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Generation of widely tunable harmonic pulses in the UV and VUV from a NIR optical parametric amplifier

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Abstract. We demonstrate the generation of tunable high-order harmonics from the interaction of the signal output of a near-infrared (NIR) traveling-wave optical parametric amplifier (OPA) with the xenon atoms of a pulsed gas jet. By tuning the OPA between 1200 and 1550 nm, we generate widely tunable pulses of ultraviolet radiation up to the ninth harmonic order, around 150 nm. The possibility to cover the VUV region thanks to the full overlap of the tunability ranges of different harmonic orders below 220 nm is also proved.

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High-order harmonic generation (HHG) is a well-studied phenomenon and, even if some of its aspects are still to be completely clarified, it seems mature enough to become a powerful tool for applications in the short-wavelength region [1]. The peculiar characteristic of the spectrum of harmonics generated from the interaction of short and intense laser pulses with the atoms of a gas jet, is the presence of a so-called *plateau*, a region where several successive orders of harmonics share an almost constant intensity despite the predictions of perturbative models. This region is terminated by an abrupt *cutoff*, where the intensities of harmonics start to drop exponentially. The extension of the plateau depends on the intensity of the pump pulses, on their wavelength, and on the ionization potential of the atom; by choosing the appropriate parameters, the entire region of the extreme ultraviolet (XUV) can be covered.

The use of extremely short pump laser pulses (less than 30 fs long) has recently extended the available range of accessible wavelengths to the soft X-rays, reaching the so-called *water window* with spectrally unresolved harmonics up to the 300th order [2]. On the other hand, simpler commercial laser systems operating at high repetition rates (generally 1 kHz), are now widely available in many laboratories around the world and, with their typical millijoule-level energy in 100-fs pulses, are already capable of efficiently generating high-order harmonics in the XUV [3]. Spectroscopic [4] and interferometric [5] applications of these new sources have already appeared and some schemes have been proposed to perform high-resolution spectroscopy in the XUV, overcoming the intrinsic broad bandwidth connected with the extremely short duration of the harmonic pulses [6].

The generation process has already been studied under very different experimental conditions and the dependence of the harmonic yield as a function of the many parameters involved has been investigated in several laboratories. High-order harmonic pulses maintain many of the characteristics of the driving laser pulses: they keep their usually short duration, the same high directionality, and a similar degree of temporal [7] and spatial [8] coherence. Moreover, if the pump laser is tunable, also the emitted harmonics are tunable in the XUV. Though this is certainly true in principle, it is not so readily feasible in practice and limits the applicability of these new sources to the few cases where a desired resonance falls close to an odd harmonic of the pump laser frequency. Even if a limited tunability has been observed thanks to a proper adjustment of the laser-jet interaction parameters [9], the availability of a tunable pump source remains the most desirable option to achieve a good spectral coverage with the emitted harmonics.

Of the high-peak-power laser systems generally used for HHG, only those based on Ti:sapphire allow a relatively broad tunability around 800 nm, but changing the emission wavelength often requires a complete realignment of the oscillator-stretcher-amplifier-compressor system. To overcome this problem some schemes have been proposed and realized [10, 11] which consist of mixing the radiation from a high-power but fixed-frequency laser with that of lower power but tunable source, usually an optical parametric generator (OPG). Due to the much lower energies of the tunable source, only one *tunable* photon of frequency ω_t usually takes part to the process, so that the final harmonic frequency is $n\omega_p + \omega_t$, where ω_p is the frequency of the strong laser and n is an even integer. The pump beam for the parametric generator is usually a frequency-doubled part of the same high-power laser, meaning that one has to start from high-energy pulses (several tens or hundreds of mJ), and only low-repetition-rate (10 Hz) systems have been used so far. Moreover, the mixing process requires a perfect spatial and temporal superposition of two beams of different wavelength in the gas jet and one has to pay particular care in the development of nontrivial optical schemes. Finally, if some special spectroscopic technique (such as the one proposed in [6], relying on a Michelson interferometer for the production of pairs of time-delayed XUV pulses to achieve a better spectral resolution) is to be adopted, then a single-color, tunable pump pulse would be definitely preferred.

In this paper we report the production of tunable ultraviolet radiation from the direct high-order harmonic conversion of the pulses from an optical parametric amplifier. We generate up to the ninth harmonic of the signal output of a three-pass OPA, tunable from about 1100 to 1600 nm. The OPA is pumped by 0.7-mJ, 100-fs pulses from a Ti:sapphire laser system operating at 800 nm and at a repetition rate of 1 kHz. The usable energy of the signal pulses in the region of interaction with the gas jet is only about 40 μJ but it is nevertheless sufficient for harmonic generation down to about 150 nm.

Other schemes have already accessed the spectral region considered in the present paper. These techniques include cascaded second-order frequency mixing in solid-state nonlinear optical materials [12] (however, limited to about 170 nm due to absorption and phase-matching constraints), resonant and near-resonant four-wave difference-frequency mixing in rare gases [13, 14], or frequency tripling in hollow fibers [15]. Such schemes may achieve higher conversion rates than those reported here, but it is important to note that the main motivation for our measurements was not the development of a new, competitive source in the UV-VUV, but rather the demonstration that HHG can become a completely tunable source, with the potential to be extended down to the XUV spectral region without major conceptual changes and relying on a relatively simple experimental setup.

As far as we know, this is the first report of HHG in a gas jet to such an order from a relatively low intensity, tunable, and high-repetition-rate source. The third and fifth harmonics of the signal output of an OPA had been observed in [10] but, in that case, about one order of magnitude higher pump pulse energies were necessary and the repetition rate of the system was only 10 Hz.

1 Experimental

Our pump source is an amplified Ti:sapphire laser system (BMI Alpha 1000) presenting a smooth doughnut-like mode in the far-field profile. The NIR parametric generator-amplifier (TOPAS) is based on three passes in a 4-mm-thick BBO type-II crystal and it has already been described in detail elsewhere [16]. The signal plus idler pulse energy at the exit of the OPA is approximately 150 μJ but the available energy in the signal pulse after some filtering and after the steering optics used to deliver a focused beam into the gas jet is only about 30–40 μJ. The duration of the NIR pulses has been measured to range between 100 and 150 fs across the tuning limits.

A telescope has been used to expand the original beam diameter of about 2 mm to diameters ranging from 5 to 10 mm, in order to obtain the smallest spot in the focal region. A 150-mm lens has been used for focusing the NIR pulses under the nozzle of a pulsed valve, injecting xenon atoms at a backing pressure of 1.5 bar into the interaction chamber. The interaction region is placed at the entrance slit position of a vacuum 1-m (ARC) monochromator equipped with a 600 lines/mm concave grating; the spectrally dispersed harmonics are detected after the exit slit by a phosphor screen–photomultiplier combination as shown in Fig. 1.

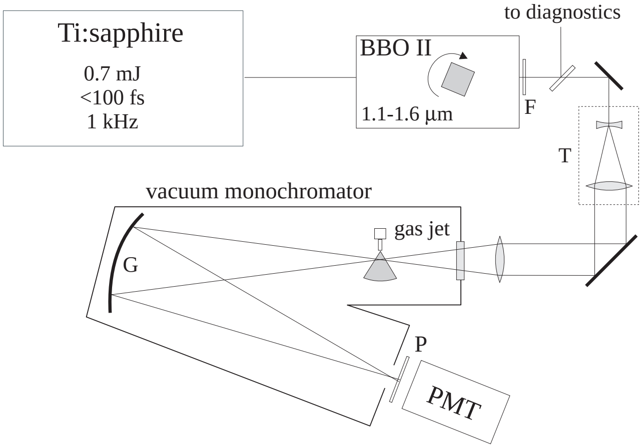

Fig. 1. Experimental setup. F is a filter to select the signal wavelengths only; T is a telescope to expand the IR beam; G is a 600 lines/mm concave grating and P is a phosphor screen that converts the VUV photons into visible photons detected by the photomultiplier PMT

2 Results and discussion

A calibration of the number of photons per pulse reaching the exit slit has been carried out by comparing the PMT signal corresponding to the third-harmonic pulses (generated by focusing the 800-nm pump beam in air) with the reading of a power-meter measuring their absolute mean power. The UV-visible conversion efficiency of the phosphor screen at the different wavelengths has also been taken into account.

The maximum parametric conversion efficiency is between 1300 and 1400 nm and the tuning curve rapidly drops for wavelengths approaching 1100 nm, whereas it decreases more gently towards the degeneracy point at 1600 nm (see Fig. 2). Accordingly, the peak harmonic yield has been ob-

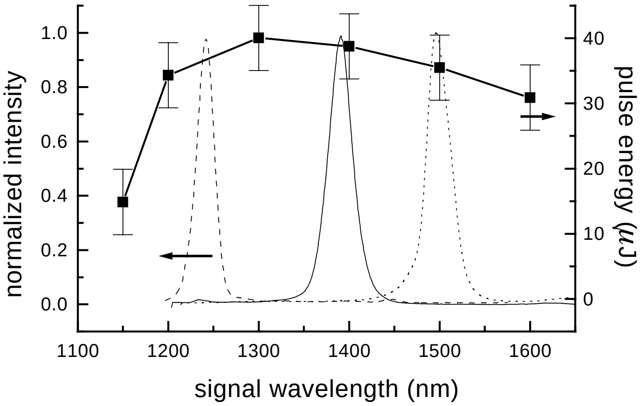

Fig. 2. Parametric conversion efficiency for different wavelengths of the signal pulse. The scale on the right indicates the energy of the pulses in the interaction region. The normalized spectra recorded at three representative wavelengths are also shown

served for pump wavelengths around 1350 nm and in these cases, up to the ninth order at about 150 nm has been efficiently produced with approximately 10^6 photons per pulse. A similar efficiency has been measured for the seventh harmonic in the full 1200–1550 nm range. The fifth and third orders present one and two orders of magnitude more photons, respectively, reaching a maximum of about 10^9 photons per pulse for the third harmonic of 1350 nm. The harmonic emission at three different and representative pump wavelengths is plotted in Fig. 3.

The conversion efficiency drops exponentially as the order increases and the 10:1 ratio of the intensities for harmonic pulses of the same order but different wavelengths (corresponding to slightly different intensities of the pump pulses from the OPA) is the evidence of the high nonlinearity of the process.

The measured spectral widths of the UV and VUV pulses range from about 14 nm for the third-harmonic pulses to about 4 nm for the ninth harmonic at 150 nm. We estimate the pulse durations to scale approximately as the inverse of the square root of the corresponding harmonic order and to range from about 50 to 100 fs, at least for the relatively low orders considered here.

Xenon has been chosen for harmonic generation from the OPA in order to maximize the UV photon yield. Lighter rare gases such as Ne or He are generally used to increase the extension of the plateau towards shorter wavelengths thanks to their higher ionization potential, but their lower polarizability also dramatically reduces the conversion efficiency. Though a static cell could have been used for harmonic generation at the wavelengths attained in the present experiment, we chose to use a gas jet in order to directly compare to the conditions normally met in HHG experiments. In view of the extension to shorter wavelengths such a choice is the only one available. As already mentioned before, it was not the purpose of these experiments to achieve the highest efficiency of generation in the UV-VUV regions, but rather to demonstrate that HHG, a technique that has already proved a very promising one to generate radiation of extremely short wavelength, can be made even more attractive thanks to the full spectral tunability provided by a tunable pump.

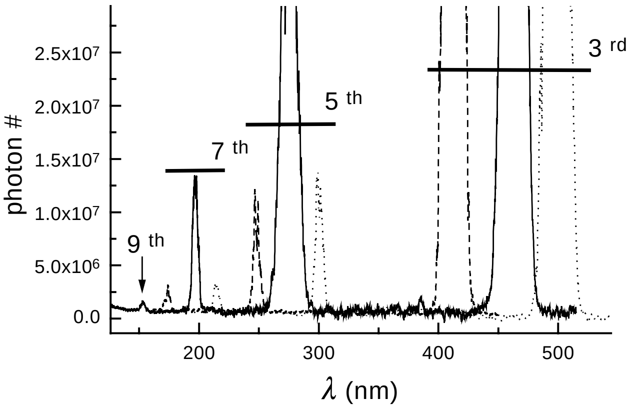

Fig. 3. Spectra of the harmonic orders generated with the OPA signal pulses at three representative wavelengths (the same shown in Fig. 2). The *solid curve* is generated with the signal at 1380 nm, the *dashed curve* is at 1240 nm, and the *dotted* one at 1496 nm

The exponential decay of the photon yield as a function of the harmonic order indicates that, with the pulse intensities available with our experimental setup, we are not able to reach the plateau region, characteristic of the high-order harmonic-generation process. Apart from the low energy at the output of the OPA, which might be substantially increased only increasing the energy of the 800-nm laser pulses, we believe that the main limitation is connected with the poor spatial quality of the beam. The non-Gaussian shape of the far-field profile of the Ti:sapphire laser is responsible for an inhomogeneous distribution at the output of the parametric amplifier. The signal–idler beams are thus far from the diffraction limit and we estimate that the waist radius is close to 100 μm, with corresponding peak intensities in the focus of less than 10^{12} W/cm^2. Note that a perfect diffraction-limited beam of the same wavelength and with a diameter of about 5 mm would focus to a waist radius of about 27 μm and to peak intensities of the order of 10^{13} W/cm^2 after a 150-mm-focal-length lens. Changing the telescope setup to different beam diameters has not improved the situation, indicating that the poor beam quality is the intrinsic limit to higher efficiencies. According to the well-established cutoff law [17], affirming that the maximum energy of the harmonic photons at the end of the plateau is approximately given by $I_p + 3U_p$, where I_p is the ionization potential of the atomic species and U_p is the so-called ponderomotive energy (proportional to the laser intensity and to the square of its wavelength), we should not expect much higher harmonic orders unless we can substantially increase the energy of the pulses or focus the same pulse energy into a diffraction-limited focal spot.

Also note that, even if we could reach the same peak intensities (of the order of 10^{15} W/cm^2) as those attainable with the 800-nm laser pulses, we should still expect a lower conversion efficiency (but in a substantially extended plateau) in this case, due to the longer wavelength of the pump [18]. An intuitive explanation for this can be attained by considering the semi-classical model for harmonic generation [17]. Here one electron of the rare-gas atom is first tunnel-ionized by the field of the laser pulse, it is then accelerated in the continuum by the same laser field and finally driven back to the ion core where it recombines to emit harmonic photons. The time the electron spends in the continuum is proportional to the wavelength and, as the electron is accelerated, its wavefunction spreads transversely. This means that for long wavelengths the wavefunction will be more spread out when it passes the core. Consequently, the recombination probability decreases and the harmonic yield becomes lower. One natural improvement to the present setup could then be the use of the recently demonstrated blue-pumped, non-collinearly phase-matched OPAs that are able to produce tunable visible pulses with sub-20-fs durations [19–21]. Even in the case of equivalent peak intensity, such sources should greatly improve the conversion efficiency thanks to the shorter wavelength used to generate harmonics and to the lower order of conversion needed to reach the same wavelength in the VUV.

Though at the moment the reported efficiencies are probably not enough for practical applications of this tunable source, it is nevertheless very interesting to note that, even with the limited 1200–1550 nm range, a complete tunability in the VUV can be obtained already from the seventh harmonic, that is for all the wavelengths below 220 nm. If the range is extended to the nominal 1100–1600 nm region, the

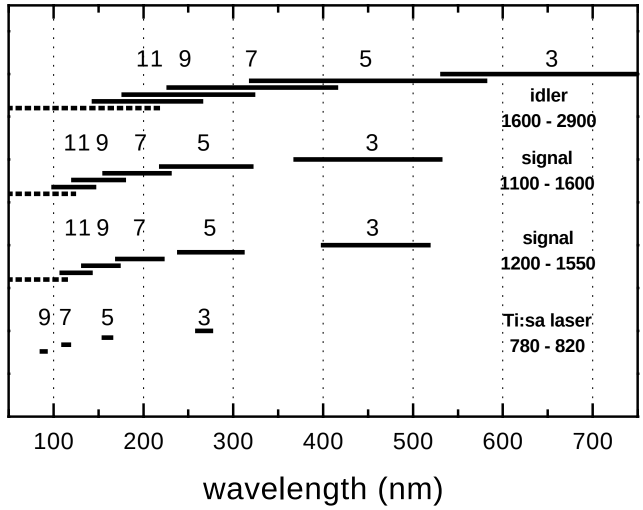

Fig. 4. Tunability ranges of the generated harmonics for different pump options. The tunability of the amplified Ti:sapphire pulses only permits a sparse coverage of the VUV range. Using the signal pulse from the OPA gives a complete tunability already below 320 nm if harmonics are generated in the full 1100–1600 nm interval. If one succeeds in generating harmonics from the idler pulse, the full spectrum, from the visible to the VUV, may be continuously covered

overlap of different orders takes place already from the fifth harmonic, or below 320 nm; for longer wavelengths, a continuous tuning may then be obtained with a more conventional fourth-harmonic generation in crystals. In Fig. 4 we show the tunability ranges of the harmonic pulses for different options of the pump source. For comparison purposes, we also indicate the wavelengths accessible by simply tuning the pump Ti:sapphire laser system over the 780–820 nm range.

The above considerations only refer to the signal pulse coming out of the OPA; if one succeeds in increasing the peak intensities, also the idler pulse (with approximately the same number of photons, but lower intensity) may start to produce harmonics. In such a case, considering its nominal 1600–2900 nm tuning range, the full tunability of the harmonics is obtained already from the third order, that is below 970 nm.

3 Conclusion

We have experimentally shown for the first time that high-order harmonic generation can be directly achieved with an intrinsically tunable pump source, such as a NIR optical parametric generator pumped at high repetition rate. We have also demonstrated the possibility of a complete coverage of the VUV region thanks to the full overlap of the tunability ranges of different harmonic orders already below 220 nm. Though the efficiencies are at the moment rather low and only medium-order harmonics have been generated, we believe that further improvements in the energy and in the spatial quality of the pump pulses may lead to the extension of the spectral coverage to the XUV and to the realization of a useful tunable source for spectroscopy in this region.

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