Generation of Coherent Soft X Rays at 2.7 nm Using High Harmonics

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Ultrafast laser pulses from a Ti:sapphire laser centered at 800 nm, with 26 fs pulse duration, were used to generate coherent soft-x-ray harmonics, at wavelengths down to 2.7 nm (460 eV) in He, and 5.2 nm (239 eV) in Ne. In He, discrete harmonic peaks are observed up to order 221, and unresolved harmonic emission is observed up to order 297. These wavelengths are well within the "water window" region of x-ray transmission. Our work represents the shortest wavelength coherent light generated to date. The harmonic cutoff from all the noble gases is consistent with analytic theory. [S0031-9007(97)04256-7]

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The discovery of high-order harmonic generation by intense laser-atom interactions provides a new method to generate coherent, ultrashort-pulse, x-ray emission [1,2]. It is important to determine the shortest wavelengths attainable from this method, both for practical applications in ultrafast x-ray dynamics, and for our theoretical understanding of the process itself. The existence of a cut-off wavelength in the harmonic generation process has been well documented both experimentally and theoretically. A variety of wavelengths and pulse durations have been used to generate harmonics, and harmonic orders up to the 135th have been observed by L'Huillier *et al.* from the two lighter noble gases using 1 ps, 1054 nm laser pulses [3]. Macklin *et al.* observed harmonics up to the 109th order using 125 fs, 806 nm pulses in neon [4]. Preston *et al.* obtained the 37th order of a 380 fs, 248 nm KrF laser from helium ions [5]. Thus, the shortest wavelength of resolvable discrete harmonics demonstrated to date is approximately 7 nm. In the work described herein, we report the generation of harmonics up to order ≈ 300, at a wavelength of 2.7 nm, which corresponds to an energy of 460 eV. These extremely short wavelength harmonics were produced using an ultrashort pulse, 26 fs, Ti:sapphire laser centered at 800 nm [6,7]. This work dramatically extends the available wavelength range of coherent, ultrashort-pulse, x rays into the "water window" region, where water is less absorbing than carbon.

The great attraction of high harmonic generation as a source of coherent x-ray pulses is that it is a simple technique. 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 [8–10]. Quantum mechanical models are needed to describe the x-ray emission accurately. However, it can be understood to a first approximation using a semiclassical theory for the motion of an ionized electron during the first optical cycle after it is ionized due to the strong laser field [11,12]. In this picture, the highest photon energy achievable is determined by the maximum kinetic energy of this electron as it returns to the nucleus:

$$h\nu_c = I_p + 3.17U_p\,, \tag{1}$$

where U_p is the ponderomotive energy, and I_p is the ionization potential of the atom. This cut-off rule has also been obtained by solving the Schrödinger equation of a single atom either numerically or analytically [13–15].

In order to show the explicit dependence of the cut-off photon energy ($h\nu_c$) on the atomic and laser parameters, we developed a simple model to predict $h\nu_c$ for a given setup. Assuming that the atom is ionized on the leading edge of a linearly polarized laser pulse and following the method developed by Chang *et al.* [16], an analytical expression of the saturation intensity I_s can be obtained. Substituting $U_p = 9.33 \times 10^{14} I_s \lambda^2$ into Eq. (1), and assuming Ammosov-Delone-Krainov (ADK) ionization rates [17], we obtain

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

where $h\nu_c$ and I_p are in eV, $a = 0.5$ (to correct an approximation in the derivation of the analytical expression of I_s), λ 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^* is the effective principle quantum number, and $C_{n^*l^*}$ can be found in [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. In Eq. (2), $C_{n^*l^*} \approx 2$, $G_{lm} = 3$ (except for He, where it is 1), and n^* varies between 0.74 for He and 1 for Xe. Equation (2) clearly shows how the cut-off photon energy changes with the laser pulse duration and wavelength, the atomic species, and the electron quantum state.

It is clear from Eq. (2) that using shorter duration laser pulses should result in the generation of higher-order harmonics. This is consistent with our previous work [18,19], where we observed harmonics up to orders 29, 41, and 61 from Xe, Kr, and Ar, with corresponding ionization potentials of 12.13, 13.99, and 15.76 eV, respectively. Using Eq. (2), we predict that harmonics up to order 27, 41, 61 should be observed from Xe, Kr, and Ar, respectively (for 100 fs excitation pulses, harmonics up to order 23, 33, and 45 are predicted). Our simple calculations and experimental observations are thus in good agreement for Xe, Kr, and Ar. In order to generate even higher harmonics, the best approach is to use Ne or He, with ionization potentials of 21.6 and 24.6 eV, respectively. From Eq. (2), under our experimental conditions, we expect to observe harmonics up to order 163 and 333 from Ne and He, which would correspond to wavelengths of 4.9 nm (253 eV) and 2.4 nm (518 eV), respectively. As a comparison, for 100 fs excitation pulses, harmonics up to order 119 and 239 should be observed for Ne and He, respectively, assuming the same peak intensity (which requires more energy).

To observe high harmonics from Ne and He, we used a setup shown in Fig. 1. The Ti:sapphire laser system used for the experiments can generate 26 fs pulses with a center wavelength of 800 nm [7]. A 1 cm diameter beam is focused onto the gas target using a 1 m focal length curved mirror, which produces a $\sim 100\ \mu$m 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 approximately 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 (MCPs). This setup is superior to our previous one [18,19], which required us to scan the spectrometer to acquire a spectrum. 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.

For the experimental conditions described above, the harmonic spectra observed from Ne and He exhibit discrete, resolvable, harmonic peaks up to the 155th and 221st order, respectively. Figure 2(a) shows the harmonic spectrum of Ne, while Fig. 2(b) shows the case of He, using an efficient x-ray grating optimized around 50 Å. In Fig. 2(b), the cutoff at the 221st order is instrumental, and due to a beam block placed in the spectrometer to eliminate scattered light from low-order harmonics and the fundamental light. Two sets of harmonic spectra are shown for He [Fig. 2(b)], corresponding to slightly different gas pressures. The highest-order harmonics (top curve) are enhanced for slightly lower gas pressures, as would be expected from phase-matching considerations. The variations in the signal near the *C* edge in Fig. 2(b) are most likely due to carbon contamination on our channel plate. We observed similar contamination on our gratings and x-ray optics, and from our simulations, contamination on the channel plate would lead to such an oscillatory response.

In order to verify experimentally that the harmonic emission is indeed due to short wavelength light, we placed a 0.4 μm carbon filter between our spectrometer

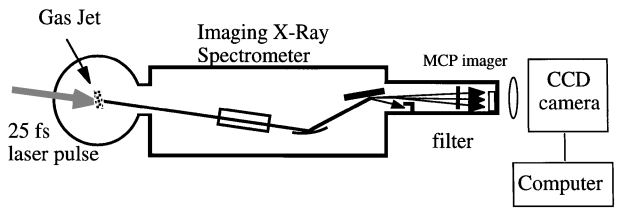

FIG. 1. Experimental setup for high-harmonic generation.

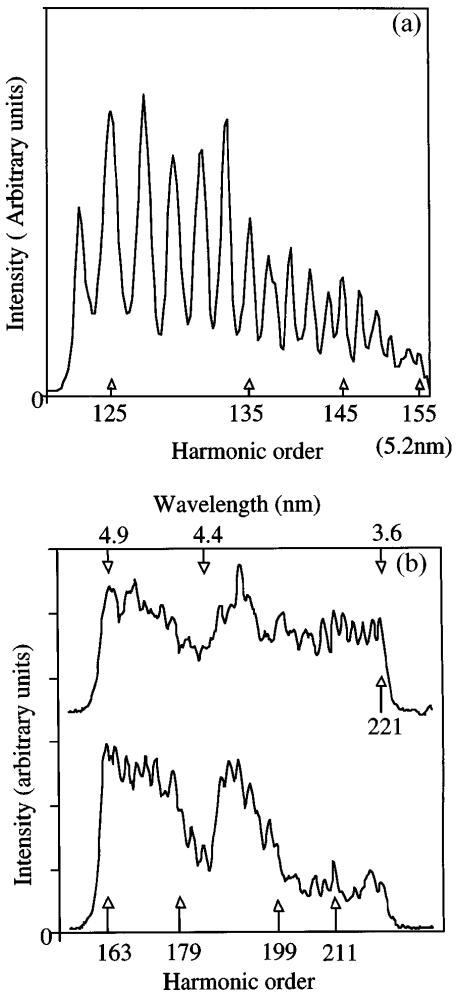

FIG. 2. Discrete harmonic emission from (a) neon and (b) helium in the cut-off region for 25 fs excitation pulses. The *y* axis is on a linear scale.

and detector. The filtered harmonic emission spectrum, on a log scale, is shown in Fig. 3(a). The position of the carbon 4.37 nm absorption edge can clearly be seen, as well as the discrete harmonics, and plateau region. This also allowed us to verify our spectrometer calibration, using the position of both the carbon and boron absorption edges. To observe shorter wavelength radiation from He, we used a grating optimized for shorter wavelengths, which allowed us to block the fundamental beam without simultaneously obscuring the harmonic radiation. However, the lower efficiency of this grating results in much lower signal-to-noise ratio. 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. 3(b). At the highest photon energies (harmonic order 297), we estimate that we generate at least several hundred photons/harmonic peak/pulse. This may be an underestimate, since we use best case estimated efficiencies for our x-ray optics and detectors. At lower photon energies, the harmonic flux is up to 10^7 times higher, depending on the energy region.

Figure 4 shows a comparison of the observed cut-off photon energy for 26 fs excitation pulses, and the predictions of Eq. (2), for all the noble gases. The dependence of the cut-off harmonic order on ionization potential (atomic species) is approximately cubic. The observed cut-off 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 experiments 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. In the case of He, there are several possible explanations for the small discrepancy between theory and experiment. First, we have not definitely observed the cutoff. Second, the laser intensity in the gas medium may be well below the intensity that we used in the calculation, due to the defocusing of the laser beam induced by the ionization. Third, Eq. (2) does not take into account propagation effects—the phase mismatch induced by the free electrons may play a significant role for efficient production of harmonics below 2.7 nm. Finally, the ADK approximation and/or other assumptions made in deriving Eq. (2) may not be valid in the case of helium, since technically ADK is valid only for large values of n^* [17].

The pulse duration of the harmonics we produce is predicted to be less than 3 fs [18–20], since the emission occurs on the rising edge of a 26 fs pulse. We have as yet measured only the pulse duration in the ultraviolet region, where the duration is expected to be longer [21]. More detailed calculations have also determined that using 26 fs excitation pulses to drive high-harmonic emission, we are just beginning to enter a new nonadiabatic regime, where the response of the system depends on the time-history (pulse shape) of the excitation [19,20].

The observed harmonic spectra also change dramatically with the chirp of excitation laser pulse, which can easily be varied by adjusting the separation of the stretcher gratings. As expected, the harmonic peaks shift to 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 [18], but in this case the shift is larger ($\times 2$) and it can cover four

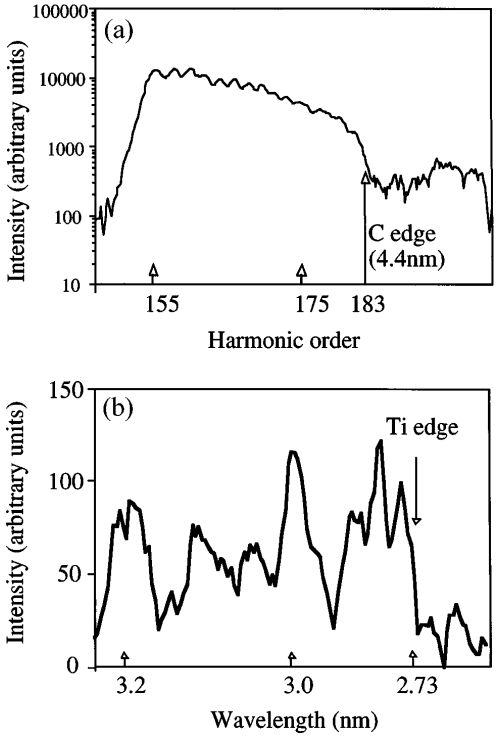

FIG. 3. Harmonic emission from Helium, (a) filtered through a 0.4 μm carbon filter, and (b) filtered through a 0.2 μm titanium filter.

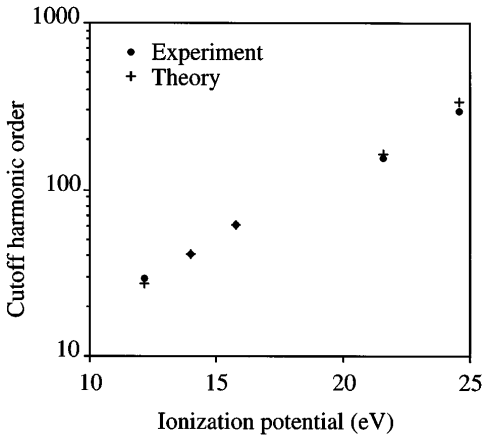

FIG. 4. Comparison of predicted [Eq. (2)] and observed cut-off photon energies for harmonic generation in the noble gases (on a log scale).

harmonic orders (two peaks). For transform-limited excitation pulses, the highest harmonics near cutoff are clearly resolved. However, for the lower orders in the midplateau region, the peaks are more difficult to resolve [as shown in the top curve of Fig. 2(b)]. Similar spectral structure for harmonics generated with transform-limited ultrashort driving pulses was predicted by the recent numeric simulations of Schafer and Kulander [22]. Finally, by increasing either the gas jet pressure or laser energy, the harmonics in the midplateau can merge to form a complete "x-ray continuum" source. Very recently, radiation at wavelengths as short as 4.4 nm was observed by another group using very short driving laser pulses [23]. In this case, the non-transform-limited bandwidth of the driving pulses precludes the observation of discrete harmonic orders.

In 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. Our shortest observed wavelength to date of 2.7 nm is the shortest wavelength coherent light generated to date. These x-ray pulses are possibly a few femtoseconds in duration. 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 kHz 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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