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Study of few-optical-cycles generation of high-order harmonics

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Abstract

The high-order harmonic generation process was studied using laser pump pulses of different duration, down to 7 fs. The time profile of this kind of pulse corresponds to a few optical cycles at the fundamental frequency, and therefore a cycle-to-cycle variation of the maximum amplitude occurs. These facts violate the adiabatic approximation of the generation process, giving rise to new aspects of the physical process. The comparison of results with pulses in the two regimes are reported and discussed. The modeling of the complete generation and propagation process in three dimensions is also presented.

1. INTRODUCTION

The generation of high-order laser harmonics using gas targets provides a unique source of high brightness, coherent extreme ultraviolet radiation, due to its table-top size and very short pulse duration (Corkum, 1993; L'Huillier & Balcou, 1993; Chang *et al.*, 1997; Protopapas *et al.*, 1997). The introduction of high-peak power few-optical-cycle laser pulses (Nisoli *et al.*, 1996, 1997) has extended the scenario of the harmonic generation process, giving rise to a number of open questions to be addressed both from an experimental and a theoretical point of view (Schnürer *et al.*, 1998; Brabec & Krausz, 2000).

In this work, we report on the generation of harmonic radiation in noble gases by using laser pulses of a duration of 35 fs down to 7 fs. The target of the work is the investigation of the influence of pulse duration on the spectral properties of harmonic radiation (viz. the shape of the harmonic spectra and the linewidths of the harmonic peaks) in the multi-optical-cycle and in the few-optical-cycle temporal regime of the driving pulse.

We also present a three-dimensional numerical model, which takes into account nonadiabatic effects, typical of the few-optical-cycle regime.

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2. EXPERIMENTAL SETUP: LASER SOURCE, INTERACTION CHAMBER, AND SPECTROMETER

As a pump source we used a Ti:sapphire laser system with chirped-pulse amplification, based on a nine-pass confocal amplifier stage and a prism compressor. This system generates a 25 fs laser pulse, with up to 0.8 mJ total energy. Self-phase modulation in argon-filled hollow-core fiber (Nisoli *et al.*, 1996, 1997) is exploited to broaden the pulse spectrum, resulting in an output beam of about 780 nm central wavelength and 400 nm bandwidth. The hollow fiber is 60 cm long and has an inner diameter of 500 μm. Then, the beam undergoes six bounces onto the chirped mirrors, which compensate for the chirp introduced by the hollow fiber and precompensate the dispersion originating from the material between the compressor and the gas jet. After the compression, the linearly polarized pulse has a duration of 7 fs and an energy up to 350 μJ.

The laser pulse is focused into the interaction chamber by a concave mirror ($f = 250$ mm) through an 0.5-mm-thick fused silica window. For a target we used a gas jet, pulsed into the chamber by an electromagnetic valve, producing a jet whose diameter was 0.8 mm and operating with a valve opening time of 400 μs. As target elements, several gases among the noble gases were used. The gas pressure in the interaction region was estimated to be 50 mbar. The esti-

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mated laser peak intensity in the interaction region was about 8×10^{14} W/cm^2.

Harmonic radiation was measured by means of different systems: the first for the investigation on the harmonics beam properties and for the single-shot studies, and the second for the high-resolution study of the harmonics spectral peaks.

The first is a grazing-incidence spectrometer using a varied line spacing grating in order to produce an erected focal image, that is, acquired by a bidimensional detector (Poletto *et al.*, 2001*a*, 2001*b*). The optical setup adopts a toroidal mirror as first optics, to provide at the tangential focusing on the entrance slit of the spectrometer. On the sagittal plane, this mirror is designed to project the far-field distribution of the harmonics beam onto the focal plane. The parameter chosen for these mirrors are: incidence angle 86° and radii of 6500 mm in the tangential plane and 14.7 mm in the sagittal plane. The grating is mounted at 87° of incidence, has an average groove density of 1200 gr/mm and a radius of 5649 mm. A reproduction of the optical setup in the tangential as well as in the sagittal plane is presented in Figure 1.

The second setup is a grazing-incidence (86°), Rowland mounting monochromator, based on a platinum-coated, 300-grooves/mm, spherical grating (2 m radius of curvature),

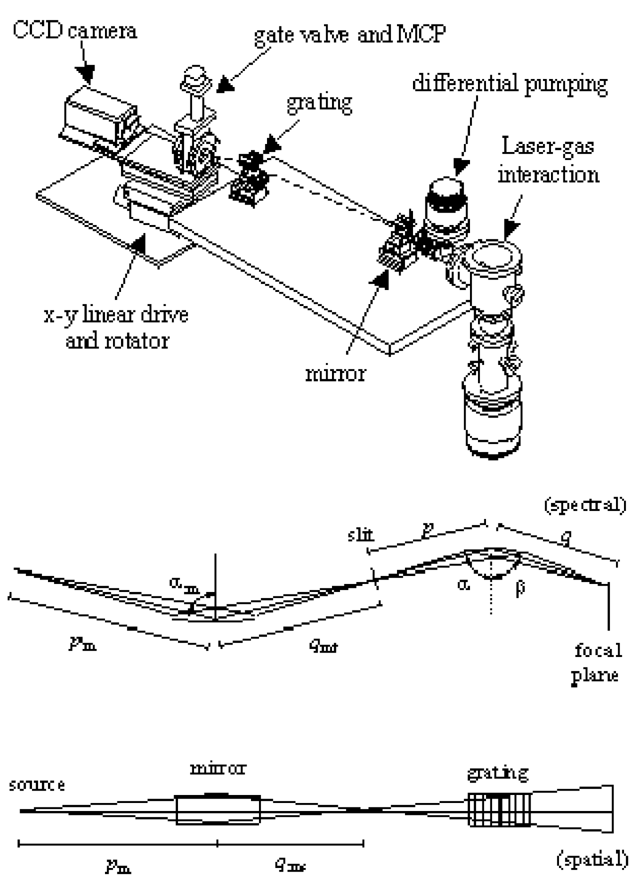

Fig. 1. Scheme of the experimental setup (upper). The representation of the rays in the spectral and sagittal or spatial plane (bottom).

designed for broadband efficiency in the 50–5 nm spectral range. Again, a toroidal mirror (incidence angle 83.5°, and radii of 2.8 m in the tangential plane and 60 mm in the sagittal plane) focuses the harmonic beam onto the monochromator entrance slit in the tangential plane. It also provides focusing in the sagittal plane in a position at the center of the diffracted spectrum to maximize the collected flux. In fact, by correcting the mounting astigmatism, the two-component optical instrument achieves high sensitivity and high spectral resolution (500–1500). Finally, the detector is a channel electron multiplier with bare glass photocathode, with variable gain up to the photon counting regime.

3. SPECTRAL AND SPATIAL OBSERVATIONS

The acquired spectra shown resolved harmonic peaks at photon energies up to about 130 eV and 160 eV using 30 fs and 7 fs, respectively. The partial overlapping of the harmonic tails gives rise to a broad pedestal for short wavelengths.

A focal-plane intensity profile taken by the flat-field spectrometer and corresponding to harmonics generated in helium starting from order 29 is shown in Figure 2. The laser pulse duration in this case was of 7 fs. The major evidence is the broadening of the spatial profile as harmonic order increases.

In the cutoff region, the divergence of the harmonics was observed to decrease, as does the integral of the line. Moreover, the spatial profiles are bell shaped for all the acquired spectra, as a result of the present conditions of generation.

Taking advantage of the well-resolved harmonic spectra even the in case of sub-10-fs driving pulses, we have analyzed the wavelength dependence of the harmonic linewidths in the case of different duration of the excitation pulses.

All the parameters of the harmonics' spectral profiles, such as central λ, integral, width, and asymmetry, depend on the geometry of the laser-gas interaction as well as on the pump intensity and spectrum. The evidence of this can be found in many studies of the spectral shift due to ionization (Miyazaki & Takada, 1995; Zhou *et al.*, 1996; Altucci *et al.*, 2000; Nisoli *et al.*, 2000), or the predictions on the harmonic intensity taking into account the propagation in the medium (Salières *et al.*, 1998). To enlighten the role of the pump duration, we observed the spectrum of the harmonics emission generated by a laser pulse of various durations, from 7 to 35 fs.

A remarkable broadening of the spectral linewidths was observed when decreasing the pump duration. This is shown in Figure 3, where the comparison of the total width at 1/e, γ, versus the harmonics wavelength of representative spectra for pump duration of 7, 20, and 35 fs is reported. The spectra are selected for the highest intensity by varying the interaction parameters. The estimate of the linewidth has been carried out by fitting the spectral distribution of a single harmonic with an analytical profile. The contribution

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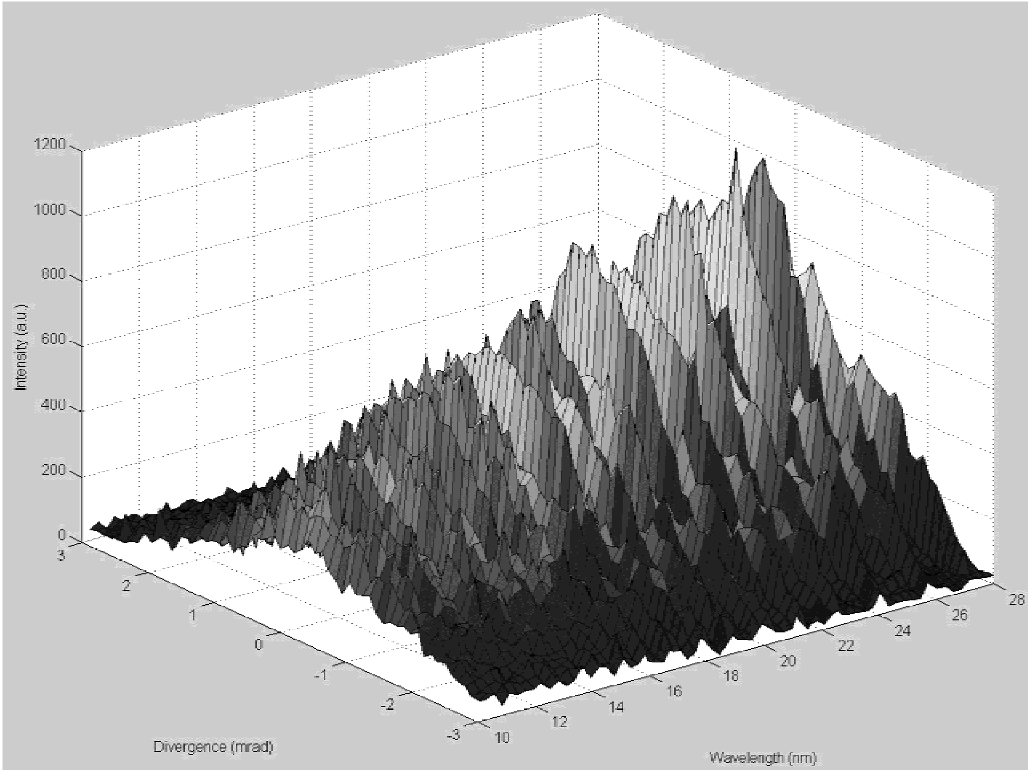

Fig. 2. Harmonics spectrum in helium, using 7 fs pulse. The spatial scaled is converted in beam divergence by using a ray-tracing model of the spectrograph.

of the instrumental function was then quadratically subtracted. This latter was estimated by means of the ray tracing of the optical setup in the operative conditions.

The experimental discrete spectrum for various pump-pulse durations was fitted by using a power law for the qth harmonic linewidth versus the harmonic wavelength as $\gamma_q = C\lambda_q^{\alpha_\tau}$. This choice has been made because the limitation in the linewidth is posed by the time-bandwidth inequality (Akhmanov *et al.*, 1992). By considering the fundamental harmonic with duration τ_1 and bandwidth γ_1, the width of the harmonics of order q that gives $\gamma_q = \gamma_1$ has a corresponding value of the exponent $\alpha_\tau = 2$. For a scaling with smaller α_τ the harmonics' profiles tend to merge for high q. On the other hand, for $\alpha_\tau > 2$, the profiles of lower harmonics have a larger value of the ratio of width on their separation than the higher ones.

The fit of the exponent for three different durations was obtained by using the least squares method and is reported in Figure 3, together with the experimental points. The resulting values for α_τ are 1.8, 2.5, and 3.3 for pump pulses of duration of $\tau = 35$, 20 and 7 fs, respectively. The value of α_{20fs} and α_{7fs}, being greater than 2, are representing the discreteness of the cutoff. For the 35-fs spectrum, the width of the low order harmonics is relatively small, and then the spectrum does not eventually merge. The different results found here are related to the generation process, which is in distinct regimes for the 35-fs case, where adiabatic approximation can be still considered reasonable, to the 7 fs case, which is nonadiabatic. Another factor which comes into play is the chirp of the pump pulse, which was found to be relevant for the harmonics spectra (Chang *et al.*, 1998). In the present experiment, we are prosecuting the investigations in order to complete the understanding of this aspect.

The linewidth curves for different durations in the plot are remarkably distinct and converge toward a value smaller than the instrumental function. This limit also expresses a lower bound for the time duration of the harmonics in the cutoff. Indeed, the minimal duration of a Gaussian profile can be obtained from its spectral linewidth, and it corresponds to the transform-limited case (Rullière, 1998). For the 7- and 35-fs pumps, the estimated values in the cutoff are 1.8 and 5.4 fs, respectively, while the duration of the pump-laser optical cycle is of 2.6 fs. The observed line profile results from the integration of emission along the interaction medium, where its spectrum can be modified. Nevertheless, the value measured for 7 fs is in agreement with the prediction of Lewenstein et al. for the single-atom emission (Lewenstein *et al.*, 1994). According to these authors, in the cutoff region, the generation involves a single electron trajectory, driven by the laser field for a fraction of $4.08/2\pi$ of the optical cycle. In the present condition, this value is 1.68 fs, almost identical to our limit. On the other hand, the 7-fs result was obtained with a pump pulse which does not have a transform-limited spectral distribution that can be reflected on some

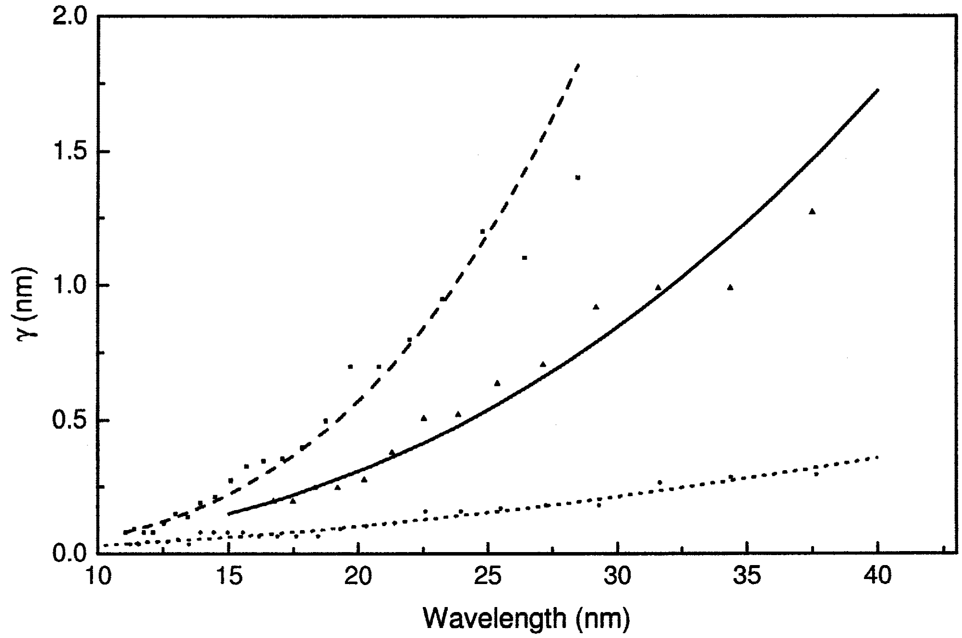

Fig. 3. Spectral linewidth in the case of different pulse durations. The curves are the fit for the three durations, dashed (7 fs), continuous (20 fs), dotted (35 fs).

modulation present in the harmonics (Chang *et al.*, 1998). Moreover, the linewidth that may be associated to the single-atom emission is modified by the propagation as it happens for other characteristics as the beam divergence and total yield (Lewenstein *et al.*, 1995; Salières *et al.*, 1995).

4. 3D MODELING OF THE GENERATION PROCESS

The experimental results have been modeled, combining the different processes involved (Corkum, 1993): the interaction of the laser with the target atom, the atom ionization, the evolution of the electron in the laser field in a Volkov state and the generation of the harmonics radiation; this latter has been propagated as a three-dimensional scalar solution of the Maxwell equations. The model takes into account the nonadiabatic effects originating from the cycle-to-cycle field amplitude variation (Priori *et al.*, 2000; Villoresi *et al.*, 2000).

We find that the emitted spectrum from single-atom, pumped by a few-cycles pulse, appears with no recognizable harmonics peaks. On the other hand, by considering the harmonics field after the propagation, the spectrum shows a discrete structure, as derived from a selection of the relevant frequencies. This remarkable change is attributed to phase mismatch due to longitudinal and radial variations of both intensity and phase of the driving pulses; we found that these effects are correctly described only in a three-dimensional geometry of the model.

The numerical model has been used to reproduce the spectra for the different experimental conditions as well as target gases used in the experiment. The comparison of the harmonic spectrum in neon with 7 fs between experiment and model is presented in Figure 4. The cutoff position was well reproduced as well as the line profile of the harmonic peaks. The physical result which appears from this analysis is that in the present geometry, the sub-10-fs harmonics are emitted with a discrete spectrum. This result is likely due to the relatively long interaction length of the laser with the pulsed gas jet. A detailed comparison of the spectral width was also performed for other pulse duration, finding a good agreement (Priori *et al.*, 2000).

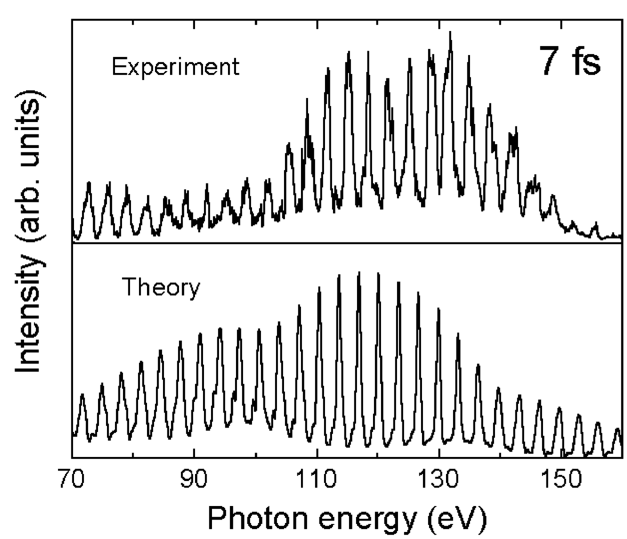

Fig. 4. Comparison between experiment (upper) and model (lower) in the spectra generated in neon by a 7-fs pulse.

5. CONCLUSIONS

The generation of high-order harmonics using a few-optical-cycles laser pulse has been reported. Remarkably different results have been found for both the spectral extension and the linewidth of the harmonic peaks. The model which include the nonadiabatic interaction of the laser field with the atoms and a three-dimensional propagation of both the harmonic and the fundamental beams in the gas have resulted in a successful description of the experimental measurements.

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