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High harmonic generation in relativistic laser–plasma interaction[a)]

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High harmonics generated due to the scattering of relativistic electrons from high intensity laser light is studied. The experiments are carried out with an Nd:Glass laser system with a peak intensity of 2×10^{18} W cm^{-2} in underdense plasma. It is shown that, at high intensities, when the normalized electric field approaches unity, in addition to the conventional atomic harmonics from bound electrons there is significant contribution to the harmonic spectrum from free electrons. The characteristic signatures of this are found to be the emission of even order harmonics, linear dependence on the electron density, significant amount of harmonics even with circular polarization and a much smaller spatial region over which these harmonics are produced as compared to the atomic case. Imaging of the harmonic beam shows that it is emitted in a narrow cone with a divergence of 2 to 3 degrees. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1470167]

I. 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, routinely produce multiterawatt pulses of subpicosecond duration, which can be focused to obtain peak intensities of 10^{19} W cm^{-2}. At these ultrahigh intensities, electrons quiver with velocities close to the velocity of light and the motion of even free electrons becomes highly nonlinear. As predicted many years ago, the nonlinear motion of relativistic electrons should lead to the emission of harmonics, a process known as nonlinear Thomson scattering.[1–3]

Over the years several efforts were made to detect this process. However, only in the late 1990's were the first unambiguous signatures of this process detected in experiments where a high intensity laser pulse interacted with underdense plasma. It was shown that the second and third harmonic emitted from the plasma had the characteristic angular distributions, as predicted by theory, and scaled linearly with the number density, as expected since the process is incoherent.[4] 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, with possible applications to processes occurring in astrophysical plasmas. 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 (micron 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 ultraviolet (XUV) radiation can be obtained.[5] 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. Combined with the fact that the calculated conversion efficiencies are expected to be of the order of 10^{-4}–10^{-5}, which is comparable to what is attained in harmonic generation from atoms, it would be feasible to use this XUV radiation for time resolved and imaging experiments in chemistry and biology. There remain issues related to the source size and coherence of these harmonics and these are the subjects of current study.

This paper is organized as follows. In Sec. II, we briefly describe the basic physics of the interaction of free electrons with light fields in the regime when the motion becomes highly nonlinear. A brief description is also given of the relevant processes which can generate high harmonics in underdense processes, and the efficiency of various processes is described with a brief review of previous results. The characteristic features expected of the harmonics that are generated from free electrons are pointed out. These will serve as tests of nonlinear Thomson scattering. Sec. III provides details of the experimental setup for studying high order harmonic emission from the plasma. In Sec. IV results are presented on our studies and their consistency with relativistic Thomson scattering is verified. Section V presents our conclusions and directions for future work.

II. 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. As the field strength increases, the electron starts to quiver at relativistic intensities in a nonlinear orbit. The primary quantity of interest is the normalized field strength a_0 given by

$$a_0=\frac{eE}{m\omega c}\sim10^{-9}\sqrt{I(\mathrm{W/cm^2})}\lambda(\mu\mathrm{m}).$$

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For values of $a_0 \ll 1$ the electron motion is linear. The nonlinear regime is accessed when a_0 approaches unity, which corresponds to an intensity $\approx 10^{18}\ \mathrm{W\,cm^{-2}}$. The ultrarelativistic limit corresponds to $a_0 \gg 1$. In this paper, we will be primarily interested in the case $a_0 \approx 1$. It can be shown that in this case the electron moves in the well-known figure-eight orbit. A consequence of this figure-eight motion is that the electron radiates at integer multiples of the fundamental frequency with a radiation pattern which has a characteristic angular distribution. There are several characteristics of this process which are of interest, since they serve to provide signatures of nonlinear Thomson scattering. First, unlike the atomic case, even harmonics of the laser should be produced even for an isotropic medium. Also substantial harmonic generation should occur even when circularly polarized light is used. For an incoherent process, the emitted radiation should 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 due to bound electrons has been a well-studied process for a long time. It has been shown to be a very efficient process (conversion efficiencies range from 10^{-7}–10^{-5})[5,6] for the generation of vaccum ultraviolet (VUV) radiation. It is well known that atomic harmonics are produced at much lower intensity ($\approx 10^{13}\ \mathrm{W\,cm^{-2}}$) and their yield scales as I^n until saturation is reached because of the 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 very different characteristics from free electron harmonics, making it easy to distinguish between the two processes. First, from symmetry considerations it can be shown that gases do not produce even harmonics and neither is there any harmonic emission when circularly polarized light is used.[7] Second, since these harmonics are produced by rescattering of electrons from the ion from the ion core, the harmonic yield should scale quadratically with the gas density.[5] It is also important to note that atomic harmonics are produced in a large intensity range. Therefore, for a tight focusing geometry with Gaussian beams, the atomic harmonics should be produced in a much larger spatial region as compared to the harmonics from free electrons, which would be emitted only from regions where the highest intensities are accessed. Moreover, the high harmonics from bound electrons are phase matched and a large amount of ionization tends to destroy the phase matching. Thus at high intensities, well above the ionization 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 produced due to relativistic Thomson scattering was on the low order (second and third) emission in the visible and ultraviolet range. Definitive signatures for this process were obtained both for incoherent and coherent emission.[8] In the former case, the unique angular distributions of the harmonics and in the latter, preionized plasma, was used to prove that the harmonics observed were indeed produced by free electrons. The fact that harmonics were produced when the incident laser beam was circularly polarized was also taken to be evidence for this. Interest in studying the generation of high order harmonics stems from primarily two reasons. On the one hand, nonlinear processes in the plasma from bound electrons can produce low order harmonics.[9] As discussed previously, high order harmonics produced due to nonlinear Thomson scattering will have very specific characteristics and it can be expected that simple experimental tests would serve to show whether short wavelength radiation is indeed produced by this process. The promise of an efficient source of XUV radiation has also spurred further research.

III. EXPERIMENTAL SETUP

The experiments were performed with a hybrid Titanium Sapphire–Neodymium Glass laser system that produced pulses of 400 fs duration at 1.053 μm with a maximum peak power of 5 TW. The 50 mm diameter beam was focused onto the front of a supersonic gas jet with an f/3.3 gold coated parabolic mirror. The focal spot had a diameter of 10–12 μm [full width at half maximum (FWHM)] and had a Gaussian like profile, which contained about 60% of the total energy of the pulse. A large diameter ($\approx 100\ \mu$m) spot contains the remaining 40% of the pulse energy. The peak intensity accessed is $2\times 10^{18}\ \mathrm{W\,cm^{-2}}$, which corresponds to an $a_0 \approx 2$.

It is known that under the conditions of our experiment there is a strong self-channeling of the laser beam in the plasma, and the production of a high energy electron beam with a small angular spread.[10,11] These 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 fluorescent ("LANEX") screen. At peak powers >2 TW a plasma channel of 1 mm 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. It is to be noted that the Raleigh range is ≈300 μm. The electron beam has an angular divergence of 1 to 2 degrees 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 ±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 about 100 μm.

High harmonic emission from the plasma is measured using a Saye-Namioka spectrometer with a range of 200–30 nm. This covers the harmonic range of 6–30 for a 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 reciprocal linear dispersion of the spectrometer is $8.3\ \mathrm{A\,mm^{-1}}$ and the resolution of the system is ≈0.3 nm. The spectrometer is configured so that it acts as an imaging system with a magnification of 1:1, i.e., the gas jet is on the entrance plane of the spectrometer (located at ≈1 m from the grating) and the detector, [microchannel plate (MCP)] is on the exit (object) plane of the spectrometer. Thus the spectrometer provides wavelength resolution in the horizontal

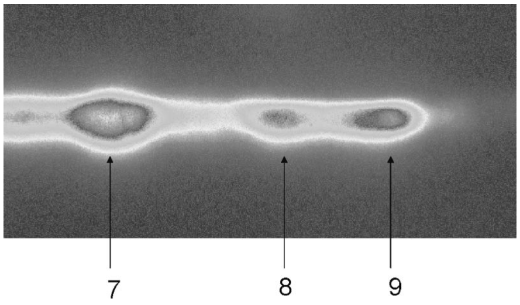

FIG. 1. Spectrum of high harmonics from Helium at an intensity of $2 \times 10^{18}\ \mathrm{W\,cm^{-2}}$ and electron density of $10^{19}\ \mathrm{cm^{-3}}$. Shown are the seventh, eighth, and ninth harmonics.

plane and spatial information in the vertical plane. Harmonics are detected using an imaging MCP (dual plate +phosphor screen) coupled to a high sensitivity high dynamic range CCD (charge-coupled device) camera. In the current experiment, the laser beam is directed along the spectrometer axis and the gas jet and the MCP are located at the object and image planes, respectively. This is the so-called slit free geometry because 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 laser beam to enter the spectrometer, as well as differential pumping. 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. Because of the mismatch in the focal lengths of the spectrometer and the parabola the ideal magnification of 1:1 is not attained although the imaging condition is still satisfied.

IV. RESULTS AND DISCUSSION

Figure 1 shows the spectrum of high harmonics obtained from underdense helium plasma at intensity of $2 \times 10^{18}\ \mathrm{W\,cm^{-2}}$ and for linearly polarized light. Because of the high resolution of the spectrometer it is possible to obtain images of only a few harmonics for a given laser shot and in order to see the complete spectrum many laser shots are required to span the entire range of the grating. From the figure it is apparent that in addition to the seventh and ninth harmonics, which would be expected to arise from bound electrons, there is a clear signal coming from the eighth harmonic. It was checked that this disappears at lower intensities ($<10^{17}\ \mathrm{W\,cm^{-2}}$) for all gas densities. The emission in the even order was much weaker than that in the odd orders. For the case shown here the eighth order was six times smaller than the seventh order and eight 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 weaker than the odd ones. 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. However, this in itself is not sufficient or conclusive and further tests were carried out to verify this hypothesis.

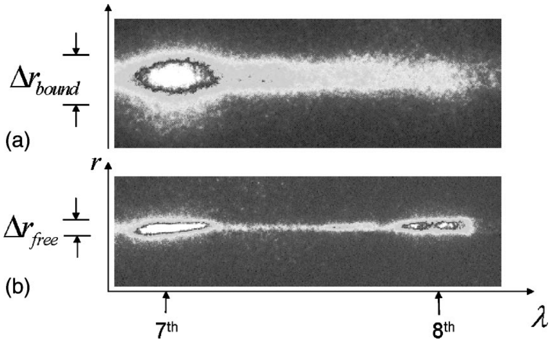

FIG. 2. Spatial profile of the high order harmonics (a) $I = 5 \times 10^{16}\ \mathrm{W\,cm^{-2}}$, $n = 10^{17}\ \mathrm{cm^{-3}}$ and linear polarization. The transverse extent of the image (FWHM) is ≈ 0.5 mm. (b) $I = 5 \times 10^{17}\ \mathrm{W\,cm^{-2}}$, $n = 10^{18}\ \mathrm{cm^{-3}}$ and circular polarization.

As described previously, the VUV spectrometer used in these experiments is configured to operate as an imaging device. Calibration of this was done by using a Helium–Neon laser beam focused on the object plane and looking at the reflected spot on the image plane using the zero order of the grating. Varying the input beam size changed the focused spot and the size of the image was recorded. It was found that the two were related in linear fashion and, therefore, the size in the vertical direction can be used to infer the actual spatial extent at the focus.

Two different measurements 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 always stronger than the even order harmonics. One may conclude that the odd orders have contributions from both the bound and free electrons while the even orders only arise from the free electrons. 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 as would be expected from the fact that bound harmonics would be produced in the focal volume when the intensity exceeds $\approx 10^{13}\ \mathrm{W\,cm^{-2}}$. 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). In order to isolate the contribution from free electrons these experiments were done at high intensity. The bound electron signal was eliminated by the use of circularly polarized light. The results are shown in Fig. 2(b). It is immediately obvious that both the seventh and eighth harmonics are present and their spatial extent is significantly smaller ($\approx 20\ \mu$m) as compared to the atomic harmonics. Figures 3(a) and 3(b) show the lineouts for two intensities under different polarization conditions, illustrating the point that the even and odd harmonics have very different spatial extents. It is also obvious that when linear polarization is used the odd harmonics will always have a larger

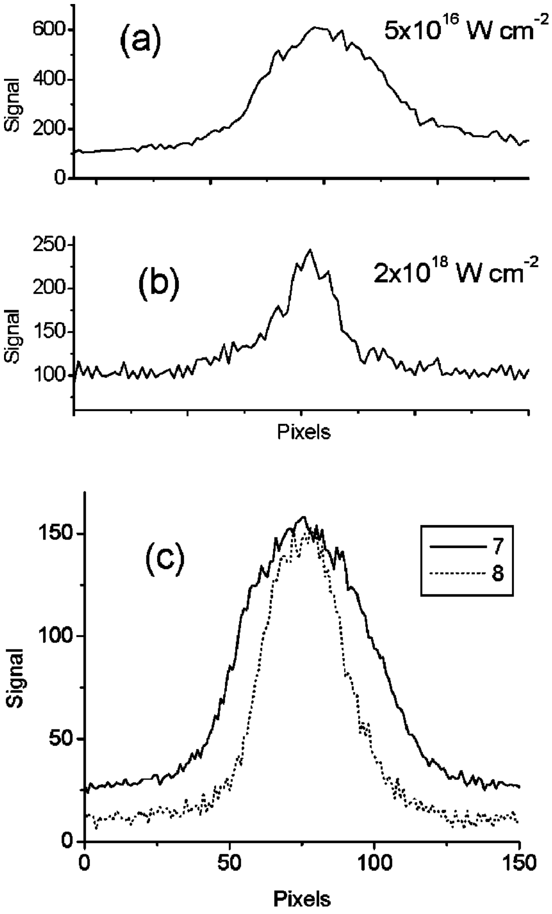

FIG. 3. Lineouts showing the transverse extent of the seventh harmonic (a), (b) data corresponding to Fig. 2 (c) $I=2\times10^{18}$ W cm^{-2}, $n=10^{19}$ cm^{-3} and linear polarization.

spatial extent than the even harmonics, since the focal volume in which the former is produced is larger than for the latter. Figure 3(c) shows the spatial extent of the seventh and eighth harmonics at $I=2\times10^{18}$ 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 as dramatic as when circular polarization is used. In fact the seventh harmonic has a width (FWHM) of 70 μm, while the eighth harmonic has a spatial extent of 30 μm. 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. This will be illustrated later when we consider the loss of phase matching due to ionization of the medium.

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 density, respectively. Thus they should scale as n^2, where n is the gas density. On the contrary, if the free electron scattering is considered to be an incoherent process from an ensemble of independent noninteracting electrons, the dependence should be linear with gas density. Figure 4 shows the results obtained for the 11th and 12th harmonics in Ar for various gas densities. In the former case, there is a quadratic dependence initially, while in the latter case, it is linear. Both signals saturate and then decay, a fact that is known to be due to ionization induced defocusing of the laser beam.[12] 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 to 30. However, it was not possible to obtain any systematic trends on in this factor because of the low repetition rate of the laser used in these experiments.

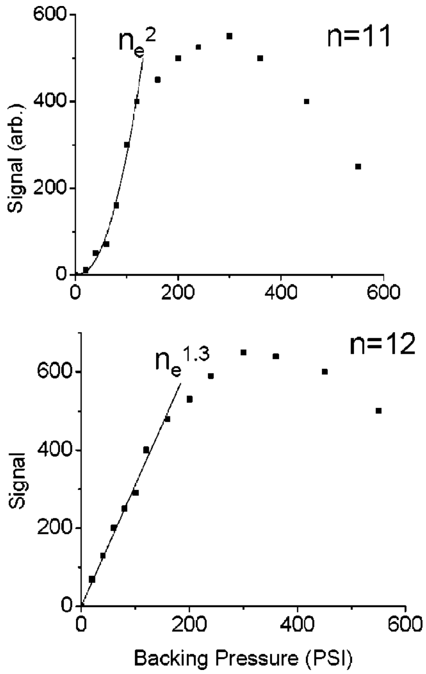

FIG. 4. Dependence of high harmonic generation on the number density. The odd harmonic scales quadratically with the density while the even shows a dependence which is close to linear.

Thus far we have hypothesized and tentatively established that the high harmonics seen in our experiments arise from the radiation due to the acceleration of electrons moving at relativistic speeds in the figure-eight motion. If this is the case, then one can obtain more definitive evidence by altering the free electron density. Ideally, a preionized plasma should be used with counter-propagating laser beams.[8] 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 that would naturally alter the density of free electrons in the focal region. To this end, we used 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 Ar 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 Ar is used as compared to 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 there are no wavelength shifts for the various harmonic orders when different gases are used. The MCP was operated at different voltages for the

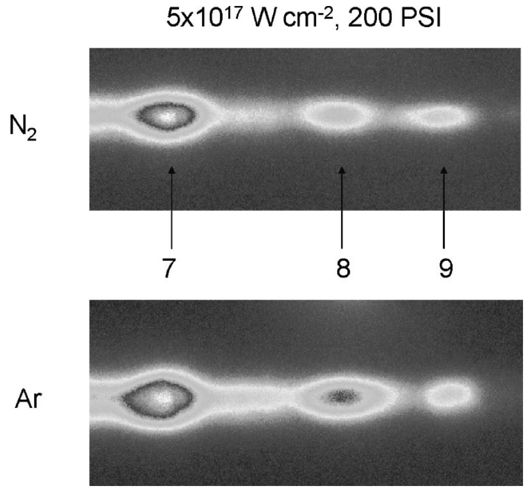

FIG. 5. Harmonic generation in high-*Z* gases. $I=5\times10^{17}\ \mathrm{W\,cm^{-2}}$, $n=10^{18}\ \mathrm{cm^{-3}}$, and linear polarization.

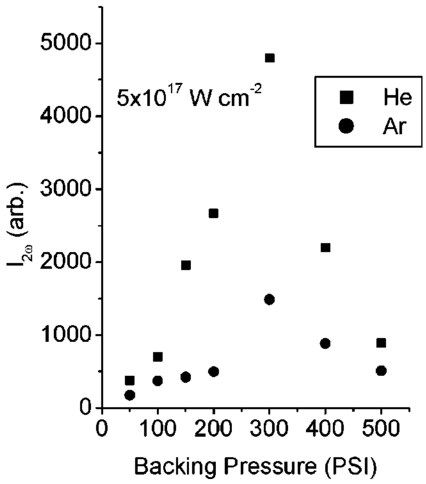

FIG. 7. Efficiency of second harmonic emission in He and Ar. While both show the same qualitative behavior the Ar produces significantly less second harmonic as compared to He.

three gases and thus only a relative comparison can be made for the corresponding spectra. At the intensities used in our experiment, He is completely ionized, while N_2 and Ar would also undergo significant ionization—as such, the electron density in the latter case would be substantially larger as compared to that when He is used. Excessive ionization would produce significant defocusing of the beam and an intensity of $5\times10^{17}\ \mathrm{W\,cm^{-2}}$ was found to be optimal.[12] 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 Ar (≈0.6 mm) as compared to He (≈0.9 mm).

The data show that, in the case of He, 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 Ar to the extent that there is more emission in the eighth as compared to the ninth. It is easy to rationalize this data on the basis of our model that the even harmonics are produced entirely due to the free electrons. As the free electron density increases when higher-Z gases are used, it leads to enhancement of scattering processes involving them. 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. As a result, there should be an increase in the ratio of the even to odd orders and this is indeed observed in the experiments.

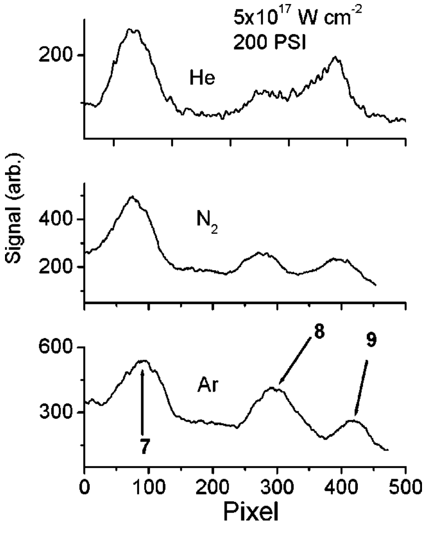

FIG. 6. Lineouts corresponding to the conditions of Fig. 5. Note the enhancement of the even order as compared to the odd as the free electron density is increased when higher atomic number gases are used.

All of 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 in 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 due 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. Figure 7 shows the results obtained from He and Ar as a function of the number density. It is clear that under the conditions for Fig. 7 and generally at every density, the Ar produces less second harmonic than He. The shorter channel in Ar as compared to He confirms the fact that the extent of self-focusing is smaller and, therefore, the peak intensity reached is reduced for high-Z gases. Moreover, the second harmonic scales as the n^2, unlike the even order harmonics, which scale linearly with n. Based on the fact that the even harmonics and conversion to second harmonic are inversely 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. However, the angular distribution of the harmonics is an important characteristic which needs to be studied. In order to 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

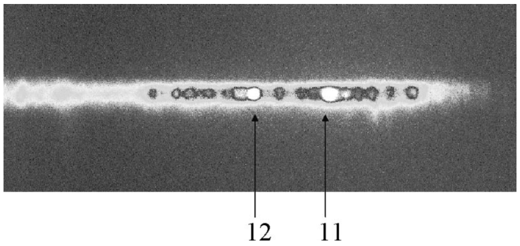

FIG. 8. Image of the high-order harmonics after diffraction through a 200 μm slit. The horizontal axis corresponds to wavelength while the vertical axis to the spatial extent of the harmonics. The calculated angular divergence for the harmonics shown is about 2.4 deg.

8 cm behind the slit. Based on a calibration using a test beam it would be possible to calculate the exact angular divergence of the harmonics. 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. Based on the measured magnification of the system, it is found that the harmonic beam has size of about 3 mm on the slit. This corresponds to a divergence of 3 deg at the sixth harmonic which decreases to ≈ 2 deg at the 20th harmonic. The harmonics are thus emitted in a forward direction with very small angular divergence.

V. 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 significant amount of VUV light. The emitted radiation is produced as a beam with very small angular divergence. The production of this beam probably results from the fact that high energy electrons play a significant role in the scattering process. While the conversion efficiency into high order harmonics has not yet been measured, based on data for atomic harmonics based on data for atomic harmonics, it is estimated to be $\approx 10^{-7}$.

ACKNOWLEDGMENTS

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