Measured laser-beam evolution during high-order harmonic generation in a semi-infinite gas cell

Nicole Brimhall, John C. Painter, Nathan Powers, Gavin Giraud, Matthew Turner, Michael Ware, Justin Peatross

Dept. Physics and Astronomy, Brigham Young University, Provo, UT 84602, USA
peat@byu.edu

Abstract: We report on direct measurements of self-guiding of 800 nm, 30 fs, 5 mJ laser pulses used to generate high-order harmonics in 80 torr helium. We track the spatial evolution of the laser pulses as they propagate several centimeters near the focus under conditions suitable for harmonic generation. The laser is observed to focus, diverge, and refocus. This behavior is accompanied by a flattop beam profile. Both of these features are absent when the laser is focused in vacuum. We also observed a 4 nm spectral blue shift in the center of the laser beam near the focus in contrast with no spectral shift at wider radii.

©2007 Optical Society of America

OCIS codes: (190.4160) Multiharmonic generation.

References

1. J. C. Painter, M. Adams, N. Brimhall, E. Christensen, G. Giraud, N. Powers, M. Turner, M. Ware, and J. Peatross, "Direct observation of laser filamentation in high-order harmonic generation," Opt. Lett. **31,** December 1 (2006).
2. J. Sutherland, E. Christensen, N. Powers, S. Rhynard, J. Painter and J. Peatross, "High harmonic generation in a semi-infinite gas cell," Opt. Express **12**, 4430-4436 (2004).
3. Y. Tamaki, J. Itatani, Y. Nagata, M. Obara, and K.Midorikawa, "Highly efficient phase-matched high-order harmonic generation in self guided laser beam," Phys. Rev. Lett. **82**, 1422–1425 (1999).
4. E. Takahashi, Y. Nabekawa, and K. Midorikawa, "Generation of 10 μJ coherent extreme-ultraviolet light by use of high-order harmonics," Opt. Lett. **27**, 1920-1922 (2002).
5. E. Takahashi, Y. Nabekawa, T. Otsuka, M. Obara, and K. Midorikawa, "Generation of highly coherent submicrojoule soft x-rays by high-order harmonics," Phys. Rev. A **66**, 021802-1-4 (2002).
6. E. J. Takahashi, Y. Nabekawa, and K. Midorikawa, "Low-divergence coherent soft x-ray source at 13 nm by high-order harmonics," Appl. Phys. Lett. **84**, 4-6 (2004).
7. V. Tosa, E. Takahashi, Y. Nabekawa, and K. Midorikawa, "Generation of high-order harmonics in a self-guided beam," Phys. Rev. A **67**, 063817-1-4 (2003).
8. H. T. Kim, I.J. Kim, V. Tosa, Y. S. Lee, and C. H. Nam, "High brightness harmonic generation at 13 nm using self-guided and chirped femtosecond laser pulses," Appl. Phys. B **78**, 863–867 (2004).
9. V. T. Platonenko, V. V. Strelkov, G. Ferrante, "Off-axially phase-matched high-order harmonic generation in an extended medium," J. Opt. Soc. Am. B **19**, 1611-1619 (2002).
10. E. T. J. Nibbering, G. Grillon, M. A. Franco, B. S. Prade, and A. Mysyrowicz, "Determination of the inertial contribution to the nonlinear refractive index of air, N2, and O2 by use of unfocused high-intensity femtosecond laser pulses," J. Opt. Soc. Am. B **14**, 650-660 (1997).
11. S. Carusotto, E. Iacopini, E. Polacco, F. Scuri, G. Stefanini, and E. Zavattini, "Measurement of the Kerr constant of He, A, O2, N2, H2, and D2," Nuovo Cimento **5D**, 328-338 (1985).
12. S. L. Chin, S. A. Hosseini W. Liu, Q. Luo, F. Théberge, N. Aközbek, A. Becker, V. P. Kandidov, O. G. Kosareva, and H. Schroeder, "The propagation of powerful femtosecond laser pulses in optical media: physics, applications, and new challenges," Can. J. Phys. **83**, 863-905 (2005).
13. S. Kazamias, F. Weihe, D. Douillet, C. Valentin, T. Planchon, S. Sebban, G. Grillon, F. Auge, D. Hulin, and P. Balcou, "High order harmonic generation optimization with an apertured laser beam," Eur. Phys. J. D **21**, 353 (2002).
14. S. Champeaux and L. Berge, "Femptosecond pulse compression in pressure-gas cells filled with argon," Phys. Rev. E **68**, 066603 (2003).

#77784 - $15.00 USD Received 5 December 2006; revised 22 January 2007; accepted 5 February 2007

1. Introduction

We recently reported on self-guiding (or filamentation) of a laser pulse in a helium-filled cell under conditions well suited for high-order harmonic generation [1]. In this related article, we present the detailed spatial evolution of the laser pulse energy distribution as it propagates within the gas cell. We also document the evolution of the brightness of the associated high-harmonic emission as the distance from the focusing mirror to the exit surface of the gas cell is varied. These observations indicate that the self-guiding behavior of the laser is favorable to phase matching.

This conclusion is supported by our previous work [2], where we probed the focal region of a helium-filled semi-infinite cell (i.e., a cell where the gas extends from the focusing optic to an exit foil at the focus) with counter-propagating pulses. We observed that the coherence lengths for harmonics up to the 91st of the 800 nm laser extended over many millimeters. This was unexpectedly long, since the natural diffraction of a free laser beam should introduce phase mismatches that limit the coherence lengths to a fraction of a millimeter. The extended phase-matching suggests that laser self-guiding plays a role, which is consistent with reports made by Tamaki et al. [3] based on the observation of strong harmonic emission from thick gas cells.

Some of the highest harmonic pulse energies reported to date have been produced in gas cells under self-guiding conditions [3-7]. Tamaki et al [3] reported enhancements of up to 40 times for the 49th harmonic in a neon cell. The enhancements in harmonic emission were associated with a narrowing of the divergence angle of the transmitted laser beam, which they cited as evidence for laser self guiding. Takahashi [6], Tosa [7], Kim [8] and coworkers also investigated the role that laser self-guiding plays in enhanced phase matching of high harmonics generated in xenon-, argon-, and neon-filled cells.

Platonenko et al. [9] explored theoretically high-harmonic phase matching in a laser undergoing filamentation via the Kerr effect. Under certain conditions they found favorable phase matching conditions for harmonics off axis. However, Tosa et al. [7] considered the Kerr effect inconsequential in their experiments (using xenon- and neon-filled cells); instead they proposed a radially abrupt plasma boundary as the primary mechanism for self guiding of the laser.

In this article, we report on direct measurements of the spatial evolution of a laser near the focus inside a helium-filled gas cell under conditions ideal for harmonic generation. The measurements show evidence of self-guiding and even refocusing of the laser energy, which is characteristic of Kerr-style filamentation. However, the laser power used in the experiment is well below the critical power for filamentation [10-12]. The model proposed by Tosa et al. [7] does not rely on Kerr lensing, but also does not countenance refocusing of the laser. A satisfactory explanation of our results therefore requires further investigation.

2. Setup

We employed 5 mJ, 800 nm, 30 fs pulses produced by a Ti:sapphire laser system. The f/125 focusing achieved an intensity of 1.9×10^{15} W/cm^2 (with 25% uncertainty) in vacuum and approximately half that in the helium-filled cell used for the experiments. An aperture closed to 8 mm located 2 m before the focusing mirror strongly enhanced the harmonic emission [13]. The gas-filled region extends from the focusing mirror (R = 100 cm) to the laser focus where it abruptly ends as the laser passes through a pinhole into vacuum (see Fig. 1 for a schematic of our setup). A grazing-incidence diffraction grating (1200 lines/mm with 2 m radius of curvature) separates and focuses the harmonics in one dimension onto a microchannel plate (MCP) coupled to a phosphor screen. The harmonics, produced in 80 torr helium, were observed from the 45th to the 91st order to have approximately uniform intensity. We determined the energy in an individual harmonic order to be approximately 1 nJ.

#77784 - $15.00 USD
Received 5 December 2006; revised 22 January 2007; accepted 5 February 2007

(C) 2007 OSA

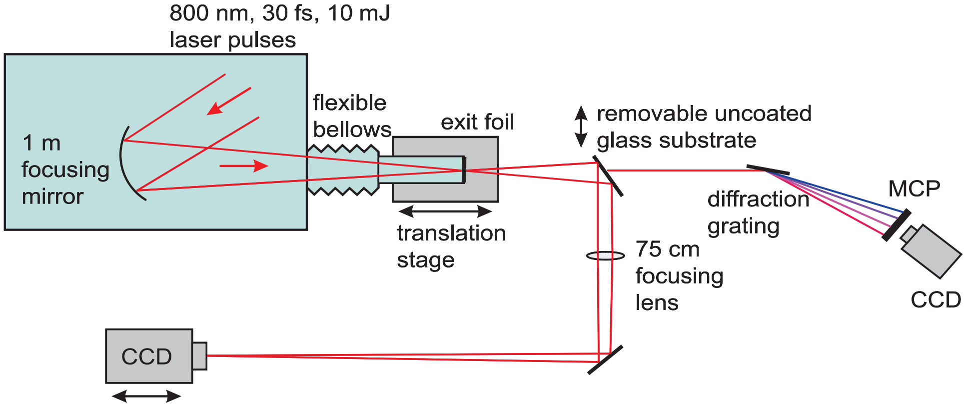

Fig. 1. Setup to image laser beam spatial profile under conditions suitable for harmonic generation.

To image the spatial profile of the laser beam under conditions suitable for producing harmonics, an uncoated glass substrate oriented at 45 degrees was inserted in the beam before the diffraction grating. The glass substrate, which functioned both as a mirror and an attenuator, could be moved in and out of the laser beam in a matter of seconds while maintaining vacuum in the harmonic detection setup. The substrate reflected approximately 2% of the laser, which was imaged by an f = 75 cm focusing lens onto a CCD camera. Neutral-density filters were used for further attenuation. The camera was positioned to image the laser at the plane of the exit foil of the gas cell. The position of the foil was scanned parallel to the laser axis along 9 cm. The axial position of the camera was also scanned to maintain an image of the laser beam at the exit foil. The magnification of the image was approximately 3x, depending on the exact location of the foil relative to the imaging lens.

3. Effect of focal position on harmonic generation and laser spatial profile

Figure 2 shows a movie of harmonics generated in 80 torr helium gas together with the evolution of the laser beam spatial profile as the foil position is varied. The left image shows the generated harmonics, the middle image shows the measured laser beam spatial profile, and the graph on the right shows a lineout of the laser beam spatial profile. The units on the laser image and beam lineout are scaled to the dimensions of the laser focus in the gas cell rather than the image size on the CCD. The z-position of the exit foil is measured relative to 100 cm from the focusing mirror, which is where the beam focuses in vacuum. Each image is the average of 10 laser shots.

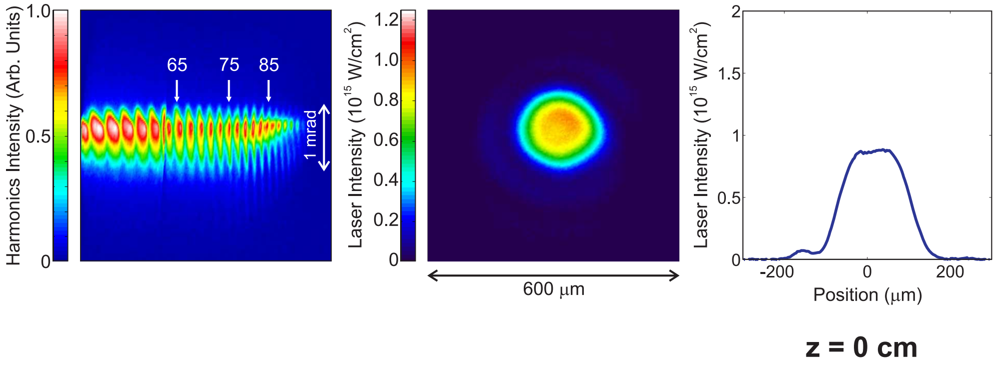

Fig. 2. Movie of harmonics (left), imaged laser beam (middle), and beam lineout (right) for harmonics generated in 80 torr helium. The z-position is measured relative to where the beam focuses in vacuum.

#77784 - $15.00 USD Received 5 December 2006; revised 22 January 2007; accepted 5 February 2007
(C) 2007 OSA

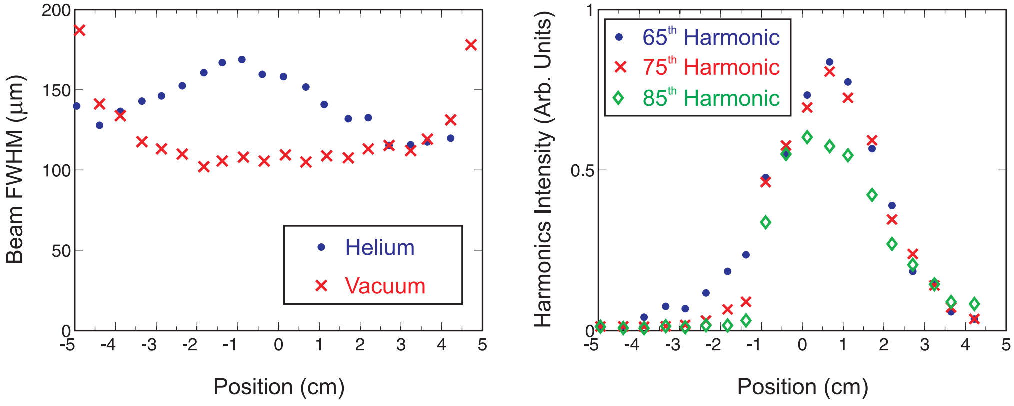

Fig. 3. (a) Diameter of the laser as it exits from the gas cell, either filled with 80 Torr helium or evacuated. The best focus in the absence of gas occurs 100 cm after the focusing mirror (Reprinted with permission from [1]; Copyright 2006 by the Optical Society of America). (b) The brightness of several harmonic orders as the foil position is varied.

Figure 3(a) plots the beam diameter in both 80 torr helium and vacuum as the foil position is varied. In helium, the width of the beam reaches a minimum near z = -3 cm, widens to reach a maximum near z = 0 cm, and then narrows again to a second minimum at z = 4 cm. In vacuum the beam goes through a single focus, as would be expected. Figure 3(b), shows the intensity of several harmonic orders at the foil positions where the beam diameter in helium was measured. The best harmonic production is observed around z = 0.5 cm, where the laser in helium is changing from diverging to converging.

Figure 4 shows a movie comparing intensity lineouts of the laser beam focused in helium and in vacuum. The beam propagating in vacuum shows a Gaussian-like profile, which reaches a single minimum width with maximum intensity at z = 0. The beam propagating in helium exhibits the double focus discussed above, and a distinct flat-top radial profile from about z = -2 cm to z = 1.5 cm. Although not immediately apparent from the lineouts, the two curves correspond to similar total energies. (The intensity at wider radii is incident on a larger area, so in two dimensions the extra intensity in the wings of the profile in helium compensates for its lower on-axis intensity).

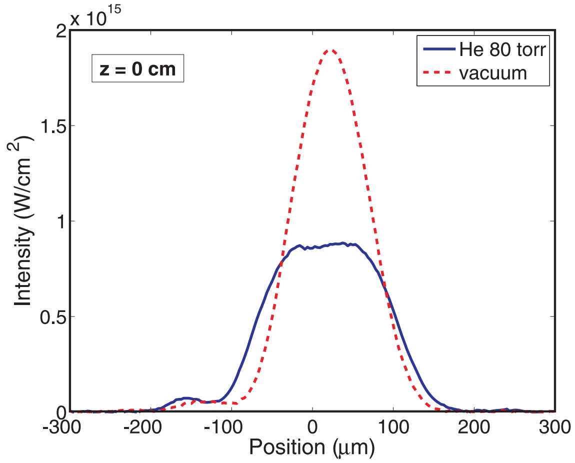

Fig. 4. Movie of lineouts of the laser focused in helium and in vacuum as the foil position is scanned longitudinally. The effective intensity assumes a uniform 30fs Gaussian envelope, even for the case of the pulse interacting with the helium.

A flat-top profile was previously associated with laser self-guiding and extended phase-matching in neon [7,8]. Tosa et al. [7] calculated the radial intensity profile of a self-guided beam and predicted that it would exhibit a top-hat profile, owing to wavefront distortions from free electrons. Kim et al. subsequently observed a radial intensity flattening in a single image produced for a laser that had traveled through a wide gas jet [8]. The top-hat profile was attributed to defocusing of the laser by free electrons at inner radii. As the on-axis energy is defocused out to wider radii, it overlaps with the less intense outer portion of the laser beam still in the act of focusing. While this explanation is consistent with the radial intensity distribution observed and laterally broadened wave fronts, it does not readily offer a reason for the good longitudinal phase matching for high harmonic generation. It also does not explain the re-convergence of our beam to a second focus, as seen in the data.

4. Effect of focal position on laser spectrum

A 50 μm fiber coupled to a spectrometer (Ocean Optics USB-2000) was used to obtain information on the laser's spectral properties. The fiber was swept through the laser image radially to test for variations in the spectrum resulting from the interaction with the helium. Because of magnification in the imaging system, the effective spatial resolution of the scan corresponded to less than 20 μm in actual focus. Figure 5 shows a movie of a Gaussian fit to the spectrum of the laser as the fiber is swept radially through the beam. A comparison is made between the cases of helium and vacuum when the foil is positioned at z = 0. In vacuum, the laser spectrum is seen to be centered at 800 nm with a FWHM of 35 nm. As expected, the vacuum spectrum exhibits no change as the fiber is moved through the laser image. For radii less than 70 μm, we observe a blue-shift of up to 4 nm of the laser focused in helium versus the laser focused in vacuum. The spectral blue-shifting of the center of the beam coincides with the onset of ionization (as suggested by a visible streak of plasma seen from the side of the focus), but other effects may also play a role [14].

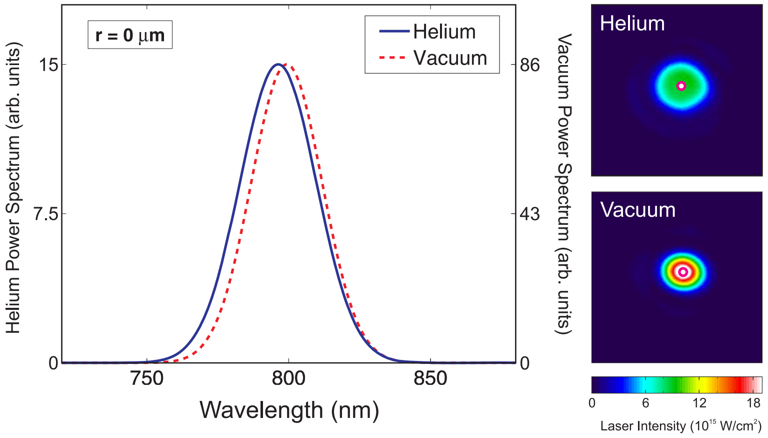

Fig. 5. Movie of the spectrum of the laser in vacuum and in helium as the fiber is scanned radially through the laser beam. The dots in the profiles on the right show the approximate position of the fiber that samples the spectrum in the beam.

5. Discussion

The double focusing observed in our laser beam as it interacts with helium is suggestive of Kerr-style self-focusing, or filamentation. Placing a partially closed aperture in the laser beam before the focusing mirror causes the observed laser focus to be smoother while at the same time dramatically increasing the high harmonic signal. This effect may support the idea of Kerr-style self-focusing in the sense that a smoother beam will tend to favor the formation of a single filament rather than competing filaments. Nevertheless, the nonlinear index in 80 torr of helium is reported to be ~ 4×10^{-22} cm^2/W [10,11], which predicts a critical power for

Received 5 December 2006; revised 22 January 2007; accepted 5 February 2007
(C) 2007 OSA

filamentation [12] in excess of 2 TW. In contrast, the power used in our experiments was an order of magnitude less. This is apparently why Tosa and coworkers [7] considered the Kerr nonlinearity to be inconsequential. However, at least in the case of helium, the nonlinear index has not been measured in two decades. Our work underscores a need to reexamine it. We speculate on the possibility that the nonlinearity of the index varies in character very near the threshold of ionization.

The development of a flat-top intensity profile in the laser focus is consistent with the predictions and observations of Tosa and Kim [7,8]. However, our observation of double foci is unexpected within their description. The brightest harmonics are attained when the exit foil of the gas cell is positioned near the middle of the self-guiding region, where the beam diameter is largest. This region of extended phase matching occurs where the laser beam changes from diverging to converging between the two foci. This is opposite in character to a conventional laser focus, which changes from converging to diverging while accompanied by the Gouy shift (known to be deleterious to phase matching).

In our view, a complete and satisfying description of processes involved in this experiment is not yet available. The interpretation of the data is complicated by the fact that the CCD camera measures energy fluence rather than intensity (technically a mislabeling of Figs. 2 and 3 in our paper). Our measurement technique is therefore unable to distinguish whether the entire pulse in time develops the flattop profile as seen in the data, or whether different temporal portions of the pulse look spatially different. This work was supported by the National Science Foundation, under grant number PHY-0457316.

Received 5 December 2006; revised 22 January 2007; accepted 5 February 2007
(C) 2007 OSA