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Temporal confinement of the harmonic emission through polarization gating

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Abstract. We show experimentally that a simple optical setup is able to reduce the duration of the harmonic emission generated by "long" (many optical cycles) laser pulses. By controlling the time-dependent ellipticity during the laser pulse, one can confine the harmonic emission to a narrow temporal window where the polarization is quasi-linear. The results obtained and their comparison with a simple model show a shortening of a factor 2 for harmonics located in the plateau region. Using shorter laser pulses of 15–20 fs duration should make it possible to isolate a single attosecond pulse.

PACS. 42.65.Ky Frequency conversion; harmonic generation, including higher-order harmonic generation – 32.80.Wr Other multiphoton processes

1 Introduction

High order harmonic generation has led recently to the first experimental demonstration of attosecond pulses [1,2]. In [1], the authors showed that the harmonic emission, under appropriate experimental conditions, takes the form of a train of attosecond pulses, with peaks (250 as) separated by half the laser period (1.35 fs in the case of a Ti:sapphire laser). This temporal structure can be understood easily within the framework of the semi-classical model [3,4], that describes the harmonic emission as a three-step process: tunneling ionization in the intense laser field, then acceleration of the freed electron and finally radiative recombination with the parent ion. Attosecond bursts are emitted at each electron-ion re-collision, with a periodicity given by half the laser period. Phase matching in the macroscopic medium helps in selecting a single burst per half cycle [5,6].

Different methods have been proposed to generate a single attosecond pulse instead of the full train. One uses the fast intensity dependence of the harmonic efficiency in the cutoff region. The generation by an ultrashort (7 fs) laser pulse has led to an isolated 650-attosecond pulse [2]. These pulses, however, have very low energy for two main reasons: first, the short laser pulses needed are obtained by hollow fiber post-compression [7], which limits their energy to less than 1 mJ. Second, the technique requires to select only the highest harmonics generated in light rare gases, for which the efficiency is low. An alternative method, based on the strong ellipticity dependence of the harmonic efficiency, would allow the generation of intense attosecond pulses. When the polarization of the laser is not linear, the re-collision probability of the electron with its parent ion, and thus the intensity of the harmonic emission, decrease rapidly with the ellipticity ϵ of the laser pulse. Experimental and theoretical studies have confirmed this fast decrease [8,9]. Using a laser pulse whose degree of ellipticity varies sufficiently rapidly in time on the scale of the laser period, would give the possibility to prevent all re-collisions except for one, thus resulting in a single attosecond burst [10]. A first attempt in this direction was performed in Lund [11], where a temporal gate was obtained by time delaying two chirped, orthogonally-polarized laser pulses. More recently, such a gate was obtained by superposing two orthogonally-polarized laser pulses with slightly different carrier frequencies [12]. However these methods do not allow a fine control of the duration of the gate. Another technique for realizing the polarization gating was independently proposed by Constant [13] and by Platonenko and Strelkov [14], that allows to vary in a continuous way the duration of the temporal gate. In theory this may lead to sub-femtosecond values of the harmonic pulse duration. A first implementation of this technique using fixed delays has recently been made in Bordeaux [15]. In this article, we present a proof of principle experiment where a simple

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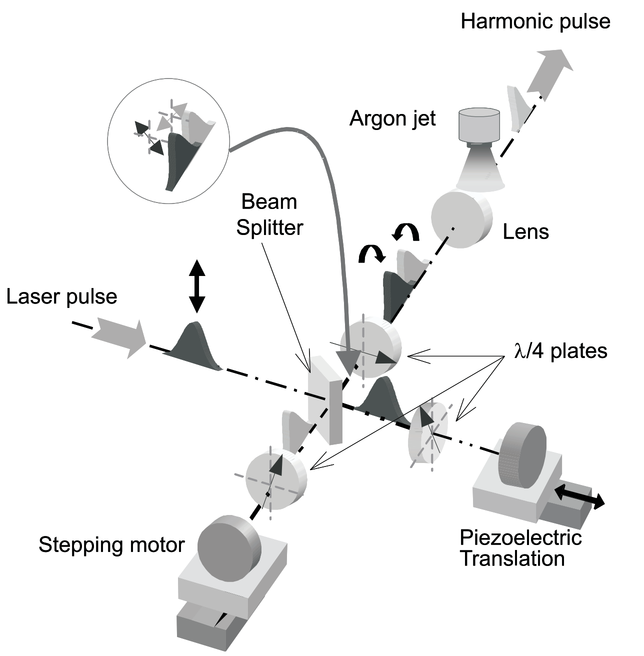

Fig. 1. Experimental setup used to generate the polarization gate. An intense IR laser pulse is split into two time-delayed, counter-circularly-polarized pulses, that are focused into an argon jet.

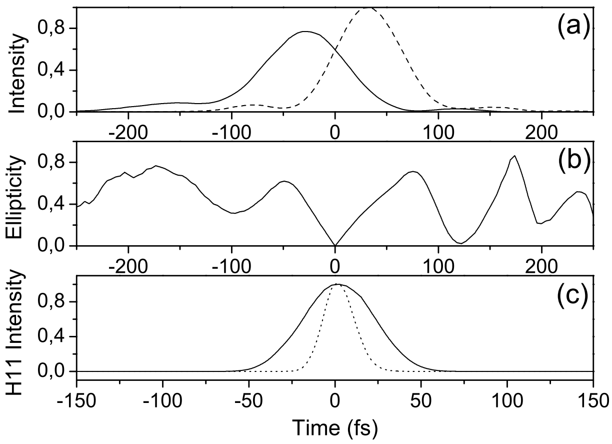

Fig. 2. (a) Temporal profile of the two laser pulses exiting the Michelson. (b) Absolute value of the ellipticity as a function of time, after the third QWP for a 60-fs delay. (c) Simulated temporal profile of H11 for a zero delay (solid line) and a 60-fs delay (dashed line).

optical scheme gives a time varying polarization resulting in a temporal confinement of the harmonic emission.

2 Experimental setup

The experimental setup is shown in Figure 1. It is based on a Michelson interferometer that produces two replicas of the input laser pulse with a delay τ adjustable with a high precision thanks to a stepping motor and a piezoelectric translation. In each arm of the interferometer is placed a zero-order quarter-wave plate (QWP): being crossed twice, the QWPs act like half-wave plates (HWPs) and they turn the polarization direction of the incoming radiation of $\simeq \pm\pi/4$. The QWPs are oriented in such a way that the two delayed pulses exit the Michelson with orthogonal linear polarizations. After the interferometer, the laser pulses cross a final third QWP that changes the linear polarizations into right and left circular polarizations respectively. The superposition of the two time-delayed, circularly-polarized pulses results in a polarization that varies from circular to linear and back to circular, thus generating a gate whose duration depends on the delay τ. Note that the polarization gate is determined here by the variation of the relative amplitude of the laser pulses, in contrast to the method used in [11] that was relying on the variation of their relative phase.

We have characterized the two laser pulses exiting the interferometer with SPIDER measurements. Their temporal profiles are shown in Figure 2a. The initial laser pulse had a duration of 50 fs, but propagation through the QWPs and the beam splitter of the interferometer broadened the pulses to about 80 fs. The two pulses were not strictly identical because of the lack of a compensator plate. With the additional information of their temporal phase, we can construct the temporal evolution of the ellipticity after the third QWP for a given delay. Its absolute value is shown in Figure 2b for a 60 fs delay. A gate of nearly linear polarization is opened when the two pulses have similar intensity. Note that another gate appears around 120 fs due to the presence of post-pulses. But the very low intensity ensures that no harmonic emission is produced there.

After the third QWP, the laser beam is focused at $f\#52$ by a 750 mm lens into a 50 torr argon jet. In order to minimize effects such as harmonic chirp, ionization-induced blue-shift and laser defocusing, that can make the interpretation of the results very difficult, we limit the peak intensity at zero delay to the rather low value of $\sim 1 \times 10^{14}$ W/cm^2. The harmonic radiation is analyzed by a plane-grating spectrometer, composed of a toroidal mirror and a grazing incidence grating, and detected with a photo-multiplier.

3 Experimental results

The first measurements consisted in recording the harmonic signal as a function of the applied delay. The solid line in Figure 3 shows the case of the 11th harmonic (H11). At zero delay, the polarization is always linear, and should correspond to the maximum conversion efficiency. This is not the case in our measurement, probably because of intensity fluctuations in the laser beam, rather than an anomalous ellipticity dependence like that reported in [16]. At large delays, the signal decreases rapidly down to zero, showing pronounced oscillations. The latter are related to the orientation of the polarization: when the delay τ varies by half an optical cycle $T/2$, the polarization plane of the laser light turns of 90 degrees at the center of the gate

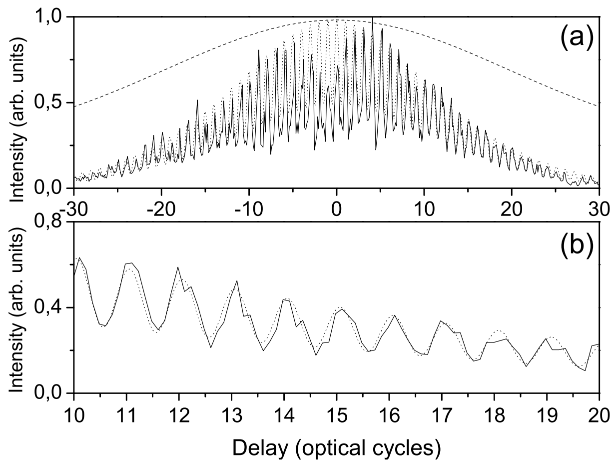

Fig. 3. (a) Variation of the harmonic signal (H11) as a function of the delay. The solid line shows the experimental trace and the short-dashed line represents the simulated curve. The long-dashed line represents the simulated intensity envelope when $\beta_q = 0$. (b) Enlargement of (a) showing the good agreement between the experimental trace and the simulated curve.

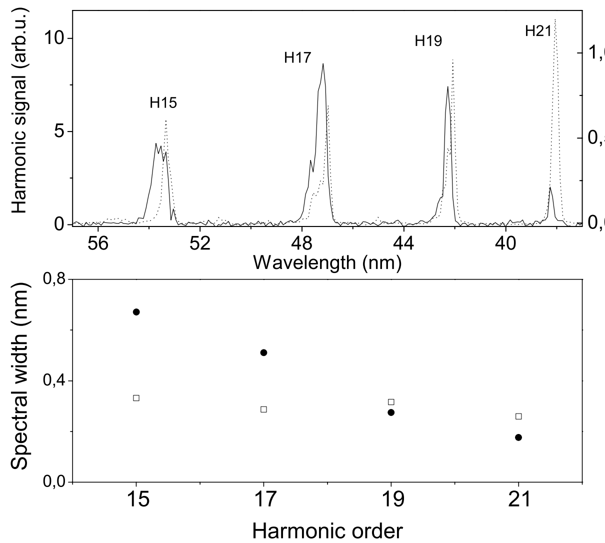

Fig. 4. (a) Experimental harmonic spectra obtained at zero delay (dashed) and 60 fs delay (solid). (b) Spectral widths at zero delay (open squares) and 60 fs delay (full circles).

(when $\epsilon = 0$) and so does the polarization plane of the harmonics. The transmission efficiency of the spectrometer is polarization-dependent (about a factor 2 difference between s and p polarizations), therefore the measured signal oscillates with period T. The decrease of the harmonic signal is the result of two effects: as the delay increases, the polarization gate becomes narrower and the laser intensity gets smaller, both resulting in a weaker generation efficiency. To determine the dominant effect, we measured the same trace as in Figure 3 but with the neutral axes of the third QWP aligned along the polarizations of the pulses exiting the Michelson. The polarization of the total pulse thus remains linear whatever the delay and we can quantify the influence of the decreased laser intensity. For a delay of 30 optical cycles, the harmonic signal is decreased by only a factor 2. Consequently, the fast decrease observed in Figure 3 is mainly the result of the polarization gating.

A signature of the shortening of the harmonic pulse could be found also in the broadening of its spectrum. However this broadening is difficult to observe owing to the fact that the harmonic pulses are not at the Fourier transform limit, but present a negative "intrinsic" chirp [17]. This chirp results in a spectral broadening, all the more important as the laser intensity is high. When the temporal gate is narrowed at large delays, the laser intensity during the harmonic generation is decreased, and so is the spectral broadening. This process can hide completely the desired effect, and even produce the opposite result of a narrowing of the spectrum at large delays, as observed in [11]. Moreover the interplay between the laser chirp and the intrinsic harmonic chirp may complicate significantly the interpretation of the results [18]. In order to minimize the influence of the intrinsic chirp, we used a low laser intensity, a heavy rare gas as generating medium (argon) and we studied low harmonic orders. Furthermore, the generating conditions were chosen so that the "short trajectory" contribution to the harmonic generation process be phase matched (gas jet placed after the laser focus) [5,6]. This was verified by measuring the far-field profile of the harmonic beam: no outer ring was observed around the central spot [19]. With these precautions, we obtained the results presented in Figure 4. A clear spectral broadening of a factor 2 is observed for the harmonics of the plateau region (H15 and H17) at 60-fs delay. This gives indication of a temporal confinement of the harmonic emission. The fact that no spectral broadening is observed for high orders may be explained by the increase of the harmonic chirp in the cutoff region when the "short trajectory" is selected [19].

4 Simulations

We have developed a simplified model based on experimental measurements of the dependence of the harmonic signal on laser intensity and ellipticity. We assume that the measured signal corresponding to harmonic q is given by

$$S_q = A \int_{-\infty}^{+\infty} I_0(t)^{q_{\mathrm{eff}}} \mathrm{e}^{-\beta_q \epsilon(t)^2} g(\theta(t)) \mathrm{d}t \qquad (1)$$

where A is a normalization constant, $I_0(t)$ the laser intensity, q_{eff} an effective nonlinear order, equal to 3 for a plateau harmonic [20] and

$$g(\theta) = 1 - \frac{1}{4}[1 + \cos(2\theta)] \qquad (2)$$

is the empirical function that expresses the efficiency of the spectrometer as a function of the orientation of the harmonic polarization plane. We assume that the harmonics are always linearly polarized along the major axis of the polarization ellipse of the fundamental: this is a good approximation as long as $\epsilon \ll 1$; when the ellipticity is important, harmonic generation is very weak, therefore the produced error is negligible. The Gaussian decrease $e^{-\beta_q \epsilon^2}$ of the harmonic signal as a function of the ellipticity ϵ depends on the harmonic order and the nonlinear medium [8,9]: β_q is a constant that increases with q and whose value lies typically between 15 and 60. In our experiments we measured a value of $\beta_q \approx 35$.

The comparison of the simulation using the above formula with the experimental-curve is shown in Figure 3. The good agreement indicates that our model, despite its simplicity, describes correctly the polarization gating. Furthermore, it demonstrates that this experimental setup allows an accurate control of the polarization gate, through the use of the piezoelectric translation. By switching off the ellipticity dependence in equation (1), we can study the influence of the decreased laser intensity on the harmonic signal at large delays. The envelope of such a trace is shown in long-dashed curve in Figure 3. It decreases much more slowly, in agreement with the measurements performed with the third QWP turned 45 degrees. Therefore it cannot explain the fast decrease of the harmonic signal. The main reason for the latter is rather the shortening of the polarization gate when increasing the delay, which demonstrates the efficiency of our polarization gating scheme. In Figure 2c are shown the simulated harmonic temporal profiles for a zero delay (solid) and a 60-fs delay (dashed). The duration of the XUV pulse is reduced by a factor of 2.3, from 49 to 21 fs, which is consistent with the measured spectral broadening of a factor 2.

5 Conclusion

In conclusion we have presented a proof of principle experiment of a new technique of polarization gating. We showed clear evidence of control of the time-dependent ellipticity of the fundamental laser beam. Both the decrease of the harmonic signal at large delay and the spectral broadening indicate a temporal confinement of the harmonic emission. The main limitation of this technique lies in the fact that, when increasing the delay, on the one hand the laser intensity during the gate — and thus the harmonic signal — decreases, and on the other hand an increasing part of the laser energy ionizes the nonlinear medium without contributing to the harmonic generation. When starting from 80 fs pulses, the delay necessary to reduce the gate duration below the optical period is so large that the harmonic efficiency is very low. The generation of an intense isolated attosecond pulse thus requires laser pulses lasting 15–20 fs, which is less demanding than the 5–7 fs necessary at present [2]. Note that a new technique of pulse post compression at 15 fs above the mJ level is currently under development [21]. Our simulations, validated by the comparison with experimental results, suggest the possibility of generating attosecond pulses with energy in the nJ range using the generating conditions that have recently allowed microjoule femtosecond harmonic emission [22].

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