

# REPORT DOCUMENTATION PAGE

Form Approved  
OMB No. 0704-0188

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1. AGENCY USE ONLY (Leave blank)      2. REPORT DATE      3. REPORT TYPE AND DATES COVERED  
FINAL REPORT - 01 Apr 93 - 30 Sep 96

4. TITLE AND SUBTITLE  
PICOSECOND X-RAY SPECTROSCOPY

5. FUNDING NUMBERS  
61102F  
2301/BS

6. AUTHOR(S)  
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AFOSR-TR-~~97~~  
97

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)  
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0135

8. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)  
AFOSR/NE  
110 Duncan Avenue Suite B115  
Bolling AFB DC 20332-8050

10. SPONSORING/MONITORING AGENCY REPORT NUMBER  
F49620-93-1-0254

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT  
APPROVED FOR PUBLIC RELEASE: DISTRIBUTION UNLIMITED

12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)

During this project, we have demonstrated the generation of coherent soft-x-ray pulses with *sub-10fs* duration. These results surpassed our own expectations. The coherent x-rays are generated by high-harmonic emission generated by focusing an intense femtosecond laser pulse into a gas jet. The increased time resolution ( $\ll 100$  fs) we have available is very important to enable the direct observation of atomic motion - we can resolve atomic dynamics (in chemical reactions and in phase transitions for example) on the timescale comparable to the first steps of the process.

14. SUBJECT TERMS

15. NUMBER OF PAGES

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT  
UNCLASSIFIED

18. SECURITY CLASSIFICATION OF THIS PAGE  
UNCLASSIFIED

19. SECURITY CLASSIFICATION OF ABSTRACT  
UNCLASSIFIED

20. LIMITATION OF ABSTRACT

19970314 046

## Picosecond X-Ray Spectroscopy

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### 2. Objectives

The objective of this project is to demonstrate the feasibility of time-resolved x-ray spectroscopy on a picosecond time-scale.

### 3. Status of Project

During this project, we have demonstrated the generation of coherent soft-x-ray pulses with *sub-10fs* duration. These results surpassed our own expectations. The coherent x-rays are generated by high-harmonic emission generated by focusing an intense femtosecond laser pulse into a gas jet. The increased time resolution ( $\ll 100$  fs) we have available is very important to enable the direct observation of atomic motion - we can resolve atomic dynamics (in chemical reactions and in phase transitions for example) on the timescale comparable to the first steps of the process.

Even more important is the fact that using very short 25fs excitation pulses, we can generate x-rays more efficiently than with longer 100fs pulses. We can also tune the wavelength of the x-ray harmonics to probe different chemical species.

### 4. New Findings and Research Highlights

We found that the process of high-harmonic generation, which can generate coherent light in the soft-x-ray region, works substantially better using the very short excitation pulses we have developed. Previous work used pulsewidths  $> 100$ fs, and generated up to the 135th harmonic. For our work however, we used 25fs excitation laser pulsewidths, and found that -

- 1) We can generate coherent soft-rays with wavelengths down to  $47\text{\AA}$ . This represents a substantial improvement over previous work, which generated harmonics down to  $70\text{\AA}$ .
- 2) We can generate x-rays significantly more efficiently than with 100 fs pulses.
- 3) The soft-x-ray pulsewidths are less than 10fs in duration.
- 4) We can continuously tune the harmonic wavelength by adjusting the chirp of the laser pulse, to completely span the soft-x-ray region of the spectrum.
- 5) Using our 25fs laser pulses to drive high-harmonic emission, we are just beginning to enter a new non-adiabatic regime of high-field physics, where the response of the system depends on the time-history (pulse shape) of the excitation.
- 6) Finally, we developed and experimentally demonstrated the fastest x-ray detector to date, with a time-resolution of 0.5ps. We are now working with Roger Falcone at Berkeley to use this detector for time-resolved x-ray studies at the Advanced Light Synchrotron Source.

These results are illustrated in Figs. 1, 2, and 3. Figure 1 shows the high-harmonic emission in argon as a function of pulse duration. Figure 2 shows the shortest wavelength coherent x-rays obtained to date. Figure 3 shows the output of the fastest x-ray detector to date. The extremely short risetime of our excitation pulses (a few optical cycles) is shorter than the tunneling time for ionization of an atom. Therefore, an atom exposed to a very fast risetime pulse will be stable against ionization up to a higher intensity than for a 100fs excitation pulse. Since ionization then occurs at a higher laser intensity, the electron can gain more energy in the laser field, and thus radiate higher harmonics. A detailed explanation of these results is given in the attached reprints.

Our results are very important for application experiments in materials science, chemistry and surface science, since we can generate a good flux of x-rays with only a few millijoules of laser energy, provided that we use a very short excitation pulse (25fs). Since we can generate millijoule-level laser pulsewidths at kHz repetition rates, we can therefore generate high-repetition-rate soft-x-ray and ultraviolet sources, with 10fs x-ray pulsewidths. This source can compete very well with synchrotron sources in terms of flux for time-resolved experiments, since the high-repetition rate of the synchrotron cannot be used when synchronized with a laser (usually kHz). Of course, it surpasses all other sources in terms of pulsewidth.

#### **5. Personnel associated with project**

Professor Margaret Murnane  
Professor Henry Kapteyn  
Dr. Jianping Zhou (recent graduate, now working at Spectra-Physics Lasers)  
Dr. Chung-Po Huang (recent graduate, now working at Robotics Vision)  
Dr. Zenghu Chang (visiting scientist from Xian Institute, China)  
Professor Ivan Christov (visiting scientist from Sofia, Bulgaria)  
Kendall Read (graduate student)  
An-Chun Tien (graduate student)  
Andy Rundquist (graduate student)  
Justin Peatross (postdoctoral researcher)  
Haiwen Wang (graduate student)  
Chengyu Shi (graduate student, no longer in group)

#### **6. Publications acknowledging AFOSR support**

1. Jianping Zhou, Chung-Po Huang, Margaret M. Murnane, and Henry C. Kapteyn, "Amplification of 26 fs, 2 TW pulses near the gain narrowing limit in Ti:sapphire," *Optics Letters* **20**, 64 (1995).
2. "Amplification of 26 fs, 2 TW pulses in Ti:sapphire," C.P. Huang, J. Zhou, H. Kapteyn, M. Murnane, to be published in to be published in *Proceedings of the SPIE*, Vol. **2377** (1995).
3. "Enhanced High-Harmonic Generation using 26 Femtosecond Laser Pulses", J. Zhou, J. Peatross, M.M. Murnane, H.C. Kapteyn, *Physical Review Letters* **76**, 752 (1996).
4. "Generation and Application of High Power Ultrashort Pulses", M.M. Murnane, H.C. Kapteyn, *Proc. SPIE* **2524**, to be published, 1995.
5. "Recent Advances in Femtosecond Laser Technology", H.C. Kapteyn, M.M. Murnane, *Proc. SPIE* **2524**, to be published, 1995.
6. "Ultrashort Light Pulses: Pushing the Limits", M.M. Murnane, H.C. Kapteyn, *IEEE LEOS Monthly Newsletter*, August 1995.

7. "Non-Adiabatic Effects in High-Harmonic Generation with Ultrashort Pulses", I.P. Christov, J. Zhou, J. Peatross, A. Rundquist, M. Murnane, H.C. Kapteyn, Review Letters **77**, 1743 (1996).
8. Z. Chang, A. Rundquist, J. Zhou, M. Murnane, H. Kapteyn, X. Liu, B. Shan, J. Niu, M. Gong, X. Zhang, "Demonstration of a Sub-Picosecond X-Ray Streak Camera", Applied Physics Letters **69**, 133 (1996).

**7(a). Presentations acknowledging AFOSR support**

1. **Invited talk**, SPIE Annual Symposium OE'LASE, San Jose, CA, February 1995. "Generation of 26 fs, 2 TW pulses near the gain-narrowing limit in Ti:sapphire."
2. **Invited Tutorial**, APS March Meeting, San Jose, CA, March 1995. "Advances in Femtosecond Lasers."
3. **Invited talk**, NSF Forum on Optical Science and Engineering, SPIE Annual Meeting, San Diego, CA, July 1995. "Generation and Applications of High Power Ultrashort Pulses."
3. **Invited Talk**, Canadian Association of Physicists Annual Meeting, Quebec 1995. "Advances in Short-Pulse Femtosecond Laser Technology."
4. **Invited Talk**, SPIE Annual Meeting, San Diego, CA 1995. "Advances in Femtosecond Laser Technology"
5. **Invited Talk**, Canadian National Research Council, Ottawa, Canada 1995. "Recent Advances in Femtosecond Laser Technology."
6. **Plenary Talk**, Australian Optical Society Annual Meeting, Brisbane, Australia, July 1995. "Advances in Femtosecond Lasers."
7. Contributed talk, Optical Society of America Annual Meeting, Portland, OR 1995. "Ultrafast X-Ray Generation with Sub-30 fs TW Pulses."
8. **Invited talk**, Euroconference on Generation and Application of Ultrashort X-Ray Pulses II, Pisa, Italy 1995. "High Intensity Laser-Matter Interactions with sub-30fs Optical Pulses."
9. **Invited talk**, NATO Advanced Research Workshop on Super-Intense Laser Atom Physics (SILAP IV) Moscow 1995. "High-Order Harmonic Generation with a 25 Femtosecond Laser Pulse."
10. **Invited talk**, AAPT Summer Meeting, Spokane, WA 1995. "Advances in Femtosecond Lasers: Applications in Research, Teaching and Business."
11. Colloquium, University of Maryland, February 1995.
12. Colloquium, University of Michigan, Ann Arbor, MI, March 1995.
13. Seminar, Lawrence Livermore National Laboratory, March 1995.
14. Colloquium, Utah State University, April, 1995.
15. Seminar, University of Washington, May 1995.

16. Colloquium, State University of New York at Stony Brook, October 1995.
17. Colloquium, University of Texas at Austin, November 1995.
18. Colloquium, University of Texas at Austin, November 1995.
19. Colloquium, Ohio State University, April 1996.
20. Colloquium, Harvard University, April 1996.
21. Seminar, Purdue University, April 1996.
22. Colloquium, Swarthmore College, October 1996.
23. Colloquium, University of California at Berkeley, Fall 1997.

#### **7(b). Technology transfer**

We have been involved in significant technology transfer efforts in our laboratory. We now have a company, Kapteyn/Murnane Laboratories L.L.C., to sell ultrashort-pulse lasers. We market lasers which can generate shorter pulses ( $< 20$  fs) than otherwise available commercially. Such short pulses have applications in chemistry, biochemistry, materials science, engineering, and high-field physics. This laser work developed from an SGER funded jointly by AFOSR. We have sold lasers to LLNL, Argonne Natl. Lab, LBL, NIST, and scientists both within and outside of the US.

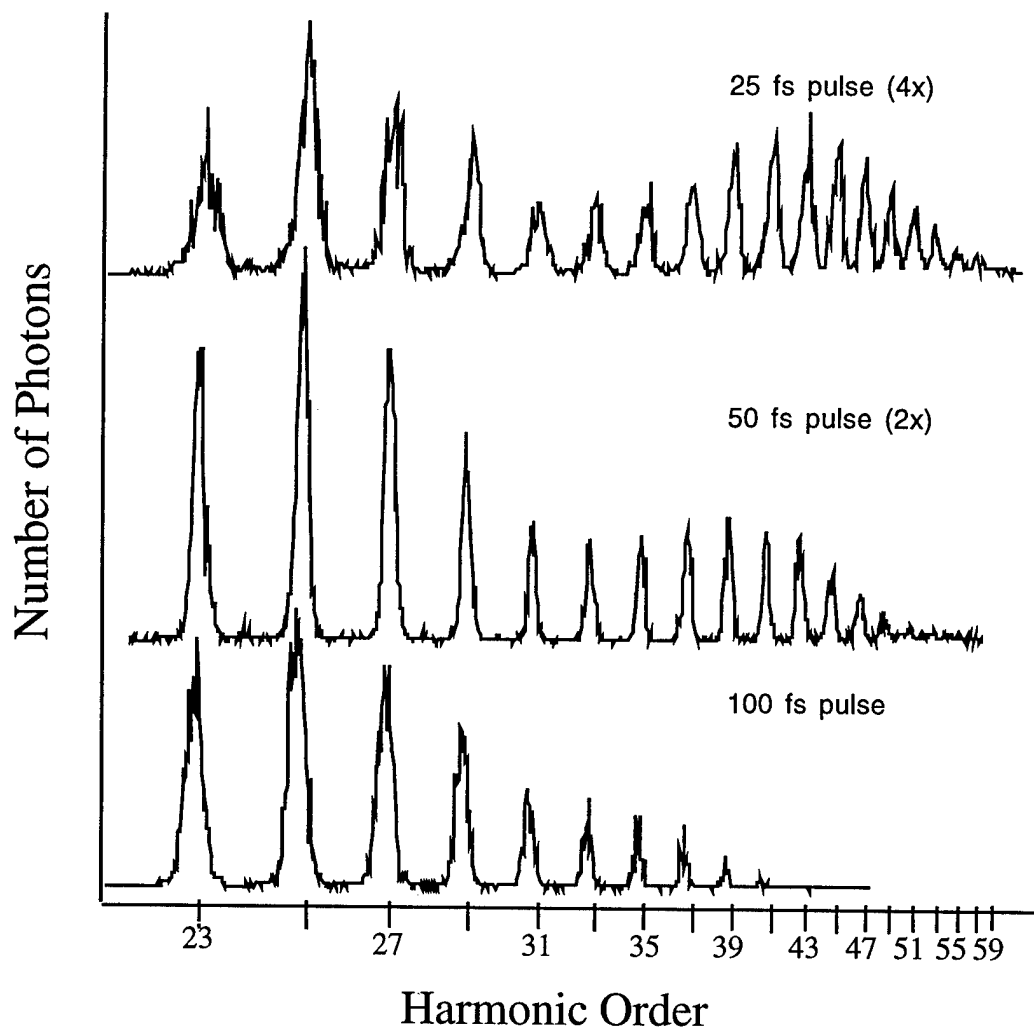
We also worked with Excel/Quantronix to commercialize our amplifier technology, and they now have a commercial product based on our design.

**8. Patents** None

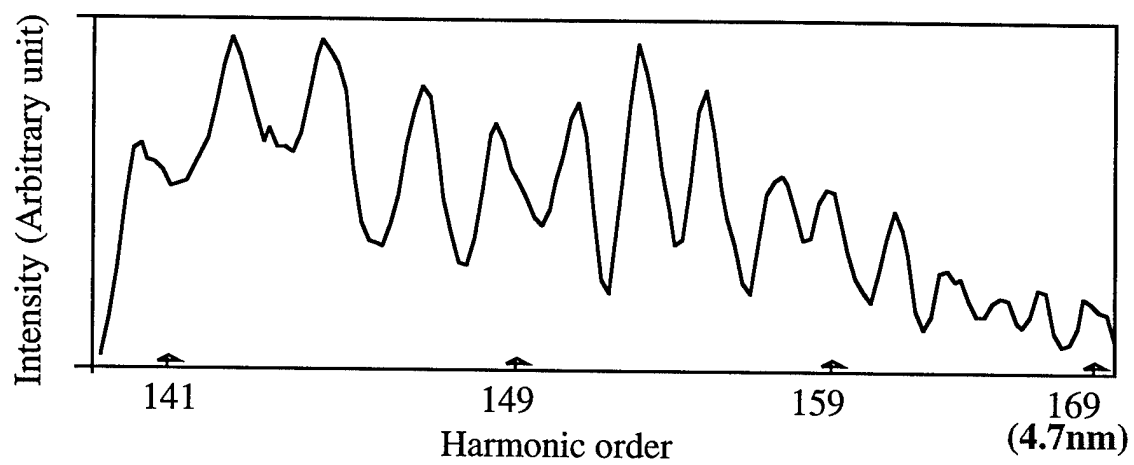
#### **9. Honors**

1995 Sloan Foundation Fellowship awarded to co-PI Prof. Henry Kapteyn

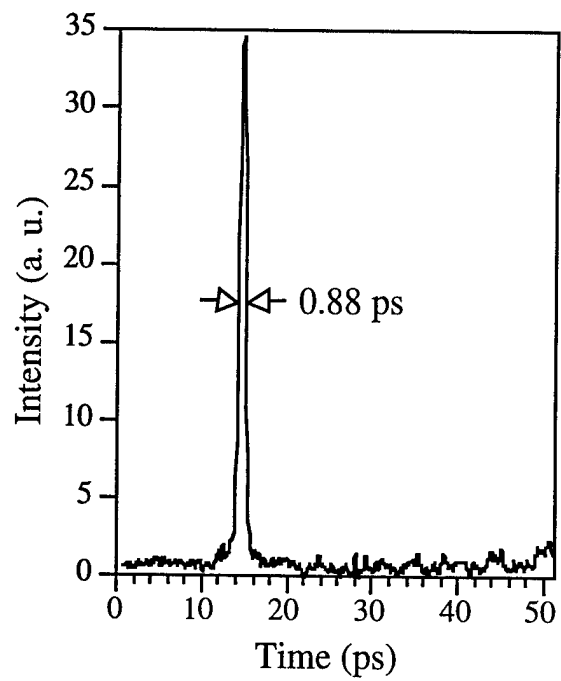
1997 Maria Goeppert-Mayer Award of the American Physical Society to P.I. Margaret Murnane



**Figure 1:** High-Harmonic emission in argon as a function of transform-limited pulsewidth



**Figure 2:** Single-shot high harmonic emission from He using a 25fs excitation pulse, showing coherent x-ray emission down to 47Å.



**Figure 3:** X-ray pulsewidth measurement using ultrafast x-ray detector

## Enhanced High-Harmonic Generation Using 25 fs Laser Pulses

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(Received 7 June 1995)

We present experimental and theoretical results on high-harmonic generation in noble gases using an 805 nm, 25 fs, titanium-doped sapphire laser. The harmonic energies observed are unexpectedly high when compared with experimental and theoretical results to date for longer excitation pulses. We observe that the efficiency of harmonic production is highest for shorter pulses. Furthermore, the wavelength of the harmonics can be tuned by adjusting the sign of the chirp of the excitation pulse, demonstrating a tunable, ultrashort-pulse, <25 fs soft-x-ray source.

PACS numbers: 42.50.Hz, 32.80.Rm, 42.65.Ky, 42.65.Re

The occurrence of a plateau in the conversion efficiency of high-order harmonic generation of intense laser pulses incident on gases has been well documented both experimentally [1–5] and theoretically [6–12]. A variety of wavelengths and pulse durations have been used to generate harmonics, and harmonic orders well exceeding one hundred have been observed by L'Huillier and Balcou from the two lighter noble gases using 1 ps, 1054 nm laser pulses [3]. Macklin, Kmetec, and Gordon observed harmonics up to the 109th order in neon using 125 fs, 806 nm pulses, which represents the shortest wavelength harmonic reported to date [4]. In heavier noble gases, which have smaller ionization potentials, the number of harmonics which can be generated is less, although they have higher conversion efficiencies. L'Huillier and Balcou observed up to the 55th and 27th harmonics from argon and xenon, respectively, with the 1 ps, 1054 nm laser pulses [3]. These observations are in agreement with theoretical predictions [10,11] that the photon energy of the highest harmonic emitted from a gas cannot exceed  $I_p + 3.2U_p$ , where  $I_p$  is the atomic ionization potential and  $U_p \sim \lambda^2 I$  is the maximum ponderomotive potential that an electron may experience prior to detachment from the atom, and  $\lambda$  and  $I$  are the laser wavelength and intensity, respectively.

Krause, Schafer, and Kulander [10] used *ab initio* calculations of the Schrödinger equation in three dimensions to show that the breadth of the plateau in the harmonic spectrum obeys this cutoff rule, where  $U_p$  is chosen at the point where the atom ionizes, even if the peak laser intensity goes higher. They also showed that this rule can be understood with a classical picture, where the electron detaches from the atom and releases energy when it is recaptured by the atom after a laser cycle [11]. The classical picture predicts that the maximum kinetic energy acquired by an electron from the field upon return to the nucleus is  $3.2U_p$ . Quantum-mechanical descriptions of high harmonic generation give similar results [8,12]. All of these

models agree with harmonic generation measurements to date, which have been made with laser pulses of duration greater than 100 fs. They also use the adiabatic assumption, where the laser intensity varies slowly with respect to an optical period.

For our work, we investigated high harmonics generated by a 25 fs, 10 Hz, 3 TW Ti:sapphire laser [13] in various noble gases [14]. The bandwidth of the pulses is 32 nm, centered at a wavelength of 805 nm, and the laser system can provide up to 70 mJ of energy per pulse, with shot-to-shot fluctuations of  $\approx 10\%$ . The laser beam is linearly polarized and focused by a curved mirror of focal length 1 m to 1.2 times the diffraction limit as measured by a charge coupled device camera, with  $f/80$  focusing. A gas target  $\approx 1$  mm thick was placed 2.5 cm after the focus, where the laser diameter was approximately  $280 \pm 50 \mu\text{m}$ . The ultrashort nature of our excitation pulses (10 optical cycles FWHM) implies that at the half maximum position of the temporal pulse envelope, the laser intensity changes by more than 25% during a single cycle. This defies the adiabatic assumption, which suggests that the atomic dipole moment undergoes quasiperiodic motion from cycle to cycle, with no dependence on the history of the pulse.

The gas target [15] consists of two thin metal plates with a  $500 \mu\text{m}$  hole drilled through them. The laser goes through the hole and interacts with the gas which enters the hole from between the plates. The device was backed continuously with pressures of 5–50 Torr, and the pressure inside the hole was about one-fifth of the backing pressure. Because the laser confocal parameter is much larger than the gas target thickness, the harmonics can be thought of as emerging from a plane so that geometric phase matching should not critically depend on the  $z$  position of the target. We placed that target after the focus mainly to increase the size of the laser spot and hence increase the signal. The harmonic signals are resolved with HIREF-SXR-1.75 monochromator (Hettrick Scientific) and measured with a

microchannel plate detector. The spectrometer, which had a resolution of  $\approx 1 \text{ \AA}$ , was scanned at a rate of  $\sim 0.02 \text{ \AA}$  per laser shot, and the harmonic signal from each laser shot was recorded. Typically,  $\approx 12\,000$  points make up a given spectrum.

For the heavier noble gases, we observed harmonics with photon energies remarkably higher than previously seen. Figure 1(a) shows harmonics generated in argon, where orders up to the 61st are visible. This corresponds to a photon energy of 93 eV. The lower harmonic orders are artificially damped by the spectrometer grating efficiency. Figure 1(b) shows harmonics up to the 41st generated krypton and harmonics up to the 29th generated in xenon. These are approximately 40% higher harmonic photon energies than have been seen previously using these gases [3]. The increase in harmonic photon energy is especially interesting from the point of view that for a given intensity the ponderomotive potential  $U_p$  of our laser is 42% less than that of the 1054 nm laser used in the experiments by L'Huillier and Balcou [3]. Thus, according to the  $I_p + 3.2U_p$  rule, one would expect to see harmonics of lesser photon energy with our laser, not greater. This suggests that for ultrashort-pulse excitation, atoms can survive to higher laser intensities before ionizing. In neon, we were unable to resolve harmonics past the 105th, but we saw light which may correspond to harmonic orders up to the 131st. As we reduced the laser intensity, the short wavelength edge gradually retreated to longer wavelengths, indicating that the shortest wavelength light

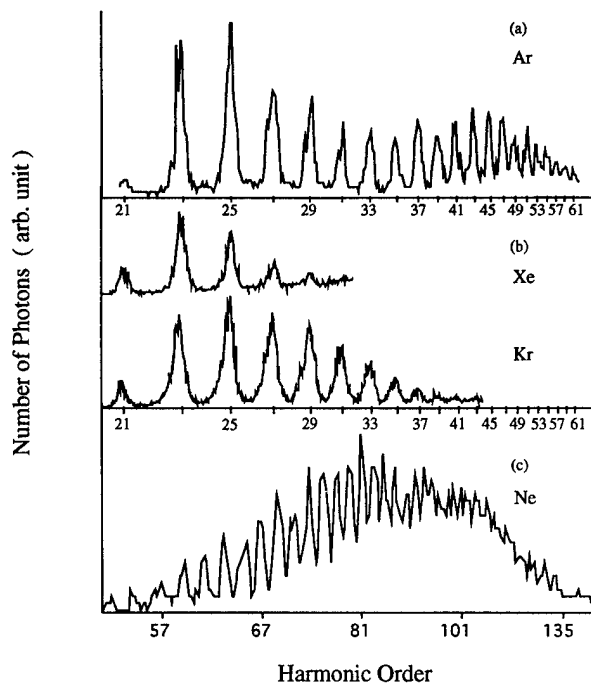


FIG. 1. Harmonic spectra generated in (a)  $\sim 5$  Torr of Ar, and (b) 2 Torr of Kr and Xe, with peak laser intensity  $5 \times 10^{14} \text{ W/cm}^2$ . (c) Harmonic spectra generated in  $\sim 10$  Torr of Ne with peak laser intensity  $2 \times 10^{15} \text{ W/cm}^2$ . The relative intensities of (a)–(c) should not be compared directly.

is real. Figure 1(c) shows harmonic spectra of Ne. The lower harmonic orders seen are artificially damped by the spectrometer grating efficiency, since we had to use a different grating to observe these shorter wavelength harmonics.

The spectra seen in Figs. 1(a)–1(c) were produced with a laser energy of 3.5 mJ, which for the focusing conditions described above corresponds to a peak intensity of approximately  $(5 \pm 2) \times 10^{14} \text{ W cm}^{-2}$ . This is significantly above ( $\times 2$ ) the point where ionization should readily occur. The possibility therefore exists that the highest harmonics might arise from ions, in which case they would not be considered to be of unexpectedly high order. To check this, we observed harmonic generation in argon as a function of gas pressure. We observed that all of the harmonic peaks decreased in strength together as the pressure was gradually reduced from 5 to 1 Torr. If the higher-order harmonic peaks were produced by ions while the lower by neutral atoms, one would expect the harmonics to scale very differently with pressure because of a changing coherence length arising from free electrons. Thus our observations suggest that the harmonic peaks all arise from neutral atoms. Our spectra are, however, integrated over space and time from an ensemble of atoms experiencing different laser intensities.

One model for estimating the intensity at which ionization occurs is barrier suppression ionization (BSI) [16]. In this picture, the outer electron suddenly escapes from the atom when the laser field suppresses the potential barrier below the field-free binding energy of the electron. This model has worked well for predicting the intensities at which atoms ionize for 1 ps, 1054 nm, laser pulses. According to this model, the intensities at which Ne, Ar, Kr, and Xe ionize are, respectively,  $8.7 \times 10^{14}$ ,  $2.5 \times 10^{14}$ ,  $1.5 \times 10^{14}$ , and  $8.6 \times 10^{13} \text{ W/cm}^2$ . The harmonic photon energies that we observed from Ar, Kr, and Xe well exceed the theoretical limit when the BSI assumption is used to determine  $U_p$ . In fact, under the BSI assumption, the harmonic photon energies would exceed  $I_p + 5U_p$  for each gas. Thus it would seem that for ultrashort pulses the BSI model is not applicable.

We investigated numerically the dependence of ionization on pulse duration for pulses of fixed peak intensity. This was done by integrating the time-dependent Schrödinger equation in one dimension. While the solution in one dimension cannot be expected to provide an exact description of ionization, it can be interesting to compare the ionization probabilities for different applied pulse durations. The field-free potential was chosen to have the form  $-1/(|x| + a)$  in atomic units [17], where  $a = 3.7 \times 10^{-2}$  a.u. The calculation showed that for our experimental conditions, atoms can experience 20% higher laser intensity before the ionization probability exceeds 10% for a 25 fs excitation pulse, compared with a 100 fs pulse. The details depend on the exact shape assumed for the leading edge of the laser pulse, which has not been experimentally determined as yet. However, in

contrast with the BSI model, the implication is that, even in the tunneling regime, neutral atoms are able to experience higher ponderomotive potentials under ultrashort-pulse illumination than is possible using longer excitation pulses, since atoms can survive higher intensities before ionizing.

The  $I_p + 3.2U_p$  rule for the highest possible harmonic photon energy comes from a classical consideration of the maximum energy that an electron can have when a laser of constant intensity pushes it back into its parent ion after having broken away during the previous cycle. Since this process requires the time scale of a laser cycle, the fact that our laser intensity changes during this time can influence the process. During the rising edge of the pulse, the relevant intensity for calculating the ponderomotive potential is higher than that at which the electron breaks away from the atom, since the field strength grows before the electron can return. This consideration alone cannot explain the anomalously high harmonic orders observed, but it does predict some enhancement.

To investigate how important the shortness of our laser pulse duration is to the total number of harmonic orders observed, we increased our laser pulse duration by introducing chirp. We did this by changing the position of the grating in our pulse stretcher prior to amplification. The beam alignment and pulse spectrum after amplification and compression remained unchanged. Calculations of the pulse propagation through our laser system suggest that the temporal profile of the pulse remains smooth as the chirp is varied.

We examined the effect of introducing various amounts of chirp, both positive and negative, on harmonic production in argon. Figure 2 shows harmonic spectra for different pulse durations at a pressure of 5 Torr. The harmonic spectrum generated by a transform-limited 25 fs pulse is shown in the middle. Above it are spectra from positively chirped pulses, and below it are spectra from negatively chirped pulses of similar durations. In all cases the peak intensity of the laser pulse was held fixed so that the longer pulses required more energy.

For almost all of the pulse durations shown in Fig. 2, we observe unexpectedly higher order harmonics than predicted by the BSI theory, which predicts that the 41st order should be the maximum observed. The photon energies in all cases are higher than those previously observed in neutral argon, although the highest observable harmonic order decreases gradually with increasing pulse duration. Another striking feature about the data in Fig. 2 is that the area associated with given harmonic peaks remains roughly the same for the different spectra. Since the laser energy is increased to keep the intensity fixed for the longer durations, the harmonic production process is most efficient for the shortest laser pulses. In other words, shorter laser pulses produce the most harmonic signal for a given laser energy, with the harmonic efficiency scaling inversely with pulse duration. This agrees with the findings of Kondo *et al.* [5] who reported on harmonics generated with a 120 fs Ti:sapphire laser. They stretched the laser pulse dura-

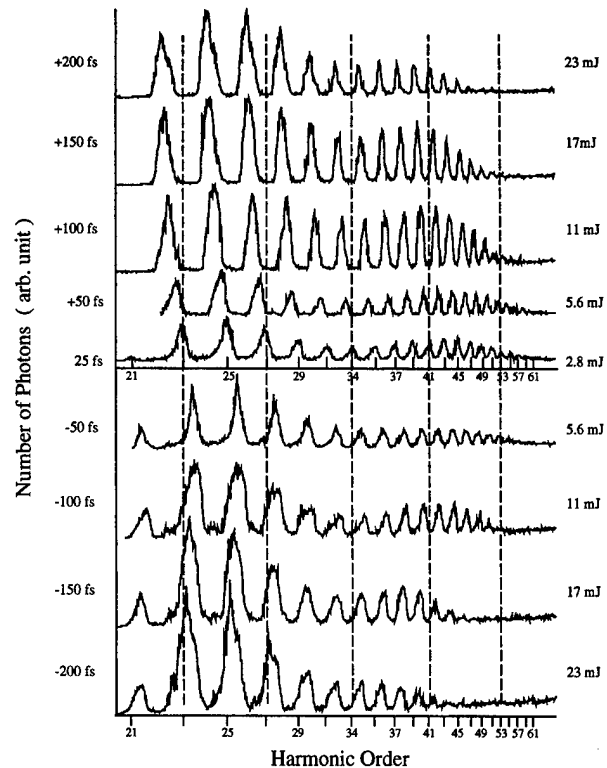


FIG. 2. Harmonic spectra generated in 5 Torr of argon for fixed peak laser intensity of  $5 \times 10^{14}$  W/cm<sup>2</sup> and for various laser pulse chirps.

tion to 750 fs by introducing chirp and increasing the laser energy to keep the peak intensity fixed. They observed no significant change in the harmonic yield over this range. Together the two sets of experiments suggest that high-harmonic conversion efficiency is roughly inversely proportional to pulse duration for pulse widths shorter than 1 ps.

In Fig. 2, a significant amount of redshifting and blueshifting of the output spectra can be seen when positive and negative chirps are introduced, respectively. This indicates that the harmonic production tends to favor the rising edge of the laser pulse and that it is unlikely that harmonic emission comes from ions. The harmonic peaks can be shifted spectrally to the degree that a continuously tunable light source is obtained for wavelengths shorter than 25 nm. The degree of blueshifting and redshifting is not the same for pulses of similar duration but with opposite chirp. For positive chirp, the redshifting of the peaks is very pronounced while the peaks remain relatively narrow. However, for the negative chirps, the harmonic peaks initially shift slightly to the blue and then broaden to approximately twice the former width. The bandwidths observed for individual peaks are sufficiently broad to support subfemtosecond soft-x-ray pulses, although it is not known as yet if the proper phase conditions are met. One possible explanation for the asymmetry observed in the shifting and broadening of the harmonic spectra as a function of laser chirp is that the negative chirp induces a

stabilizing effect against ionization. Thus, in the case of negative chirps, the atom could experience a wider range of frequencies.

Our model calculations show similar spectral shifting and broadening of the harmonics with different signs of the excitation laser chirp. The calculations show that at the point on the leading edge of the pulse profile where a negatively chirped 100 fs pulse (with the same bandwidth as a 25 fs pulse) causes 50% ionization of the atom, there is 68% ionization for a positively chirped 100 fs pulse with the same intensity. Thus, when irradiated by a negatively chirped excitation pulse, the atom survives longer and is exposed to a greater fraction of the input bandwidth than for the positively chirped case. This may explain why the harmonic peaks are broader, and shift less, for a negatively chirped excitation pulse, since harmonic emission presumably stops after ionization. Figure 3 shows calculated single-atom harmonic spectra which are less spectrally shifted for negatively chirped excitation pulses than for positively chirped pulses, in agreement with our experimental data. These results suggest that laser pulse chirp might be used to control the ionization rate of atoms, to allow the production of extremely broad bandwidth and attosecond duration x-ray pulses, with well-defined and possibly compressible chirp.

In summary, we have investigated harmonic generation using ultrashort laser pulses. In many of the noble gases, we observe unexpectedly high harmonic orders, up to 40% higher energies than have previously been pre-

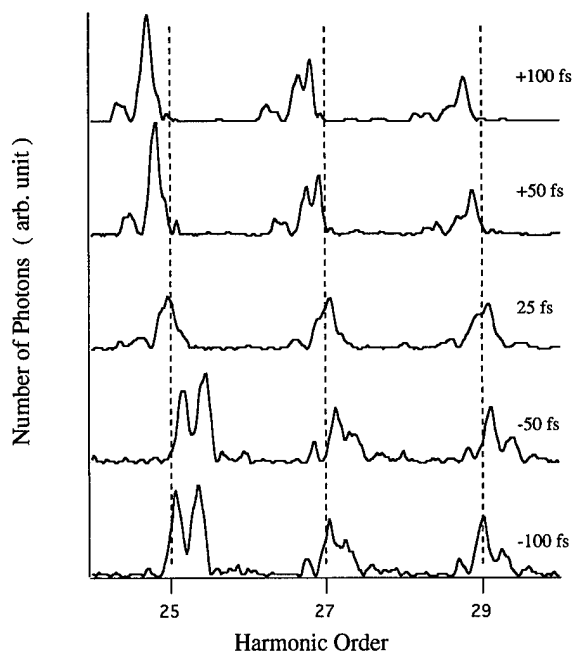


FIG. 3. Theoretical predictions for the harmonic spectra generated for various excitation laser chirp for fixed intensity and bandwidth.

dicted or observed. We also observe increased conversion efficiency of laser to soft-x-ray light by using shorter excitation pulses. Finally, we observe that the harmonic spectra are asymmetric with respect to the sign of the chirp and that the harmonics may be tuned by adjusting the chirp. Our results suggest that the BSI model is not applicable to very short excitation pulses and that the laser pulse chirp might be used to control the ionization rate of atoms. Our results have demonstrated a tunable, sub-20 fs, soft-x-ray source.

The authors wish to thank Chung-Po Huang and Chengyu Shi for invaluable help. This project was supported by the Air Force Office of Scientific Research and by the National Science Foundation. M. Murnane and H. Kapteyn acknowledge support from Sloan Foundation Fellowships.

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## Nonadiabatic Effects in High-Harmonic Generation with Ultrashort Pulses

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(Received 13 March 1996)

High-harmonic generation using ultrashort laser pulses with pulse durations 25 to 200 fs is studied theoretically and experimentally. We observe that the harmonic spectrum of argon taken with 25 fs laser pulses contains harmonics up to 20 orders higher than for 100 fs laser pulses with the same intensity. We show that this increase in harmonics is because the atom survives to higher laser intensities, due in part to the nonadiabatic response of the atomic dipole to the fast rise time of our pulse. [S0031-9007(96)01018-6]

PACS numbers: 42.50.Hz, 32.80.Rm, 42.65.Ky, 42.65.Re

The shortest x-ray pulses which can be generated to date are obtained by high-harmonic conversion of intense laser pulses in gas jets. This scheme has been used to generate harmonics up to orders exceeding 100, using either picosecond [1] or femtosecond pulses [2]. High-harmonic generation thus represents an attractive technique for generating ultrafast coherent radiation in the UV and soft x-ray region of the spectrum.

For high intensity laser illumination, the ionization of atoms occurs via tunneling through the core potential [3–5]. Once free, the electron moves in the laser field, and when the laser field reverses, the electron can return to the core with a maximum kinetic energy of  $3.17U_p$ . Here,  $U_p = E^2/4\omega^2$  is the ponderomotive or quiver energy (atomic units) of a free electron in a electric field  $E$  with frequency  $\omega$ . Thus the energy of the highest harmonic emitted from an atom of ionization potential  $I_p$  is predicted to be  $I_p + 3.17U_p$ . With the exception of our experiments, this prediction is in good agreement with experimental data to date for the width of the plateau region and the cutoff wavelength of the harmonic spectra [1,2]. Here,  $U_p$  corresponds to the maximum field that an electron may experience before ionization, even though the laser field may subsequently increase. This simple law has also been confirmed by numerical [6] and analytic calculations [7,8].

In all the experiments performed by others, laser pulse widths  $>100$  fs have been used. In the visible region of the spectrum, this implies that the field amplitude changes little between successive optical cycles. Thus when the laser pulse reaches the intensity required for tunneling, there is sufficient time for the electron to escape from the atomic core before the field increases further. We recently reported [9] the generation of high harmonics from noble gases, which for the case of 25 fs pulses, exhibited unusually high orders in comparison with previous work using longer pulses. We also reported that the harmonics could be tuned in wavelength by adjusting the chirp of the excitation pulse.

In this paper, we report new results on the direct comparison of harmonic emission as the transform-limited

pulse width of the excitation laser is varied over the range 25–200 fs. This avoids any complications due to the introduction of chirp [9]. Our experiments show that the harmonic spectrum of argon taken with 25 fs laser pulses contains harmonics up to 20 orders higher than for 100 fs excitation pulses with the same intensity. We also report on quantum calculations which consider the interaction of a 1D atom with an intense laser pulse. We show that the ionization process can be strongly affected by the ultrashort rising edge of the pulse, and that, in the presence of finite ionization rates, the atom can survive to higher laser intensities prior to ionization. The electron is then exposed to a stronger, rapidly increasing, laser field, which allows the electron to gain even more energy prior to reencountering the parent ion. Quantum mechanical calculations show a further effect for very short pulses. For rapid rise-time pulses, the phase of the atomic dipole lags that of the field, which can result in reduced ionization due to the nonsinusoidal shape of the electric field in time.

A simple intuitive picture of tunneling, described quantum mechanically by the Keldysh theory [3], can be seen as follows. In the case of a field with an amplitude  $E$ , comparable to the amplitude which suppresses the Coulomb barrier to the level of  $I_p$  (the appearance amplitude  $E_{app} = I_p^2/4$ ) [10], the barrier width is given by  $L = 2(E_{app} - E)^{0.5}/E$ . Classically, we assume the bound energy of the electron to be kinetic with velocity  $v = (2I_p)^{0.5}$ , to obtain a tunneling time

$$T_{tun} = \sqrt{E_{app} - E} / (EE_{app}^{0.25}). \quad (1)$$

On the other hand, quantum mechanically for weak fields ( $E \ll E_{app}$ ), the ratio of the tunneling time to the laser period is given by the Keldysh parameter  $\gamma = 2E_{app}^{0.25}\omega/E$ . Thus a simple classical model retains the qualitative features of the more exact calculation. In the case of argon, for example ( $I_p = 0.58$  a.u.), at a field of  $0.5E_{app}$ , we find a tunneling time of  $\approx 3$  fs. (The period of our laser light is  $\approx 2.7$  fs, while the pulse width is 25 fs.) We will show that when the tunneling time is close to an optical cycle ( $\gamma \approx 1$ ) and for rapid rise-time pulses, the electron wave

function can experience a strongly delayed response. This effect is in addition to the theoretically predicted increase of harmonic orders for short pump pulses recognized by others in the past [5,11]. However, none of the previous work considers the dynamics of wave function within a few periods of the optical field, in the transient regime where the atom responds to the nonsinusoidal individual half period of the increasing electric field, and when the Keldysh parameter is close to unity. Moreover, although many recent calculations on high-harmonic generation and stabilization of atoms used simulated pulses with a rise time of a few optical periods, followed by a region with constant amplitude to avoid the "death valley" problem of atomic ionization [12,13], only recently have lasers advanced to the point that such high-power ultrashort pulses can be generated in the laboratory [14,15].

In order to model the interaction of a single atom with an ultrashort pulse, we solve the Schrödinger equation for a 1D Coulomb potential [16]

$$i \frac{\partial \Psi(x,t)}{\partial t} = \left[ -\frac{1}{2} \frac{\partial^2}{\partial x^2} - \frac{1}{|x| + \alpha} - xE(t) \right] \Psi(x,t). \quad (2)$$

For small  $\alpha \approx 0$ , the eigenvalues and eigenfunctions of Eq. (2) approach those of the 1D hydrogen atom. (For no external field, the 1D and 3D hydrogen atom energy levels are the same, with zero probability of finding the electron at the core.) The lowest-energy state has energy of  $-0.5$  a.u. (ignoring a single infinitely bound state peaked at the core, but which does not couple to other states through dipole transitions) [16]. In contrast, when the smoothing parameter  $\alpha$  is large, this state is no longer infinitely bound, and it can interact with other states through dipole transitions. For example, the soft potential  $-1/(2+x^2)^{0.5}$  supports a ground state of  $I_p = 0.5$  a.u. which is peaked at the core [17,18]. We use a small value of  $\alpha = 0.037$  a.u., which allows the core-peaked wave function to be ignored because it is deeply bound. This modest degree of smoothing gives for the next lowest state (the *de facto* ground state) a binding energy of  $I_p = 0.443$  a.u., and the corresponding appearance field is  $0.049$  a.u. ( $I_{app} = 8.4 \times 10^{13}$  W/cm<sup>2</sup>). We calculate the harmonic spectra by using the dipole acceleration approach [18]. In order to determine the time dependence of a specific harmonic, we Fourier transform the corresponding spectral peak. The ionization probability was calculated by projecting the wave function over the first 50 bound states of the atom, since the contribution of the remaining states was found to be negligible.

Our calculations show that for weak pulses ( $E \ll E_{app}$ ) the harmonic spectra do not depend on the pulse width, since the highest intensity the atom can experience is always the peak intensity of the pulse. On the other hand, for strong pulses ( $E \approx E_{app}$ ) the ionization can depend on the pulse width. Figures 1(a), 1(b), and 1(c) show the time de-

pendence of the ionization probability, the third-harmonic field, and the phase of the atomic dipole, respectively, for 100 fs excitation. These figures show that the peak of the third-harmonic field corresponds to an ionization probability of 42%, occurring at a field amplitude of  $0.74E_{app}$ , and that the dipole phase is nearly constant. Figures 1(d), 1(e), and 1(f) correspond to excitation by a 25 fs duration pulse, of the same peak intensity. In this case, at the peak of the third harmonic the ionization probability is 32% while the amplitude of the field is  $0.86E_{app}$ . Therefore the atom is exposed to higher fields for shorter duration excitation pulses ( $<100$  fs). Because of the higher field, the kinetic energy of the electron significantly increases, and this results in the generation of more intense and higher-energy harmonic peaks.

It can also be seen from Fig. 1(f) that the phase of the dipole changes significantly for 25 fs illumination when compared with 100 fs illumination, particularly when the ionization exceeds 50%. Thus for weak fields or long pulses, the atomic dipole adiabatically follows the oscillations of the laser field, but, for ultrashort pulses of high intensity, the response of the wave function is delayed with respect to the laser field. The onset of this phase delay coincides with the stabilization of the

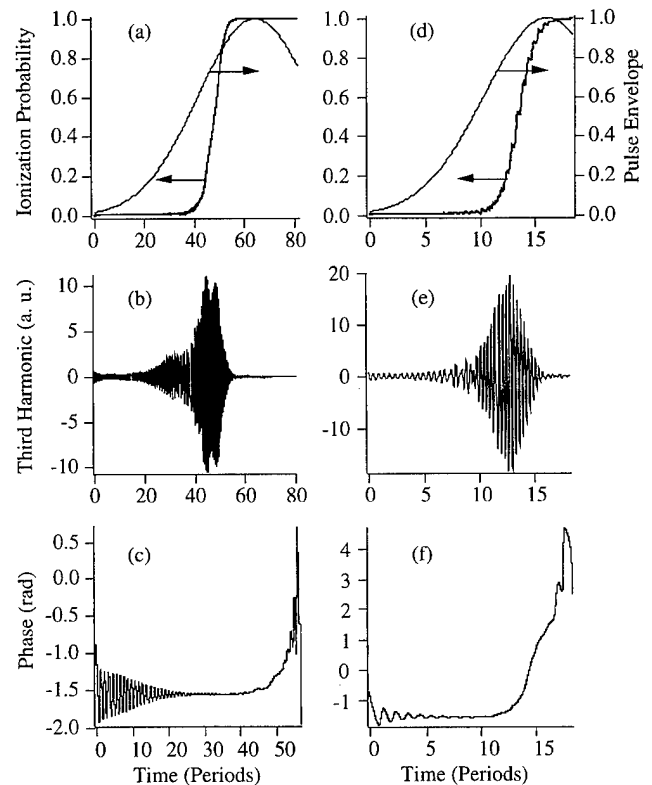


FIG. 1. Time dependence of (a) the pulse envelope and ionization probability for a 100 fs excitation pulse, with peak amplitude  $E = E_{app} = 4.9 \times 10^{-2}$  a.u.; (b) the generated third harmonic; and (c) the phase of the atomic dipole at the fundamental frequency; (d), (e), and (f) correspond to the case of a 25 fs pulse, with the same peak amplitude.

atom against fast ionization on the leading edge of the pulse. Our results can also be considered in the context of tunneling theory. For cw radiation, the tunneling ionization ( $\gamma \ll 1$ ) can be described by a "rate" which is dominated by the exponential factor [19,20]

$$W \propto \exp[-2(2I_p)^{3/2}/3E]. \quad (3)$$

For long pulses, when the laser intensity varies slowly during an optical period, one may assume from above that the atomic dipole adiabatically follows the field oscillations, with no dependence on the pulse history. In that approximation, the ionization rate depends on the instantaneous value of the field envelope  $E(t)$ , and we may substitute  $E(t)$  for  $E$  in Eq. (3). After integration of (3) over time, we find that the ionization probability is proportional to the pulse duration, which for weak fields is consistent with Fermi's golden rule. Thus the ionization probability is a function of the pulse energy (photon number), and thus for shorter pulses the ionization saturates at higher intensities than for longer pulses, as predicted previously [11].

In contrast, in the case of an ultrashort pulse ( $<25$  fs), the ionization and harmonic generation occur within a few periods on the leading edge of the pulse. Therefore it is not precise to introduce an ionization "rate," since the ionization probability is nonlinear, and may even decrease with time (see Fig. 1 and [21]). Thus the application of a quasiclassical rate like Eq. (3) to a rapid rise-time pulse is limited. Equation (3) does not consider the shape of the incident field, including the carrier frequency, which means that it is rigorously valid only for low frequencies and long pulses [3]. Moreover, Eq. (3) gives the ionization rate averaged over a half period of the carrier wave, which cannot describe the nonadiabatic effects due to the return of the electron wave packet to the core, when the particular cycle where the harmonic generation is occurring may have a nonzero integrated electric field (rapidly rising leading edge). In order to observe the pure nonadiabatic response, we calculate the unsaturated ionization of an atom irradiated by an exponentially increasing pulse given by  $E(t) = E_0 e^{t/T} \sin(\omega t)$ , where  $\omega$  corresponds to 800 nm, and  $E_0$  is chosen to ensure equal incident energy for different values of the rise time  $T$ , for each fixed number of periods in the pulse. Here equal energy means that the time integral of the square of the field has the same value in each case. First, we show in Fig. 2(a), that for a half period of the carrier wave, the ionization is almost independent of the rise time  $T$ . Then, Fig. 2(b) shows the case of an incident field consisting of one full optical period. The ionization in Figs. 2(a) and 2(b) differ since the pump field which ensures equal energy differs in time in all cases. The effect of ionization suppression due to the nonadiabatic response of the dipole can be seen by comparing the ionization of the 5 fs rise-time pulse with that of the 50 fs case, even though we have irradiated the

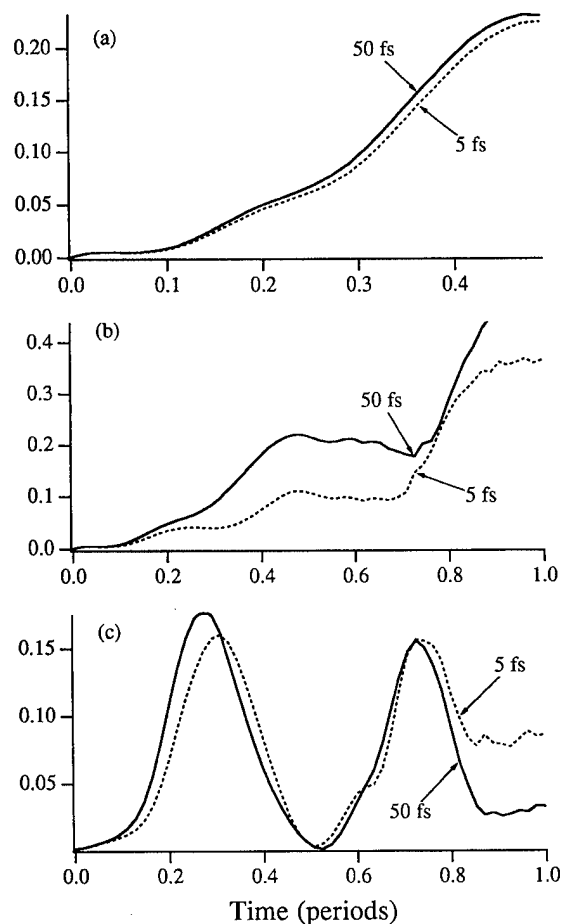


FIG. 2. Ionization probability of the model atom irradiated by an exponentially increasing field which ensures the same energy for a given number of cycles but different rise times  $T$ . (a) One half period:  $T = 50$  fs (solid),  $T = 5$  fs (dashed); (b) one full period:  $T = 50$  fs (solid),  $T = 5$  fs (dashed); (c) population of the first excited state for 50 fs (solid) and for 5 fs (dashed) rise times.

atom with the same "number of photons" in the same period of time. This difference disappears when 15 fs pulses are compared with 50 fs pulses. It is important to point out that this effect of ionization suppression appears when there is a significant ionization ( $>10\%$ ) during the first half period of the field, which ensures that there is sufficient penetration of the electron wave packet into the Coulomb barrier. Since the atom responds to each individual half period of the increasing electric field, the different depth of penetration of the electron wave packet into the potential barrier due to the large difference in amplitudes of the subsequent half periods leads to an increasing delay of dipole response with shorter pulses ( $<25$  fs), and to a decrease of the total ioniation.

In frequency space, the short rise time and broad spectrum of the pulse leads to population of intermediate states. This defies the adiabatic assumption that only the initial and final states of the electron are significant in the tunneling process, while the intermediate states play

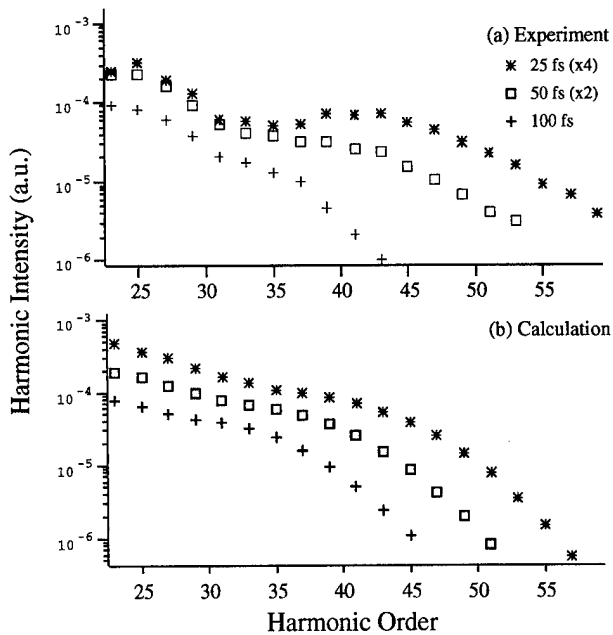


FIG. 3. (a) Experimental and (b) theoretical results for high-harmonic generation in argon by 25 fs (\*), 50 fs ( $\square$ ), and 100 fs (+) excitation pulses, with a peak intensity of  $4 \times 10^{14}$  W/cm $^2$ .

no role [3,19]. We found that the population of the first excited state for rise times of 5 fs is  $\approx 9\%$ , while for rise times of 50 fs it is less than 3.5% [see Fig. 2(c)]. Thus for very short rise times, a portion of the population is captured in intermediate states, which again leads to suppression of the ionization of the atom.

Figure 3(a) shows the experimental results for high-harmonic generation in argon by pulses of durations 25, 50, and 100 fs, and with a fixed peak laser intensity of  $4 \times 10^{14}$  W/cm $^2$ . The setup is described elsewhere [9,14]. The pulse duration was varied by varying the bandwidth of the laser to obtain near transform-limited pulses with varying duration. The results of the corresponding simulations are shown in Fig. 3(b). This simulation was done using the soft Coulomb potential  $-1/(1.40 + x^2)^{0.5}$ , which has a core-peaked ground state with a binding energy the same as that for argon, 0.58 a.u. Both experiment and theory clearly show an increase in number of harmonics with decreasing pulse duration. A comparison of these results, and those of others [1,2] made with longer pulses and much higher peak intensity, shows that the higher saturation intensities achieved by using shorter excitation pulses results in higher ponderomotive energy and more intense higher harmonics than with longer pulses. Thus the nonadiabatic behavior of the atom when illuminated by very short, intense light pulses offers the possibility for greatly enhanced generation of short wavelength radiation.

In conclusion, we have shown that we can generate significantly more high-harmonic orders using very short excitation pulses, with rise times under 100 fs. This

enhanced emission is a result of the rapid rise time of the pulse, which in the presence of the rapidly increasing field and the nonadiabatic atomic dipole response, allows the atom to survive to higher laser intensities. Thus our results indicate that the laser pulse shape itself can be used to control the ionization of atoms. It is likely that, with pulses of duration  $\approx 10$  fs, significant soft-x-ray flux in the "water window" ( $>270$  eV) may be generated.

This project was supported by the National Science Foundation and the Air Force Office of Scientific Research. I.P.C. acknowledges support from the Science Foundation of Bulgaria. H.C.K. acknowledges support from a Sloan Foundation Fellowship.

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# Amplification of 26-fs, 2-TW pulses near the gain-narrowing limit in Ti:sapphire

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Received September 8, 1994

We report the generation of 26-fs-duration pulses, with an energy of 60 mJ, from a simple multipass Ti:sapphire amplifier system. The peak power of our amplified pulses is 2 TW, and the repetition rate is 10 Hz. Our amplifier design consists of two highly doped multipass amplifiers and is simple and compact. We use an all-reflective, low-groove-density grating stretcher and compressor, combined with a relatively short material path length in the amplifier. This design allows us to minimize higher-order dispersion. The result is a laser system that generates multiterawatt transform-limited pulses, with good beam quality and low amplified-spontaneous-emission levels, at a duration near the theoretical limit imposed by gain narrowing in Ti:sapphire.

The extremely broad gain bandwidth, high saturation energy, and high damage threshold of Ti:sapphire<sup>1</sup> has made this material one of the most attractive for the generation of terawatt-power optical pulses. Recent advances in Ti:sapphire laser oscillator designs<sup>2,3</sup> have made it possible routinely to generate low-energy nanojoule pulses of  $\approx 10$  fs. With chirped-pulse amplification schemes,<sup>4</sup> these low-energy pulses can be amplified to higher energies.<sup>5-9</sup> In this Letter we show that, by using a very short seed pulse, an all-reflective stretcher and compressor, and a multipass amplifier geometry, we can minimize spectral and gain narrowing in a high-energy amplifier. The result is that we can amplify ultrashort pulses to multiterawatt levels with low prepulse and amplified spontaneous emission (ASE) levels, at durations near the gain-narrowing limit in Ti:sapphire. Our results contrast with previous ones<sup>10</sup> in which efforts to obtain terawatt peak power and 30–40-fs pulse duration by use of a regenerative amplifier system resulted in distorted spectral and pulse shapes at the shortest pulse durations. Our system design is also relatively straightforward and compact. The entire setup occupies a bit more than 4 m<sup>2</sup> of optical table space on a single optical table, making it a true tabletop terawatt system.

Our amplifier system is based on one that we reported previously<sup>5</sup> but have extended to higher energy by the addition of a second stage of amplification and with some simplifications and improvements. A diagram of the system is shown in Fig. 1. The input to the amplifier is from a self-mode-locked Ti:sapphire laser,<sup>2</sup> which produces  $\approx 10$ -fs pulses, with an energy of 5 nJ, a center wavelength of  $800 \text{ nm}$ , and a bandwidth of  $66 \pm 4 \text{ nm}$ . The ultrashort pulses propagate through a single-grating pulse stretcher, which stretches the pulses to 40–50 ps. The design of our stretcher is similar to our previous design<sup>5</sup> and uses a 15.25-cm-diameter parabolic focusing mirror (76.2-cm focal length; Edmund Scientific) to avoid spherical aberration. However, we replace the 300-groove/mm gratings in the stretcher and

compressor by 600-groove/mm gratings (40 mm  $\times$  100 mm rectangular; Milton Roy Company).

As the per-pass diffraction efficiency of these gratings is  $>90\%$ , compared with  $\sim 74\%$  for the old gratings, the energy output of the compressor was increased by  $\approx 200\%$ . The 600-groove/mm gratings also allow us to compensate for higher-order dispersion by adjusting the grating angle. With the old gratings, third-order dispersion from the grating was too small to provide compensation, and it was necessary to use an additional prism pair to adjust high-order dispersion. In the stretcher, the angle of incidence of the beam is  $13.9^\circ$ , and the effective separation is 42 cm. In the compressor, the incidence angle is  $21^\circ$  and the separation is 44 cm. We also found that the new gratings provided a more-focusable output beam, presumably because of a flatness problem with the old gratings.

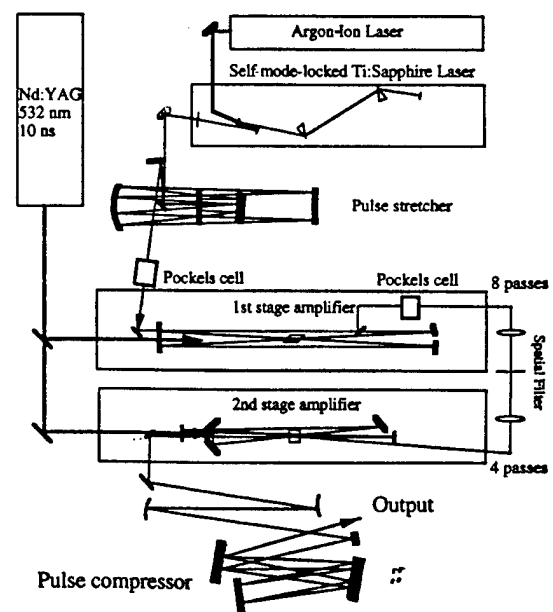


Fig. 1. Diagram of the laser system that generates 2-TW, 26-fs pulses.

Single pulses are selected at 10 Hz from the mode-locked pulse train by a KD\*P Pockels cell and crossed calcite polarizers, placed after the pulse stretcher to avoid damage to the stretcher grating owing to ASE. The preamplifier uses an 8-mm long, highly doped (0.23%; Union Carbide), Brewster-cut Ti:sapphire rod, at the focus of an eight-pass figure-eight amplifier. It is pumped longitudinally with  $90 \pm 5$  mJ of 532-nm light from a doubled Q-switched Nd:YAG laser (Continuum YG-681C). A second Pockels cell is placed after the preamplifier to reject ASE from the first amplifier, which is emitted primarily in a  $\sim 100$ -ns pulse peaked approximately 60 ns behind the amplified pulse. We achieve further reduction of ASE by passing the beam through a spatial filter before it enters the power amplifier. This spatial filter consists of two singlet lenses (40 and 50 cm) used at  $\sim f/200$  and a 200- $\mu\text{m}$  pinhole. It also serves to expand the beam for the second amplifier stage. At this high  $f$ -number, wave-front distortion effects from the lens are negligible. This refractive setup avoids astigmatism and provides for convenient adjustment of the collimation of the beam.

In our previous design we had used a saturable absorber filter (RG-850) to suppress ASE. However, detailed investigation showed that the RG-850 filter attenuates the leading red edge of the chirped pulse and introduces undesired modulation in the amplified pulse spectrum. Therefore we eliminated this filter in our present work. The amplified spectrum from the first stage is 44 nm, with an energy of 2–2.5 mJ. The ASE level is  $\sim 5\%$  before the Pockels cell and  $\sim 0.5\%$  after it. Our multipass preamplifier design has the advantage of very low ASE and prepulse levels and increased spectral throughput compared with that of regenerative amplifiers. It is therefore more suited to the amplification of very short pulses.

The power amplifier also uses a multipass configuration, but in this case flat mirrors are used and the beam is not focused through the crystal. In this amplifier and throughout the system, a combination of protected-silver and dielectric (CVI Laser Corp. TLM-1) mirrors is used, depending on the fluence on the mirror. The amplifier crystal is a 7-mm-long 10-mm-diameter highly doped (0.23%; Union Carbide) normal-incidence Ti:sapphire rod, which is antireflection coated on both sides. The crystal is pumped by 500 mJ of 532-nm light, which is relay imaged to a diameter of 5 mm onto the crystal. A high-efficiency air filter excludes dust from the entire two-stage amplifier system. Our pump fluence is approximately  $2.5 \text{ J cm}^{-2}$ . The spatial profile of the pump is approximately flat-topped. The crystal absorbs 84% of the YAG energy on the first pass, and the residual energy is backreflected into the crystal by a curved mirror. To obtain a more uniform amplified beam, we set up the mirrors so that the beam is flipped once horizontally and once vertically. To reduce nonlinear distortions in the amplifier, the beam leaves the pumped face of the crystal in the final pass of the amplifier. Given a material path length of only  $\sim 3$  mm at the highest power level, we estimate a  $B$  integral of  $<0.1$  for the last pass and  $<0.5$  for the entire system. We have never seen any evidence of

nonlinear self-focusing in the amplifier system, even with  $\sim 15$ -ps 100-mJ pulses. Following the second stage of amplification, the amplified beam has an energy of  $\sim 120$  mJ and a bandwidth of 39 nm, as shown in Fig. 2. This bandwidth is near the calculated gain-narrowing limit of 44 nm, which is the bandwidth obtained by simulation of the amplification of an infinitely broad spectrum by our estimated gain of  $\sim 5 \times 10^8$ , assuming the published gain curve for Ti:sapphire.<sup>11</sup>

After four passes through the amplifier, the beam then propagates through a beam expander and a collimator to the compressor. The beam diameter is approximately round, with an  $\sim 1.1$ -cm diameter. The throughput of the compressor is  $\sim 50\%$ , yielding a typical pulse energy of  $60 \pm 2$  mJ, with a pulse-to-pulse fluctuation of 5 mJ. We observe an  $\sim 15\%$  spectral narrowing through the compressor (mainly at shorter wavelengths), resulting in a reshaped spectral FWHM of 32 nm, as shown in Fig. 2. In the compressor, all beams are in the same plane as the dispersion, making it necessary to use the gratings off-Littrow. It seems that the observed spectral narrowing is due to efficiency modulation introduced when the gratings are off-Littrow. However, deviation of the beam out of the plane of dispersion results in a spectrally dispersed focus.<sup>12</sup> The compressor also introduces a periodic intensity modulation of  $\sim 10\%$  on the beam profile, which may be caused by a modulation of blaze angle on the 600-groove/mm gratings (it was not observed with the 300-groove/mm gratings). This modulation does not affect the focusability, throughput, or output pulse duration.

We used a single-shot autocorrelator to measure the duration of our amplified pulses. Figure 3 shows a typical single-shot autocorrelation trace of the attenuated compressed pulse, indicating a duration of  $25.5 \pm 2$  fs (assuming a  $\text{sech}^2$  pulse shape). The Fourier transform of our 32-nm spectrum corresponds to a 24-fs pulse, indicating that our pulses are nearly transform limited. The Fourier transform predicts a ratio of 1.51 between the autocorrelation width and the pulse width, close to the 1.55 for an ideal  $\text{sech}^2$  pulse.

The power amplifier is pumped by an approximately flat-topped YAG pump profile, and the profile of the amplified beam closely resembles that of the

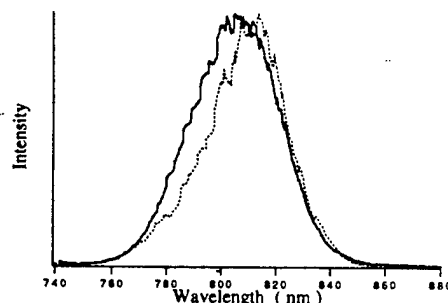


Fig. 2. Spectrum of the output pulses at full energy: solid curve, after the second amplifier, the spectrum is 39 nm FWHM with a pulse energy of 120 mJ; dotted curve, after passing through the compressor, the spectrum narrows to 32 nm, and the pulse energy is reduced to 60 mJ.

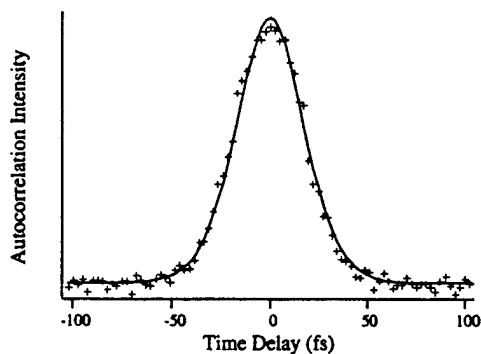


Fig. 3. Single-shot autocorrelation trace of the pulse output at full energy. Crosses, experimental data; solid curve, fit to the data, corresponding to a 26-fs pulse with a  $\text{sech}^2$  pulse shape.

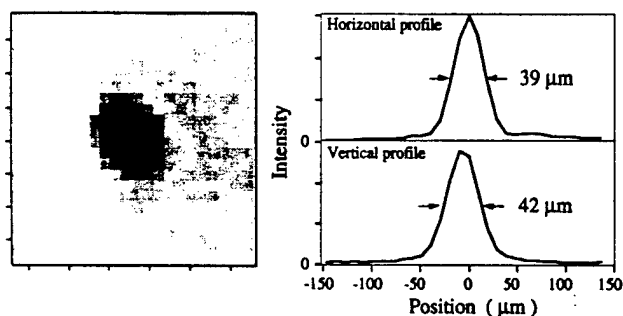


Fig. 4. Focal spot resulting from focusing a small fraction of the pulse power with a 1-m radius-of-curvature mirror. The spot size of  $40 \mu\text{m}$  FWHM is approximately diffraction limited, assuming an 11-mm-diameter flat-topped beam.

pump beam. We examined the focusability of this beam, using a 1-m radius-of-curvature mirror placed at near-normal incidence and a CCD camera. Figure 4 illustrates the results of the measurement.

An ideal flat-topped beam of this size should focus to  $\sim 37 \mu\text{m}$ , while a Gaussian beam would focus to approximately  $\sim 18 \mu\text{m}$ . We measured  $\sim 40 \mu\text{m}$ ; thus the beam focuses to 1–2 times the diffraction limit in each dimension. To verify this result independently, we also measured the transmission through a  $100\text{-}\mu\text{m}$  pinhole placed at the focus. We found that  $\geq 85\%$  of the energy was transmitted through the pinhole. Even with this relatively long focal-length optic, the peak intensity obtained is  $3 \times 10^{17} \text{ W cm}^{-2}$ . An intensity of  $10^{19} \text{ W cm}^{-2}$  would be readily achievable with  $\sim f/10$  optics.

The total ASE per pulse, measured by not triggering the first Pockels cell, is 0.3 mJ in a 7-ns pulse. This implies an ASE-to-pulse intensity contrast ratio of  $>10^7:1$ , which we also verified by using a fast photodiode. The contrast between the main pulse and the pulse out of the oscillator preceding the main pulse by 12 ns was directly measured to

be  $>10^4:1$ , and contrast measurements for individual components (two Pockels cells and the first amplifier) imply a ratio of  $>10^7:1$ . The low prepulse and ASE from this multipass amplifier contrast with those typically obtained directly from a short-pulse regenerative-amplifier system.

In conclusion, we have demonstrated the generation of 2-TW transform-limited  $26 \pm 2$ -fs-duration pulses, with an energy of 60 mJ and good beam quality, from a simple and compact two-stage multipass amplifier system. Higher-order dispersion and nonlinear effects are minimized by use of short amplifier crystals and reflective optics. The multipass design minimizes prepulse and ASE. We anticipate no *B*-integral or additional problems with scaling this system design to 1–10-J pulse energies by using future laser-pumped amplifiers. The development of such compact terawatt laser systems has significant implications for high-field physics and for coherent and incoherent x-ray and extreme-ultraviolet radiation generation.

This research was supported by the National Science Foundation, the U.S. Department of Energy, the U.S. Air Force Office of Scientific Research, and the Washington Technology Centers. The authors acknowledge generous support and assistance from George Venikouas and Milan Kokta of Union Carbide Corporation. The authors also thank Chengyu Shi for help with the autocorrelator.

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# Demonstration of a sub-picosecond x-ray streak camera

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(Received 11 March 1996; accepted for publication 6 May 1996)

A novel design, magnetically focused, x-ray streak camera was designed and tested using sub-20 fs soft-x-ray pulses generated by high harmonic emission in a gas. The temporal resolution of the camera was demonstrated to be under 0.9 ps throughout the ultraviolet to soft-x-ray wavelength region. Our streak camera represents the fastest x-ray detector developed to date. © 1996 American Institute of Physics. [S0003-6951(96)03327-X]

During the past decade, the development of ultrafast x-ray sources based on laser-produced plasmas, high harmonic emission, and synchrotrons, has advanced rapidly.<sup>1-3</sup> It is now possible to generate sub-picosecond pulses throughout the vacuum ultraviolet and x-ray region of the spectrum, and sub-10 fs pulses have been generated using high harmonic emission.<sup>4</sup> However, progress in the development of ultrafast sub-picosecond x-ray detectors has been relatively slower.<sup>5,6</sup> Although cross-correlation techniques have demonstrated femtosecond time resolution, such measurements are very difficult, and are possible only at discrete wavelengths.<sup>7</sup> The fastest x-ray streak camera measurement to date was demonstrated to be 2 ps. This measurement was limited both by the time response of the streak camera itself, and by the laser-plasma-based x-ray source.<sup>5,6</sup> In this letter we describe the design and implementation of a novel x-ray streak camera, which exhibits sub-picosecond time resolution. The time response of the streak camera was measured using ultrashort sub-20 fs high-order harmonics produced by a 25 fs laser.<sup>4,8</sup> The resolution of our camera was experimentally demonstrated to be 0.88 ps.

It is well known<sup>9,10</sup> that the temporal resolution of streak cameras is limited mainly by the transit time dispersion of the photoelectrons as they travel from the photocathode to the deflection plates. It is also limited by the spatial resolution, and the deflection speed of the streak plates. For sub-picosecond time resolution, space-charge effects may also limit the time resolution, and thus limit the dynamic range. For our work, we designed and tested a novel streak camera design to reduce the limitations on temporal resolution as much as possible. The configuration of the new x-ray streak camera is shown schematically in Fig. 1. In our camera, a pair of meander-type deflection plates is located before a magnetic focusing lens. This has several advantages: first the electron transit time from the anode to the deflection plates is minimized, as is the transit-time dispersion; second, the fast time response and high deflection sensitivity (8 cm/kV) of the meander-type deflection plates provides the possibility of high sweep speeds on the exit phosphor screen; finally, the

short transit time also reduces space-charge effects.

The electron transit time dispersion from the photocathode to the deflection plates in the camera can be evaluated analytically. In the photocathode to anode region, it is straightforward to show that the transit time difference between an electron liberated with energy  $eV_0$ , and an electron liberated with zero energy is given by<sup>10</sup>

$$t'_{pa} = \sqrt{\frac{2mV_0}{eE}} \quad (1)$$

where  $m$  and  $e$  are the charge and mass of the electron, respectively,  $eV_0$  is the initial energy of an emitted photoelectron in the axial direction, and  $E$  is the extraction field.

For x-ray photocathodes in the 100 eV to 10 KeV region, the distribution of the initial energies of the emitted photoelectrons can be expressed as<sup>11</sup>

$$N(eV_0) \propto \frac{eV_0}{(eV_0 + W)^4}, \quad (2)$$

where  $W$  is related to the photocathode material ( $W=1$  eV for KBr). The photocathode material also determines the full width at half-maximum (FWHM) of the energy distribution ( $\delta\epsilon$ ). From Eqs. (1) and (2), we obtain the transit time distribution of the electrons. The FWHM of this distribution is defined as the time dispersion, and can be shown to be

$$\delta t_{pa} = \frac{2.63\sqrt{\delta\epsilon}}{E} \text{ (ps)}, \quad (3)$$

where  $\delta\epsilon$  is in eV, and  $E$  is in kV/mm. For our camera,  $\delta\epsilon=1.1$  eV (KBr photocathode), and  $E=10$  kV/mm, resulting in a calculated time dispersion of about 276 fs.

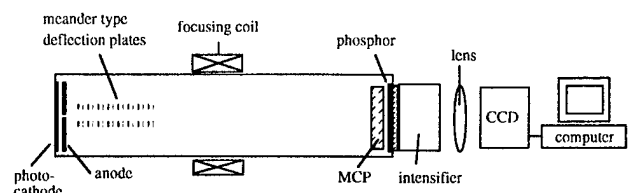


FIG. 1. Configuration of the sub-picosecond x-ray streak camera.

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be improved further by redesigning the deflection plates to achieve higher sweep speeds. Our work demonstrates that sub-picosecond time resolution experiments can now be performed using currently available synchrotron or laser-plasma sources.

This work was supported by the National Science Foundation, by the Air Force Office of Scientific Research, and by the Chinese Science and Technology Committee. The authors gratefully acknowledge the assistance of Hou Xun. Zenghu Chang acknowledges fellowship support from the Chinese Academy of Sciences. Henry Kapteyn acknowledges support from a Sloan Foundation Fellowship.

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## Ultrashort Light Pulses: Pushing the Limits

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### Introduction

During the past five years, there has been a revolution in the field of ultrafast laser technology. Femtosecond lasers today are simple and turn-key, with output powers orders of magnitude higher than were available only a decade ago. Rapid progress has also been made on the available pulse widths from these systems, and pulses as short as three optical cycles ( $\approx 8$  fs) can now be generated directly from a very simple laser oscillator. Nonlinear frequency conversion techniques can be used to extend femtosecond pulse generation throughout the visible and infrared, and new measurement techniques have been devised, which can extract the complete waveform of an optical pulse only a few cycles in duration. Thus, for ultrashort pulses in the visible and near infrared regions of the spectrum, we now understand that the pulse durations we obtain are close to the fundamental limits of operation for these systems.

Extending femtosecond pulse technology to shorter wavelengths has unique advantages. First, there is a wealth of new ultrafast science which can be explored. Ultrafast soft- and hard-x-ray pulses can be used to directly probe both long- and short-range atomic order and atomic motion, and to monitor the evolution of highly-excited systems. A second advantage of working in the vacuum ultraviolet or soft-x-ray region is that the generation of shorter, attosecond, pulses will be feasible only at such short wavelengths. This is because the required fractional bandwidth to support an ultrashort pulse is smaller in the x-ray region than in the visible region. For example, to support a 1 femtosecond pulse with a center wavelength of  $8000 \text{ \AA}$ , a FWHM bandwidth of  $\Delta\lambda \approx 6700 \text{ \AA}$  is required. In contrast,

to support the same 1 femtosecond pulse at a wavelength of  $100 \text{ \AA}$ , the required FWHM bandwidth drops to only  $1 \text{ \AA}$ . The generation of sub-100 fs x-ray pulses can be achieved using high field effects, where a super-intense, ultrashort optical pulse interacts highly nonlinearly with a medium.

### Fundamental Limits in the Visible Region

In the visible region of the spectrum, the shortest pulses produced directly from a laser were generated using a very simple titanium-doped-sapphire laser. Ti:sapphire is one of the broadest bandwidth media known. Its gain profile is sufficiently broad to support a  $\approx 3$  fs pulse, assuming that the laser components can be designed to support such a short pulse, and also assuming that no fundamental limits exist for modelocking such a broad bandwidth. In the past few years, much effort has been devoted to understanding and optimizing the performance<sup>1</sup> of femtosecond lasers, and in particular, the titanium-doped-sapphire laser.<sup>2</sup> The original Ti:sapphire lasers used a 2 cm long laser crystal, and a pair of high-dispersion SF10 prisms to compensate for linear dispersion in the laser. With this design, the shortest pulse which could be generated was  $\approx 50$  fs. We now understand that this upper limit is due to imperfect dispersion compensation in the laser.

Dispersion is extremely important in femtosecond lasers. It stabilizes the pulse in the laser, and also determines the limiting pulsewidth for a given geometry. For long-pulse (narrow-bandwidth) operation, these lasers work in a negative-dispersion regime. The lasing bandwidth and pulsewidth can be controlled by adjusting the amount of negative dispersion, by changing the amount of prism glass in the laser. The shortest pulses are obtained near the zero-dispersion point in the laser — a small amount of net-negative dispersion is needed to compensate for self-phase modulation in the laser medium. Our work at WSU has shown that the shortest pulsewidth for a given laser set-up is determined by the presence of higher-order dispersion in the laser, which will prevent frequen-

cies far from the line center from being modelocked.<sup>3</sup>

For ultrashort pulse generation, the round trip time or group delay for light in the laser cavity must be nearly frequency independent i.e.  $T(\omega) = \delta\phi/\delta\omega = T_0 = \text{constant}$ , where  $\phi$  is the total phase advance of light after one cavity round-trip.  $T(\omega)$  can be expressed as a Taylor series about the center frequency  $\omega_0$  -

$$\begin{aligned} T(\omega) &= \frac{\delta\phi}{\delta\omega} \\ &= \phi'(\omega_0) + \phi''(\omega_0) \Delta\omega \\ &\quad + \frac{1}{2} \phi'''(\omega_0) \Delta\omega^2 \\ &\quad + \frac{1}{6} \phi''''(\omega_0) \Delta\omega^3 + \dots \end{aligned}$$

where  $\phi'$ ,  $\phi''$ , etc. are the derivatives of the phase with respect to frequency. In a laser cavity, the second-order (or group velocity) dispersion term ( $\phi''$ ) leads to a linear chirp, which usually must be negative in the laser for femtosecond-pulse stabilization. This GVD term can be adjusted by adjusting the amount of prism glass in the laser. The next term, proportional to  $\phi'''$  is the third-order dispersion term, while the next term again is the fourth-order dispersion term.

We have shown in past work<sup>4-6</sup> that the presence of non-zero third-order dispersion (TOD) limits the lasing bandwidth in Ti:sapphire, and prevents the generation of sub-50 fs pulses. In a series of experiments, we gradually reduced the TOD in a Ti:sapphire laser, by reducing both the material in the cavity, and choosing the prism glass type to minimize TOD. As we reduced the crystal length from the standard 2 cm length, to 9 mm, then 4.5 mm, and finally 2 mm, while choosing optimum prism materials to minimize TOD, the minimum pulsewidth we obtained from the laser decreased from 32 fs, to 17 fs, to 11 fs, and finally  $\approx 10$  fs.

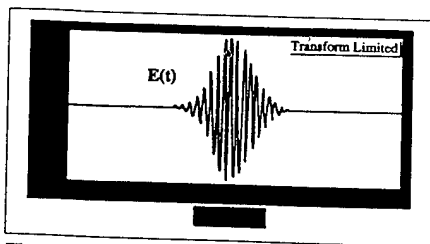
In order to generate even shorter pulses from solid-state lasers, the third-order dispersion error must be compensated. A very simple method to overcome this limit is to operate the Ti:sapphire laser at a center wavelength of 850 nm, where, using a

fused silica prism pair and a short 2 mm Ti:sapphire crystal, simultaneous near-zero second- and third-order dispersion can be obtained. In recent work, we used such a laser design to explore the fundamental limits of modelocking.<sup>7</sup> In addition to operating the laser near 850 nm to eliminate TOD, we also replaced the dielectric mirrors with metallic silver reflectors, to eliminate any effects due to the finite reflectivity bandwidths of dielectric mirrors. We found that the shortest pulse we could obtain from this laser was  $\approx 8$  fs, even though we could obtain very broad bandwidths of  $\geq 170$  nm FWHM. We now understand that having eliminated TOD in the laser, we are limited by the next error term, or fourth-order dispersion (FOD). The shortest pulses of 8 fs are obtained when a "soliton-like" balance exists between self-phase modulation and net negative higher-order dispersion in the cavity.

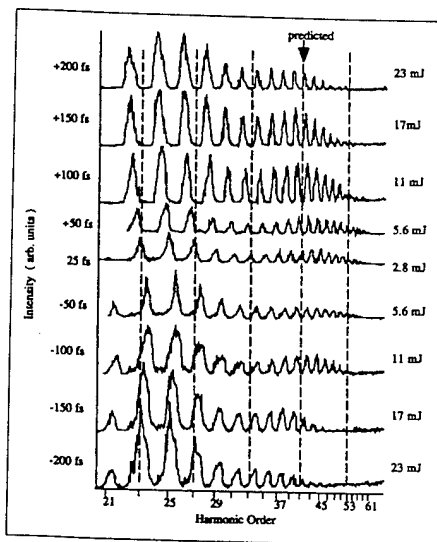
Modeling of solid-state femtosecond lasers has now advanced to such a point that we can now self-consistently model the laser to obtain theoretical bandwidths and pulsewidths in excellent agreement with experiments.<sup>7-9</sup> In work performed with Ivan Christov from Sofia University, we confirmed that the shortest pulsewidth from Ti:sapphire is determined primarily by the value of non-compensated FOD. Since this is a technological rather than a fundamental limit, we can expect to be able to generate shorter pulses when more advanced dispersion-compensation schemes are developed. In fact, given that our model can reproduce our experimental observations, we can now use it to predict the shortest-obtainable pulse duration. By introducing an ideal FOD value, we predict that stable, soliton-like, pulses with pulse durations under 5 fs are possible from Ti:sapphire.

#### Direct Observation of a Femtosecond Pulse Waveform

In recent work, we collaborated with Drs. Rick Trebino and Ken DeLong at Sandia National Laboratories to measure the complete pulse shape of a pulse of only 13 fs in duration.<sup>10</sup> For this work, we used the technique of second-harmonic-generation Frequency Resolved Optical Gating (FROG) that Sandia developed. A short pulse is split into two and recombined with a variable time-delay between the two pulses. These two pulses cross in a frequency-doubling crystal, and the second harmonic signal is detected. In the standard auto-



▼ Figure 1. Retrieved electric field waveform of a 13 fs pulse.



▼ Figure 2. Harmonic spectra generated in 5 torr of argon, for a fixed peak laser intensity, and for various excitation-pulse chirps. The predicted line shows the theoretically predicted and experimentally observed highest harmonic in argon to date, prior to our work.

correlation technique used to measure femtosecond pulses, the intensity of the SHG light is recorded as a function of delay. However, in SHG FROG, the spectrum of the frequency-doubled light is measured as a function of delay between the two beams. This gives sufficient information that a unique amplitude and phase for the pulse can be deconvolved from the data. The information obtained from the FROG technique is much superior to that obtained from traditional autocorrelation techniques, which do not yield a unique shape for the laser pulse. Fig. 1 shows the reconstructed data in the form of the electric field  $E(t)$  of a 13 fs pulse, the shortest waveform measured to date. This measurement is equivalent to an oscilloscope with a bandwidth  $> 100$  THz! Many applications of ultrashort pulses can use this technique to optimize the excitation pulse for experiments. In our case, we used FROG to verify our understanding of the fundamental limits of ultrashort-pulse solid-state lasers described above.

#### Femtosecond to Attosecond X-Ray Pulses

Thus far we have discussed fundamental limits of operation for laser oscillators, which can deliver pulses of several nJ energy, and peak powers on order of MW. By amplifying these low energy nJ pulses by factors of  $10^8$  in compact amplifiers, it is possible to generate pulses with peak powers of several TW. These amplifiers also based on Ti:sapphire, since it is an excellent amplifier material because of its high energy storage capacity, long upper level lifetime, and high thermal conductivity. Usually, the pulsewidth increases somewhat in the amplification process, and typically we generate 20 - 25 fs TW pulses. [Zhou, 1995 #175] These pulses can produce electric fields so intense that they instantaneously strip the electrons from atoms, or they can generate intensities in excess of that which would be produced if the entire solar flux incident on the earth were focused onto the head of a pin! Obviously, when such high-intensity pulses irradiate an atomic gas for example, the atoms are driven highly nonlinearly.

The shortest x-ray pulses which can be generated to date are obtained by high-harmonic conversion of these intense laser pulses in gas jets. The intense laser pulse ionizes the outer electron in an atom, by suppressing the Coulomb barrier binding the electron to the atom. Once free, the electron moves in the field of the laser, and when the laser field reverses, can emit high-harmonics if it collides and recombines with the parent ion. This scheme has been used to generate harmonics in excess of order 100,<sup>11,12</sup> thus up-shifting a femtosecond pulse from the visible into the soft-x-ray region.

For our most recent work,<sup>13,14</sup> we investigated high-harmonic generation using very short, intense, 25 fs excitation pulses. We found that we can generate 50% higher energy harmonics from gases using our ultrashort excitation pulses, than have previously been generated using longer, 100 fs, excitation pulses. In addition, we can generate these harmonics more efficiently, since the efficiency increases with decreasing excitation pulse duration, as shown in Fig. 2. We believe that the rapid risetime of our pump pulses allows the just-ionized electron to gain more energy from the laser field, which then leads to higher harmonic generation than with longer pulses. Also, we can generate harmonics more efficiently because we need less energy with a 25 fs pulse to reach the ionizing intensity for an atom. Most of the har-

monic radiation from a single atom arises from a single encounter of the returning electron with the atom. Therefore, the pulsewidth is expected to be considerably shorter than the exciting laser pulse, and possibly attosecond in duration. Since we have used the shortest excitation pulses to date, we believe that we have generated the shortest x-ray pulses to date, of at most 20 fs. We can also tune the wavelength of the harmonics by controlling the shape of the laser pulse, as shown in Fig. 2. The pulse shape is controlled by adjusting the dispersion compensation in the amplifier, to adjust the chirp and shape of the output. Therefore, our results demonstrate that we can generate tunable, sub-20 fs, and possibly sub-fs, x-ray pulses. Now all that remains is to develop the technology to characterize and use these x-ray pulses.

### Conclusion

Work to date has shown that by careful design of femtosecond solid-state lasers, sub-10 fs pulses can easily be generated directly from a laser. The fundamental limits of operation of these lasers are well understood, and sub-5 fs pulses will be generated directly from a laser when more sophisticated

fourth-order dispersion-compensation schemes are developed in the future. At present, we have reached a technological, rather than a fundamental limitation on the operation of these systems. The availability of intense ultrashort pulses allows femtosecond science technology to be extended into the x-ray region. Sub-20 fs x-ray pulses have already been generated, and these sources will soon be used to probe a broad range of dynamic processes in nature.

### Acknowledgments

We gratefully acknowledge support for this research from the National Science Foundation, the Air Force Office of Scientific Research, the Washington Technology Centers, and the Department of Energy.

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## VP's REPORT

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Waynant (FDA) were appointed to the Committee earlier this year. Tom and Ron bring to the position considerable experience in standards development for both optical devices and systems. Also, Tom has agreed to serve as the LEOS representative to the Optics and Electro-Optics Standards Council (OEOSC). Other individuals interested in joining the committee are welcome and LEOS members from industry are particularly encouraged to contact Breck Hitz, Dan Botez (Technical Council Chair), or myself for further information.

Several other appointments have been made that should be brought to your attention. Ken Pedrotti (Rockwell International) was appointed as one of LEOS' two representatives to the Solid State Circuits Council (SSCC). Ken's term on the Council will extend through 1998 and our other representative is R. L. Van Tuyl (Hewlett-Packard). Also, John Reintjes (Naval Re-

search Laboratory) has accepted an appointment as one of three LEOS representatives to the Joint Council on Quantum Electronics.

One of the priorities for the second half of 1995 is establishing an e-mail technical service for LEOS members. Our motivation is to provide a means by which any LEOS member can seek assistance with a technical problem or question — in short, to make the considerable expertise existing within the Society available to our membership. Several possible options for this service are currently being considered and your comments and suggestions would be most appreciated.

At the May meeting of the Board of Governors, Art Guenther described a study that is being undertaken by the recently formed National Research Council Committee on Optical Science and Engineering (COSE). Chaired by Charles Shank of the Lawrence Berkeley Laboratory, the Committee is charged with surveying the field of optical science and engineering, examining progress made in the last decade, and projecting "... the

future impact of OS&E on societal needs in the short (3-5 years) and long terms (5-20 years)." Since the Committee intends to "develop a vision for the future and identify some 'grand challenges' that could give the field direction and focus efforts in areas that have potential for benefit to society," your comments are welcome. If you are interested in more of the details regarding the study or wish to express your views, you may contact the Committee by e-mail (COSE@NAS.EDU), FAX (202-334-2791), or phone (202-334-3520).

I am constantly reminded of the concerted effort by a large number of individuals that is necessary to support the extensive programs of LEOS. Over the past three years, I have had the privilege of working with many outstanding individuals and wish to express my thanks to all of you and, in particular, to Dan Botez, Technical Council Chair, the Chairs and members of our technical subcommittees, the Board of Governors and the LEOS Executive Office for the sacrifice in time that each of you has made on behalf of the Society.