Time-gated high-order harmonic generation

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(Received 10 April 2003; published 6 October 2003)

We generate high-order harmonics by using an 800-nm fundamental pulse whose polarization evolves with time. Controlling the ellipticity modulation of the fundamental field allows us to continuously confine the harmonic emission from an estimated minimum value of 7 fs (1 fs$=10^{-15}$ s) up to more than the 35 fs input pulse duration. Depending on the observed harmonic, the harmonic spectrum can show either a narrowing or a broadening when the ellipticity is changed in good agreement with an effective confinement of the high-order harmonic generation.

DOI: 10.1103/PhysRevA.68.043804 PACS number(s): 42.65.Ky, 32.80.Wr

High-order harmonic generation (HHG) has long been predicted to be a potential source for attosecond pulsed emission [1,2]. It was a major experimental breakthrough when it was demonstrated that the harmonics are effectively emitted in a train of attosecond pulses [3] from which a single attosecond burst of XUV light can be extracted [4]. So far, only state of the art 5–7 fs intense fundamental pulses [5,6] allowed the production of isolated subfundamental optical cycle XUV pulses [4] after the selection of near cutoff harmonics [7]. An alternative way to generate isolated XUV attosecond pulses [2] is to rapidly modulate the polarization of the fundamental pulse and use the strong ellipticity dependence of HHG [8] for confinement. If its polarization remains linear for a time much shorter than the pulse duration, the harmonic emission can then be strongly confined. In this way, even relatively long pulses ($\sim$15 fs) could be used for single attosecond pulse emission. Furthermore, we show here that modulating the polarization allows continuous control of the XUV pulse duration. A first attempt of HHG with a polarization modulated pulse was performed in LUND [9] by using nonlinear effects to control the fundamental ellipticity. A confinment of the HHG was then observed but the strong nonlinearity involved in the polarization modulation process made it hardly controllable for the emission of a single attosecond pulse. Similarly, another test was recently performed with two orthogonally polarized laser pulses with different frequencies [10] and showed harmonic confinement but was hardly downscalable for attosecond pulse emission.

In this paper we introduce a linear technique for controlling the ellipticity modulation and we present experimental evidence of a continuously controllable temporal confinement of HHG down to a minimum duration of $\sim$7 fs by using a 35-fs fundamental pulse. This confinement is observed by studying the harmonic spectrum while controlling the ellipticity of the fundamental field. We first present the experimental technique used to create a flat top intensity profile pulse with a time-dependent polarization. We then describe the experimental setup and experimental findings for both the plateau and cutoff harmonics and show that these observations are in good agreement with an effective controllable confinement of HHG. Finally, we conclude with the possible implications of this technique.

The linear method used here to modulate the ellipticity of the fundamental pulse consists in transmitting femtosecond pulses through two quartz quarter wave plates. The first thick quartz plate splits the $\tau_0=35$ fs incoming linearly polarized pulse in two, delayed, linearly polarized pulses with perpendicular polarization (Fig. 1). At a central wavelength of 800 nm, the 1.05-mm-thick plate results in a delay, $\delta t=31.3$ fs, in between these two pulses and a dephasing of 47 $\pi/2$ (multiple-order quarter wave plate). The outcoming field is circularly polarized at the time when the two perpendicular fields have the same amplitude. When the incident polarization is at 45° of the axis of the first plate ($\theta_1=45°$), the total field is therefore circularly polarized at the center of the outgoing pulse. At the beginning and end of this pulse, the field is linearly polarized. Using a multiple order quarter wave plate allows us, therefore, to create a pulse having a polarization that continuously evolves from linear to circular and back to linear [11]. Transmitting this pulse through an additional zero-order quarter wave plate (with its axis at $\theta_2=45°$ of those of the first plate) changes the circular field in linear and the linear field in circular. The combination of these two plates (with $\theta_1=\theta_2=45°$) changes an input linearly polarized pulse into a polarization modulated pulse whose polarization changes from circular to linear and back to circular (Fig. 1).

While this polarization evolution strongly depends on the dephasing between the two outgoing pulses, the intensity profile (transmitted energy per unit of time) of the output

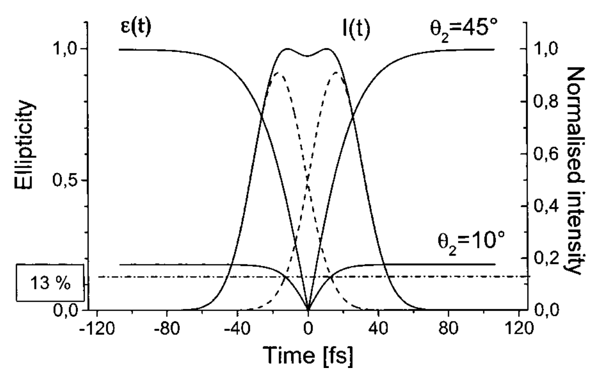

FIG. 1. Pulse intensity profile (full line) and time-dependent ellipticity for two angles ($\theta_2=45°$ and $\theta_2=10°$) of the zeroth-order quarter wave plate. The crossed polarized 35 fs Gaussian pulses separated by 31.4 fs are shown (dotted lines). The 13% ellipticity defining the gate width is indicated.

pulse only depends on the delay, δt. For $\delta t \approx \tau_0$, the intensity is close to maximum at the center of the pulse and remains constant during τ_0, creating therefore a pulse with a flat top intensity profile (Fig. 1).

The temporal domain in which the electric field is close to linear (ellipticity smaller than 13%) defines a temporal gate inside which HHG can occur efficiently. We choose this 13% criterion since we observed that a (constant) 13% ellipticity of the fundamental field reduced by 50% the emission efficiency for the plateau harmonics (order $q=19$ and 27) generated in argon as compared to harmonics generated with a zero ellipticity. The width of this temporal gate, τ_G, is imposed by the duration τ_0 of the input pulse (supposed as Gaussian in this paper), the delay δt, and by the angles θ_1 and θ_2. For $\theta_1=\theta_2=45°$ and $\delta t \approx \tau_0$ (which corresponds to the so-called "narrow gate" in this manuscript) this width is equal to $\tau_0/5$ ($\tau_g=7$ fs for $\tau_0=35$ fs). Controlling the width of this gate without any other changes allows us to observe the effect of a temporal confinement of HHG on the harmonic spectra. Changing the angle θ_2 allows us to change this gate width without changing the output pulse intensity profile. Indeed, at the center of the outgoing pulse, the polarization is linear for any angle θ_2 while at other times during the pulse the degree of ellipticity depends on θ_2. Changing θ_2 changes the time at which the 13% ellipticity criteria is achieved and hence the gate width. In the case of $\theta_2=0$, the output field is always linearly polarized and HHG can occur throughout the full output pulse (this configuration $\theta_1=45°$, $\theta_2=0°$ will be referred to as "large gate"). Simply varying θ_2 by rotating the zero-order quarter wave plate allows us, therefore, to vary continuously the gate width from a minimum value ($\tau_0/5$) to the full pulse ($\sim 2\tau_0$) without changing significantly the intensity profile of the outgoing pulse.

This analysis is valid only for well-defined wavelengths at which the two plates are quarter wave plates. However, multiple order wave plates are chromatic and this analysis holds only if the field instantaneous frequency is constant, i.e., if the two output pulses are Fourier limited short pulses. Since this technique is linear, it is only important that the pulses are unchirped in the HHG cell and the input pulse can be chirped to compensate for the dispersion of the plates. This precompensation is possible only if one can neglect the dispersion difference between the two axes of the plate which is indeed negligible here (for a 35 fs unchirped input pulse, the output pulse duration differs only by 0.02 fs if the pulse propagates along the slow or fast axis). For shorter pulses (~ 15 fs), thinner plates (0.5 mm) can be used and the same analysis holds.

In the present experiment we checked that the output pulse duration was minimum where harmonics are generated by inserting a doubling BBO crystal after the wave plates and the focusing lens (see the text below). The compressor was tuned so that the doubled signal was maximum and hence the pulse had its minimum duration in the HHG cell.

Apart from the two quartz wave plates, the setup used for HHG was standard. After passing through the plates, the 800-nm beam was focused by a 1.5-m focal length lens onto a 15-mm-long gas cell (filled with argon) located at focus.

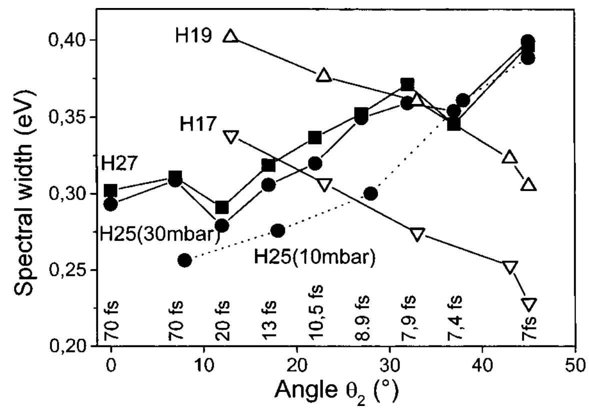

FIG. 2. Evolution of the spectral width of the plateau ($q=17$ and 19) and near cutoff ($q=25$ and 27) harmonics as a function of the zeroth-order quarter wave-plate angle (θ_2). The large and narrow gate, respectively, correspond to $\theta_2=0°$ and $\theta_2=45°$. The theoretically estimated gate width is indicated. Harmonics are generated with 1.1 mJ in 30 mbar of argon (full lines) and 1.4 mJ, 10 mbar of argon (dotted line).

The harmonic beam was analyzed by a grazing incidence XUV spectrometer ($f=1$ m toroidal mirror and flat grating, 470 g/mm, deviation $=165°$) equipped with a back illuminated charge-coupled device (CCD) camera (protected from the IR stray light by an 1000-Å-thick aluminum filter mounted on a mesh). The observed harmonic order ranged from 15 to 35. We concentrate on results obtained with a low pulse energy (1.1 to 1.4 mJ after compression) and a low argon pressure (9 and 30 mbar) in order to avoid spectral modifications due to ionization. The only parameter was the gate width controlled by changing θ_2.

For the plateau harmonics, we could observe a spectral narrowing of the individual harmonics when the gate width was decreased (Fig. 2) as was already observed in [9]. This is in contradiction with the intuitive thinking that a temporal confinement of the HHG should lead to an increase of the harmonic spectral width but it can be explained by considering the influence of the intensity dependent atomic phase. In contrast, for the cutoff harmonics, we could observe a spectral broadening of the harmonics when the gate width was decreased (Fig. 2). For the 29th harmonic, for instance, the spectral width (estimated with a Gaussian fit) changed from 300 meV for the large gate configuration to 400 meV for the narrow gate.

These observations were robust and all the cutoff harmonics showed such a spectral broadening. Reducing the pressure from 30 to 9 mbar (while increasing the energy from 1.1 to 1.4 mJ to maintain a reasonable level for the harmonic signal) allowed us to observe the same effect (Fig. 2). At this low energy level, this result confirms the negligible effect of ionization of the medium on the spectral modifications and that the observed changes are induced by the temporal confinement. Furthermore, for these low energies and low pressures, the spectrum of the fundamental pulse was not changed by propagation in the generating gas medium.

To further check these observations, we also selected two harmonics (25th and 27th) and changed the laser energy in order to put these harmonics either in the plateau region

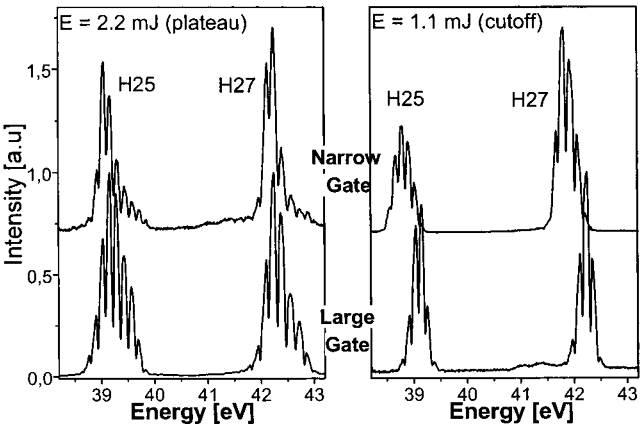

FIG. 3. Spectra of the 25th and 27th harmonics generated in 30 mbar of argon in the narrow (top) and large (bottom) gate configuration. (a) When these harmonics are in the plateau. (b) When they lie in the cutoff region. The rapid modulations in the spectra are due to the mesh supporting the Al filter.

($E_{\mathrm{laser}}=2.2$ mJ) or close to the cutoff ($E_{\mathrm{laser}}=1.1$ mJ). Similarly to our previous observations, when the harmonics are in the plateau, their spectral width decreases when the gate width is reduced (Fig. 3). When those harmonics are in the cutoff region, their spectral width increases when the gate width is decreased (Fig. 3).

We also followed the efficiency of HHG as a function of the gate width and this efficiency did not dramatically change. Usually for the plateau harmonics, the efficiency decreased when the gate width was decreased but for the cutoff harmonics, the behavior was more complex and there were even some low-energy specific configurations where the HHG efficiency was larger in the narrow gate configuration than in the large gate configuration.

The results presented here are consistent with a temporal confinement of the HHG induced by the modulation of the fundamental polarization and with the current understanding of high-order harmonic generation, especially by considering the crucial effect of the intensity-dependent atomic dipole phase [12]. For the plateau harmonics two quantum paths (''short'' and ''long'') are involved in the HHG process and it was shown that they can be considered separately to explain experimental findings [13]. For the cutoff harmonics, these two quantum paths merge into a single one.

For a given harmonic, the phase of the atomic dipole is proportional to the laser intensity and evolves as $\varphi^{(i)}=\alpha^{(i)}I$, where I is the laser intensity [13]. For the plateau harmonics, $\alpha^{(1)}=2.5$ cm^2/W (with I in units of 10^{14} W/cm^2) for the short quantum path and $\alpha^{(2)}=25$ cm^2/W for the long quantum path. For the cutoff harmonics, $\alpha^{(c)}=12$ cm^2/W. For a pulse with intensity $I(t)$, the phase of the harmonic dipole is $\varphi_q=-q\omega_0 t-\alpha I(t)$, and the corresponding instantaneous harmonic frequency, $\omega_q(t)=q\omega_0+\alpha dI/dt$, changes with time. To estimate this frequency change, one needs to know the intensity profile (considered here as Gaussian) and peak intensity of the pulse. Observing the 33rd harmonic implies that the peak intensity is at least equal to 1.86×10^{14} W/cm^2 (calculated by using the cutoff law [14]) and we consider in the following a peak intensity of 2×10^{14} W/cm^2 (a perfect focussing of the 1.1 mJ pulse by the 1 m lens with a beam size $W=8$ mm would lead to a peak intensity of $\sim10^{15}$ W/cm^2).

In the large gate configuration, the plateau harmonics can be generated as I changes in the leading and falling edge of the pulse, and ω_q changes significantly during the emission process. For instance, the 17th harmonic can be generated for $I>6\times10^{13}$ W/cm^2 (for times -38 fs$<t<38$ fs) and its instantaneous frequency ω_{17} changes by ~2.4 eV. Note that this 2.4 eV is an overestimate of the harmonic spectral width since the generation efficiency changes with I as well as α. Still, this width is so large that this effect should be predominant and impose the plateau harmonic spectral width. Confining the plateau harmonic emission to a few femtoseconds would result in confining the possible values of dI/dt during emission and therefore a narrowing of the spectrum as observed here and as was already observed and fully detailled in a former experiment [9]. For the short quantum path, the instantaneous frequency change is much smaller and a temporal confinement of the harmonic generation should lead to a broadening of the spectrum (as we recently observed in an other experiment [15]) which is inconsistent with our observation. However, the relative efficiency between the two quantum paths emission strongly depends on the generation configuration (gas pressure and laser focusing geometry) and the long quantum path seems to be the dominant one here (this is further confirmed since here the plateau harmonics are naturally spectrally broader than the cutoff harmonics). A temporal confinement of the plateau harmonic emission results therefore in a spectral narrowing as observed here, and the minimum bandwidth (0.26 eV) is consistent with a temporal confinement of ~7 fs (estimated, for Gaussian pulses, through the relation $\Delta E\ \delta t_{\mathrm{hhg}}=1.8$, with ΔE in eV and δt_{hhg} in fs). Note that if the confinement was on the order of 1 fs, one should also observe a spectral broadening for these plateau harmonics.

For the cutoff harmonics, the situation is different since $\alpha^{(c)}$ is smal and these harmonics can only be generated around the peak of the pulse where dI/dt is small. The intensity-dependent frequency shift is therefore smaller than for the plateau harmonics. As the HHG is confined to a few fs, the spectrum of these harmonics should be enlarged as observed here.

To estimate the enlargement due to confinement, we considered that the cutoff harmonics have a natural bandwidth ΔE_0 that can be due to the laser bandwidth, the atomic phase induced frequency chirp, and nonlinear effects in the gas medium. The confinement of the harmonic emission to δt_{hhg} also enlarges the bandwidth by ΔE_{conf} (with $\Delta E_{\mathrm{conf}}\ \delta t_{\mathrm{hhg}}=1.8$). Considering ΔE_0 and ΔE_{conf} as independent leads to a total width ΔE of $\Delta E=\sqrt{\Delta E_0^2+\Delta E_{\mathrm{conf}}^2}$. For the cutoff harmonics [see Fig. 2(b)], the bandwidth changes from 0.3 (large gate) to 0.4 eV (narrow gate) which corresponds to $\Delta E_{\mathrm{conf}}=0.26$ eV, consistent with a temporal confinement of 7 fs. Again, this is in good agreement with the estimated minimum gate width, $\tau_g=7$ fs.

The simultaneous observation of a spectral narrowing for the plateau harmonics and a spectral broadening for the cutoff harmonics as the gate width is decreased are therefore consistent with an effective temporal confinement of the HHG inside the polarization controlled temporal gate. Although the spectral modifications are only an indirect signature of confinement and the exact duration of the emission cannot be directly measured here, these experimental observations are consistent with a confinement inside a temporal gate having a minimum duration of ∼7 fs as expected by theory.

Using a simple linear setup to modulate the ellipticity led to experimental observations consistent with a controllable confinement of the high-order harmonic emission for both the plateau and cutoff harmonics. The essence of this technique is to continuously control the harmonic emission from a maximum value down to a small fraction (∼1/5) of the input pulse duration. This opens new perspectives for the use of widely available ''long'' fundamental pulses in the attophysics domain and using high energy sub-15 fs pulses [5,16] should lead to the emission of an isolated attosecond pulse [17]. Furthermore, continuously controlling the number of attosecond pulses in an attosecond pulse train could ultimately be used directly to perform pump probe experiments by using directly the different attosecond pulses as pumps and probes which would remove the need to create two identical replica with XUV optics.

We gratefully acknowledge discussions with Thomas Brabec and P. B. Corkum. This study was supported by the Région Aquitaine, the European Regional Development Funds (FEDER), and the European network ''ATTO'' No. HPRN-2000-00133.

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