective effect modifying the dependence on density, there is currently no understanding of the underlying cause. However, it was not possible to obtain any systematic trends for this factor because of the low repetition rate of the laser used in these experiments. However, that the dependence is significantly smaller than quadratic lends credence to the fact that the emission observed is an incoherent addition of independent scattering events.

Thus far we have hypothesized and tentatively established that the high harmonics seen in our experiments arise from the radiation that is due to the acceleration of electrons moving at relativistic speeds. If this is the case, one can obtain more definitive evidence by altering

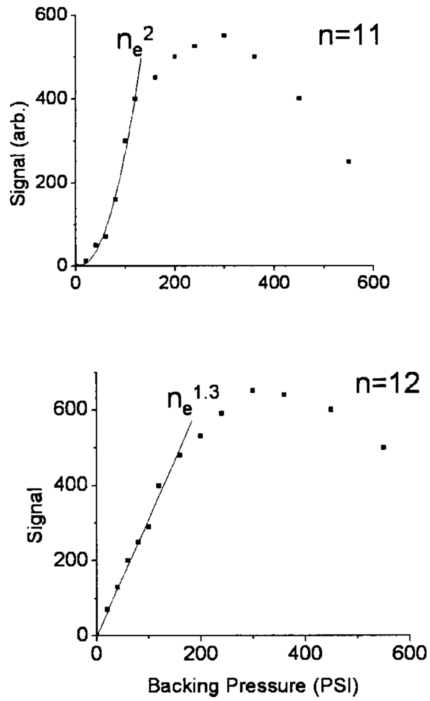

Fig. 4. Dependence of high-harmonic generation on the number density. The odd harmonic scales quadratically with density, while the even shows a dependence that is close to linear.

the free-electron density. Ideally a preionized plasma should be used with counterpropagating laser beams. However, this experiment is complicated because of the requirements of splitting a high-power beam and matching the foci in the interaction region. A simpler approach is to use different gases, which would naturally alter the density of free electrons in the focal region. To this end we used molecular nitrogen and argon in addition to helium and compared the spectra obtained in the three cases.

Figure 5 shows the spectra obtained when N_2 and argon are used for the case of the seventh, eighth, and ninth harmonics. It is clear from the figure that there is an enhancement of the eighth relative to the seventh and ninth harmonics when argon is used as compared with N_2. A more accurate comparison can be made by means of the lineouts obtained from these spectra. Figure 6 shows the lineouts obtained for the three gases for identical laser conditions and the same backing pressure for the gas jet. It should be noted that the MCP was operated at different voltages for the three gases, and thus only a relative comparison can be made for the corresponding spectra. At the intensities used in our experiment helium is completely ionized, while N_2 and argon would also undergo significant ionization; thus the electron density in the latter case would be substantially larger than that when helium is used. Excessive ionization would produce significant defocusing of the beam, and an intensity of 5×10^{17} W cm^{-2} was found to be optimal. It should be noted that at this intensity the channeling of the laser beam in the plasma is strong, but it was found that the plasma channel was significantly shorter in N_2 and argon ($\approx$0.6 mm) as compared with helium ($\approx$0.9 mm). It is known that the extent of self-focusing of the beam in the plasma is reduced beyond a certain density, again because of excessive ionization.

The data show that in case of helium the odd harmonics are strongest and the even order (eighth) is significantly weaker. When N_2 is used there is a significant enhancement of the even harmonic, and this effect is heightened in the case of argon to the extent that there is more emission in the eighth as compared with the ninth. It is easy to rationalize these data on the basis of our model that the even harmonics are produced entirely because of the free electrons. As the free-electron density increases when higher-*Z* gases are used, it leads to enhancement of scattering processes involving them. A weaker channel is evidence of the higher electron density at the focus. However, on the basis of channeling data it is clear that this large electron density leads to a defocusing of the beam. The consequent loss of intensity results in a lower harmonic emission in all orders, moreover, there is significant loss of phase matching for the atomic harmonics, too, resulting in lower emission in the odd or-

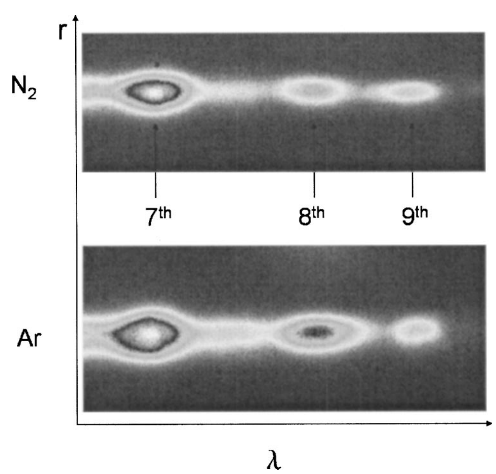

Fig. 5. Harmonic generation in high-*Z* gases; $I = 5 \times 10^{17}$ W cm^{-2}, $n = 10^{18}$ cm^{-3}, linear polarization.

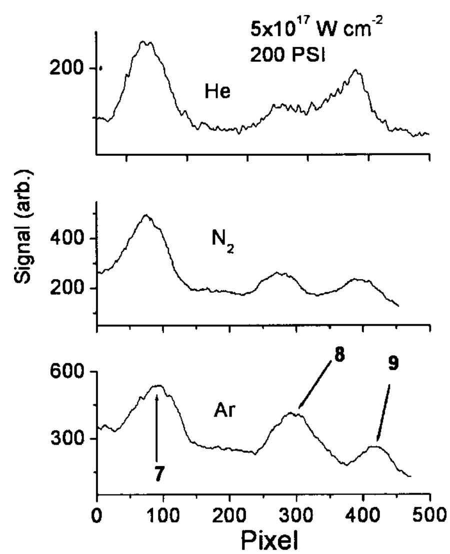

Fig. 6. Lineouts corresponding to the conditions of Fig. 5. Note the enhancement of the even order as compared with the odd as the free-electron density is increased when higher-atomic-number gases are used.

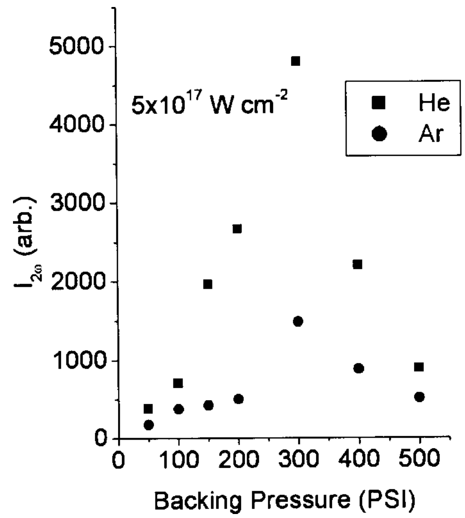

Fig. 7. Efficiency of second-harmonic emission in helium and argon. While both show the same qualitative behavior, the argon produces significantly less second harmonic than does helium.

ders. Thus we have a situation where the production of even-order harmonics is enhanced, while that of the odd orders is reduced, as one goes from low-Z to high-Z gases. Moreover, the entire process should decrease in efficiency. The experimental data shown are completely consistent with the above results.

All the above experiments show that the harmonics observed in our experiments are indeed produced from free electrons. However, it is possible that they may also be generated from nonlinear mixing processes for the plasma. This is improbable because the nonlinear coefficients for higher-order mixing processes are extremely small. However, it is well known that the plasma generates the second harmonic very efficiently owing to the gradient. It is therefore important to rule out this as a possible mechanism. To this end experiments were done to measure the amount of second-harmonic light. In these experiments the second-harmonic light was collected in the forward direction over the entire cone angle of the laser. It is known that the second harmonic is emitted in a cone, which is smaller than the cone angle of the fundamental, and thus this experimental arrangement ensures 100% collection efficiency for emission at this wavelength. Figure 7 shows the results obtained from helium and argon as a function of the number density. It is clear that under the conditions for Fig. 7 and generally at every density the argon produces less second harmonic than does helium. The shorter channel in argon as compared with helium is consistent with this because a lower intensity is reached. Moreover the second harmonic scales as n^2, unlike the even-order harmonics, which scale linearly with n. Thus the second-harmonic emission is not correlated with the emission of even-order harmonics, and they follow completely different scaling laws. Because the plasma gradient and the even harmonics are not correlated, we can rule out nonlinear mixing as a possible mechanism for the generation of high-order harmonics.

Thus far we have discussed the overall spectral features of the harmonics. Much of the discussion has been qualitative, though the evidence presented points strongly to the fact that we are indeed seeing high-order harmonic generation from free electrons. However a direct comparison with theoretical predictions would be satisfying. Theoretically the angular distribution of these harmonics is well known and would provide a simple and direct method for comparing experiment and theory. To let us look at the angular distribution of the emitted beam of harmonic radiation, a slit was placed in the object plane of the spectrometer. The gas nozzle was moved 8 cm behind the slit. The idea is that the harmonics would be imaged on the slit. The size and location of the slit was chosen based on the fact that the fundamental beam completely fills the slit. From experience one would expect the harmonics to have a smaller divergence than the fundamental. The system was calibrated by illuminating the slit with a helium–neon beam and recording the size of the image on the MCP plane. The input beam size was varied, and a plot was made of the vertical size of the spot on the MCP screen as a function of the input beam size. From the size of the image on the MCP, after correction for the wavelength, the size of the beam on the slit could be inferred (see the discussion in the experimental setup). Since the size of the beam on the slit and the distance of the slit from the center of the gas jet is known, it is easy to obtain the divergence of the harmonics. By this method in principle one obtains the complete angular profile of the harmonic beam. Figure 8 shows the pattern obtained on the MCP in this case for the 11th and the 12th harmonics. The multiple lobes correspond to single slit diffraction of the high-order harmonics. On the basis of the measured magnification of the system, it is found that the harmonic beam has a divergence of $\sim 3°$ at the 6th harmonic, which decreases to $\approx 2°$ at the 20th harmonic. The harmonics are thus emitted in a forward direction with very small angular divergence.

At first sight there is a significant discrepancy between experiment and theory. It is well known that the harmonics peak off axis, and it is well known that there can be no radiation other than the fundamental along the forward direction. Figure 9 (dotted curve) shows the calculated spectrum for a typical case (sixth harmonic). Does this mean that some process other than Thomson scattering is leading to the observed results? We should keep in mind that the spectrum depicted in Fig. 9 is for the case in which the electrons are initially at rest and the emission cone for both electrons and the laser beam has been neglected. These are drastic simplifications, and their

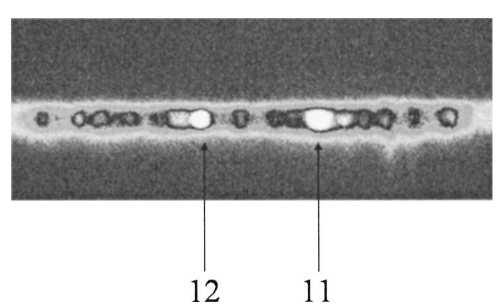

Fig. 8. Image of the high-order harmonics after diffraction through a 200-μm slit. The calculated angular divergence for the harmonics shown is $\sim 2.4°$.

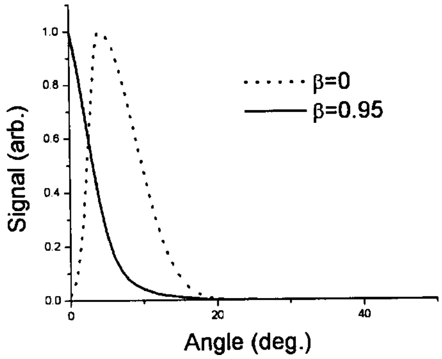

Fig. 9. Spatial profile of harmonics for the case in which the electron starts at rest (dotted curve) and for an initial velocity of 0.95*c* (solid curve).

validity must be reconsidered. First, it is well known that under the conditions of our experiment a well-collimated, high-energy electron beam is produced.[16] In fact this is used to optimize the plasma conditions. From independent experiments it is known that this electron beam has a typical energy that lies in the range 100 keV–10 MeV with a most probable energy of ~1 MeV. The physics underlying the production of this high-energy electron beam has been discussed elsewhere.

As a first step the angular distribution of harmonics with a nonzero initial electron velocity needs to be calculated. This is a simple extension of the results of Sarachik and Schappert,[3] and the results have been available for a number of years.[18] For an electron starting with an initial velocity v_0 the coordinate and velocity are given by the following expressions:

$$\mathbf{r}(\eta) = \mathbf{r}_0 + \frac{1}{k}\int_{\eta_0}^{\eta} \frac{\gamma_0 m_0 \mathbf{v}_0 + e\mathbf{A}(\eta')}{\gamma_0 m_0 c\left(1 - \dfrac{\mathbf{k}\cdot\mathbf{v}_0}{kc}\right)} \mathrm{d}\eta' + \frac{\mathbf{k}}{k^2}\int_{\eta_0}^{\eta} \frac{\dfrac{1}{2}\left[\dfrac{e\mathbf{A}(\eta')}{\gamma_0 m_0 c}\right]^2 + \left[\dfrac{e\mathbf{A}(\eta')}{\gamma_0 m_0 c}\right]\cdot\left(\dfrac{\mathbf{v}_0}{c}\right)}{\left(1 - \dfrac{\mathbf{k}\cdot\mathbf{v}_0}{kc}\right)^2} \mathrm{d}\eta', \tag{6}$$

$$\boldsymbol{\beta}(\eta) = \frac{\boldsymbol{\beta}_0 + \dfrac{e\mathbf{A}(\eta)}{\gamma_0 m_0 c} + \hat{\mathbf{k}}\left\{\dfrac{\dfrac{1}{2}\left[\dfrac{e\mathbf{A}(\eta)}{\gamma_0 m_0 c}\right]^2 + \left[\dfrac{e\mathbf{A}(\eta)}{\gamma_0 m_0 c}\right]\cdot\boldsymbol{\beta}_0}{1 - \hat{\mathbf{k}}\cdot\boldsymbol{\beta}_0}\right\}}{1 + \left\{\dfrac{\dfrac{1}{2}\left[\dfrac{e\mathbf{A}(\eta)}{\gamma_0 m_0 c}\right]^2 + \left[\dfrac{e\mathbf{A}(\eta)}{\gamma_0 m_0 c}\right]\cdot\boldsymbol{\beta}_0}{1 - \hat{\mathbf{k}}\cdot\boldsymbol{\beta}_0}\right\}}, \tag{7}$$

where $\gamma_0 = (1 - {\beta_0}^2)^{-1/2}$ depends on the initial electron velocity. As before, the radiation spectrum is to be calculated by use of Eq. (4). Our calculation also takes into account the profile of the electron beam and the fact that the laser has a divergence angle of 30°. Figure 9 (solid curve) shows the radiation spectrum obtained when the electron energy is 1 MeV. The calculated width is ~3.5°, which is excellent agreement with the measured width of 2.8°.

5. CONCLUSIONS

In this paper we have described new results on the generation of high harmonics in underdense plasmas. It has been shown that relativistic Thomson scattering produces a significant amount of very UV light. The emitted radiation is produced as a beam with very small angular divergence. The production of this beam is shown to result from the fact that high-energy electrons produced in the plasma play a significant role in the scattering process. Although the total conversion efficiency into high-order harmonics has not been measured yet, from data for atomic harmonics it is estimated to be $\approx 10^{-7}$. Since the conversion efficiency scales as γ,[6] it will be possible to obtain a kilo-electron-volt x-ray beam by using a higher-energy electron beam from a short-pulse high-intensity laser.

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