Direct observation of laser filamentation in high-order harmonic generation

John C. Painter, Mark Adams, Nicole Brimhall, Eric Christensen, Gavin Giraud, Nathan Powers, Matthew Turner, Michael Ware, and Justin Peatross

Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84602

Received August 9, 2006; revised September 8, 2006; accepted September 15, 2006; posted September 18, 2006 (Doc. ID 73880); published November 9, 2006

We investigate the spatial evolution of a laser pulse used to generate high-order harmonics (orders ranging from 45 to 91) in a semi-infinite helium-filled gas cell. The 5 mJ, 30 fs laser pulses experience elongated focusing with two distinct waists when focused with $f/125$ optics in 80 Torr of helium. Extended phase matching for the generation of harmonics occurs in the region between the double foci of the laser, where the laser beam changes from diverging to converging. © 2006 Optical Society of America

OCIS codes: 190.4160, 260.5950, 190.7110.

Efficient laser high-order harmonic generation in gases has been demonstrated in jets,[1] hollow waveguides,[2,3] and cells.[4] The energy of the harmonics depends on the product of the gas density and the coherence length, to the extent that this product does not exceed the reabsorption limit.[5] In a jet, higher densities compensate for relatively short coherence lengths. Conversely, waveguides can dramatically extend the coherence length, but typically at lower densities for similar conversion efficiencies. A significant benefit to the waveguide approach is the ability to effectively utilize less energetic laser pulses. Conversion efficiencies as high as 10^{-4} have been reached for harmonic orders in the teens generated in xenon with a Ti:sapphire laser (in both a jet[1] and a gas cell[4]).

Some of the highest harmonic pulse energies reported to date have been produced in gas cells. Takahashi *et al.* produced 4.7 μJ pulses of the 13th harmonic in a xenon-filled cell by using 16 mJ laser pulses with a loose focusing geometry.[4] They also produced 0.3 μJ pulses of the 27th harmonic in argon by using 20 mJ laser pulses[6] and 25 nJ pulses of the 59th harmonic in neon by using 50 mJ laser pulses.[7] This last result is to our knowledge the highest harmonic order previously characterized for energy in a study to maximize conversion efficiency.

Several studies indicate that laser self-guiding enhances harmonic production in gas cells. Tamaki *et al.*[8] 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,[7] Tosa,[9] and Kim[10] and coworkers have also investigated the role that laser self-guiding plays in enhanced phase matching of high harmonics generated in xenon-, argon-, and neon-filled cells or wide jets. Their conclusions are based primarily on observations of harmonic production and plasma recombination light, as well as simulations of laser propagation. In this Letter, we present what are believed to be the first measurements of the spatial evolution of the laser within a helium-filled cell under conditions well suited for high-harmonic generation. These measurements show direct evidence of laser filamentation.

Figure 1 shows the high harmonics generated in our setup after they have been dispersed and focused in one dimension by a curved grating onto a microchannel-plate assembly coupled to a phosphor screen and a CCD. Harmonics ranging from the 45th to 91st generated in 80 Torr of helium are seen to carry nearly uniform brightness and lie within a 1 mrad divergence-angle cone. Figure 2 shows a schematic of our experimental setup. A partially closed iris is placed in the laser beam path upstream of the focusing mirror, which has been shown to strongly enhance harmonic brightness.[11] The image of harmonics seen in Fig. 1 was obtained from a single 30 fs, 5 mJ, 800 nm laser pulse. The helium interacts with the laser in what we call a semi-infinite gas-cell geometry, where helium occupies the entire region from the focusing mirror to an exit foil near the laser focus. The region beyond the exit foil is evacuated. The peak intensity of the laser inside the gas cell is estimated to reach $\sim 1.5 \times 10^{15}$ W/cm^2.

We measured the energy of the harmonics using a Si/Zr photodiode (AXUV-100, IRD) placed behind a 0.2 μm thick Zr filter (Lebow). The entire beam (laser and embedded harmonics) was sent into the energy detector. We confirmed that the filter–diode combination did not respond to the laser energy and that recombination light from ionized gas did not contribute to the measured signal. The characteristic transmission and response curves for the filter and photodiode dictated that the combination responded to harmon-

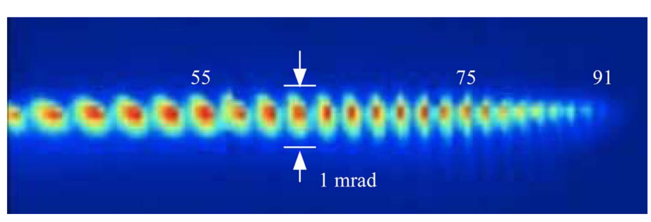

Fig. 1. (Color online) High harmonics produced in 80 Torr helium dispersed by a grating onto a detector.

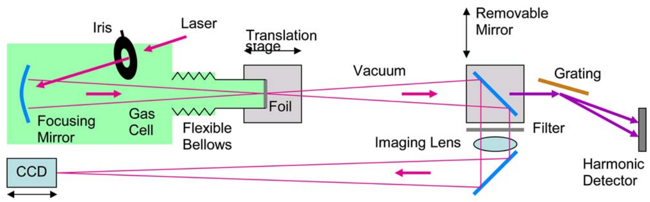

Fig. 2. (Color online) Production of high harmonics in a semi-infinite gas cell with variable exit location. Automated insertion of a mirror allows either the high harmonics or the residual laser to be observed.

ics from the 41st to well beyond the 91st, which is the highest harmonics we observed for our conditions. Under the (reasonable) simplifying assumption that the harmonics in the observed range are uniformly bright, together with knowledge of the spectral response of the filter–diode combination, the energy in individual harmonics was found to be approximately 1 nJ. These harmonic orders extend well above the 59th harmonic, which was the highest order previously measured.[7]

We observed the evolution of the laser's spatial profile in the harmonics-generating region using an imaging system designed to accept the residual laser pulse at full power (see Fig. 2). The laser was reflected into the imaging system using an uncoated glass substrate as a mirror temporarily placed in the path of laser after the cell. The substrate was moved in and out of the beam using automated positioning (requiring only a few seconds). It reflected about 2% of the light at 45°. Neutral-density filters were used for further attenuation. A thin BK7 lens ($f=75$ cm) imaged the laser spot onto a CCD camera with a magnification of about 3.5 (the exact value depending on the location of the imaging plane). To track the evolution of the laser profile within the gas, the position of the gas-cell exit foil was scanned axially along the beam, and the CCD camera position was adjusted to keep the plane of the foil in focus. The laser was used to drill an ~400 μm hole through the foil before each experiment. The axial translation system for the foil was aligned to keep lateral movement of the exit hole to less than ~25 μm over the full range of translation (~10 cm).

Figure 3 shows the evolution of the laser beam diameter (full width at half-maximum) at the position of the exit foil as it is scanned along the axis of the laser. Images were obtained at full laser power both with and without helium (80 Torr) present in the cell. A clear difference between the two cases can be seen. The laser undergoes double focusing in the presence of the helium, a behavior characteristic of filamentation.[12]

By far, the strongest high-harmonic production occurs when the foil is placed approximately midway between where the two foci occur (see Fig. 3). In this region, the laser beam changes from diverging to converging, exactly opposite to what occurs at the focus of a normally diffracting laser beam with its detrimental Gouy shift. We speculate that, as the laser changes from diverging to converging, the wavefronts undergo a phase shift approximately opposite in character to the Gouy shift and that this may play a role in elongating the coherence length.

In previous work,[13,14] we reported on extended phase matching for high-order harmonic generation under similar conditions. In that measurement, we used counterpropagating light pulses to suppress harmonic production in selected regions within the focus. The experiments demonstrated that phase matching takes place over many millimeters in a helium-filled semi-infinite cell. This extended coherence length, which is many times longer than what is expected for a freely diffracting laser beam, was linked to dramatic enhancements in the harmonic output. The extended phase-matching region observed with the counterpropagating technique coincides with the portion of the laser beam that exhibits the anomalous diffraction properties reported in this Letter.

We characterized the reabsorption length for the harmonics using a secondary gas cell placed well after the laser focus, through which the harmonics passed on the way to the detector. The reabsorption lengths were found to agree with theoretical estimates.[15] For harmonic orders 45–91 generated in 80 Torr helium, the absorption length was found to be ~7 mm, on par with the distance over which harmonics were observed to be produced. The production length for high harmonics therefore appears to be at or near the reabsorption limit.

A comparison of the laser radial profile with and without gas present is shown in Fig. 4. As the laser interacts with the gas, its profile evolves into a top-hat shape from its original near Gaussian shape.[9,10] The top-hat profile is most pronounced near the location responsible for best harmonic emission (position 100 cm), which is where the images in Fig. 4 were recorded.

We measured the spectrum of the laser light as a function of position in the focus of the laser at the exit foil. This was done using a 50 μm fiber, which was scanned in the image plane of the beam imaging setup. The magnification and resolution of the setup granted effectively 20 μm sampling resolution within

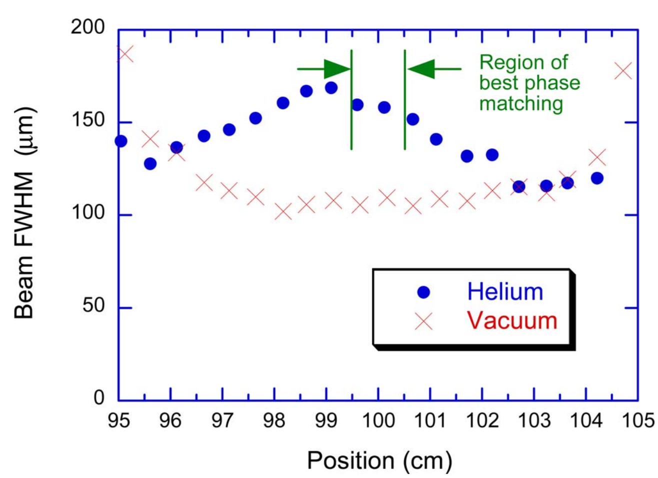

Fig. 3. (Color online) Diameter of the laser as it exits from the gas cell, either filled with 80 Torr helium or evacuated. The best focus in the absence of gas occurs 100 cm after the focusing mirror.

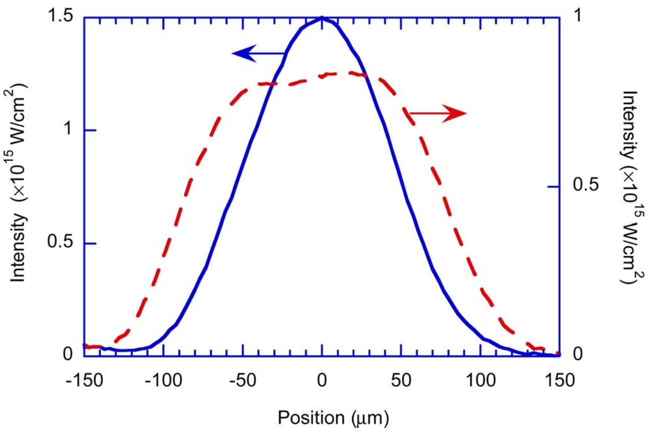

Fig. 4. (Color online) Laser intensity profile at full power captured 100 cm after the focusing mirror. The image is obtained with (dashed curve) and without (solid curve) 80 Torr helium present in the cell.

the actual laser focus. Again, neutral-density filters were used to attenuate the laser energy. The spectrum exhibited an approximately Gaussian shape with a 30 nm full width at half-maximum. Under conditions for best harmonic production, we saw a 4 nm spectral blueshift in the radial center of the laser relative to the spectrum at radii larger than 90 μm, where the spectrum did not shift (remaining similar to when the laser is focused in vacuum).

In conclusion, we have recorded direct evidence of laser filamentation under conditions optimized for generating high harmonics in a semi-infinite helium-filled gas cell. The harmonics emerge within a sub-1-mrad divergence angle and are phase matched over many millimeters. The region where the laser beam changes from diverging to converging between double foci gives rise to the extended phase matching. The occurrence of multiple foci and the fact that the first focus is pulled closer to the focusing mirror are characteristic of self-focusing and filamentation.[12] In addition, the observed improvement when an aperture is partially closed on the beam might be explained through the removal of competition between multiple filaments.[12] However, in considering the Kerr effect, the optical path difference (between an on-axis high-intensity portion of the laser and an off-axis low intensity portion of the laser) is given by $n_2 Il$, where $n_2 = 4 \times 10^{-22}$ cm^2/W is the nonlinear Kerr index[16,17] in 80 Torr helium, $I = 1 \times 10^{15}$ W/cm^2 is the laser intensity, and $l \sim 5$ cm is an effective propagation distance. This suggests an on-axis wavefront displacement of approximately 0.02 μm. This is similar to but distinctly below the estimated wavefront shift $\rho^2/R = 0.05$ μm necessary to change a diverging beam into a converging one, where $\rho = 75$ μm is the beam radius and $R = 10$ cm is an estimated wavefront radius of curvature. This estimate is essentially contained within the derivation of the critical power for self-focusing,[12] compared to which our laser power falls well short. It may be worth re-examining the published value for n_2 in helium,[16,17] which has received little attention for two decades. Tosa *et al.* considered the Kerr effect inconsequential in their experiments with xenon- and neon-filled cells; instead they proposed a radially abrupt plasma boundary as the primary mechanism for self-guiding of the laser.[9,10]

Our work was supported by the National Science Foundation under grant number PHY-0457316. J. Peatross's e-mail address is peat@byu.edu.

References

1. J.-F. Hergott, M. Kovacev, H. Merdji, C. Hubert, Y. Mairesse, E. Jean, P. Breger, P. Agostini, B. Carre, and P. Salieres, Phys. Rev. A **66**, 021801(R) (2002).
2. A. Rundquist, C. G. Durfee III, Z. Chang, C. Herne, S. Backus, M. M. Murnane, and H. C. Kapteyn, Science **280**, 1412 (1998).
3. Y. Tamaki, Y. Nagata, M. Obara, and K. Midorikawa, Phys. Rev. A **59**, 4041 (1999).
4. E. Takahashi, Y. Nabekawa, and K. Midorikawa, Opt. Lett. **27**, 1920 (2002).
5. E. Constant, D. Garzella, P. Breger, E. Mevel, C. Dorrer, C. L. Blanc, F. Salin, and P. Agostini, Phys. Rev. Lett. **82**, 1668 (1999).
6. E. Takahashi, Y. Nabekawa, T. Otsuka, M. Obara, and K. Midorikawa, Phys. Rev. A **66**, 021802(R) (2002).
7. E. J. Takahashi, Y. Nabekawa, and K. Midorikawa, Appl. Phys. Lett. **84**, 4 (2004).
8. Y. Tamaki, J. Itatani, Y. Nagata, M. Obara, and K. Midorikawa, Phys. Rev. Lett. **82**, 1422 (1999).
9. V. Tosa, E. Takahashi, Y. Nabekawa, and K. Midorikawa, Phys. Rev. A **67**, 063817 (2003).
10. H. T. Kim, I. J. Kim, V. Tosa, Y. S. Lee, and C. H. Nam, Appl. Phys. B **78**, 863 (2004).
11. S. Kazamias, F. Weihe, D. Douillet, C. Valentin, T. Planchon, S. Sebban, G. Grillon, F. Auge, D. Hulin, and P. Balcou, Eur. Phys. J. D **21**, 353 (2002).
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, Can. J. Phys. **83**, 863 (2005).
13. J. R. Sutherland, E. L. Christensen, N. D. Powers, S. E. Rhynard, J. C. Painter, and J. Peatross, Opt. Express **12**, 4430 (2004).
14. J. Peatross, J. R. Miller, K. R. Smith, S. W. Rhynard, and B. W. Pratt, J. Mol. Spectrosc. **51**, 2675 (2004).
15. http://www-cxro.lbl.gov/
16. E. T. J. Nibbering, G. Grillon, M. A. Franco, B. S. Prade, and A. Mysyrowicz, J. Opt. Soc. Am. B **14**, 650 (1997).
17. S. Carusotto, E. Iacopini, E. Polacco, F. Scuri, G. Stefanini, and E. Zavattini, Nuovo Cimento D **5**, 328 (1985).