Generation of Coherent Soft-X-Ray Radiation Extending Far Beyond the Titanium *L* Edge

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Coherent soft-x-ray radiation up to photon energies of 700 eV is obtained by focusing several-mJ, 10-fs near infrared laser pulses into a He gas jet. The observed nearly constant photon yield over several hundred eVs may be attributed to nonadiabatic self-phase matching, originating from a substantial ionization within a fraction of the optical cycle of the driving laser pulse.

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A bright laboratory x-ray source in the wavelength range of several nanometers is expected to revolutionize biological microscopy, allowing the study of biological samples in their natural environment with high contrast and resolution. Shorter wavelengths also open the way to generate shorter single attosecond pulses than demonstrated in the 13-nm spectral range [1,2]. Practically useful photon fluxes in the few-nanometer range were previously delivered by synchrotrons or x-ray lasers pumped by multikilojoule laser sources [3], both available at large-scale facilities only. In the past decade, coherent soft x rays have been produced from atomic gases ionized by femtosecond laser pulses at photon energies reaching the carbon *K* edge (284 eV) and the titanium *L* edge (454 eV) [4–7] at yields insufficient for practical use. Since these first demonstrations, several research groups improved the pump laser [8–11], and investigated phase matching schemes [12–14] to increase the x-ray photon yield. Recently, it has been demonstrated that quasiphase matching (QPM) in a modulated hollow-core fiber [15] gives rise to an enhanced photon yield near the carbon *K* edge [16].

It has been suggested that the efficiency of coherent x-ray production from ionizing gases can also be substantially increased by another method, which has been referred to as nonadiabatic self-phase matching (NSPM) [14,17]. The method relies on rapid ionization with few-cycle pulses of duration around 10 fs or shorter, which tends to lower the phase velocity of the driver pulse and helps match it to that of the x rays emitted by the atoms. For lower photon energies (< 100 eV), few-cycle drivers extended the coherence lengths to the order of the x-ray absorption length, resulting in x-ray production limited by the reabsorption of the generated radiation in the gas jet [18–20]. For photon energies above 200 eV, the coherence length becomes much shorter than the absorption length, providing room for QPM or NSPM to enhance the photon yield.

In this Letter, we report that—by exploiting the latter mechanism—coherent soft-x-ray radiation can be produced up to 700 eV, constituting the highest-photon-energy coherent laboratory source to date.

The ultrashort driving laser pulses are generated with an all-solid-state laser system based on Ti:sapphire. At the output of the amplifier, we have obtained 3-mJ pulses with a duration of $\sim$10 fs at a repetition rate of 1 kHz [11]. The beam was focused into a He gas jet using a broadband dielectric mirror with a focal length of 150 mm. The beam diameter (FWHM, full width at half maximum) in the 0.4-mm-long gas jet was approximately 50 μm, resulting in an estimated peak intensity of 8×10^{15} W/cm^2. To improve phase matching, the geometrical phase shift [21] has been exploited by moving the gas jet out of the focus. Maximum x-ray yield was achieved approximately 1 mm behind the beam waist of the pump beam. The x-ray beam launched into a spectrometer 1.1 m downstream from the source. The spectral characterization of the soft x rays was performed by a 1-m grazing-incidence scanning monochromator equipped with a 300 grooves/mm platinum-coated grating (248/310G, McPherson). The signal at the output of the monochromator was detected by an uncoated channeltron (4715G, Kore Technology) and a lock-in amplifier (5209, Signal Recovery).

Figure 1 shows a measured spectrum when the extreme-ultraviolet/soft-x-ray beam was passed through a 1-mm aperture. In the inset of Fig. 1, well-resolved harmonic lines are presented in the low energy range on a linear scale. The harmonics merge to an irregularly modulated part due possibly to the unstabilized carrier-envelope phase and partly to the limited resolution (~ 0.1 nm) of the monochromator. The spectrum in Fig. 1 shows rapidly dropping harmonic intensity up to 200 eV, followed by a plateau ending in a sharp cutoff at about 700 eV. The accuracy of determining the cutoff energy was around 5% due to the reduced spectral resolution in this spectral range. Towards higher energies, the signal starts increasing once again. This signal is attributed to scattering from the zeroth order of the monochromator.

To confirm the wavelength scale of the monochromator, several edge filters were introduced in the x-ray beam (Fig. 2). By inserting a filter consisting of 300-nm-thick titanium on a 200-nm parylene foil, we were able to

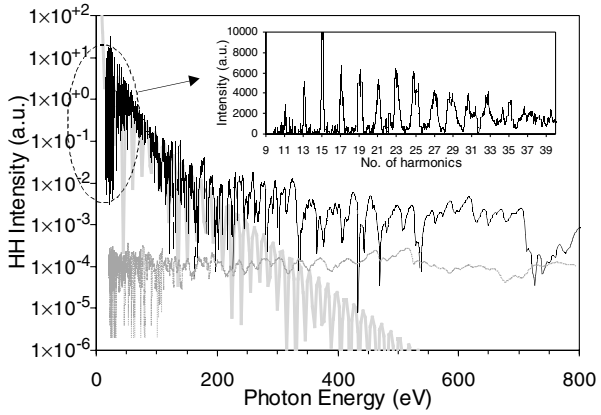

FIG. 1. Measured spectrum of coherent extreme-ultraviolet and soft-x-ray radiation emitted by a gas of He atoms (black line) ionized by 10-fs, 780-nm laser pulses having a peak intensity of 8×10^{15} W/cm^2. The thin and thick gray lines depict the background measured without gas flow and the scaling of the soft-x-ray production yield with photon energy as estimated on the basis of the adiabatic approximation $I_N \sim (\chi^{(1)} I_1)^N \sin^2(\pi l/2l_c)/(\pi l/2l_c)^2$ [22], where N, l, and l_c are the harmonic order, interaction length, and coherence length, respectively. $\chi^{(1)} I_1$ was chosen to fit the low energy part to the measurement. Inset: High-order harmonic lines in the low-energy, extreme-ultraviolet part of the emitted spectrum.

clearly resolve the carbon K edge (284 eV) and titanium L edge (454 eV). Insertion of a 100-nm Al filter permitted calibration at longer wavelengths owing to the appearance of the aluminum L edge at 73 eV.

For studying the high-energy part of the emitted spectrum with improved contrast, a 100-nm-thick titanium filter on 100-nm-thick parylene foil was used. The photon yield in the several-hundred-eV range was maximized by moving the jet with respect to the focal spot and by varying the backing pressure. Figure 3 depicts a series of measured spectra at different gas pressures. The photon yield above the Ti L edge was found to be maximum at a backing pressure of 250 mbar, exceeding those achieved at either 150 or 400 mbar by a factor of 2. Optimum flux beyond 450 eV came at the expense of a small shift of the cutoff to lower energies, but it remained far above the oxygen K edge (543 eV), at the range of 700 eV. Decreasing the pressure below 100 mbar reduced the signal below the background noise in the high-energy range. The carbon K and titanium L edges were clearly observed in all recorded spectra; however, they are not as sharp as the edges shown in Fig. 2 because of the thinner filter and the somewhat larger integration time of the lock-in amplifier. To compare the yield of our source with other sources, we estimated the number of photons after optimization at various energies. Taking into account the sensitivity of the channeltron, the diffraction efficiency of the grating and the effect of the slits in the monochromator, we estimated the following photon numbers in a 5% spectral band: 5×10^8 photons/s at 100 eV, 1×10^8 photons/s at 200 eV; 2×10^7 photons/s at carbon K edge; 2×10^6 photons/s at the nitrogen K edge; 1×10^6 photons/s at the titanium L edge; 9×10^5 photons/s at oxygen K edge and 2×10^5 photons/s over 700 eV. These values were corrected for the transmission of the filter combination used.

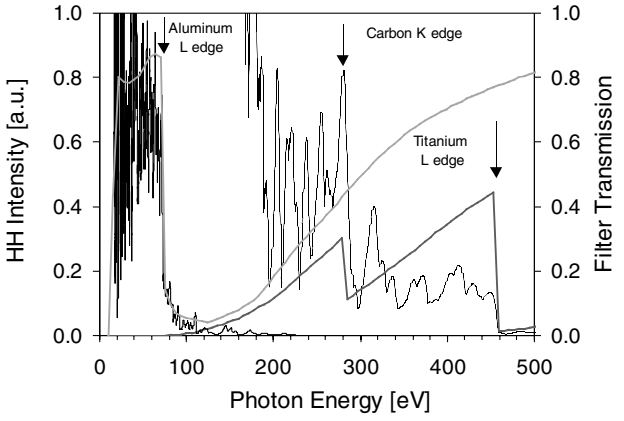

FIG. 2. Measured coherent extreme-ultraviolet and soft-x-ray spectrum from He (produced under the experimental conditions summarized in the caption of Fig. 1) filtered by a 100-nm-thick aluminum foil (0–200 eV range of the spectrum) or a 300-nm-thick titanium filter carried by a 200-nm parylene foil (200–500 eV range). Spectral transmission curves of the Al (thick gray line) and Ti + parylene (thick black line) filters are also shown.

The possibility of plasma radiation making a significant contribution to the measured signal was ruled out by moving the entrance slit of the monochromator off the optical axis and observing no signal (even in zeroth order). This is the first laboratory source generating coherent radiation up to 700 eV, and its photon flux exceeds that of previously demonstrated sources [4–6] by several orders of magnitude in the water window.

The cutoff energy of high-order harmonic generation emitted by individual atoms is given by $h\nu_{\max} = W_p + 3.2U_p$, where W_p and U_p are the ionization potential of

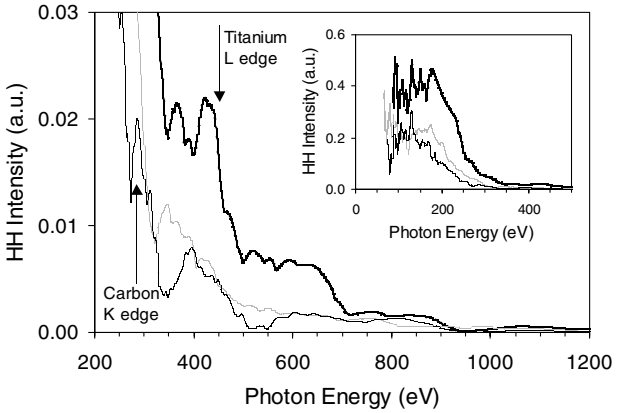

FIG. 3. Soft-x-ray spectra recorded at three different backing pressures: 150 mbar (thin line), 250 mbar (thick line), and 400 mbar (thin gray line).

the atom and the ponderomotive potential in the optical field, respectively. Under our experimental conditions (pulse duration $\sim$10 fs, peak intensity $\sim 8 \times 10^{15}$ W/cm^2), this formula yields maximal photon energy of $\sim$1.5 keV. Depletion of the ground state by ionization, free electron plasma induces rapid dephasing [12,21,23,24,22] for increasing photon energies currently prevent us from generating this radiation efficiently from macroscopic volumes.

Under our experimental conditions, the helium atoms are ionized at a fractional ionization rate of $> 30\%$/cycle near the pulse center, creating a substantial nonadiabatic contribution to the self-induced phase of the driving laser pulse. This contribution is opposite in sign to that caused by the refractive index of a free electron gas and, hence, extends the propagation length over which the high-energy x-ray emitted from individual atoms can grow coherently in the macroscopic interaction medium [14,17]. This mechanism has been termed nonadiabatic self-phase matching and predicted to set in if few-cycle pulses strongly ionize the atomic gas medium. The most striking implication of NSPM is that of a soft-x-ray yield nearly constant over a photon energy range where production efficiency is predicted to rapidly roll off according to the scaling of the adiabatic (or quasistatic) coherence length with photon energy.

The thick gray line in Fig. 1 depicts the predicted soft-x-ray yield versus photon energy inferred from the adiabatic scaling law of the coherence length, which is in striking contrast with the plateau observed up to 700 eV. At the highest photon energies observed, the adiabatic coherence length matches the 0.4-mm length of our interaction volume at pressures lower than 1 mbar. According to adiabatic phase matching, the x-ray yield should saturate upon increasing the pressure above this value. In reality, our high-energy photon yield increases by orders of magnitude as the pressure is raised from a few mbar up to > 100 mbar. These observations strongly suggest that NSPM plays a decisive role in achieving the results reported above, because the coherence length is no longer a correct measure of the harmonic growth.

Ionizing helium with few-cycle (10 fs, 780 nm), high-energy (2.5 mJ) laser pulses, we have generated well-collimated coherent soft x rays up to photon energies of 700 eV by exploiting nonadiabatic self-phase matching of the driving laser wave to the generated x rays. Owing to its practically useful fluxes up to 700 eV, the demonstrated source paves the way towards the development of a compact x-ray microscope for studying biological and biochemical samples *in vivo*. Extension of the spectral coverage to the 0.5–2 keV range will open the door to investigating magnetic processes with fs and sub-fs time-resolved spectroscopy.

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