Coherent 0.5-keV X-Ray Emission from Helium Driven by a Sub-10-fs Laser

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(Received 29 July 1997)

Helium atoms ionized by intense few-cycle light pulses in the barrier suppression regime emit spatially coherent extreme ultraviolet continuum extending to photon energies greater than $E_{\mathrm{ph}} = 0.5$ keV ($\lambda < 2.5$ nm). The high-energy end of the continuum in the range of $E_{\mathrm{ph}} \geq 0.2$ keV ($\lambda \leq 6$ nm) was characterized spectrally over a considerable dynamic range using energy-dispersive detection. The sub-10-fs laser pulse duration was found to be crucial for generating radiation with the highest photon energies at the low (<0.5 mJ) pump energy levels used in the experiments. The single-atom quantum theory of high-order harmonic generation combined with Maxwell's wave equation provides a satisfactory account for the experimental observations. [S0031-9007(98)05814-1]

PACS numbers: 42.65.Ky, 41.50.+h, 42.50.Hz, 42.65.Re

This Letter reports the generation of coherent x rays with photon energies in excess of 500 eV (corresponding to wavelengths shorter than 2.5 nm) in a helium gas jet irradiated by near-infrared sub-mJ, sub-10-fs optical pulses at a 1-kHz repetition rate. This radiation constitutes the shortest-wavelength coherent light reported to date. The x rays are emitted in a well-collimated beam with a divergence that decreases with increasing photon energy. The 1-kHz repetition rate of the laser driver allowed the use of highly sensitive energy-dispersive x-ray spectrometry (EDS), which permitted high-dynamic-range ($\approx 10^3$) spectral characterization of a high-harmonic source in the sub-keV range for the first time. These measurements provide insight into a new regime of extreme ultraviolet (XUV) high-harmonic (or continuum) generation in which the atoms undergo ionization with their Coulomb barrier suppressed below the ground-state energy level during the interaction responsible for high-energy XUV emission. Comparison of the observed spectra with results of theoretical simulations points to the key role of dephasing during propagation.

Coherent harmonic emission from atomic ensembles exposed to ultrashort laser pulses has been previously observed up to photon energies many times higher than the potential energy of bound electrons in the atomic Coulomb field [1–3]. In a semiclassical picture, this phenomenon has been understood in terms of tunnel ionization of the atom, followed by acceleration of the electron in the external field and recombination with its parent ion [4,5]. These processes take place over an optical period and are repeated quasiperiodically in laser pulses that comprise many oscillation cycles, leading to the emission of high harmonics of the driving field. The highest harmonic photon energy is given by $(E_{\mathrm{ph}})_{\max} \approx I_p + 3.2U_p$, where I_p is the ionization potential of the atom, and U_p is the ponderomotive potential of the laser field [3–5]. Here U_p in eV is approximately given by $0.93 \times 10^{-13} I_s \lambda^2$, where λ is the carrier wavelength in μm and I_s is the saturation intensity in W/cm^2, which the atoms can be exposed to before the ground state is depleted; i.e., the ensemble is fully ionized [6]. Since $(E_{\mathrm{ph}})_{\max}$ is higher for shorter pulses, the highest-energy discrete harmonics (200 eV in Ne, 240 eV in He) have been generated with the shortest driver pulses available (25 fs) [7,8] at a 10-Hz repetition rate.

Recently 0.1-TW, 5-fs pulses at a repetition rate of 1 kHz became available in the near infrared [9]. These intense light "transients" opened up the way to an entirely new regime of strong-field atomic processes in that (i) the laser field can exceed the barrier-suppression limit well before the ground state is depleted, and (ii) the emission of the highest-energy photons can be confined to a fraction of a *single* oscillation cycle. These features are evident from Fig. 1, which depicts the evolution of the (linearly polarized) electric field in a 5-fs pulse at $\lambda = 0.75$ μm having a peak intensity of 4×10^{15} W/cm^2 together with the Ammosov-Delone-Krainov (ADK) tunnel ionization rate [10] induced by the pulse in helium. Striking consequences include the generation of *single* [11] attosecond XUV pulses [12–14], the extension of coherent XUV generation to shorter wavelengths [11], and the coalescence of high-order harmonics to a quasicontinuum [11,14]. In fact, recent experiments using the novel 5-fs driver for irradiating helium and a wavelength-dispersive spectrograph (WDS) demonstrated the generation of coherent XUV continuum extending beyond the K edge of carbon at 280 eV (i.e., into the water window) [15,16]. This paper presents the first XUV high-harmonic spectra at photon energies greater than 300 eV over a substantial dynamic range. The data obtained with an EDS system allow comparison with existing models of high-harmonic generation (HHG) for the first time in this wavelength range and reveal

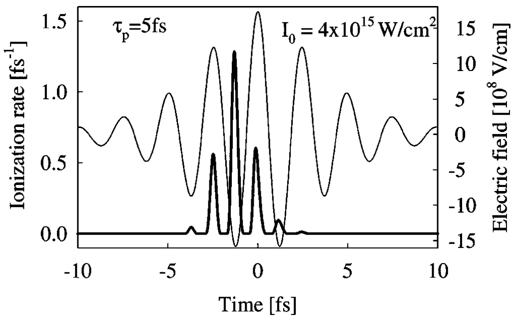

FIG. 1. Evolution of the electric field in a linearly polarized 5-fs laser pulse of a peak intensity of 4×10^{15} W/cm^2 carried at $\lambda = 0.75$ μm (thin line) and the ionization rate induced by this pulse in helium (thick line). The electric field amplitude should be contrasted with the barrier suppression field of $E_{\mathrm{bs}} = 1.03 \times 10^9$ V/cm for helium.

the crucial role of the sub-10-fs pump pulse duration for near-keV coherent x-ray generation at submillijoule energy levels.

The single-atom quantum theory of HHG developed by Lewenstein *et al.* [17] recovers the semiclassical interpretation outlined above and proved successful in accounting for a broad range of experiments performed with many-cycle laser pulses [18]. This approach was recently extended to the few-cycle regime by incorporating ground-state depletion based on the ADK ionization rate and using Maxwell's wave equation for describing propagation of the fundamental and XUV fields [11]. Figure 2 plots the XUV spectrum emerging from a He ensemble irradiated by an intense 5-fs near-infrared laser pulse [Fig. 2(a)] together with the single-atom response [Fig. 2(b)], as predicted by this model. The single-atom response (power spectrum of dipole acceleration) exhibits a nearly constant spectral intensity up to 600 eV, in agreement with the prediction of the above cutoff law for I_s inferred from Fig. 1. By contrast, the XUV radiation emerging from the interaction region is subject to a dramatic decrease with increasing photon energy due to a rapidly decreasing coherence length with increasing photon energy in the high-pressure target. As a consequence, the experimentally observed cutoff energy is often lower than that of the single-atom response [18] and is, in general, significantly affected by the sensitivity of the detection system.

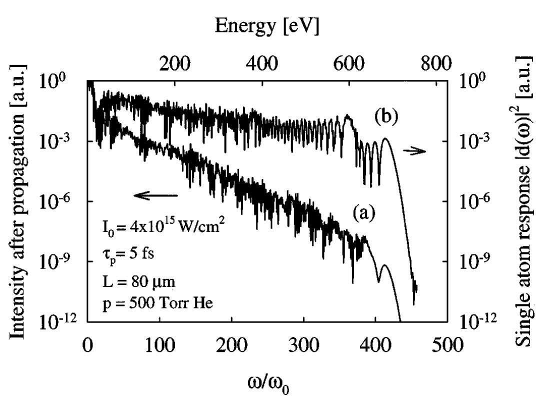

FIG. 2. Dipole radiation spectrum of He exposed to a 5-fs laser pulse carried at $\lambda = 0.75$ μm as obtained from the quantum theory of Lewenstein *et al.* and near field XUV spectrum emitted by the ensemble. 1D propagation of the fundamental and XUV pulses through the 80-μm-thick target is calculated using Maxwell's wave equation.

The experimental setup is schematically shown in Fig. 3. The interaction region between the laser beam and the helium gas is formed by a thin nickel tube, the inner diameter (0.6 mm) of which was squeezed to ≈ 50 μm. $f/30$ optics focuses the laser beam through a 0.3-μm-thick fused silica window onto the squeezed section of the tube. The laser drills holes in the tube wall, the diameters of which are precisely matched to that of the beam, minimizing gas load to the surrounding vacuum ($p_{\mathrm{chamber}} < 1$ Torr). The tube is backed continuously with pressures of 4–5 atmosphere, resulting in an estimated target pressure of ≈ 500 Torr. The high pressure is dictated by the "geometric" coherence length associated with tight focusing of the pump laser [6], which was necessary to achieve the required peak intensities with our present source. In the experiment described below the fluence on the optical axis at the target is (35 ± 5) J/cm^2. The 0.1-TW, sub-10-fs, 1-kHz Ti:sapphire laser system ($\lambda \approx 0.77$ μm) is described in detail elsewhere [9].

The XUV radiation exiting the target collinearly with the laser beam hits a cooled (77 K) lithium-drifted silicon crystal detector (Z-MAX, NORAN Instruments) 35 cm downstream of the source. Thin metal foils deposited on nickel meshes and placed in front of the 400-nm diamond window of the detector block the laser radiation. Because the foils are not completely free from tiny pinholes, their apertures have to be confined to a few hundred micrometers and at least a couple of them separated by some 5 cm have to be used in order to achieve the required rejection of the fundamental. The alignment precision of these apertures on the optical axis is limited by the large laser beam diameter (≈ 10 mm) at the apertures, which compromises the XUV throughput. The energy resolution (FWHM) of the EDS detection system at the carbon K_α line is ≈ 90 eV. The distribution of dark counts (gas off, laser blocked) peaks at around 100 eV and rapidly rolls off for increasing energies. No dark counts are observed beyond 200 eV. For the filter combinations described below this behavior does not change when the laser is unblocked except that the noise

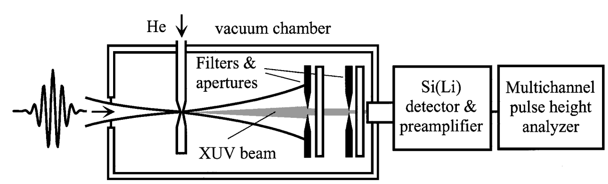

FIG. 3. Schematic of the experimental setup. Explanations are given in the text.

counts increase by some 10%. The spectra shown below are corrected for these noise counts. Neither grazing the nickel tube wall deliberately with the laser beam nor using argon as the target material results in any detectable signal, indicating the negligible role of incoherent plasma emission in the data presented below.

For correct EDS analysis the probability of two or more x-ray photons striking the detector for any given laser shot must be sufficiently low to avoid sum peaks (pileups). This can be ensured by keeping the count rate at sufficiently low levels and discriminating against the high-flux low-energy ($<$250 eV) photons by employing suitable combinations of metal foils. To determine the maximum count rates at which pileups can be safely rejected, a nickel mesh is introduced in the XUV beam at varying angles in front of a couple of 350-nm titanium foils to serve as a variable neutral filter. A comparison of the spectra obtained at count rates ranging between 6.7 and 76 counts/sec reveal that a safe rejection of pileups calls for a probability of detecting a soft x-ray photon per shot not greater than 1%. This critical count/shot probability is dependent on the shape of the spectrum and is expected to increase with increasing rejection of low-energy with respect to high-energy photons [19]. In the measurements reported below the ratio of low-energy to high-energy counts $N_{250\text{-eV}}/N_{500\text{-eV}}$ is kept below 10^3 and the count/shot probability below 1% for safe pileup rejection.

Previous measurements of the XUV beam profile by scanning a knife-edge across the beam yielded a divergence of $<$1 mrad and $<$0.5 mrad at 120 eV and 280 eV, respectively [16], giving evidence of an excellent spatial coherence. Repeating the knife-edge measurements in combination with EDS, the tendency to a decreasing divergence with increasing energy has also been observed beyond 280 eV. The dependence of the high-energy x-ray yield on the pulse duration has been investigated by varying the extent of spectral broadening in the pulse compressor [9]. The significant decrease in the experimentally observed cutoff as pulse length increases (Fig. 4) is commensurate with our simulations. In addition to ground-state depletion directly limiting the high-frequency dipole moment through the cutoff law, the higher background free electron density leads to a reduced coherence length for longer driver pulses. We conclude that efficient production of coherent x-ray photons in excess of 400 eV calls for pulses of 10 fs or shorter in duration.

For comparison with the prediction of the theoretical model (Fig. 2) the x-ray spectrum generated by 7-fs pulses has been measured with two filter combinations having distinctly different characteristics. The integration time has been chosen to yield approximately the same signal in the 180–200 eV range for both filter combinations listed in Fig. 5. The differences in the measured spectra mirror the calculated differences in the filter-set transmittances to a resonable accuracy, providing a self-consistency check. To compare experiment with theory, the spectrum depicted

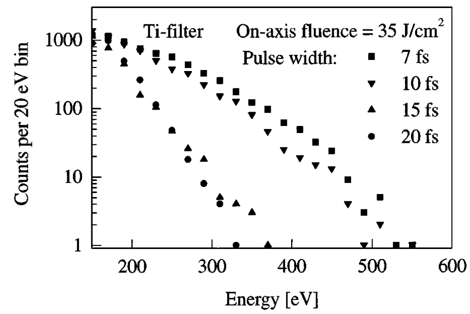

FIG. 4. Energy spectra transmitted through 700 nm titanium for different pulse durations. Notice that the titanium L edge at 470 eV is clearly visible in the spectra generated with the 7-fs and 10-fs pulses.

by plot (a) in Fig. 2 has been multiplied by the transmittance of the filter sets (including window transmittance) and convolved with the detector response. Considering the uncertainty in the thickness of the filters (known with an accuracy of 0% to 20%) and deviations between the experimental and simulation parameters the theoretical prediction (dashed lines in Fig. 5) agrees reasonably with the experimental results. This agreement provides conclusive evidence for the dramatic effect of the energy-dependent coherence length on the emitted coherent sub-keV radiation, which is reflected by Fig. 2(a).

The counts appearing at photon energies greater than 470 eV (the L edge of titanium) in Figs. 4 and 5 result from limited detector resolution. In order to gain access to the spectral region beyond 470 eV the titanium foils have been replaced by a 200-nm aluminum and 400-nm boron foil. The transmitted x-ray spectrum generated by 7-fs pulses is shown for $E_{\text{ph}} > 300$ eV in Fig. 6 along with the filter transmittance (including the window) versus energy. The data shown in Fig. 6 yield a photon flux at around 550 eV which is by more than 3 orders of magnitude lower than that at 350 eV, again, in reasonable agreement with the theoretical prediction [Fig. 2(a)]. Although reliable assessment of the absolute photon flux is not possible with

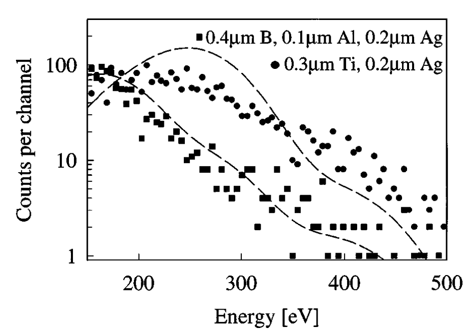

FIG. 5. Energy spectra transmitted through different filter combinations for 7-fs pump pulses. Correction of the near-field XUV radiation spectrum from Fig. 2 for the detector response and respective filter (and window) transmittances result in the dotted lines (without using fit parameters).

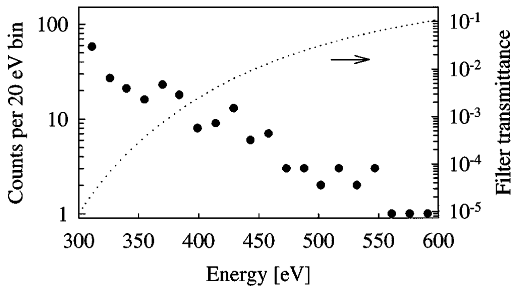

FIG. 6. High-energy end of the XUV spectrum for 7-fs pump pulses (9×10^5 shots) transmitted through 400-nm boron and 200-nm aluminum.

the present setup, considering the losses introduced by the meshes and the apertures along with the uncertainties in the filter thicknesses, a previous estimate of 10^3 photons per shot in a 10% bandwidth at 300 eV evaluated from the WDS work [16] appears to be realistic. This implies that only a few photons are produced at energies in excess of 500 eV by a single laser pulse, in spite of the fact that the single-atom dipole response [Fig. 2(b)] radiates at these energies almost as intensely as for much lower energies. As a consequence, increasing the coherence length for the highest-energy x rays by means of some (quasi) phase matching techniques to compensate for the dispersive effects of free electrons may be required to produce practically useful photon fluxes in the 500 eV regime. Present models for the atomic nonlinear response and collective growth of the XUV field appear to provide a good starting point for consideration of such schemes. On the other hand, an x-ray yield in the vicinity of the carbon K edge at 280 eV of approximately 10^6 photons/sec in a 10% bandwidth is already sufficient to prompt consideration of practical applications for this coherent water-window emission.

In summary, we have demonstrated that sub-10-fs near-infrared laser pulses allow the generation of coherent x rays with photon energies in excess of 500 eV at submillijoule energy levels. These pulses are available at a kHz repetition rate, permitting efficient implementation of EDS. This allowed the first high-dynamic-range spectral characterization of a high-harmonic source in the sub-keV range. Single subfemtosecond pulses from an improved kHz-rate sub-10-fs-laser-driven XUV source together with EDS will pave the way towards the extension of nonlinear optics and time-resolved spectroscopy into the x-ray regime.

The authors thank S. Sartania and M. Lenzner for invaluable help. This research has been supported by the Austrian Science Fund Grants No. P11109 and No. Y44-PHY.

Note added.—After the submission of this paper, harmonic generation at 2.7 nm was reported using 26-fs, 20-mJ pulses at a 10-Hz repetition rate [20]. The reported water-window x-ray yield/pulse is comparable to that obtained by low-energy sub-10-fs pulses in the present work.

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