Generation of Coherent, Femtosecond, X-Ray Pulses in the "Water Window"

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***Abstract*— We report experimental and theoretical results on high-harmonic generation with 25-fs laser excitation pulses. The shortest wavelength we observe, at 2.7 nm, is well within the "water window" region of X-ray transmission. In the case of all the noble gases, we obtain excellent agreement between theoretical predictions for the highest harmonic photon energy generated and our experimental observations. We also obtain excellent agreement between theory and experiment for the highest photon energy generated as a function of laser pulsewidth between 25 and 100 fs. Finally, we observe that the individual harmonic peaks near the cutoff are well resolved for positively chirped pump pulses, but are unresolved in the case of negatively chirped excitation pulses.**

***Index Terms*— Attosecond pulse, high-order harmonics, water window X-ray.**

I. Introduction

COHERENT X-ray sources are attractive because of their potential applications in many fields of science and technology. A convenient and simple method used to generate X-ray radiation with good spatial and temporal coherence is high-harmonic conversion of focused laser beams in gases [1]–[3]. However, the shortest wavelength discrete harmonic observed until very recently was approximately 7 nm. These harmonics were generated using laser pulses of 100 fs and longer [2], [3]. Recent advances in laser technology however, have led to harmonic generation using high-power ultrashort pulses with pulse durations in the range of 5–25 fs [4]–[8]. Using ultrashort 25-fs Ti:sapphire pulses at 800 nm, for example, harmonics up to order 299 have been generated, which extends the useful range of coherent radiation down to 2.7 nm (450 eV) [5].

In this paper, we introduce a simple analytical formula for calculating the cutoff harmonic wavelength, as a function of pulsewidth and atomic species. The formula explicitly shows that the use of shorter laser pulses will lead to the generation of shorter-wavelength coherent harmonic radiation in the "water window" region of the soft X-ray spectrum (4.4–2.3 nm). We also report experimental results on the generation of harmonics up to order 297, at a wavelength of 2.7 nm, which corresponds to an energy of 454 eV. These extremely short wavelength harmonics were produced using an ultrashort-pulse 25-fs, Ti:sapphire laser centered at 800 nm [9], [10] We demonstrate experimentally that the wavelength of the individual harmonics in the 3–5-nm range can be tuned by chirping the laser pulses. The individual harmonic peaks near the cutoff are well resolved for positively chirped pumping pulses and become randomly distributed and unresolved for negatively chirped laser pulses. We believe that this behavior may be due to intensity-dependent contributions to the harmonic phase.

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II. Theory

In a typical high-harmonic generation experiment, a high-peak-power femtosecond laser pulse is focused into an atomic gas, and the highly nonlinear interaction of the laser light with the atoms results in the emission of coherent high-order harmonics of the laser in the forward direction, as a low divergence X-ray beam. Quantum mechanical models are needed to fully describe the X-ray emission process during HHG accurately [6], [7], [11]–[13]. However, to a first approximation, quantum models can be used to describe the ionization process, while semiclassical theories can be used to describe the motion of the just-ionized electron during the first optical cycle after the ionization due to the strong laser field [14]. In this picture, on the rising edge of a high intensity laser pulse, the ionization of atoms occurs via tunneling through the core potential. 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 $3.17U_p$. Some fraction of these electrons will undergo stimulated recombination with the core, and release their energy as high harmonics. Here, $U_p = E^2/4\omega^2$ is the ponderomotive or average quiver energy (atomic units) of a free electron in an electric field E of frequency ω. The maximum kinetic energy of $3.17U_p$ corresponds to an electron released at an optimum phase of the driving field—electrons released at any other phase within the optical cycle collide with the atom at lower energy, or do not collide at all. Therefore, the energy of the highest harmonic emitted from an atom of ionization potential I_p is predicted to be:

$$h\upsilon_c = I_p + 3.17U_p. \tag{1}$$

This cutoff rule has also been obtained by solving the Schrödinger equation of a single atom either numerically or analytically [12], [15], [16].

TABLE I
HIGH-HARMONIC GENERATION PARAMETERS FOR THE NOBLE GASSES, FOR USE WITH (2)

	I_p (eV)	n*	$C_{n^*l^*}$	G_{lm}
He	24.59	0.74	2.06	1
Ne	21.56	0.79	2.06	3
Ar	15.76	0.93	2.03	3
Kr	13.99	0.99	2.01	3
Xe	12.13	1.06	1.97	3

In order to show the explicit dependence of the cutoff photon energy (hu_c) on the atomic and laser parameters, we derived a simple formula based on (1) to predict hv_c [17], [18]

$$hv_c = I_p + \frac{0.5 I_p^{(3+a)} \lambda^2}{(\ln(0.86\tau 3^{2n^*-1} G_{lm} C_{n^*l^*} I_p)/(-\ln(1-p)))^2} \tag{2}$$

where hv_c and I_p are in eV, $a = 0.5$ (to correct an approximation in the derivation of the analytical expression), l is the laser wavelength in μm, and τ is the FWHM of the pulse in fs. Here, p is the ionization probability for defining the saturation intensity (which is chosen to be 0.98 for our calculation), $n^* = (13.6/I_p)^{1/2}$ is the effective principle quantum number and $C_{n^*l^*} \approx 2$ can be found in reference [17]. $G_{lm} = (2l+1)(l+|m|)!/6^{|m|}|m|!(l-|m|)!$, where l and m are the orbital and magnetic quantum numbers of the outermost electron. $G_{lm} = 3$ for p state and $G_{lm} = 1$ for s state electron. Values of $I_p, n^*, C_{n^*l^*}$, and G_{lm} for the noble gasses are summarized in Table I. Equation (2) clearly shows how the cutoff photon energy changes with the laser pulse duration and wavelength, the atomic species, and the electron quantum state.

III. EXPERIMENT

The Ti:sapphire laser system used for the experiments can generate TW-level, 25-fs, pulses with a center wavelength of 800 nm [10]. A 1-cm diameter laser beam is focused onto the gas target using a 1 m focal length curved mirror, which produces a ~100-μm diameter focal spot. The gas nozzle diameter is 1 mm, while the gas pressure was approximately 8 torr (at the interaction region) for these experiments.

Typically, 20 mJ of laser energy is used to generate the harmonics, corresponding to an intensity of 6×10^{15}W/cm^2 at the focus. The X-rays are dispersed using a flat-field soft X-ray spectrometer, and then detected using an image intensifier with a pair of microchannel plates (MCP's). The spectrally dispersed image is recorded using a cooled charged-coupled device (CCD) camera connected to a computer. It is essential to block the fundamental laser beam inside the spectrometer to prevent the generation of a significant ion background at the detector. X-ray filters must also be placed in front of the MCP to block the very bright scattered low-order harmonics.

IV. COMPARISON BETWEEN EXPERIMENT AND THEORY

Equation (2) demonstrates that the cutoff photon energy is inversely proportional to the square of the logarithm of the laser pulse width, so it is clear that using shorter duration laser pulses should result in the generation of higher order harmonics. This is clearly shown in Fig. 1, which plots the experimental observations and theoretical predictions [using (2)] for an Argon atom, for pulse durations of 25, 50, and 100 fs, respectively. These results are also consistent with our previous work, [4] and with fully quantum mechanical calculations. Equation (2) also indicates that the cutoff photon energy should be proportional to the cube of the ionization potential of the atom. Experimentally, we generated harmonics up to 29, 41, 61, and 155 from Xe, Kr, Ar, and Ne, with corresponding ionization potentials of 12.13, 13.99, 15.76, and 21.56 eV respectively. Using (2), we predict that harmonics up to order 27, 41, 61, and 163 should be observed from Xe, Kr, Ar, and Ne respectively. Our simple calculations and experimental observations are, thus, in very good agreement.

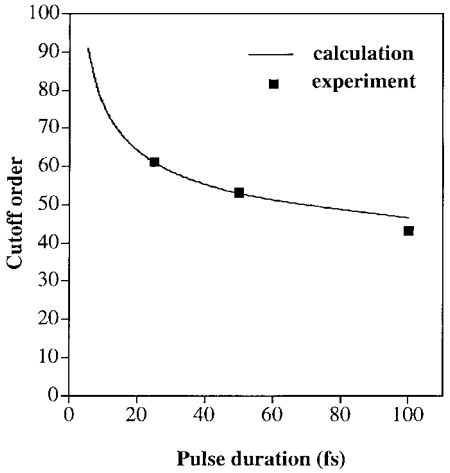

Fig. 1. Dependence of the cutoff order on laser pulse duration for argon.

To generate even higher harmonics, we used helium gas with an ionization potential of 24.59 eV. At first, it might not seem obvious that significantly higher harmonics would be expected from He, given that the ionization potential is only slightly higher than in Ne. However, since the cutoff photon energy is proportional to the cube of the ionization potential, a small difference in I_p will end up with a big difference in hv_c. Moreover, the outermost electron of He is in an s-state rather than a p-state as in the other noble gasses. Since the term G_{lm} for an s electron is three times less than for an p electron, s electrons generate higher harmonic photon energies. Physically this is because s electrons are more difficult to ionize than p electrons, even in the case of comparable ionization potentials. From (2), under our experimental conditions, we expect to observe harmonics up to the 333rd order from helium, which would correspond to a wavelength of 2.4 nm (518 eV). Experimentally, we observe discrete harmonic peaks in He up to the 221st order, as shown in Fig. 2(a). The cutoff at the 221st order is instrumental, due to a beam block placed in the spectrometer to eliminate scattered light from low-order harmonics and the fundamental light. The variations in the signal amplitude near the C edge are most likely due to carbon contamination on our microchannel plate detector.

The results shown in Fig. 2(a) were obtained with a grating optimized for 5 nm. To observe shorter-wavelength radiation

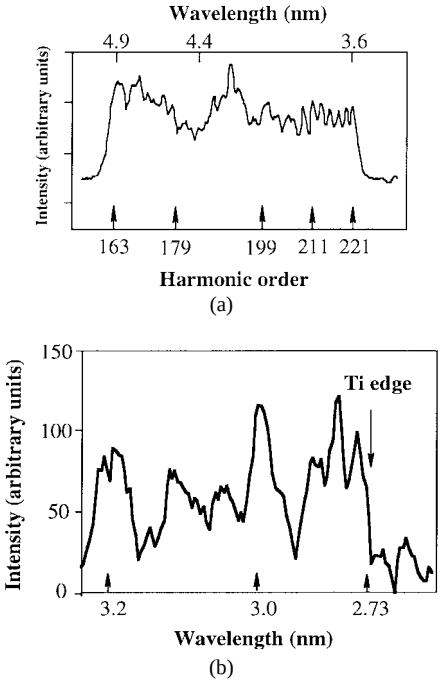

Fig. 2. (a) Discrete harmonic emission from Helium near the cutoff region, for 25-fs excitation pulses, using a grating optimized for 5 nm. (b) Harmonic spectra from filtered through a 0.2-μm titanium filter ($l > 2.7$ nm), using a grating optimized for shorter wavelengths.

from He, we used a second grating with higher groove density, optimized for shorter wavelengths. This allowed us to block the fundamental beam without simultaneously obscuring the harmonic radiation. The lower efficiency of this grating results in much lower signal-to-noise ratio (SNR). Nevertheless, we can observe harmonic radiation transmitted through a 0.2-μm Ti filter, terminating for wavelengths shorter than the Ti edge at 2.73 nm, as shown in Fig. 2(b) [5]. At the highest photon energies (harmonic order $>$200), we estimate that we generate at least several hundred photons/harmonic peak/pulse.

Fig. 3 shows a comparison of the observed cutoff photon energy for 25-fs excitation pulses, and the predictions of (2), for all the noble gasses. The dependence of the cutoff harmonic order on ionization potential (atomic species) is approximately cubic. The observed cutoff harmonic order is in excellent agreement with theory in all cases except for He, where there is still a slight 10% difference between our experimental observation (297th) and the theoretical prediction (333rd). The good agreement between theory and experiment is most likely because our gas densities are sufficiently low and our pulses sufficiently short that propagation effects do not play a major role in determining the output. Very recently, radiation at wavelengths as short as 4.4 nm was also observed by another group using very short driving laser pulses [8]. In this case, the nontransform-limited bandwidth of the driving pulses and high gas pressures precludes the observation of discrete harmonic orders.

The pulse duration of the harmonics is predicted to be extremely short, because the emission occurs only on the rising edge of the pulse [7], [19]. For 25-fs excitation pulses, harmonics are predicted to be generated during $\approx$3 half-cycles of the laser pulse (i.e., 3.5 fs) as shown in Fig. 4(a), during

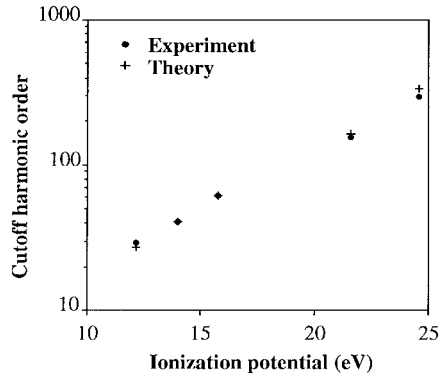

Fig. 3. Comparison of predicted [from (2)] and observed cutoff photon energies for harmonic generation in the noble gasses (on a log scale).

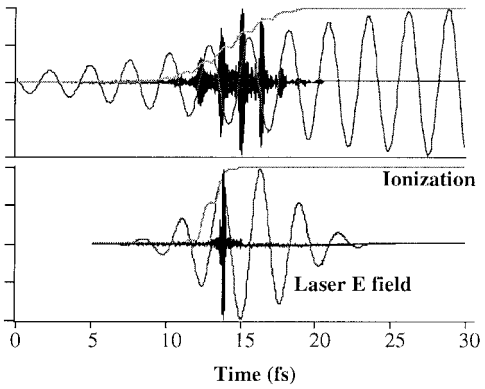

Fig. 4. Time dependence of the laser field (bold line), ionization probability (dashed line), and harmonics for (a) 25-fs excitation pulses and harmonics 61–81 and (b) 5-fs laser pulse and harmonics 61–81. The peak laser intensity is 1.1×10^{15} W/cm^2. Note that the fundamental and the harmonic fields are normalized in arbitrary units because of the very large difference between the corresponding absolute values.

which time the ionization probability varies from 10% to $\approx$100%. During this time also, the amplitude of the incoming pulse changes by $\approx$30%. For very short or excitation pulses ($\approx$5 fs), a *single-cycle* of the excitation pulse can drive harmonic emission over a range of adjacent harmonic orders. As a result, the temporal coherence of the harmonics is dramatically improved compared with longer excitation pulses. In addition, more efficient X-ray pulses, with durations as short as 100 as, can be emitted under the proper focusing and gas jet pressure conditions, as shown in Fig. 4(b). However, in order to generate an attosecond X-ray pulse, harmonics in a narrow wavelength range ($\sim$10 harmonic peaks) must be energy selected in a nondispersive way, such as using an appropriate thin-film filter. Geometrical pulse broadening must also be avoided by spatial aperturing and high-quality optics.

The observed harmonic spectra also change dramatically with the chirp of excitation laser pulse. The laser chirp can easily be varied by adjusting the separation of the stretcher gratings. As expected, the harmonic peaks shift toward longer wavelengths for positive chirp, when the leading edge of the pulse is redder than the trailing edge. This result is qualitatively similar to our results with argon [4], but in this case the shift is larger ($\times$2) and it can cover four harmonic orders (two peaks). For transform-limited excitation pulses, the highest harmonics

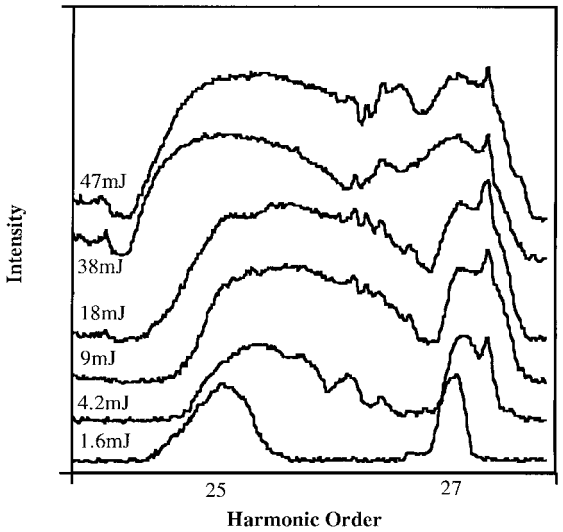

Fig. 5. Spectral broadening and blue-shifting of the 25th harmonic in argon, as a function of incident laser energy. The cutoff on the right is due to a spectrometer aperture.

near cutoff are clearly resolved. However, for the lower orders in the plateau region close to the cutoff, the peaks are more difficult to resolve [also shown in Fig. 2(a)]. This smearing also occurs when the pump pulse is negatively chirped. It is interesting to note that similar spectral structure for harmonics generated with transform limited ultrashort driving pulses has been predicted in recent numeric simulations by Schafer and Kulander for Argon gas [13].

Finally, by increasing either the gas jet pressure or laser energy, the harmonics in the mid-plateau can merge to form a complete "X-ray continuum" source, as shown in Fig. 5. This behavior may be due to self-phase modulation (SPM) and/or ionization-induced blue shifts of the excitation laser in the gas jet. It is unlikely that these effects are due to volume effects arising from different interaction and emission volumes as the laser energy is increased. The blue shift observed experimentally might arise from an increasing interaction volume if the excitation laser were positively chirped. However, we can show that the emission volume remains small since our spectrometer is imaging, which therefore spatially images in one direction, while spectrally dispersing in the perpendicular direction. It is also possible that the spectral broadening is due to some intrinsic single atom effects due to different possible trajectories of the emitting electron. However, this explanation is unlikely since the broadening disappears if the gas density is reduced. Further work is in progress to understand these effects.

V. Conclusion

We have generated coherent X-ray pulses at wavelengths of 2.7 nm, which is well within the "water window" region between 4.4 and 2.3 nm, where water is less absorbing than carbon. These X-ray pulses are possibly a few femtoseconds in duration, and can be tuned continuously in wavelength by adjusting the laser chirp. We obtain excellent agreement between theory and experiment for the variation of harmonic orders as a function of pulsewidth, and the variation of harmonic orders as a function of gas species, Therefore, using ultrashort excitation pulses, coherent, tunable, femtosecond, X-ray beams can be generated throughout the soft-X-ray region. In the future, this very compact femtosecond X-ray source, driven by kilohertz repetition rate lasers, may be very important for applications such as imaging through aqueous solutions, or time-resolved photoelectron spectroscopy of organic molecules and solids.

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Prof. Murnane honors include the American Physical Society Simon Ramo Award, an Alfred P. Sloan Foundation Fellowship, a National Science Foundation Presidential Faculty Fellowship, and the 1997 Maria Goeppert-Mayer Award of the American Physical Society. She is member of the American Physical Society, the Optical Society of America, and the Association for Women in Science.

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