Invited Paper

Angular distribution of high-order harmonics generated in the tunneling regime

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ABSTRACT

The angular distribution of high-order harmonics produced in a jet of xenon gas is studied using a 1.053μm wavelength laser. All the harmonics in the 9th to 25rd range display approximately equal angular widths with cone angles that are approximately half that of the laser. These widths are significantly larger than would be predicted by lowest order perturbation theory.

1. INTRODUCTION

The study of highly non-linear processes in an intense laser field has become a topic of great interest. Among the possible effects is harmonic production of the laser field. In the past few years a number of groups have studied the production of high order harmonics.[1-5] The observation of the 53rd harmonic of a 1 μm wavelength laser has been observed in Ne.[4] In atomic gases, the dipole approximation predicts the presence of odd harmonics only. Recent work on the role of phase matching in high order harmonic generation[6, 7] has led to an explanation of the relative intensities of the harmonics and predictions for the far-field intensity distributions. We report here on preliminary measurements of the angular(far field) distribution of high-order odd harmonics (9th through 25rd) and we find the results to be inconsistent with predictions of lowest order perturbation theory (LOPT).

General features of the harmonic spectra are in agreement with previous experiments.[1-5] A plateau in the harmonic intensities is observed in xenon with a cutoff at the 25th harmonic. The laser intensity required to produce high-order harmonics is approximately the same as for these other groups. As expected, the harmonics are present when linearly polarized light is used and absent with circularly polarized light.

2. EXPERIMENTAL SETUP

The laser used for these experiments is a Nd:glass laser which uses the chirped pulse amplification and compression technique to produce high-intensity laser pulses. It has been described in detail elsewhere.[8, 9] Briefly, a 100-Mhz train of ~50 ps pulses is generated by a Nd:YLF oscillator. These pulses are injected into a single-mode optical fiber where the pulses are temporally stretched and frequency chirped due to group-velocity dispersion and self-phase modulation. The pulses are further stretched by a pair of anti-parallel expansion gratings[10] to a pulse length of ~300 ps. A single pulse is selected from the train and injected into a regenerative amplifier where the energy is increased to ~1 mJ. This pulse is further amplified with two additional Nd:phosphate-glass amplifiers to a maximum of ~100 mJ. The maximum energy is limited by the damage threshold of the compression gratings. The pulse is then recompressed with a double-pass through a 1700 l/mm grating pair. The laser is operated at the

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fundamental wavelength of 1.053 μm, with a nominal pulse width of 1 ps and a bandwidth of ~15 Å. Using f/20 optics (60-cm focal length lens) intensities ranging from 10^{13} to mid-10^{14} W/cm^2 were obtained. We have previously performed low gas density experiments to measure the degree of ionization versus laser intensity.[11, 12] These experiments show that intensities of $2X10^{14}$ W/cm^2 are sufficient to produce Xe^{2+}.

The harmonics are produced in a jet of xenon gas. The gas is introduced into a vacuum chamber using a Lasertechnics model LPV pulsed nozzle. The nozzle is directed into the mouth of a 2" diffusion pump which keeps the chamber pressure from rising above 10^{-5} Torr when the jet is fired. The laser is focused approximately 0.5mm from the nozzle opening. Two nozzle sizes were used: 1000μm and 300μm. No harmonics were seen with the 1000μm nozzle since the confocal parameter of the laser was less than the width of the jet. Phase matching conditions for harmonic generation require the interaction length to be less than the confocal parameter of the focused beam.[13]

The backing pressure for the gas jet is approximately 1 atmosphere. Xenon can form dimers at these high pressures resulting in a broadband fluorescent emission which spans the wavelength range of the harmonics. This signal cannot be detected with our spectrometer since its low collection efficiency reduces the signal to noise ratio to the point where the fluorescence light has an intensity at our below the noise level.

A calibration of the gas pressure at the laser focus was performed by measuring the visible light emission perpendicular to the direction of laser propagation. The backing pressure of the jet was varied and the scattered signal measured. The entire chamber was then backfilled to various pressures which produced similar signal levels. The spatial extent of the gas is clearly less in the case of the gas jet which means there are fewer atoms from which the light can scatter. If low laser intensities are used, however, most of the scattered light will come from a very short region near the beam waist so the back-filled chamber should give only a slightly larger signal than the gas jet would at equal pressures. We estimate the output pressure of the gas jet at the laser focus to be ~5 ($\times 3^{\pm 1}$) torr (particle density ~ 10^{17} cm^{-3}).

The spectrometer used in these experiments is shown schematically in Fig.1. It consists of a slit with approximate dimensions of 100μm × 2.5cm followed by a bare gold wire transmission grating. The slit is aligned to be parallel to the grating wires. The grating consists of 0.5μm diameter gold wires which are separated by 1μm (center to center). Following the grating is a microchannel plate (MCP) image intensified phosphor screen (Galileo model 8081). The microchannel plate intensifier is not UV enhanced so it cannot detect harmonics lower than the ninth. The screen is positioned colinearly with the laser beam. The light incident on the slit and grating is dispersed by the grating and produces spectral lines on the phosphor screen. The spectral content of the light can be measured in the direction perpendicular to the grating lines and the angular distribution of the harmonic lines can be measured parallel to the slit. Thus we simultaneously obtain information about the spatial distribution as well as the spectral content of the light.

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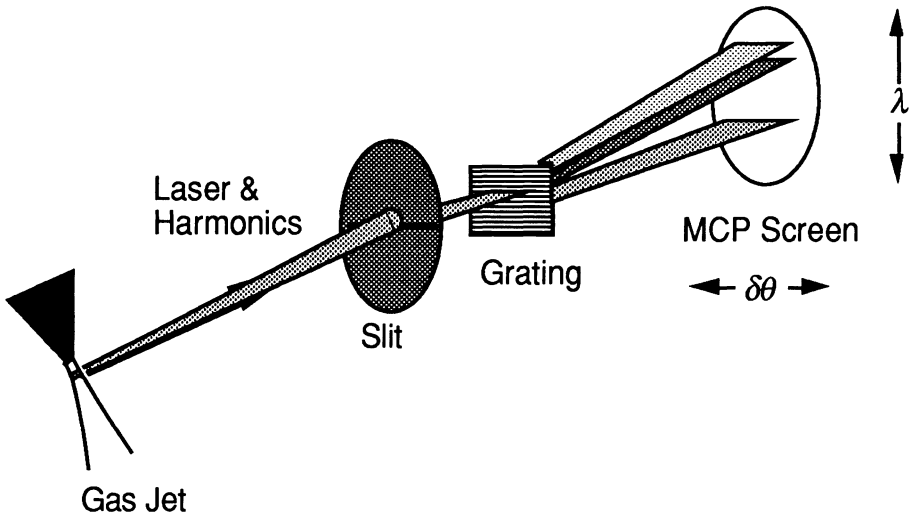


Fig.1 The spectrometer configuration. Laser and harmonic signals pass through a 100μm slit and are dispersed by the gold wire transmission grating. The spectral lines are detected with an MCP intensified phosphor screen. The frequency spectrum is measured in the vertical direction and the angular distribution is measured in the horizontal direction.

Measurements of the harmonic distribution have also been done with the slit and the transmission grating removed so that the laser and harmonic beams directly strike the surface of the MCP. We were able to compare the spatial distribution of the harmonics to that of the laser by pulsing the gas jet on every other shot. Although the MCP is insensitive to 1μm wavelength light we were able to see a signal from the laser itself by increasing the bias voltage on the plates from 1.0kV to 1.8kV (a gain increase of ~1000). The extremely high intensity of the laser was sufficient to create a small signal. When the slit and transmission grating were in place, however, the laser beam alone would not create a signal on the MCP using comparable voltages. This is due to the large amount of 1.053μm light which is reflected by the grating.

4. RESULTS

The far-field distributions of the 11th, 15th, and 19th harmonics of xenon are shown in Fig. 2. The peak heights have been normalized so that their widths can be compared. The spatial structure could be due to many things such as defects in the grating or MCP, density variations in the gas jet, structure in the incident laser beam, harmonic generation due to resonances, or phase matching effects. Some of these effects could be reduced by signal averaging, but the harmonics in Fig. 2 represent data from a single laser shot. We are performing further studies to determine if the structure is a significant feature of the the data shown.

To obtain information about the relative harmonic intensities and about the envelope of the harmonic emission, we have averaged the spectra from a number of shots. The spectra were spatially integrated to obtain the relative harmonic intensity for Xe at $2X10^{14}$ W/cm^2 as shown in Fig. 3. The apparent 27th harmonic is probably detector noise. The low frequency cutoff is due to the detector response. As can be seen, a plateau is observed to the 19th harmonic and then a gradual decrease to the 25th.

The shot-averaged envelope of the far-field distribution of the 13th harmonic is shown in Fig. 4. The FWHM angular width is 24 mrad compared to 38 mrad for the incident laser beam.

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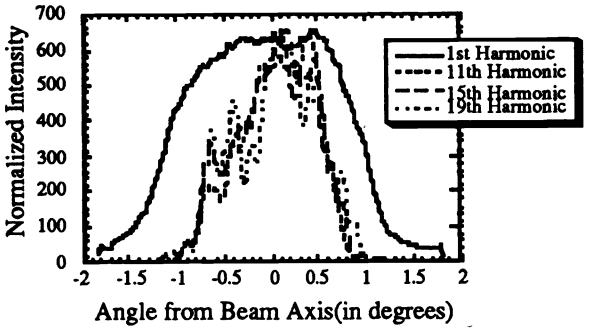


Fig. 2 Experimental measurement of the spatial extent of high harmonics relative to that of the incident laser. The peak heights have been normalized so that their widths could be compared. Note that the laser beam profile represents a different laser shot from the other three curves.

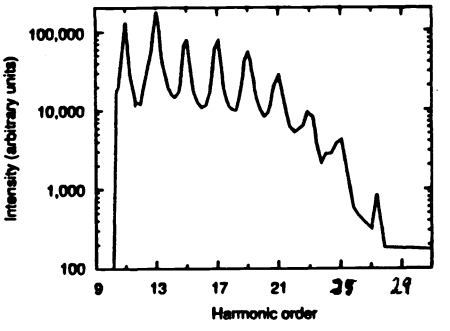


Fig. 3 Spatially integrated harmonic spectrum for Xe at $2X10^{14}$ W/cm^2. The intensity is in arbitrary units.

Fourier transform methods can be used to calculate the beam profile at the focal and detector planes[14, 15] (if cylindrical symmetry is assumed, Hankel transforms are used). If the harmonics are assumed to have an I^N dependence as predicted by LOPT, the spatial distribution of the harmonics at the focal plane can be calculated by raising the laser beam profile to the Nth power. The beam profile at the detector can then be calculated by performing an inverse Fourier transform. This method of calculating the focal spot size is in good agreement with the actual value which was independently measured. The results of these calculations are shown in Figs. 5 for an incident super-Gaussian laser beam profile. Clearly the LOPT prediction does not coincide with the experiment.

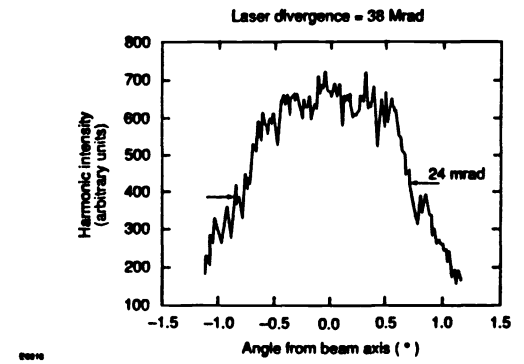


Fig.4 Multiple shot averaged far-field distribution of the 13th harmonic in Xe at $2X10^{14}$ W/cm^2.

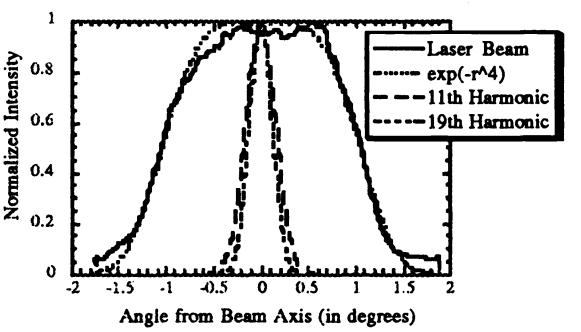


Fig.5 Comparison of the laser beam spatial profile to a super-Gaussian and the calculated harmonic widths using the super-Gaussian profile as described in the text..

The observed far-field width of the harmonics is much larger than predicted by LOPT. In addition to the effects of phase-matching[6,7] it is possible that the laser light is undergoing self-focussing or refraction in the plasma generated by ionization. We can easily estimate the angular deflection due to refraction by noting that the index of refraction for electromagnetic waves in a plasma is given by $n(\omega) = ck / \omega = \sqrt{1 - \omega_{pe}^2 / \omega^2}$,[16] where ω_{pe} is the plasma frequency. Using the small angle approximation for the wave propagation, we find that the angular

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divergence of the beam is given by $\delta\theta \approx 0.5\left(\omega_{pe}^2 / \omega^2\right)(L / r_n)$, where, r_n is the density gradient length, and L is the length of the plasma. Because of high nonlinearity of the ionization process, the plasma density gradient length can be much shorter than the focussed beam radius. If we take r_n=1 μm, L=1 mm, and a plasma density of 10^{17} cm^{-3}, we find that the angular deflection of the 13th harmonic is 0.3 mrad, much smaller than observed discrepancy between to experiments and the LOPT prediction. Refraction will, though, limit the minimum size of any observable structure in the far-field intensity distribution.

5. SUMMARY

We have presented measurements of the angular distributions of high-order harmonics which were created in xenon gas. The angular widths are less than that of the laser, but wider than the widths predicted by LOPT. Refraction of the electromagnetic waves in the plasma caused by ionization is not large enough to explain the observed widths, but may become important at higher electron densities.

6. ACKNOWLEDGMENTS

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7. REFERENCES

(a) Also Department of Physics and Astronomy, University of Rochester.

1 A. McPherson, G. Gibson, H. Jara, U. Johann, T.S. Luk, I.A. McIntyre, K. Boyer, and C.K. Rhodes, "Studies of multiphoton production of vacuum-ultraviolet radiation in rare gases," J. Opt. Soc. Am. B, 4, 595-601 (1987).

2 X.F. Li, A. L'Huillier, M. Ferray, L.A. Lompre, and G. Mainfray, "Multiple-Harmonic Generation in Rare Gases at High Laser Intensity," Phys. Rev. A, 39, 5751-5761 (1989).

3 L. A. Lompre, A. L'Huillier, M. Ferray, P. Monot, G. Mainfray, and C. Manus, "High-order harmonic generation in xenon: intensity and propagation effects," J. Opt. Soc. Am. B, 7, 754-761 (1990).

4 A. L'Huillier, L.A. Lompre, G. Mainfray, and C. Manus, High Order Harmonic Generation, International Conference on Multiphoton Processes V, Ed. G. Mainfray and P. Agostini. (Paris, France: 1990) 45.

5 N. Sarukura, K.Hata, T. Adachi, and R. Nodomi, "Coherent soft x-ray generation by the harmonics of a ultra-high power KrF laser," Phys. Rev. A, 43, 1669-1672 (1991).

6 A. L'Huillier, K.J. Schafer, and K.C. Kulander, "Theoretical aspects of intense field harmonic generation," J. Phys. B., to be published (1991).

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7 A. L'Huillier, K.J. Schafer, and K.C.. Kulander, "High-Order Harmonic Generation in Xenon at 1064nm: The Role of Phase Matching," Phys. Rev. Lett., 66, 2200-2203 (1991).

8 P. Maine, D. Strickland, P. Bado, M. Pessot, and G. Mourou, "Generation of Ultrahigh Peak Power Pulses by Chirped Pulse Amplification," IEEE J. Quant. Elect., QE-24, 398 (1988).

9 Y.-H. Chuang, D.D. Meyerhofer, S. Augst, H. Chen, J. Peatross, and S. Uchida, "Pedestal Suppression in a Chirped Pulse Amplification Laser," J. Opt. Soc. Am. B, 8, 1226-1235 (1991).

10 O.E. Martinez, "3000 Times Grating compressor with Positive Group Velocity Dispersion: Application to Fiber Compensation in the 1.3-1.6 μm Regime," IEEE J. Quantum Electron, QE-23, 59-64 (1987).

11 S. Augst, D. Strickland, D. D. Meyerhofer, S. L. Chin, and J. H. Eberly, "Tunneling Ionization of Noble Gases in a High-Intensity Plasma," Phys. Rev. Lett., 63, 2212 (1989).

12 S. Augst, D. D. Meyerhofer, D. Strickland, and S. L. Chin, "Laser ionization of noble gases by Coulomb-Barrier suppression," J. Opt. Soc. Am. B, 8, 858-867 (1991).

13 J. Reintjes, C.Y. She, and R.C. Eckardt, "Generation of Coherent Radiation in the XUV by Fifth and Seventh Order Frequency Conversion in Rare Gases," IEEE J. Quantum Electron, QE-14, 581-596 (1978).

14 A. Seigman, Lasers, (Mill Valley, Ca: University Science Books, 1988)

15 J.W. Goodman, Introduction to Fourier Optics, (New York: McGraw-Hill, 1988)

16 W.L. Kruer, The physics of laser plasma interactions, (Redwood City, Ca: Addison-Wesley, 1988)

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