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LETTER TO THE EDITOR

Single attosecond pulse production with an ellipticity-modulated driving IR pulse

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Abstract

We theoretically study attosecond pulse production via high-harmonic generation using a driving laser pulse with a time-dependent ellipticity. The theoretical approach produces results that agree with our experimental data obtained using 35 fs driving laser pulses and is further used to study the generation of single attosecond pulses with shorter laser pulses. We find an equation for the duration of the temporal window created by the time-varying driving laser polarization in which high-harmonic emission can occur. We formulate the necessary requirements concerning the driving laser field in order to confine the high-harmonic emission in the form of a single attosecond pulse. Indeed, we show that using incident 12 fs laser pulses single attosecond pulses can be produced for certain carrier-envelope phase (CEP) values of the driving pulse. For 6 fs incident laser pulses, single attosecond pulses are produced for all values of the CEP (the intensity of the attosecond pulse still depends on the actual value of the CEP). If implemented with state-of-the-art 5 fs laser pulses, this technique can even lead to the production of sub-100 as pulses.

Attosecond pulse production is one of the most fascinating applications of high-order harmonic generation (HHG) in gases. It has great practical and fundamental interest as a way of obtaining the shortest electromagnetic pulses created by now. It was shown both theoretically [1–3] and experimentally [4–6] that appropriate phase-locking of different harmonics generated in gas targets with intense femtosecond pulses results in a train of VUV pulses of attosecond duration. The successful generation of isolated attosecond pulses is an important issue since single attosecond pulses are more promising for applications in time-resolved spectroscopy.

In this letter, we consider a way of producing single attosecond pulses based on HHG using an intense laser pulse with time-dependent ellipticity [7]. HHG efficiency is highest for linearly polarized driving laser fields and rapidly decreases with increasing driving ellipticity.
Temporal modulation of driving laser ellipticity confines the VUV emission to a temporal ‘gate’ where the polarization is very close to linear, as shown in recent experiments [10–14]. The duration of this gate defines the envelope of the whole attosecond pulse train resulting from the HHG. If the gate is short enough, the emission of one single attosecond pulse is possible [7, 15–17].

Another way of producing isolated attosecond pulses is to use linearly polarized few-cycle (∼5 fs long) driving pulses and to spectrally select the highest frequency (cut-off) VUV. As this VUV is emitted at the peak of the fundamental intensity, so in such short driving pulses this emission is confined to a subfemtosecond duration [18].

The technique considered in this paper, however, has two advantages. First, it makes it possible to generate single attosecond pulses using longer driving laser pulses rather than using state-of-the-art 5 fs pulses. Besides, in the latter method the lower the harmonic order, the worse is the temporal confinement; even using the 5 fs pulse confinement of the VUV emission down to the single attopulse is possible only for the cut-off VUV. In contrast, using the fundamental with the time-modulated ellipticity provides the temporal confinement of both plateau and cut-off VUV, which is the second advantage of the method. Practically, this advantage means the possibility of obtaining a single attosecond pulse with a broad spectral width. Then using a spectral filter one can select a desired spectral range, to optimize the pulse duration or for a specific application. Thus this technique implies wide possibilities of the attosecond pulse control.

In this work, we compare calculated and measured harmonic spectra generated with relatively long ellipticity-modulated driving laser pulses. The observed agreement validates our theoretical approach. We then present results of calculations made in the time domain for shorter driving pulses and determine the necessary requirements on the latter in order to generate isolated attosecond pulses using ellipticity gating.

We calculate the single-atom response to the driving laser field using the theory of HHG developed in [19]. We generalize this theory for the case of an arbitrarily polarized laser field. The approach and some of its results are briefly presented in [20]. We take into account propagation and diffraction of the harmonic radiation in a way close to that suggested in [21] as well as re-absorption of the harmonics by the target gas, which is quite important for the harmonic orders considered in this work [22].

In our experiments, the driving laser field with a time-varying ellipticity is obtained using two quartz wave plates (one multiple-order $\lambda/4$ and the other zero-order $\lambda/4$) as described in detail in [11, 20]. The properties of the field are controlled via the thickness of the multiple-order wave plate that introduces a delay (denoted from here on as $\delta$) between the two orthogonally polarized components of the incident field, as well as via the orientation of the zero-order wave plate (characterized from here on by the angle $\beta$). One important advantage of this method is that it is possible to continuously vary the ellipticity of the laser field just by rotating the zero-order wave plate without changing the intensity of the pulse [11].

In the temporal centre of the ellipticity-modulated pulse (considered below as $t = 0$), the field polarization is almost linear for every $\beta$. Around the pulse centre, the ellipticity $\varepsilon(t)$ varies quasi-linearly with time as

$$\varepsilon(t) = -\alpha \cos(2\beta)t + O(t^3). \quad (1)$$

The factor $\alpha$ depends on the shape and duration $\tau$ of the incident pulse as well as on the delay $\delta$ introduced by the multiple-order wave plate. For an incident Gaussian pulse, we have

$$\alpha = 2 \ln 2 \frac{\delta}{\tau^2}. \quad (2)$$
Figure 1. Measured (solid line) and calculated (dotted line) spectra for the seventeenth harmonic generated in the ‘large gate’ (a) and ‘narrow gate’ (b) configurations. The driving laser wavelength is 800 nm, the peak intensity at the focus is $1.4 \times 10^{14}$ W cm$^{-2}$, the initial pulse duration is $\tau = 35$ fs and the delay is $\delta = 31.4$ fs. Harmonics are generated in a 15 mm long cell located at the focus and filled with Ar with a backing pressure of 30 mbar. Spectra are measured and calculated in the far field and integrated over the emission angle. Every curve is renormalized to its maximum.

From (1), we see that for $\beta = 45^\circ$, $135^\circ$, etc (the so-called ‘large gate’ configuration), the ellipticity remains zero throughout the entire pulse, thus producing a large temporal window for harmonic emission [11]. On the other hand, for $\beta = 0^\circ$, $90^\circ$, etc (the so-called ‘narrow gate’ configuration), the ellipticity rapidly changes with time such that the polarization becomes close to linear only for a short time interval around the pulse centre. From equations (1) and (2), we see that shorter pulses and longer delays are necessary in order to obtain higher rates of change in ellipticity. However, the driving laser intensity at the pulse centre decreases with increasing delay, so we use delays close to or slightly longer than the incident pulse duration itself.

In our experiments, we use $\tau = 35$ fs and $\delta = 31.4$ fs; other experimental conditions are described in [11]. In figure 1, we present experimental and calculated spectra for the seventeenth harmonic generated in argon. In the experiment, the spectrum obtained in the ‘large gate’ configuration exhibits a redshifted satellite and almost no blueshifted one. Although less pronounced, the asymmetry of the satellites is reproduced in the calculated spectra. In the ‘narrow gate’ configuration, both measured and calculated spectra are broadened and red-shifted. Thus, quite fine experimentally measured features can be reproduced by our calculations (a more detailed comparison can be found in [20]). These observations validate our theoretical approach for VUV generation using a time-dependent driving laser field. In what follows, we use this theoretical approach to obtain results directly in the time domain. For these calculations, we consider propagation in the generating medium but we only consider on-axis VUV emission that can be experimentally selected downstream by inserting an aperture in the far field, thus only selecting the central part of the VUV beam.

Calculations show that under our experimental conditions, we can continuously shorten the generated train of attosecond VUV pulses from 36 fs in the ‘large gate’ configuration down to 8 fs in the ‘narrow gate’ configuration [20]. Using shorter incident pulses in our calculations
we reduce the train duration, and finally, using 10 fs incident fundamental pulse and $\delta = 13$ fs, we obtain in the ‘narrow gate’ configuration (under a certain value of carrier-envelope phase (CEP), namely, using the notation of [20], under $\varphi = \pi / 2$) a single attosecond pulse as shown in figure 2(a) (dotted line). The duration of this pulse is about 200 as, which is longer than the Fourier-limited duration of 120 as for this pulse. This is due to the intrinsic VUV chirp of attosecond pulses generated via HHG [5, 23], which can nevertheless be compensated for [5, 24, 26]. Although we are dealing with quite modest driving laser intensities and relatively low harmonic orders, attosecond pulses produced with this method are shorter than those obtained by spectral selection of cut-off harmonics generated with a linearly polarized pulse [25]. Note that the selection of on-axis emission practically removes the contribution to the attosecond pulse train from the long quantum path as well as from secondary recollisions [20]. The extent to which the extra VUV emission is suppressed determines the contrast of the attosecond pulses in the train. In our calculations, the contrast is about $10^{-2}$. However, it can be improved by changing the experimental conditions.

It is well known that for the majority of applications, the carrier-envelope phase (CEP) plays an important role for few-cycle laser pulses (below 7 fs at 800 nm). As can be seen in figure 2, our results are sensitive to the CEP of even longer driving pulses. The point is that the duration of the gate obtained for such pulses is comparable to the duration of an optical cycle. Therefore, the specific temporal evolution of the laser field (determined by the CEP) has a direct effect on the VUV emission. By averaging the VUV emission over all possible values of the driving laser CEP, we obtain the ‘gate’ envelope also presented in figure 2. We can now accurately define the gate duration as the FWHM of the envelope. To stress the effect of the delay $\delta$ on the gate duration, we show in figure 2 the results obtained in the case of equal incident pulse durations but different delays.

If the gate duration is several optical cycles or more, the ellipticity varies relatively slowly in time and so the VUV generation efficiency changes adiabatically with ellipticity. The dependence of VUV generation efficiency on ellipticity is close to Gaussian (experimental measurements [8, 9] of this dependence are well reproduced by our theory [20]). At the centre of the gate, the variation of ellipticity is close to linear (see equation (1)), thus in this case, the temporal envelope of the gate is also close to Gaussian.

From here on, we denote by $\varepsilon_{tr}$ the value of driving laser ellipticity for which VUV generation efficiency is decreased by half. The gate duration can be found as the time interval during which the ellipticity modulus is less than $\varepsilon_{tr}$. From equations (1) and (2), we obtain the duration of this interval by keeping only the linear part of equation (1):

$$\Delta = \frac{\varepsilon_{tr} \tau^2}{\delta |\cos 2\beta| \ln 2}.$$  (3)

Naturally, the gate duration can be approximated to this only in the case where VUV generation is limited by the driving laser ellipticity and not by its intensity, thus roughly for $\Delta^2 < \tau^2 + \delta^2$. Note that for $\beta = 0^\circ$, 90$^\circ$, etc (‘narrow gate’), equation (3) coincides with the gate duration found in [17] by using two circularly polarized pulses with a delay between them [16].

For gate durations much larger than an optical period, equation (3) with $\varepsilon_{tr} = 0.15$ agrees very well with the calculated duration. However, if the fundamental polarization varies very rapidly (gate duration comparable to an optical cycle), the VUV generation efficiency does not vary adiabatically with ellipticity and the latter cannot be accurately defined. In figure 2, we see that when this is the case, the gate envelope is not Gaussian any more but has a flat top. Strictly speaking, equation (3) is not applicable in this case. In order to check if equation (3) gives at least a correct estimation, we calculate the gate duration for different
Figure 2. Calculated VUV intensity (photon energy ranging from 25 to 45 eV) generated in the 'narrow gate' configuration for an incident pulse duration $\tau = 10$ fs and delays $\delta = 13$ fs (a) and $\delta = 18$ fs (b) for two values of the driving laser carrier-envelope phase (CEP) differing from one another by $\pi/2$ (solid and dotted lines), as well as the temporal gate envelope (dashed line). The driving field intensity at the pulse centre is $2.2 \times 10^{14}$ W cm$^{-2}$. The VUV intensity is calculated on-axis resulting in the selection of the short path component. Every curve is renormalized to its maximum.

$\tau$, $\delta$, $\beta$ and at driving intensities of $2.2 \times 10^{14}$ W cm$^{-2}$ and $3 \times 10^{14}$ W cm$^{-2}$. The agreement in this case is surprisingly good if the gate duration is comparable to or even shorter than an optical cycle (down to 1 fs). The calculated duration is slightly shorter than that predicted by equation (3) but the deviation is typically less than 10 to 15 per cent. Also, the calculated duration has a soft dependence on the driving intensity, namely it is slightly shorter at higher intensities.

Based on the fact that the attosecond pulses in the train are separated by approximately half an optical cycle, we can now formulate the requirements concerning the driving laser field in order to achieve single attosecond pulse production via an ellipticity gating. (Henceforth, we consider that single attosecond pulses are generated if any other pulses produced in the train are at most half as intense as the main pulse). If the duration given by equation (3) is less than an optical cycle (for instance, for a 'narrow gate' with either $\tau = 15$ fs and $\delta = 20$ fs, $\tau = 12$ fs and $\delta = 12$ fs or $\tau = 10$ fs and $\delta = 13$ fs), a single attosecond
pulse can be generated for a particular range of values of the CEP. For instance, in conditions of figure 2(a) this range is \( \varphi = 0.7–2.9 \). The shorter the gate duration, the wider the range becomes. If the duration (3) is shorter than half an optical cycle (for instance, for a ‘narrow gate’ with either \( \tau = 10 \) fs and \( \delta = 18 \) fs or \( \tau = 6 \) fs and \( \delta = 6 \) fs), an isolated attosecond pulse is generated for all values of CEP and changes in the latter result only in fluctuations in the attosecond pulse intensity as shown in figure 2(b). (Strictly speaking, the single attopulse is generated under every CEP except one, under which two attopulses with equal intensity are generated. However, this intensity is lower than the intensity of a single attopulse generated under all other CEPs, so this case is practically unimportant).

Using a ‘narrow gate’ with \( \tau = 5 \) fs and \( \delta = 5 \) fs, at an intensity of \( 3 \times 10^{14} \) W cm\(^{-2}\) (the short pulse duration prevents ionization even at this intensity) and selecting VUV radiation with photon energies higher than 25 eV, we obtain isolated and chirped 200–240 as pulses (depending on CEP). Using such state-of-the-art driving laser pulses, a VUV burst as short as 90 as could be produced after compensating for the VUV chirp. Further pulse shortening could be achieved by increasing the VUV spectral width, for instance, by using Ne or He as the target gas.

In conclusion, in this paper we validate our theory of VUV generation with arbitrarily polarized driving laser pulses by comparing calculated results with experimentally measured VUV spectra obtained with ellipticity-modulated 35 fs pulses. We apply our theory to the study of attosecond pulse generation with shorter driving laser pulses, find an equation describing the duration of the VUV generation temporal window (gate) created via this modulation of driving laser ellipticity, and formulate the necessary requirements on the driving laser field in order to generate single attosecond pulses. In particular, using the delay equal to the incident pulse duration one can obtain the generation of a single attopulse for a certain range of values of the CEP using incident pulses as short as 12 fs, and with 6 fs driving pulses single attosecond pulses are generated for all values of the CEP, albeit the attosecond pulse intensity still depends on the CEP. Applying the above technique using 5 fs driving laser pulses could lead to the successful generation of isolated sub-100 as pulses.

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