Generation of isolated attosecond pulses by spatial shaping of a femtosecond laser beam

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Abstract. We present a new method for generating isolated attosecond pulses via high-order harmonic generation in gases. It relies on using collective effects to achieve transient phase-matching which provides both a high efficiency and a strong temporal confinement under specific conditions. By controlling the spatial shape of the fundamental beam and the geometry of the laser–gas interaction, this transient phase matching leads to the generation of isolated broadband attosecond pulses with long driving pulses (10–20 fs) even without controlling their carrier envelope phase. Such laser pulses are becoming available at high energy levels and our approach offers a route to increase the energy of isolated attosecond pulses by orders of magnitude as compared to existing sources.

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1. Introduction

By controlling the microscopic atomic response to a strong field excitation, within the high-order harmonic generation (HHG) process in gases, it is now possible to generate isolated attosecond pulses [1]–[4]. This requires state-of-the-art laser systems delivering 5–7 fs fundamental pulses with their carrier-envelope phase (CEP) actively controlled [5]. Such laser systems are not widely available and only deliver low-energy pulses at the few hundred microjoule level with limited long-term stability. Due to the current conversion efficiency of the harmonic process, this restricts the extreme ultraviolet (XUV) attosecond pulse energy to the nanojoules or sub-nanojoules level, which is an important limitation for applications. On the other hand, it is possible to generate high-order harmonics at the microjoules level [6, 7] with high-energy laser systems, albeit delivering longer pulses and with no control of the CEP. Using such high-energy laser systems to generate isolated attosecond pulses would significantly increase the attosecond pulse energy and lead to the invention of promising techniques [8, 9]. Besides, increasing the energy of isolated attosecond pulses will clearly help access the field of nonlinear physics induced by attosecond pulses which is actually a major challenge.

In this paper, we present a new approach for generating isolated attosecond pulses that is compatible with using relatively long (10–20 fs) laser pulses. Our approach relies on controlling collective effects involved in HHG to confine the XUV-emission by transient phase matching. As HHG is a coherent emission, its efficiency depends nonetheless on the single-atom response, but also on the phase-matching conditions that control the macroscopic response. The macroscopic emission is maximized when the phase matching is achieved [10, 11], i.e. when the phase velocity is the same for both the XUV and the fundamental infrared (IR) fields. These phase velocities are determined by the neutral gas density, the intrinsic harmonic phases (also called ‘atomic phases’), the geometrical conditions (amplitude and phase profiles of the laser field in the interaction region and geometry of the gas target) [12, 13] and the electron density, which strongly affects the IR phase velocity [14]. The XUV emission is therefore very sensitive to the free-electron density in the medium. As ionization accompanies HHG and changes during...
the pulse, the phase-matching conditions vary during the laser pulse [15]–[17]. A transient phase-matching can be obtained in specific conditions and the XUV-emission can be optimized and confined to the temporal window where phase matching occurs.

When the transient phase-matching evolves very quickly (i.e. typically at the timescale of the time interval between the two consecutive attosecond pulses in a train that is about half an optical cycle of the fundamental), an isolated attosecond pulse can be efficiently generated. This situation can be achieved in the tunnel regime, since the ionization rate naturally increases by steps near the maxima of the field [18]. The heights of the ionization jumps can be adjusted by adapting the pulse peak intensity, its time profile and by choosing the proper gas. Therefore, the temporal evolution of the phase matching can be controlled. Moreover, as the key parameter to maximize HHG is \( \delta k L \) (where \( L \) is an effective medium length over which the emission takes place and \( \delta k = k_q - qk_0 \), the wave-vector mismatch, \( k_q \) being the wave vector of the \( q \)th harmonic, and \( k_0 \) the wave vector of the laser field), the confinement can further be controlled by a proper choice of the medium length and the gas pressure. Eventually, a global optimization of the process requires that transient phase-matching is achieved as the atomic response is maximized.

This picture contains the essence of our confinement technique but holds only in a one-dimensional (1D) geometry. It is not directly experimentally applicable since usually the intensity of the laser beam varies radially and therefore the ionization is not uniform throughout the beam. This position-dependent ionization leads to position-dependent timing of the (local) phase-matching window. The duration of the total XUV-emission is then lengthened and becomes longer than the window of transient phase-matching that would occur in a 1D geometry. Moreover, in the far field, due to on- and off-axes phase matching, the fine features of the XUV-pulse time profile may vary over the XUV-beam section [19, 20]. Furthermore, the space-dependent ionization can act as a diverging lens that affects the beam propagation and limits the HHG efficiency [21].

To overcome these limitations, we use a laser beam shaped in such a way that the intensity at the target almost does not vary radially up to a certain distance from the beam axis where it drops rapidly to zero (‘flat-top’ laser beam). The flat-top radial intensity profile provides simultaneous ionization throughout the central part of the beam that is compatible with a well-controlled transient phase matching. Also, with long pulses, the optimization of the HHG efficiency was demonstrated using similar quasi-flat-top beams [22]–[24].

In this paper, we first describe a technique to get a flat-top femtosecond beam at the target and describe our approach for simulating HHG and transient phase matching. We show that using a flat-top laser beam can lead to a single-attosecond pulse generation when adequate geometrical conditions and fundamental pulse duration are selected. We introduce an effective coherence length that allows us to emphasize the role of phase-matching gating by presenting transient phase-matching maps. We then discuss how the technique can be applied to relatively long (20 fs) pulses. We conclude by highlighting the unique potential of this approach.

2. The flat-top laser beam

Spatial shaping of laser beams is well established with monochromatic lasers, whereas femtosecond lasers are by essence polychromatic. Still, even with femtosecond laser beams, it is possible to obtain a quasi-flat-top laser beam at the focal point by using a simple experimental
setup (presented in figure 1). This setup consists of two concentric cylindrical glass plates of similar thicknesses (an annular glass plate with a circular hole (diameter: $\phi_{\text{plate}}$) at its center and the complementary plate, each transmitting a specific part of the beam), a diaphragm and a focusing lens (or mirror). The central part of the collimated fundamental beam crosses the central plate, whereas the outer part of the beam (annular part) crosses the outer plate. The dephasing, $\delta \varphi$, between them is fixed by the thickness of each plate. The energy ratio between these two parts is determined by the central-hole diameter as compared to the beam size ($W_{1/e^2}$ is the radius of the incident Gaussian beam at $e^{-2}$ of the top intensity) and can be further adjusted by an additional iris (of diameter $\phi_{\text{iris}}$).

The two resulting co-propagating beams are then focused and the shape of the laser beam nearby the focus is determined by the former parameters ($W_{1/e^2}$, $\phi_{\text{plate}}$, $\delta \varphi$ and $\phi_{\text{iris}}$) and the position $z_1 < z < z_2$ ($z_1$ and $z_2$ are $z$-coordinates of the target edges) at which the beam profile is observed as compared to the focus position ($z = 0$ defines the focus position for $\delta \varphi = 0$). These parameters can be varied to optimize the field properties at the target. Experimentally, $\delta \varphi$ can be controlled by tilting one of the plates whereas the energy ratio between the inner and outer parts of the beam can be adjusted by varying the diameter of the iris.

In figure 2, we present both the calculated intensity (into vacuum) and phase profiles for the following parameters: incoming pulse energy 170 $\mu$J, pulse duration 10 fs, incident Gaussian
Figure 2. Calculated intensity (a) and phase distribution (b) for the flat-top beam obtained with the setup shown in figure 1.

beam size $W_{1/e} = 1$ cm, iris diameter $\phi_{\text{iris}} = 2.8 \times W_{1/e}$, plate diameter $\phi_{\text{plates}}/W_{1/e} = 1.9$, $\delta \varphi = -\pi$, focal length $f = 1$ m and $z$ between $-1.9$ and $-1.1$ mm. This parameter set allows us to obtain:

- a quasi-flat-top radial intensity profile;
- an intensity slightly decreasing on the axis along $z$, but approximately constant off the axis;
- a small (geometrical) phase variation along $z$ except at the very periphery of the beam.

In the following subsections, we will discuss how these properties influence the attosecond pulse generation.
3. Numerical method

The phase of the fundamental field and its evolution with propagation needs to be known, as it defines the harmonic emission and the phase matching conditions. Writing the fundamental field as

$$\tilde{E}(r, z, t - z/v_g) \exp(ik_0z - i\omega_0t) + \text{c.c.}$$

(where $v_g$ is the group velocity), one obtains the following equation [25] for the complex amplitude $\tilde{E}$

$$\left(2k_0^2 \frac{\partial}{\partial z} + \nabla^2_\perp\right) \tilde{E} = -2k_0^2 \frac{\Delta n_{nl}}{n_0} \tilde{E},$$

(1)

where $n_0$ is the linear refractive index of the neutral gas, $\Delta n_{nl}$ is a nonlinear addition to the refractive index and $k_0 = n_0\omega_0/c$. This equation can be solved numerically (see, for instance [26], where we studied the HHG with the self-channeled laser beam). However, for a thin and low-pressure target, the following approximation can be used. The field amplitude can be written as

$$\tilde{E} = E_0 \exp[i\theta(r, z, \xi)],$$

(2)

where $\xi = t - z/v_g$ and $E_0$ the amplitude in absence of the self-action satisfying the equation

$$\left(2k_0^2 \frac{\partial}{\partial z} + \nabla^2_\perp\right) E_0 = 0.$$  

(3)

Substituting equation (2) in (1) and taking equation (3) into account, we have

$$2k_0^2 \frac{\partial \theta}{\partial z} + (\nabla_\perp\theta)^2 - i\nabla^2_\perp\theta = 2k_0^2 \frac{\Delta n_{nl}}{n_0}.$$  

(4)

Considering that at the front edge of the target ($z = z_1$) $\theta$ is equal to zero, its transverse derivatives can be neglected for short propagation lengths. Then, one obtains the following approximate equation:

$$\theta = \frac{\omega_0}{c} \int_{z_1}^{z} \Delta n_{nl} \left[E(r, z', \xi)\right] dz'.$$  

(5)

Let us estimate up to which propagation length this description is valid. The transverse derivatives in equation (4) can be neglected if

$$(\nabla_\perp\theta)^2, \quad \nabla^2_\perp\theta \ll 2k_0^2 \frac{\Delta n_{nl}}{n_0}.$$  

(6)

After propagation over a distance $L$, the phase shift at the axis can be estimated from equation (5) as

$$\tilde{\theta} \approx \frac{\omega_0}{c} \Delta \tilde{n}_{nl} L,$$  

(7)

where $\Delta \tilde{n}_{nl}$ is a typical value of $\Delta n_{nl}$ at the axis. Assuming $(\nabla_\perp\theta)^2 \approx \frac{\tilde{\theta}^2}{\rho^2}$ and $\nabla^2_\perp\theta \approx \frac{\tilde{\theta}^2}{\rho^4}$, where $\rho$ is a typical transverse size of the beam at the target, one obtains from equations (6) and (7) the following limitations for the propagation length:

$$L \ll L_{\text{lim}} = \rho \sqrt{\Delta \tilde{n}_{nl}},$$  

(8)

$$L \ll b,$$  

(9)
where $b$ is the confocal length. Physically, the description of the field with equations (2) and (5) means that we take into account the nonlinear phase self-modulation, but we neglect the spatial modification of the intensity due to self-action. Correspondingly, requirements (8) and (9) mean that this modification is negligible only up to some propagational length, and for the longer ones the self-action of the beam leads to modification of the intensity profile.

The requirements (8) and (9) are fulfilled for the target thickness considered here, so we use the field (2) where the nonlinear phase-shift (5) is calculated numerically taking into account plasma formation and Kerr nonlinearity. For typical experimental parameters, $L$ is limited by the requirement (8), but not by the requirement (9), because $L_{\text{lim}} < b$. Note that, generally, a flat-top laser beam is less affected by defocusing than a Gaussian beam since the electron density follow the intensity profile and the uniform electron density does not act as a diverging lens. So, for a flat-top beam our approach can be valid even for the target thickness exceeding $L_{\text{lim}}$.

The harmonic single-atom response was calculated as in [3, 27, 28] using the theory described in [29]. The latter approach utilizes the ionization rate calculated via numerical resolution of the 3D Schrödinger equation for a model Ar atom [30]. It also takes into account the Coulomb modification of the free electronic wave packet after ionization. The theoretical calculation of the attosecond pulses in the single-atom response agrees very well with the numerical results [29]. We also checked experimentally that the macroscopic XUV response agrees well with our simulations for optimized generation of isolated attosecond pulse [27, 31], in particular, by CEP-stabilized laser pulses [3]. Overall, this theoretical description has been experimentally validated even for the generation of isolated attosecond pulses [3]. An important feature of our approach is the possibility to separate the contributions of different quantum paths within the frame of the single-atom response. Technically, this allows integration of the rapidly oscillating single-atom dipole to be avoided when calculating the medium response, thus separation of the contributions makes the calculation easier. Moreover, this separation allows the phase matching to be studied separately for different quantum paths and helps in interpreting the results.

4. Isolated attosecond pulse generation with 10 fs IR laser pulse

The simulated attosecond pulses generated in argon with the flat-top laser beam described above (and shown in figure 2) and a 0.75 mm (hard edge) thick target is presented in figure 3. The gas pressure is 60 mbar, and the center of the jet is placed 1.5 mm before the focus (thus $z_1 = -1.875$ and $z_2 = -1.125$). We show the intensity of the XUV with frequencies exceeding $32\omega_0$ (~50 eV), whereas the cutoff is around 60 eV. As we are considering a realistic flat-top profile here, phase matching slightly varies spatially in the gas target (albeit much less than with a Gaussian beam), which induces changes in the attosecond pulse train structure across the XUV beam section after propagation as shown in [19, 20]. Therefore, in order to increase the temporal contrast of the isolated attosecond pulse, we select the XUV light emitted in a reduced solid angle (inside a limit angle) as can be done experimentally by placing a diaphragm in the XUV far-field. For the used parameters, the limit angle is set to $2.8 \times 10^{-3}$ rad, so that about half of the emitted XUV energy is inside the integration area. Under these conditions, an attosecond pulse is predominant and only weak pre- and post-pulses are present.

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Figure 3. Simulated attosecond pulses generated in argon by the 10 fs flat-top laser field presented in figure 2 with a relatively thicker target (0.75 mm) and a very thin layer (7.5 µm); the dashed line presents the laser intensity temporal profile. The XUV intensity with frequencies exceeding $32\omega_0$ is presented. One can see that a train of several attosecond pulses is generated with the thin layer while a single attosecond pulse is enhanced by the transient phase matching when the thick target is used.

To highlight the role of the transient phase-matching, figure 3 also shows the calculated XUV-emission from a hundred times thinner target. The intensity generated in the latter is multiplied by $10^4$ (i.e. by the square of the thickness ratio) to use the same scale. For the thin target, transient phase matching does not affect the emission which follows the single-atom response and the XUV-emission contains several attosecond pulses. We obtain a $\sim 5$ fs attosecond pulse train, shorter than the (10 fs) laser pulse duration because of the nonlinear response of the atomic medium. Here, the depletion of the gas medium, which is about 20% after the pulse, does not significantly shorten the attosecond pulse train. This appears on figure 3 as the attosecond pulse train envelope is centered around $t = 0$ for the thin target. In contrast, for the thick target, the macroscopic XUV-emission is strongly confined by the transient phase matching and only one attosecond pulse is generated efficiently near the peak of the pulse.

5. Transient phase-matching maps

More physical insight on how transient phase matching shapes the XUV temporal pulse structure can be extracted from a detailed analysis of phase-matching maps [11] presenting the coherence length

$$L_{coh}(r, z, \xi) = \frac{\pi}{|\delta k|}.$$  (10)
Here, $\delta k$ is the wave-vector mismatch
\begin{equation}
\delta k = k_q - k_{q}^{\text{pol}},
\end{equation}
where the $q$th harmonic wave vector is $k_q = n_q q \omega_0 / c$, and the polarization wave vector is
\begin{equation}
k_q^{\text{pol}} = \nabla \left[ q k_0 z + q \varphi_{\text{las}}(r, z, \xi) + \varphi_{\text{at}}(r, z, \xi) \right].
\end{equation}
Here, $\varphi_{\text{at}}(r, z, \xi)$ is the intensity-dependent phase of the harmonic atomic response, and $\varphi_{\text{las}}(r, z, \xi)$ is the phase of the laser field. The latter is a sum of the vacuum and the medium-induced terms:
\begin{equation}
\varphi_{\text{las}}(r, z, \xi) = \varphi_0(r, z) + \theta(r, z, \xi).
\end{equation}

The phase $\varphi_0(r, z)$ is presented in figure 2. The variation of the phase-matching conditions in time is controlled by the atomic phase $\varphi_{\text{at}}(r, z, \xi)$ and the time-dependent laser phase $\theta(r, z, \xi)$ given by equation (4).

However, practically, it is certainly impossible to present the 4D map of $L_{\text{coh}}(r, z, \xi)$. One needs therefore to consider some simplified quantity. Here, we introduce an effective coherence length, which describes the phase-matching properties after integration over propagational direction:
\begin{equation}
L_{\text{coh}}^{\text{eff}}(r, \xi) = \frac{\pi}{2} \max_{z_1 < z < z_2} (\Phi),
\end{equation}
where
\begin{equation}
\Phi = \left| \int_{z_1}^{z_2} \exp \left[ i \delta k_z (z_2 - z') \right] dz' \right|,
\end{equation}
where $\delta k_z$ is $z$-projection of the mismatch given by equation (11).

If $\delta k_z$ is constant along $z$ and $z_2 - z_1 > L_{\text{coh}}$, then the effective coherence length is equal to the coherence length for the on-axis emission. If $\delta k_z$ varies along $z$, then the integral (15) can be estimated with a stationary phase method and one sees that the equation (14) gives the size of the region which contributes to the coherent emission. Thus, the suggested definition (14) really describes the effective length of the coherent on-axis generation both for the constant and the varying $\delta k_z$.

The atomic phase $\varphi_{\text{at}}$ for a given harmonic in equation (12) can be approximated [32] as
\begin{equation}
\varphi_{\text{at}}(r, z, \xi) = -\tau U_p(r, z, \xi),
\end{equation}
where $U_p$ is the ponderomotive energy that scales linearly with the intensity and $\tau$ is a travel time (both in atomic units) for the corresponding quantum path. For a plateau harmonic, there are two paths essentially contributing to its generation, both having travel times smaller than the laser period. They are denoted as the short and the long paths, according to their travel times. The difference of travel times in equation (16) leads to the difference in phase matching for different quantum paths.

In figures 4(a) and (b), we present calculated effective coherence lengths for the short and the long quantum paths as functions of $r$ and $\xi$. More precisely, we present $L_{\text{coh}}^{\text{eff}}$ for the two extreme values of the travel time $\tau$ in equation (16): $\tau = 0$ corresponds to the short path and $\tau = 2\pi / \omega_0$ corresponds to the long one. In the figure, the light regions correspond to high coherence length and thus to good phase-matching. To identify the region where the
Figure 4. The effective coherence lengths calculated for the short (a) and long (b) quantum paths, and the far-field XUV intensity (c) simulated for the 0.75 mm target. Other parameters are the same as in figures 2 and 3. The simultaneous transient phase matching at the axis and at the periphery for the short path provides intense isolated attosecond pulse generation. The dotted lines in graphs (a) and (b) show the radius for which the laser intensity is half of its maximum.
high-frequency atomic response is most essential, we show with the dotted line the radius at which the laser intensity is half of its maximum.

We see that for the short path a transient phase-matching ‘gate’ occurs simultaneously for all radii $r$. For the long path this is not the case because the laser intensity varies along the $z$-direction on the axis, but not at the periphery; this variation does not influence the phase matching for the short path, but affects it for the long one due to strong dependence of the long-path atomic phase on the laser intensity (see equation (16)).

Correspondingly, the short path provides a single attosecond pulse with weak satellites (see figure 4(c)), whereas the long path provides an attosecond pulse which is generated later and has stronger satellites. In particular, contribution of the short path to the main attosecond pulse is an order of magnitude higher than that of the longer one, and thus the latter does not make the attosecond pulse essentially longer here. So, the fine features of the intensity variation in our target allow the separation of the emission from the different quantum paths. A little more (not very effective) suppression of the long-path contribution is achieved when selecting the far-field XUV intensity over a smaller solid angle.

6. Robustness against CEP variations

In figure 5, we present the simulated attosecond pulses generated by laser pulses with different CEP, $\varphi_{\text{CEP}}$. This phase defines the position of the laser field with respect to its envelope, and the incident (i.e. before focusing and propagation) field strength considered in the calculation is proportional to $\cos(\omega_0 t + \varphi_{\text{CEP}})$. One can see that a single attosecond pulse is generated for a wide range of $\varphi_{\text{CEP}}$. The contrast (ratio between the main pulse and the strongest satellite intensities) exceeds 2 for all CEPs except for the range $\varphi_{\text{CEP}} \in (\pi/12, 5\pi/12)$. Thus, under these conditions, a single attosecond pulse is predominant for about 67% of the possible CEP values, and even without CEP control isolated attosecond pulses can predominantly be obtained. Experimentally, a post-selection of the pulses showing a continuous spectrum corresponds to selecting isolated attosecond pulses and would not lead to a strong reduction of the repetition rate.

The wide CEP range for the isolated attosecond pulse generation is partly explained by the fact that the temporal window of the phase-matched generation is linked not only to the laser pulse envelope, but also to the CEP because the field controls peculiarities of the ionization dynamics. This is an important difference from the ‘intensity’ [5] and ellipticity [3, 4] gates which both are linked to the envelope of the laser pulse. Correspondingly, these methods require shorter pulses and better CEP stability; both the ‘intensity’ and the ellipticity gates are applicable to generate isolated attosecond pulses with CEP-stabilized pulses [5, 28]. For instance, using the ellipticity gate with 10 fs incident pulse and 10 fs delay provides isolated attosecond pulse generation for only about 40% of the possible CEP values [28].

Using 10 fs pulses to generate isolated attosecond pulses is still a rather stringent constraint since it implies using post-compression of the laser pulses and for this pulse duration, the pulse energy is so far limited to about 5 mJ [33]. Only a few OPCPA-based laser systems actually deliver sub 10 fs pulses with more than 10 mJ [34, 35]. Using 20–25 fs long driving pulses to generate attosecond pulses would not only be more convenient, but would also allow the use of laser systems of much higher energy as delivered now by 10 Hz high-energy lasers even up to the 100 TW class.
Figure 5. (a) Intensity of the attosecond pulses simulated for different values of the CEP of the laser pulse. The target thickness is 0.75 mm, the other parameters are the same as in figure 3. (b) Attosecond pulse intensity profile generated for two specific CEP values.

7. Isolated attosecond pulse generation with 20 fs laser pulse

Using the transient phase-matching scheme with longer pulses is indeed possible. Combining a laser field and its relatively weak second harmonic (SH) as a pump allows the periodicity of the attosecond pulses generated via HHG to be increased from the laser half-cycle to its one cycle [36]–[40]. This provides additional time for changing the phase-matching conditions, and thus allows the use of longer laser pulses for single attosecond pulse generation.

Our simulations (see figure 6) show that attosecond pulse trains are generated with 20 fs pulses for one- and two-color fields in a thin target, whereas an isolated attosecond pulse is generated with a two-color field in the (0.75 mm) thick target after selecting frequencies greater than 37 eV. The IR laser beam is the same flat-top beam as above, and the SH beam is a Gaussian beam focused by the same lens. The SH beam radius at $z = 0$ is $W_{\text{SH,1/e^2}} = 85 \mu m$ and its
Simulated attosecond pulses generated by a 20 fs flat-top laser field alone in a thin target, and by the flat-top laser beam and a weak second-harmonic Gaussian beam in the thick and the thin targets. The XUV intensity with frequencies exceeding $24\omega_0$ ($\sim 37$ eV) is presented.

We see that in the presence of the SH field, the attosecond pulses are not just more rare, but also more intense. This occurs because the ionization is enhanced at every second laser half-cycle, when the two fields add up. In the thick target, one of the attosecond pulses in the train is selectively enhanced by the transient phase matching.

We have also investigated the influence of the CEP on the attosecond pulse time structure in the two-color configuration, keeping the same relative phase $\varphi_{\omega_0-2\omega_0}$ (see figure 7). When the relative phase is fixed, one should consider $\varphi_{\text{CEP}} \in [0, 2\pi]$ (and not $[0, \pi]$), because the periodicity of the emission is an IR optical cycle for the two-color field. It is clearly observed that an isolated attosecond pulse is predominant even for a wider CEP range than for 10 fs, one-color field (figure 5). Indeed, a single attosecond pulse is generated for about 80% of the possible CEP values. Moreover, the contrast is also better than that with the 10 fs one-color IR pulse.

Let us now discuss the spectral width and the duration of the obtained attosecond pulses. Generally, the transient phase-matching gating allows the generation of broadband attosecond pulses since the phase-matching condition depends only weakly on the harmonic order (the phase mismatch is mainly imposed by the phase variation of the IR field since for the harmonics, the refraction index is very close to unity). Thus the phase-matching gating is effective both for the plateau and the cut-off XUV.

Note however that for low-order harmonics, the absorption length is shorter than the target thickness considered here. Correspondingly, the phase mismatch is accumulated along a shorter
Figure 7. Attosecond pulses generated by two-color field under different CEPs of the 20 fs IR laser pulse. The relative phase between the IR and the second harmonic is fixed so that peaks of the IR field coincide with those of the SH (see text for more details). The XUV intensity with frequencies exceeding $24\omega_0$ (a), (b) and $12\omega_0$ (c) are presented.

length and the transient phase-matching confinement is less efficient than that for the higher harmonics.

In order to study the influence of low-order harmonics but also to closely match to realistic experimental conditions, we have calculated attosecond pulses when selecting XUV frequencies higher than $12\omega_0$ (as can be experimentally transmitted by an Al filter) and using in the XUV far-field a diaphragm smaller by a factor of 1.5 as compared to the previous value to improve the contrast (see figure 7(c)). We observe that the contrast is nevertheless not as good as that in figure 7(b), because the lowest order harmonics are less confined by the transient

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phase-matching. But, on the other hand, the attosecond pulse is significantly shorter because of the broader bandwidth (170 as against 300 as).

Another way to broaden the spectra of the attosecond pulses would be to increase the cutoff by increasing the peak intensity but with this approach, one cannot arbitrarily change the peak intensity without affecting the transient phase-matching window. Roughly, the generation is phase-matched when the neutral gas dispersion is compensated with the dispersion of the free-electrons appearing due to ionization. In our conditions, the phase-matching window occurs before the laser pulse maximum (see figures 3–6), and the laser intensity that efficiently generates XUV is lower than the peak intensity. The cutoff has therefore to be estimated at that time, and is found to be lower than the maximum cutoff. This intensity depends on the pulse intensity temporal profile, and the gas type. The shorter the laser pulse, the higher this intensity, and thus higher the cutoff in the XUV spectrum. A higher cutoff results in broader spectra and shorter attosecond pulses. For instance, in our conditions, the single-atom response cutoff approximately occurs at the 41st harmonic, whereas in the spectrum generated in the thick target the cutoff is near $38\omega_0$ for the 10 fs pulse (for parameters of figure 5) and near $34\omega_0$ for the 20 fs pulse (for parameters of figure 7). Correspondingly, by using the 10 fs laser pulse, we obtain shorter attosecond pulses than using the 20 fs one (see figures 5 and 7(a)). For a given pulse duration and interaction geometry, the maximum achievable bandwidth is therefore limited by the cutoff at the time of phase matching. In return, this implies that transient phase matching is robust against shot-to-shot intensity variation, as soon as the peak intensity is larger than the transient phase-matching intensity.

8. Perspectives

In this subsection, we discuss the perspectives of the ‘transient phase-matching gating’ approach and address an overview of the further studies.

In figure 2, one can see that in our conditions, the region where the transverse laser intensity distribution remains flat-top is limited in the $z$-direction (less than 1 mm). This ‘flat-top length’ limits the thickness of the target for which the transient phase matching is simultaneous at the axis and at the periphery of the beam. We have chosen quite a modest energy of the laser pulse to validate the principle of the method. With more energetic pulses, one can afford longer focal lengths thus keeping the same intensity in a longer ‘flat-top length’ and use a thicker target. As the confinement effect varies with $\delta k L$, using longer targets would lead to higher contrast and make the method valid for longer laser pulses.

For instance, using a 30–40 fs single-color pulse instead of the 10 fs pulses considered here would induce smaller $\delta k$ steps from one half-cycle to the next. A good temporal contrast in the attosecond pulse can be preserved by keeping an equivalent $\delta k L$ and hence by using a longer medium. In practice, the medium length can be extended up to a limit of a few times the absorption length [15]. In the case considered here (figure 3, $\omega_{XUV} > 32\omega_0$, 60 mbar of argon) the absorption length is about 6 mm. Thereby, the medium length considered before (750 $\mu$m) can be increased by a factor of 10–20 (provided the flat-top length is long enough, which in return calls for high-energy lasers) and this can compensate for the reduced $\delta k$ steps. However, using longer pulses would result in achieving this transient phase-matching at a time when the intensity is lower than that in the presented calculations. This implies that the single-atom response is reduced in the phase-matching window and results in a lower XUV yield and a less-extended cutoff. Still, a single-attosecond pulse emission seems possible when using longer
IR pulses as delivered by standard high-energy laser systems. However, the longer the medium, the more sensitive phase-matching gets to the ionization rate and the transverse uniformity of the flat-top beam gets more crucial in that case to ensure a constant ionization yield. Therefore, a better control of the flat-top profile can be necessary as can be achieved with more sophisticated spatial shaping techniques. In any case, our simulations showed that it is rather easy to find sets of parameters compatible with the emission of isolated attosecond pulses based on transient phase-matching gating.

As we mentioned, this approach is compatible with high-energy laser pulses. For instance, using a 10 mJ laser pulse and considering a harmonic generation efficiency of the order of $10^{-5}$ per harmonic (typical for argon), one can expect to get an attosecond pulse with an energy on the order of $1\mu J$ ($10\text{ mJ} \times 10^{-5} = 0.1\mu J$ per harmonic; summing over 10 harmonics gives $1\mu J$ per attosecond pulse). Even higher efficiencies could be obtained since this typical efficiency is estimated with the Gaussian beams where the optimization cannot be achieved everywhere throughout the beam. Indeed, our simulations showed that the net optimized efficiency was always comparable or higher by using the flat-top beam as compared to a Gaussian beam.

In the approach of the ‘transient phase-matching gating’ developed here, the width of the temporal window for the phase-matched generation is controlled not by the laser pulse duration itself but by the steepness of the rising front of the pulse. By making the pulse asymmetric one can create a rising front that is sharper than that in the symmetric one; so, by using an asymmetric laser pulse one could obtain an isolated attosecond pulse with even longer laser pulses than the ones considered here.

The role of dephasing between the laser and the second harmonic will be the subject of further research. In particular, the SH field could be a way to control the efficiency of each quantum path. Moreover, optimal focusing conditions of the SH could emphasize the generation from the central part of the beam and thus depreciate the contribution from the wings where the deviation from the ideal flat-top profile makes the phase-matching window longer.

We observed that with this technique, it is possible to produce broadband attosecond-isolated pulses with relatively long (10–20 fs) intense pulses. When available, using even shorter pulses can still be attractive in the sense that ultra broadband attosecond pulses could then be obtained. Similarly, controlling the CEP would also help in stabilizing the output of the system. This approach will clearly remain interesting even if the laser technology keeps evolving so quickly.

Finally, it must be stressed that using a flat-top laser beam is a way of efficiently using high-energy laser pulses and getting optimum interaction intensity without using focusing optics with prohibitively long focal lengths. The approach suggested here for spatial shaping using two transparent plates is clearly a rough one but it can easily be improved by using more sophisticated adaptive optics such as deformable mirrors. Generally, the flat-top profile allows all the laser energy to be concentrated in the high-intensity central part without losing energy in the weak-intensity wings. This property is very promising for applications of such beams in strong-field physics.

9. Conclusions

We have shown in our simulations that isolated attosecond pulses can be efficiently generated by high-order harmonic emission gated by ultrafast transient phase matching. A crucial
requirement is to reach the phase matching at the same time throughout the generating medium. This is realized using a flat-top radial intensity profile that can be achieved with the simple technique we have proposed. The calculation is first performed for moderate input laser energy and a 10 fs IR pulse and thus on a rather thin gas target which has already allowed us to demonstrate the possibilities of this approach. The main features of the method are the possibility of isolating a single attosecond pulse when using relatively long driving pulses, its robustness against CEP fluctuations and the fact that the confinement occurs when the emission is maximized both on the microscopic and macroscopic levels ensuring thereby an efficient emission of the attosecond pulses. A second set of simulations is performed with longer IR laser 20 fs pulses when adding its second-harmonic field. It also successfully shows that a single attosecond pulse can be obtained through two-color transient phase matching. Finally, an isolated 170 as pulse can be obtained when frequencies above 12ω₀ are selected which corresponds to harmonics transmitted by an Al filter thus approaching realistic experimental conditions as close as possible.

The results of such calculation can be favorably scaled-up to larger energy laser systems even with slightly longer pulses. Favorable parameters to reach microjoule energy level in a single attosecond pulse could be achieved by using realistic long focal length focusing optics, a longer gas cell, adding temporal pulse shaping and/or optimizing the overlap (both in time and space) of the IR pulse and its second harmonic fields.

Forthcoming research in high-energy post-compression, CEP stabilization of high-energy lasers, high-energy OPCPA-based systems and high-energy short pulsed lasers will offer the possibility to test the different alternatives proposed in this paper. This would provide isolated attosecond pulses at high energy that are intense enough to induce nonlinear transitions. Therefore, a new important step in attophysics would be reached as the whole range of pump/probe techniques would become accessible. In particular, this will allow XUV/XUV pump/probe experiments with an attosecond resolution which is actually a major challenge and is not accessible with any other XUV source.

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