Generation of tunable isolated attosecond pulses in multi-jet systems

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\textbf{Abstract.} We theoretically investigate how the generation of attosecond pulses from high-order harmonics can be controlled by using a specially designed sequence of gas jets. We demonstrate that quasi-phase-matching provided by such a multi-jet system can be limited to a sub-femtosecond time window, while adjusting the multi-jet structure allows tuning of the central frequency of the generated isolated attosecond pulse.

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1. Introduction

A powerful source of short coherent extreme-ultraviolet (XUV) and x-ray pulses would find many applications in science and technology. While the most intense pulses in this spectral range are delivered by free-electron lasers, the shortest pulses down to the attosecond regime [1]–[3] are currently obtained by means of high-harmonic generation (HHG) in gases. Although these pulses have already been used in time-resolved experiments [4, 5], their applicability is limited by the relatively low intensity as well as by the lack of tunability—currently it is impossible to tune the photon energy of attosecond pulses. Controlling the HHG process has been considered as a possible way to overcome these deficiencies.

A possible approach to control HHG and, consequently, the generation of attosecond pulses is shaping the driving laser pulse [6]–[9]. A second approach focuses on controlling the HHG process itself rather than controlling the driving pulse. In this case, it was shown that the conversion efficiency can be improved by quasi-phase-matching (QPM) in a counter-propagating beam configuration [10], in a diameter-modulated hollow-core fibre [11, 12] or by using a system of a few gas jets placed one after another along the propagation direction [13]. Recently, it has been shown that QPM can be used to selectively phase-match contributions from either short or long electron trajectories driven by a long laser pulse [14].

In this paper, we show that multi-jet systems (MJSs) may, beyond enhancing the harmonic photon flux, also allow efficient control of the spectral and temporal structure of the harmonic field generated by a few-cycle laser pulse. We demonstrate that QPM provided by a specially designed MJS is able to enhance selected harmonics generated within a sub-femtosecond time window. Our approach opens the door for the generation of single attosecond pulses with enhanced flux and tunable photon energy.

The purpose of an MJS is to improve phase-matching, i.e. to increase the length over which the harmonic field can coherently build up. This is achieved by limiting the thickness of each gas jet \( L_g \) to the coherence length \( L_c \) for a selected harmonic frequency. Between the jets, the laser and harmonic fields propagate in vacuum, and their spatio-temporal profiles change owing to diffraction. If the width of the vacuum region \( L_v \) is chosen properly, phase-matched propagation can be re-established at the beginning of the next jet.

The mechanism by which an MJS controls the time structure of the harmonic field, can be described within the three-step model [15, 16]. In this model, an outer-shell electron tunnels through the potential barrier formed by the combined Coulomb and laser fields, then moves almost freely in the laser field, returns back to the parent ion, and finally recombines to its initial state emitting a photon with energy equal to the final kinetic energy of the electron plus the ionization potential. In general, electrons returning along several trajectories can emit photons with the same energy. At the recollision moments of these trajectories, ‘bursts’ of harmonics are radiated. Different bursts, generated at different moments during the laser pulse, experience different phase-matching conditions while propagating in an MJS because these conditions are usually determined by the concentration of free electrons, which grows from the front of the laser pulse towards its tail.

2. Numerical simulations

In order to simulate the generation of high harmonics (HH) and their coherent build-up in an MJS, we used a three-dimensional (3D) non-adiabatic model [9], which provides the laser...
field \( E_L \) propagating through the ionized gas, the single-atom dipole response \( P_{\text{nl}} \) estimated in the strong field approximation using the Lewenstein model \([17]\) and the electric field of the generated harmonics \( E_H \) (due to the cylindrical symmetry of the light beams, the propagation is effectively reduced to two spatial coordinates). The model also accounts for the absorption and dispersion of harmonics as they propagate. In all the simulations presented in this paper, the propagation of the laser pulse was performed in all spatial dimensions. However, to accelerate the simulations, the generation and propagation of harmonics were simulated on the beam axis only, with the exception of the results shown in figure 5(d), for which off-axis harmonics were simulated as well.

Central to our approach is the algorithm that automatically builds an optimal MJS for a selected burst of harmonics. Simulating the propagation of the laser and harmonic fields, the on-axis intensity of a selected harmonic burst was controlled after each step \( \Delta z \). Whenever the intensity of the burst increased, the step was considered as a part of a jet; otherwise, the propagation was repeated with the zero (or strongly reduced) gas pressure assigning the step to the inter-jet space. As a result, we obtained an MJS optimized for the on-axis near-field intensity of the selected harmonic burst together with the harmonic field emerging from the MJS.

In order to maximize coherent harmonic build-up in a series of gas jets, it is important that the laser pulse should not be distorted too much as it propagates over this length. This implies that the intensity of the laser field must be as small as possible in order to avoid strong ionization, since the interaction with plasma defocuses and blue-shifts the laser pulse \([1, 9]\). Recently developed sources of phase-stabilized few-cycle pulses with a carrier wavelength of \( \lambda_0 = 2.1 \mu m \) \([18]\) offer a possibility to generate high-order harmonics without using high laser intensities because the ponderomotive energy for this wavelength is seven times larger as compared to the ponderomotive energy provided by conventional 0.8 \( \mu m \) pulses with the same intensity.

In our simulations, we assumed a 15 fs 1.5 mJ Gaussian pulse with a central wavelength of 2.1 \( \mu m \), which was focused 5 mm after the first jet to a beam waist of \( w_0 = 92 \mu m \). The initial on-axis peak intensity of this pulse was equal to \( 6.6 \times 10^{14} \) W cm\(^{-2} \). This intensity ionizes about 0.5% of He atoms and generates harmonics with the cutoff at 770 eV, which corresponds to a harmonic order of 1300 (H1300). The He pressure in each jet was assumed to be 200 Torr.

### 2.1. Build-up of harmonic bursts in a single jet

We first present results obtained in one single jet, our attention being focused on the trajectories contributing to H301 (177 eV, well in the plateau). Figure 1 shows the time dependence of the initial on-axis single-atom dipole response. It was obtained by performing the inverse Fourier transform of \( P_{\text{nl}}(\omega) \) filtered by a rectangular spectral window from \( 280\omega_0 \) to \( 320\omega_0 \), where \( \omega_0 = 2\pi c/\lambda_0 \) is the carrier laser frequency, \( c \) being the vacuum speed of light. Each burst of radiation in figure 1 corresponds to an electron trajectory that was identified from a classical calculation \([19]\). We singled out the trajectories that significantly contributed to the harmonics in the selected spectral range. From the electron emission and recollision moments one can identify the long (L) and the short (S) trajectories.

The harmonic bursts generated by different trajectories can be separately observed and analysed while advancing the propagation, as shown in figure 2. In this figure the intensities of the bursts are plotted as functions of the propagation distance \( z \). We observe two different regimes. Harmonic fields \( E_{H2} \), \( E_{H3} \) and \( E_{H4} \), generated earlier in the pulse, demonstrate the first
Figure 1. The time dependence of the HH emission in the spectral range from $280\omega_0$ to $320\omega_0$. Each burst of radiation corresponds to an individual trajectory labelled as long (L) or short (S), according to the excursion time of the electron. Arrows indicate birth and recollision times of long electron trajectories.

Figure 2. A single-jet simulation showing the build-up of the harmonic intensity for some of the trajectories (the labels are the same as in figure 1).

regime: they exhibit regular oscillatory behaviour with a large and constant coherence length $L_c$, similar to the Maker fringes reported in [20].

Harmonic fields generated by trajectories like L7 and L9 demonstrate the second regime: after a few rapid oscillations, which indicate a small $L_c$, there is a sharp increase in $E_{H}^{L7}$ and $E_{H}^{L9}$, indicating a highly coherent growth of harmonic radiation. This increase occurs when the contribution to the harmonic phase induced by the distortion of the laser pulse upon propagation compensates for the contribution caused by the refractive index of the plasma, which results in
Figure 3. Harmonic intensities $|E_{H}^{L7}|^2$ (black solid line) and $|E_{H}^{L3}|^2$ (dashed red line) as functions of the propagation distance. The pressure profiles of the MJSs are also shown: the upper level corresponds to 200 Torr, the lower one corresponds to 20 Torr.

transient self-phase-matching for one of the trajectories [21]. This mechanism, which is similar to non-adiabatic self-phase-matching [13, 22], leads to an increase of the HH signal by orders of magnitude.

From figure 2, one can also infer the temporal structure of the harmonic field in the case of a single jet: after 2 mm of propagation, harmonic emission is composed of two dominant bursts of comparable intensities generated by trajectories L7 and L9, as well as much less intense satellites corresponding to trajectories L3 and S4. The emission times of these bursts are basically those shown in figure 1.

2.2. MJSs

Next, we present results obtained in MJSs. We used our algorithm to design systems for enhancement of harmonics around H301 providing phase-matching either for the contribution from trajectory L3 or, alternatively, for the contribution from trajectory L7. To identify the harmonic bursts, we again used a rectangular spectral window centred at H301 and spanning over 40 harmonic orders. The structures of the obtained MJSs as well as the intensities of the corresponding harmonic bursts are shown in figure 3. For trajectory L7, we started the optimization procedure only after the self-phase-matching set in, while for trajectory L3 the algorithm was started from the beginning of the propagation. In these MJSs, the intensities $|E_{H}^{L3}|^2$ and $|E_{H}^{L7}|^2$ increase approximately as $z^2$, which is evidence of phase-matched harmonic generation. In particular, after 2 mm of propagation, $|E_{H}^{L3}|^2$ and $|E_{H}^{L7}|^2$ increase by factors of 55 and 35, respectively, as compared to a single 2 mm thick jet (the harmonic intensities in figures 2 and 3 are measured in the same units).

It is specific to HH generated by few-cycle driver pulses that phase-matching conditions may significantly vary within a driver pulse. When an MJS is designed for a specific trajectory,
Figure 4. On-axis harmonic spectra obtained after 1 mm of propagation in two different MJSs, both designed to enhance $E_{L7}^{12}$, but in spectral windows centred either at H301 (a) or at H451 (b). The inset in (a) shows the spectrum after 1 mm of propagation in a continuous jet.

the fields emitted by remaining trajectories are not phase-matched, so that their intensities remain comparable to those observed in the absence of QPM. An example is provided by figure 3, where we show the build-up of the contribution from trajectory L7 in the MJS optimized for trajectory L3, as well as the intensity of trajectory L3 in the MJS optimized for trajectory L7. Final intensities of these contributions are comparable to those shown in figure 2, and they are more than one order of magnitude smaller than the intensities of their counterparts obtained under optimal phase-matching conditions.

We also applied the optimization procedure for a short trajectory, like S4, and obtained a regular multi-jet structure with an average jet thickness of $L_g = 130 \mu m$ and an average separation between jets of $L_v = 155 \mu m$. However, due to the small dipole response for this trajectory, even after 1 mm of propagation in the MJS, the intensity $|E_{S4}^H|^2$ was still an order of magnitude lower than $|E_{L7}^H|^2$ generated in the same MJS.

Figure 4 demonstrates that the photon energy, at which an MJS efficiently enhances harmonics, can be tuned by adjusting the parameters of the MJS. We designed two MJSs to phase-match harmonic bursts originating from trajectory L7. This trajectory was chosen because after 250 $\mu m$ of propagation the contribution from L7 is larger than the contributions from other trajectories, so that ensuring phase-matching for this harmonic burst results in an attosecond pulse with smaller satellites. The first MJS was designed for enhancement of harmonics around H301. In this design, the average jet thickness was approximately equal to the average jet separation: $L_g = L_v = 55 \mu m$. The second MJS was designed for the coherent build-up of harmonics around H451, which was achieved with $L_g = 60 \mu m$ and $L_v = 30 \mu m$.

While the harmonic spectrum generated in a single 1 mm thick jet exhibits the usual plateau with a large modulation depth between individual harmonics (inset in figure 4(a)), each of the MJSs generates a spectrum in which a group of about 50 harmonics around either H301 or H451
Figure 5. Attosecond pulses generated in three different MJSs. The trajectory and the harmonic order chosen for phase-matching is specified for each case. Panels (a), (b) and (c) contain the near-field on-axis time-dependent intensity of the pulses, while panel (d) shows the far-field power, calculated for a pinhole of 100 µm diameter placed 40 cm behind the interaction region.

rises above the plateau. Such an amplified structure has a relatively small modulation depth and a full-width at half-maximum (FWHM) of approximately \(42\omega_0\). These spectral features indicate that one of the generated harmonic bursts is much more intense than the others. Indeed, by filtering these groups of harmonics with the same spectral window as the one used to design the MJS and evaluating the inverse Fourier transform we obtained the harmonic bursts shown in figure 5. The dominant HH emission occurs at the emission moment of the specific trajectory that was chosen for optimization, as seen from comparison with figure 1. The FWHM of the pulses is around 185 as (more precisely, \(0.025T_0\) for L7 and \(0.028T_0\) for L3, where \(T_0 = 2\pi/\omega_0 = 7\) fs is the period of oscillations of the laser field). Figure 5 demonstrates the possibility to select, by using an appropriate MJS, a single harmonic burst and to suppress the others. Moreover, the possibility to select the emission moment earlier or later within the laser pulse may give control over other characteristics of the emitted field, like the harmonic chirp. At this point, we mention that the emission time within the pulse can be additionally tuned by tuning the carrier-envelope phase (CEP), as we assumed CEP-stabilized pulses. The peaks in figure 1, generated with a cosine pulse (CEP = 0), will shift to later times by \(T_0/4\) if the driving pulse is a sine one. Calculations show that in this case, the contribution from trajectory L7 gives a less intense signal and experiences a smaller coherence length as it develops in a higher plasma density, while trajectory L5 yields the most intense harmonic burst at the end of the jet. Using the CEP as a controlled parameter provides an additional degree-of-freedom for designing MJSs.
The first MJS calculations [13] aimed at maximizing the harmonic intensity in a certain spectral window, making no distinction among different trajectories. This method ensures an improvement in the harmonic intensity, but it does not guarantee control over the temporal characteristics of the emitted radiation, unless there is a dominant trajectory that gives an overwhelming contribution to the total HH field. Also, $L_v$ was obtained using the Gouy phase shift of the unperturbed beam. However, the laser field does suffer distortions during the propagation in a plasma, and it develops a phase shift in vacuum that is different from the Gouy phase shift. Additionally, since the phase of the single-atom harmonic emission depends on the action associated with classical electron trajectories, the optimal values of $L_v$ and $L_g$ are trajectory-dependent (see figure 3), and they are sensitive to how the intensity and the CEP of the laser pulse change as the pulse propagates. Most importantly, the self-phase-matched harmonic generation, if it takes place, should not be interrupted by a vacuum region, as was the case in [13]. Our approach is to use the 3D model to simulate the pulse propagation and to exploit the trajectory-selective self-phase-matching occurring in the first jet. With this method, we could combine the benefits provided by self-phase-matching (the temporal and spectral selectivity and an increased conversion efficiency) with that provided by QPM, demonstrating that effects like the non-adiabatic self-phase-matching can improve conventional phase-matching mechanisms, such as QPM.

In our simulations, we used the near-field harmonic intensity for automatic MJS generation. Even though it is straightforward to generalize our approach using the far-field intensity of a selected harmonic burst to optimize an MJS, this would make the simulations much more time consuming. We found, however, that an MJS built-up using only the on-axis harmonic field can still show good temporal selectivity when the generation of HH in this structure is simulated in 3D. Analysis of our 3D results shows that QPM efficiently enhances the HH radiation in a narrow region of a few micrometres around the beam axis. Outside this region, the HH intensity in the chosen spectral window quickly decreases, which is not surprising since the MJS is optimized for on-axis harmonics, while, off axis, the phase-matching conditions change due to the radial variations of the laser intensity and phase. For the MJS designed to select trajectory L7, we assumed a pinhole of 100 $\mu$m diameter placed 40 cm away from the interaction region and applied the same rectangular spectral filter from $280\omega_0$ to $320\omega_0$. In the time domain, we obtained (see figure 5(d)) an intense pulse at $t = 9T_0$, which is generated by trajectory L7, and also less intense double peaked emissions at $t = 8T_0$ and $8.5T_0$, which correspond to trajectories L3 + S4 and L5 + S6, respectively. Even though the contrast is not as good as that in figure 5(b), the power contained in harmonic burst L7 is three times larger than the total power contained in the remaining bursts, which is remarkable since only near-field harmonics on the beam axis were used to design the MJS. The size of the pinhole was chosen from the analysis of radial distributions of energies contained in individual bursts (figure 6). At distances $r = 30, 100$ and $150\mu$m from the axis, the contribution from trajectory L7 dominates over contributions from other trajectories, but the overall harmonic intensity is small for $r > 50\mu$m, as shown in figure 6(b).

We also analysed how an imposed minimum length of the first gap and a gradual change of the gas density at jet boundaries influence the harmonic signal. This was done in two steps: in the first calculation we assumed rectangular gas density profiles and set the thickness of the first jet to 266 $\mu$m, which maximized the output from this jet. The following vacuum region was required to be at least 300 $\mu$m thick, and then the automatic MJS build-up was started. This procedure increased the thickness of the first vacuum region to 374 $\mu$m and generated a
succession of gas and vacuum regions shown in figure 7. The intensity of the harmonic burst generated by trajectory L7 in dependence on the propagation distance is shown by the dashed line. In the second calculation, we used the obtained gas density profile and substituted the abrupt changes of the density at jet boundaries by linear gradients. In addition, a background pressure of 40 Torr between the jets was used to approach realistic experimental conditions. One can observe that the coherence of the process is affected, but not destroyed: the signal continues to grow in each gas jet, but it decreases slightly between jets due to the destructive interference with harmonics generated in the inter-jet regions.

The dimensions of our multi-jet designs present a significant challenge to the available technology. Thicker jets separated by a larger distance can probably be obtained by a more careful choice of parameters, such as the position of the laser focus, the beam waist, the gas pressure, etc. In our simulations, we were able to cover only a limited range of the parameters, and there is certainly room for further improvement. Also, in our design procedure, we required that the harmonic intensity should monotonically increase within each jet, while the separation between the jets was set to the minimal distance that restored phase-matching. These requirements simplified our simulations, but they are not absolutely necessary to achieve the spectral and temporal selectivity of QPM because phase-matching is retained if $L_g$ and $L_v$ are increased by amounts that change the phase difference between the selected harmonic burst and the atomic dipole response by an integer multiple of $2\pi$. Another technical difficulty could arise from the fact that the MJS scheme generates slightly different gas thicknesses for successive jets. This drawback can possibly be overcome by individually adapting the gas pressure in each jet. Varying the pressure in a particular jet has an effect similar to that of varying the jet thickness [14], and this can be used for fine tuning of multi-jet structures.
Figure 7. The influence of gas density gradients and background pressure on QPM. The ‘ideal’ MJS with abrupt changes of the gas density and the zero background pressure was designed to phase-match the contribution from trajectory L7 to harmonics around H301. The ‘real’ MJS has density gradients at jet boundaries and a background pressure of 40 Torr.

3. Conclusions

We studied the HHG in MJSs that were designed to control the harmonic output in selected temporal and spectral ranges. For a few-cycle pulse, we demonstrated the selective enhancement of a harmonic burst generated by a specific electron trajectory in a certain optical cycle. This can be achieved by building a MJS that favours phase-matching for the chosen harmonic burst, especially if this burst is pre-selected by self-phase-matching occurring in the first gas jet. Our analysis reveals that one can exploit the QPM mechanism to obtain a single attosecond pulse in a spectral range that can be chosen \textit{a priori}. This opens a route to a source of single attosecond pulses with a tunable carrier frequency.

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