RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 51, NUMBER 6 JUNE 1995

Amplification of xuv harmonic radiation in a gallium amplifier

T. Ditmire,[1] M. H. R. Hutchinson,[2] M. H. Key,[3,5] C. L. S. Lewis,[4] A. MacPhee,[4] I. Mercer,[2] D. Neely,[5] M. D. Perry,[1]
R. A. Smith,[2] J. S. Wark,[3] and M. Zepf [3]
[1]*Laser Program Lawrence Livermore National Laboratory, P.O. Box 808, L-443, Livermore, California 94550*
[2]*Imperial College of Science, Technology and Medicine, Prince Consort Road, London SW7 2BZ, United Kingdom*
[3]*Department of Physics, Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom*
[4]*School of Mathematics and Physics, Queens University of Belfast, Belfast BT7 1NN, United Kingdom*
[5]*Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire, United Kingdom*
(Received 21 December 1994)

We report the observation of amplification of an xuv harmonic pulse in an xuv laser. The 21st harmonic of a frequency-doubled 1.3-psec neodymium chirped-pulse-amplification laser pulse was injected into the gain region of an 18-mm-long Ga XXII x-ray amplifier. A gain of approximately 3 was recorded for the 21st harmonic, relative to the unamplified 19th and 23rd harmonics when the harmonic wavelength was tuned to overlap the 25.11-nm $J=2-1$ laser transition.

PACS number(s): 42.65.Ky, 42.55.Vc

The development of sources of intense, coherent xuv radiation has involved both xuv lasers and harmonic generation from optical frequency lasers. With the availability of intense pulses from chirped-pulse-amplification (CPA) lasers [1], extremely high-order harmonics of the fundamental laser frequency have been observed [2,3] at wavelengths below 7.0 nm [4]. In the plateau region, conversion of laser energy to xuv radiation with an efficiency up to 10^{-7} has been achieved [5]. When produced under weak focusing conditions and at intensities sufficiently low that phase matching is not destroyed by plasma formation, these harmonic sources appear to exhibit a high degree of spatial coherence [6,7]. Since the harmonics are produced only during the laser pulse, the xuv radiation has a pulse duration determined by the duration of the driving laser pulse. As a result, the xuv pulses can be produced at essentially any duration ranging from a few tens of femtoseconds to hundreds of picoseconds, depending on the drive laser source.

Availability of kilojoule nanosecond laser pulses has made it possible to create xuv lasers based on electron collisionally pumped laser schemes in neonlike [8] and nickel-like [9] ions and in recombination schemes [10]. These lasers operate by amplification of spontaneous emission, and the plasmas are produced from solid-slab, thin-foil, and fiber targets. Most recently xuv laser action in a gas target irradiated by an ultrashort pulse [11] and in a capillary discharge [12] has also been demonstrated. These sources have produced amplified spontaneous emission in the range 43–400 nm, with output energy as high as several millijoules at 15.5 nm [13], with a pulse duration of several hundred picoseconds. Used as sources of amplified spontaneous emission (ASE) with no cavity, these lasers have inherently low spatial and temporal coherence. However, recent advances in soft-x-ray optics have enabled double-pass configurations and injection seeding between amplifiers, resulting in improved spatial coherence [14,15].

By using a suitable collisionally pumped laser plasma as a true amplifier for independently produced high-order harmonic radiation, high-energy xuv pulses with a duration determined by the harmonic pulse can be achieved in principle. In this paper, we report an observation of amplification of a high harmonic from an infrared CPA laser in a collisionally pumped x-ray amplifier. This was achieved by injecting harmonic radiation from a frequency-doubled Nd:glass CPA laser into a neonlike gallium plasma. The 21st harmonic of the Nd:glass laser was tuned to give a spectral overlap with the $J=2-1$ transition of Ga XXII at a wavelength of 25.11 nm.

For these experiments, the high-energy Vulcan laser at the Rutherford Appleton Laboratory was configured to operate with one arm in the short-pulse CPA mode for the generation of harmonics and three arms to provide long-pulse pumping beams for the x-ray amplifier. The experimental setup is shown in Fig. 1. A 1-nJ, 850-fsec duration pulse from a Nd:LMA additive pulse mode-locked oscillator was stretched using a two-grating dispersive delay line, amplified to 10–30 J in one arm of the Vulcan Nd:glass laser chain, and then recompressed using a single-pass, two-grating system to a final pulse length of 1.3 psec [full width at half maximum (FWHM)]. The beam was then frequency doubled

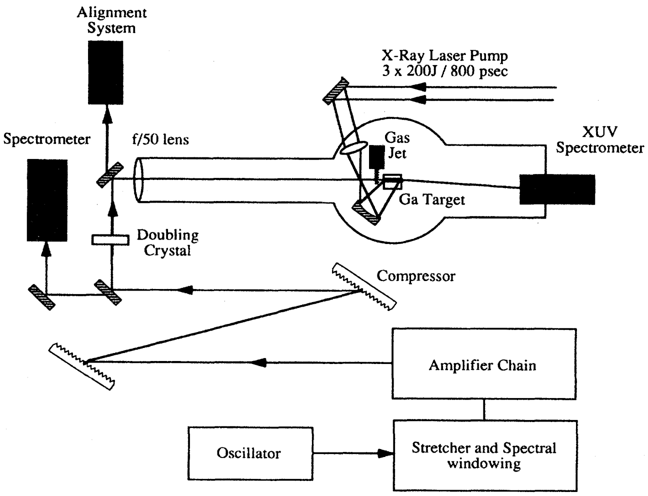

FIG. 1. Experimental setup for the injection of high harmonics into a neonlike Ga XXII plasma.

1050-2947/95/51(6)/4337(4)/$06.00

in a 4-mm-thick KD*P crystal, with a conversion efficiency of 30%, giving up to 5 J of second-harmonic light in a 7-cm-diam beam, which was focused with an $f=3.5$ m singlet lens ($f/50$) to an approximately $4x$ diffraction-limited spot achieving a peak intensity of $\sim 5\times 10^{15}$ W cm^2. The confocal parameter was approximately 12 mm. This focal geometry was chosen as an optimal compromise between maintaining good harmonic conversion efficiency, which requires a weak focus and a large focal spot size, and reasonable spatial overlap of the harmonic and the 100-μm Ga gain region. At focus the 527-nm beam traversed 1 mm of helium gas at an atom density of $\sim 1\times 10^{19}$ cm^{-3} produced by a supersonic gas jet [16].

The wavelength of the 21st harmonic must exactly match that of the 25.11-nm gain line of the soft-x-ray amplifier. The short-pulse Nd:LMA laser oscillator used to seed the CPA arm of the Vulcan operated at a center wavelength of 1055.4 nm and a bandwidth of 1.4 nm. Light entering the amplifier chain was tuned by spectral windowing, using a knife edge placed in the Fourier plane of the grating stretcher. This, in conjunction with gain, pulling in the glass amplifier chain determined the output spectrum of the amplified pulse.

The Ga laser amplifier was pumped by three 1054-nm beams, each providing 200 J in an 800-psec pulse to pump the soft-x-ray amplifier. Ga was chosen as the x-ray laser amplifier because of the very close overlap of one of the $J=2-1$ lines with the 21st harmonic of the frequency-doubled Nd:glass laser. The target was a vacuum-evaporated stripe of Ga on a glass substrate. The stripe was 600 nm thick, 200 μm wide, and up to 18 mm long. This target was irradiated from one side with three overlapped line foci produced by an off-axis lens-mirror system [17]. The pump beams were aligned such that both ends of the target were well illuminated. The separation of the gas jet and the entrance to the gallium amplifier was 20 mm, which is comparable to the Rayleigh range of the harmonic beam. This allowed for good coupling of the harmonics into the approximately 80-μm-diam gain region of the laser plasma, while being sufficiently far removed to avoid preionization of the gas jet by thermal xuv radiation from the plasma.

To align accurately the harmonic beam with the x-ray laser gain region, we used a surrogate stripe target with a 200-μm pinhole positioned at the end closest to the gas jet and centered 100 μm away from the surface of the Ga, the position of the expected gain region. Low-power shots along the harmonic beam line were imaged through the pinhole with an on-axis lens and a charge-coupled device (CCD) camera. The optimum position of the surrogate Ga target was referenced with a separate telescope system, and the surrogate was then replaced with a Ga stripe target for the amplification experiments.

The radiation from the harmonic beam was analyzed with an axial 1200-mm^{-1} flat-field grating xuv spectrometer run in the slitless mode and placed 2 m axially from the gas jet to avoid damage from the copropagating 527-nm laser radiation. Spectra were recorded with either a time-integrated, space-resolving xuv-sensitive back-illuminated CCD camera, or an x-ray streak camera with a low-density CsI photocathode. The CCD camera and grating spectrometer were calibrated with a known xuv source [18,19], enabling an absolute measurement of the incident and amplified harmonic energy. For x-ray laser or amplified harmonics shots, the instrument was shifted slightly off axis to allow for refraction by the density gradient in the gallium plasma. For the 18-mm plasma lengths used in the experiment we expect this shift to be on the order of 5 mrad. All stray light from the laser and emission from the plasma at wavelengths below 17.3 nm were rejected by a combination of a 400-nm-thick aluminum filter and reflections from a pair of 8° grazing-incidence gold mirrors placed at the spectrometer input.

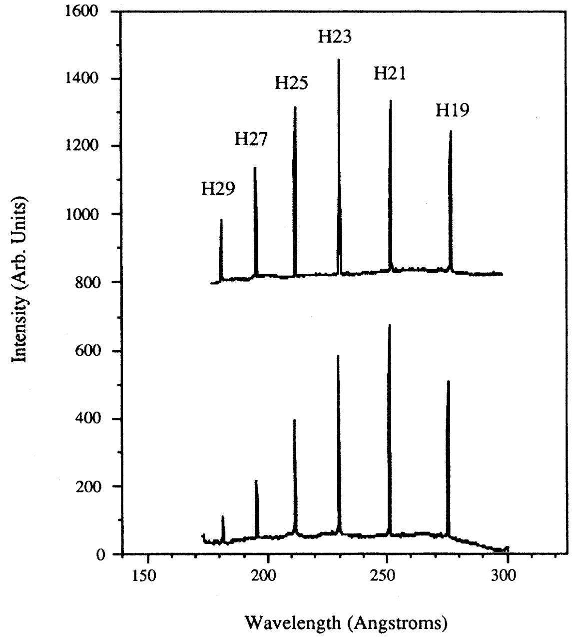

FIG. 2. Time-integrated harmonics spectra from He gas targets at an intensity of 4×10^{15} W/cm^2 showing little shot-to-shot variation in the relative size of the 19th, 21st, and 23rd harmonics. One spectrum is offset in intensity for clarity.

Lasing in Ga XXII had only been observed in one previous experiment [20], with few details and no gain coefficient reported. However, we expected that neonlike gallium would exhibit properties similar to the germanium XXIII system, its neighbor in the periodic table. This system has been extensively studied experimentally and computationally [21–23]. Hence, we anticipated that the 800-psec pump pulse would produce significant gain on the two gallium $J=2-1$ laser lines for approximately 400–600 psec, allowing relatively simple timing of the harmonic beam to the x-ray laser pump pulse [22].

The spectrum of the xuv harmonics produced in helium at a focused intensity of 10^{15} W/cm^2 is shown in Fig. 2. The measured energy of the 21st harmonic at 25.1 nm was between 0.5 and 2 nJ. The upper wavelength is limited by the spectrometer, the lower by the plateau cutoff of the harmonics. The efficiency of harmonic production is estimated to be $\sim 10^{-9}$, which is substantially less than has been previously demonstrated for optimum conditions in the plateau region [5]. We attribute this difference to the $4x$ diffraction-limited character of our beam, which reduced the coherent power to $\frac{1}{16}$ of the total and therefore reduced the conversion efficiency relative to that for a Gaussian beam. The maximum shot-to-shot variation of the ratio of the 21st harmonic to the 19th

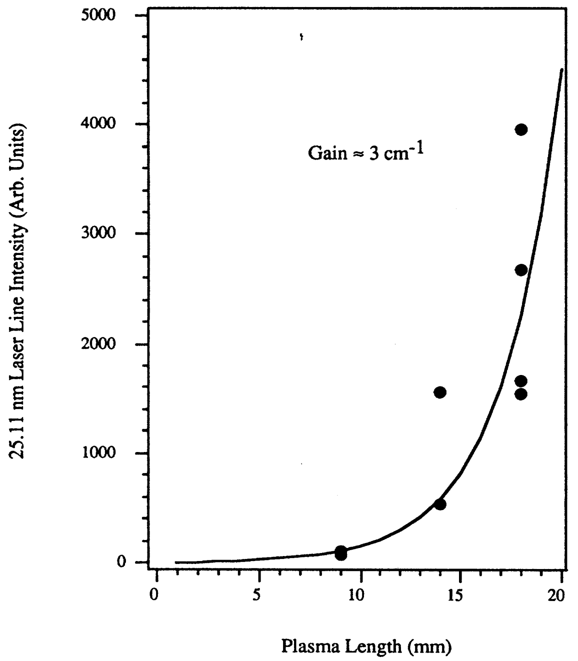

FIG. 3. Neonlike gallium $J=2-1$ 25.11-nm line intensity as a function of plasma length fitted to a Linford gain curve with a gain coefficient of ~3 cm^{-1}.

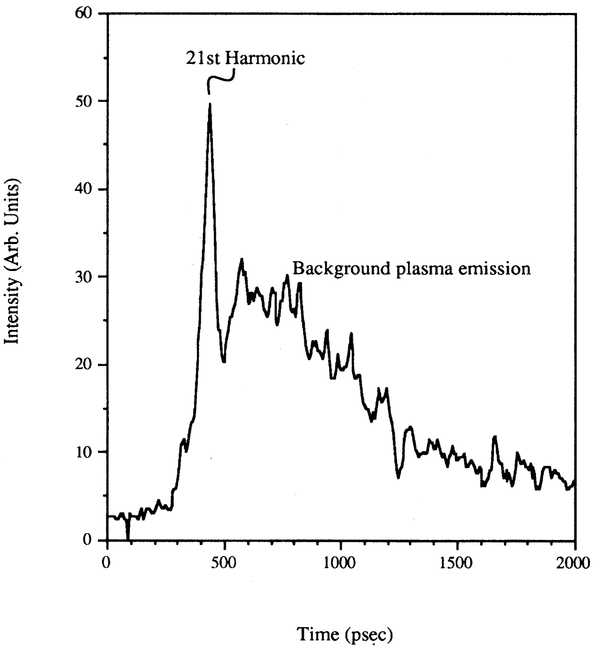

FIG. 4. Time evolution of the background plasma emission at 25.11 nm from an 18-mm Ga target, showing the amplified 21st harmonic signal at early time.

and 23rd harmonics was found to be on the order of ~20%, which allowed us to use this ratio as a relative intensity calibration of the 21st.

The gain of the Ga laser was measured using the axial flat-field spectrometer to determine the length dependence of the two gallium $J=2-1$ laser lines at 25.11 and 24.7 nm for plasmas up to 18 mm. Figure 3 shows the 25.11-nm line intensity as a function of plasma length, fitted to the gain formula of Linford *et al.* [24], with a gain coefficient of approximately 3 cm^{-1}. The time-integrated output of the 25.11-nm line for an 18-mm-long plasma was 10–20 nJ. With the harmonic beam injected, time-integrated spectra of combined harmonic and plasma emission clearly show the harmonic radiation visible above the continuum emission for orders other than the 21st (which is covered by the laser line). On these shots there was spatial breakup of the harmonic radiation into two lobes, which suggests that some of the harmonic radiation is being refracted by the plasma.

In order to distinguish the amplified 21st harmonic radiation from the longer-lived background plasma emission, we used a streak camera to time resolve the spectra. The combination of 100-psec/mm sweep rate and 0.3-mm slit width gave a temporal resolution of ~30 psec. Figure 4 shows the time evolution of the signal at 25.11 nm from an 18-mm Ga target shot with a clearly visible intensity spike at an early time from the injection of the 21st harmonic into the plasma 150 psec before the peak of the plasma emission. We find that the ratio of time histories at 25.11 and 24.7 nm (the positions of the two $J=2-1$ laser lines) is close to unity except at the point in time when the harmonic is injected. Studies on Ge show that the gain on the two $J=2-1$ lines follow each other closely over a wide range of conditions [19], and we see no intensity spike in the 24.7-nm time history corresponding to that at 25.11 nm.

In Fig. 5, we compare time-resolved spectra from a harmonic shot with no plasma present and a harmonic spectrum after traversing an 18-mm Ga plasma. These data represent the shot exhibiting the highest gain on the 21st harmonic. For

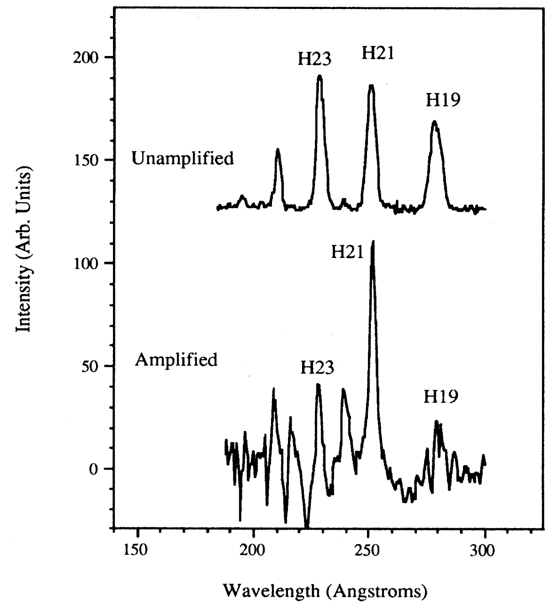

FIG. 5. Comparison of (background subtracted) harmonic spectra with and without a Ga XXII plasma present. Spectral data are integrated over a 30-psec time window and the unamplified spectrum is offset in intensity for ease of comparison.

these data, the streak camera was offset from the harmonic axis to account for the refraction of the harmonics by the Ga plasma and to sample only the harmonics that had traversed the Ga gain region. The harmonic was tuned to give optimum spectral overlap with the 25.11-nm laser line. These two lineouts were averaged over a 30-psec time window, the resolution of the streak camera. The instrument and filter response have been deconvolved and the background, found from averaging 30-psec intervals on either side of the harmonic signal, has been subtracted. There is a clear enhancement of approximately 3 of the 21st harmonic relative to the 19th and 23rd harmonics in the case where the harmonic propagates through the amplifying plasma.

We can estimate the expected gain for the 21st harmonic based on the measured 3-cm^{-1} gain coefficient for the 25.11-nm Ga XXII laser line and the coupling efficiency between the harmonic and the plasma amplifier. For complete coupling between the harmonic and the 18-mm x-ray amplifier, we would expect a small signal gain of $\exp[\alpha l] \approx 220$. However, this will be reduced by both the spectral and spatial overlap of the harmonic with the gain region of the plasma. Based on previous measurements of harmonic linewidths for 140-fsec pulses at 825 nm [25] we estimate the linewidth of the 21st harmonic to be ≈ 0.01 nm (FWHM), which is well below the resolving power of our xuv spectrometer. Measurements of the linewidths of similar collisionally pumped neonlike x-ray lasers can be used to infer the linewidth of the similar Ga laser [26]. From this work we estimate that the linewidth of the Ga XXII, $J=2-1$ transition is ≈ 0.005 nm. Thus, with optimum spectral overlap of the harmonic to the laser line, we calculate that the coupling efficiency will be $\approx 25\%$, assuming a convolution of Gaussian spectral profiles. This lowers the maximum gain to ~ 50. The observable harmonic gain is further reduced because only a fraction of the harmonic energy is coupled into the plasma since the spatial extent of the harmonic exceeds that of the gain region. We estimate from far-field measurements that the beam waist of the harmonics at the plasma is ~ 300 μm, while the diameter of the gain region is only ~ 80 μm. This further reduces the observable gain to ~ 3.5.

In conclusion we have demonstrated the injection and amplification of high-order harmonic radiation in a collisionally pumped xuv amplifier. A net energy gain of 3 was achieved for the 21st harmonic of a frequency-doubled Nd:glass laser in a neonlike gallium plasma at 25.11 nm. The measured gain is in good agreement with that calculated when the spatial and spectral overlap of the harmonic with the amplifier is accounted for. In an optimized experiment using a more tunable source and better mode matching between the harmonic source and amplifier, considerably higher amplification could be possible. We believe that this can provide a method of achieving short-pulse operation of soft-x-ray lasers with improved spatial coherence for use in applications such as ultrashort pulse probing of laser plasmas. In addition, this technique can be used for direct measurements of the spatial and temporal distribution of gain in x-ray laser plasmas.

We gratefully acknowledge the efforts of the Rutherford Appleton Laboratory target preparation group and the Vulcan laser operators. This work was supported by an EPSRC grant and the U.S. Dept. of Energy, under Contract No. W-7405-Eng-48. R. A. Smith is supported by the EPSRC.

---

[1] P. Maine *et al.*, IEEE J. Quantum Electron. **24**, 398 (1988).
[2] A. L'Hullier and Ph. Balcou, Phys. Rev. Lett. **70**, 774 (1993).
[3] J. J. Macklin *et al.*, Phys. Rev. Lett. **70**, 766 (1993).
[4] M. D. Perry and G. Mourou, Science **264**, 917 (1994).
[5] T. Ditmire, J. K. Crane, H. Nguyen, L. B. DaSilva, and M. D. Perry, Phys. Rev. A **51**, R902 (1995).
[6] J. W. G. Tisch, R. A. Smith, J. E. Muffett, M. Ciarrocca, J. P. Marangos, and M. H. R. Hutchinson, Phys. Rev. A **49**, R28 (1994).
[7] P. Salières, T. Ditmire, K. S. Budil, M. D. Perry, and A. L'Huillier, J. Phys. B **27**, L217 (1994).
[8] D. Mathews *et al.*, Phys. Rev. Lett. **54**, 110 (1985).
[9] B. J. MacGowan *et al.*, Phys. Rev. Lett. **59**, 2157 (1987).
[10] C. Chenais-Popovics, R. Corbett, C. J. Hooker, M. H. Key, G. P. Kiehn, C. L. S. Lewis, G. J. Pert, C. Regan, S. J. Rose, S. Sadaat, R. Smith, T. Tomie, and O. Willi, Phys. Rev. Lett. **59**, 2161 (1987).
[11] B. E. Lemoff *et al.*, Phys. Rev. Lett. **74**, 1574 (1995).
[12] J. J. Rocca *et al.*, Phys. Rev. Lett. **73**, 2192 (1994).
[13] L. B. DaSilva, B. J. MacGowan, J. A. Koch, R. A. London, S. Mrowka, and J. H. Underwood, Opt. Lett. **18**, 1174 (1993).
[14] A. Carillon, H. Z. Chen, P. Dhez, L. Dwivedi, J. Jacoby, P. Jegle, G. Jamelot, Jie Zhang, M. H. Key, A. Kidd, A. Klisnick, R. Kodama, J. Krishnan, C. L. S. Lewis, D. Neely, P. Norreys, D. O'Neill, G. J. Pert, S. A. Ramsden, J. P. Raucourt, G. J. Tallents, and J. Uhomoibhi, Phys. Rev. Lett. **68**, 2917 (1992).
[15] G. F. Cairns, C. L. S. Lewis, A. G. MacPhee, D. Neely, M. Holden, J. Krishnan, G. J. Tallents, M. H. Key, P. N. Norreys, C. G. Smith, J. Zhang, P. B. Holden, G. J. Pert, J. Plowes, and S. A. Ramsden, Appl. Phys. B **58**, 51 (1994).
[16] M. D. Perry, C. Darrow, C. Coverdale, and J. K. Crane, Opt. Lett. **17**, 523 (1992).
[17] I. N. Ross *et al.*, Appl. Opt. **26**, 1584 (1987).
[18] L. B. DaSilva (private communication).
[19] G. Kiehn *et al.*, Proc. SPIE **831**, 150 (1987).
[20] T. N. Lee *et al.*, Bull. Am. Phys. Soc. **33**, 1920 (1988).
[21] T. N. Lee *et al.*, Phys. Rev. Lett. **59**, 1185 (1987).
[22] D. M. O'Neill, C. L. S. Lewis, D. Neely, J. Uhomoibhi, M. H. Key, A. MacPhee, G. J. Tallents, S. A. Ramsden, A. Rogoyski, E. A. McLean, and G. J. Pert, Opt. Commun. **75**, 406 (1990).
[23] P. B. Holden *et al.*, J. Phys. B **27**, 341 (1994).
[24] G. J. Linford *et al.*, Appl. Opt. **13**, 379 (1974).
[25] T. Ditmire, K. S. Budil, J. K. Crane, H. Nguyen, and M. D. Perry, in *Ultrafast Phenomena IX*, edited by P. F. Barbara, W. H. Knox, G. A. Mourou, and A. H. Zewail, Springer Series in Chemical Physics Vol. 60 (Springer-Verlag, Berlin, 1994).
[26] J. A. Koch *et al.*, Phys. Rev. A **50**, 1877 (1994).