Regulating the higher harmonic cutoffs via sinc pulse

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Abstract

We theoretically investigate the generation of higher harmonics and the construction of a single attosecond pulse (ASP) by means of two oppositely polarized sinc-shaped driver pulses. In comparison to a few-cycle Gaussian pulse of the same energy, here we observe a significant broadening in the bandwidth of the XUV/soft-x-ray supercontinuum spectrum in the synthesized pulse. Furthermore, we observe that the harmonic cutoff and its corresponding intensity follow a well-defined scaling with the delay parameter between the two pulses. In principle, this delay can easily be tuned on an optical bench. The typical nature of the synthesized pulse ensures the generation of a single ASP instead of a pulse train. In this case, we obtain a single ASP with a duration of \(\sim 27\) attoseconds in the XUV/soft-x-ray regime of the electromagnetic spectrum. Depending on the delay parameter we observe an enhancement in some satellite harmonics. The proposed setup promises a highly tunable source of energetic photons, wherein the energy of the photons can easily be controlled from the XUV to the soft x-ray regime by simply changing the delay between two oppositely polarized sinc-pulses.

Keywords: higher harmonic generation, attosecond pulse, sinc pulse, strong-field-approximation, TDSE

(Some figures may appear in colour only in the online journal)

1. Introduction

The last decade has witnessed rapid development in the field of higher harmonic generation (HHG), both on the experimental and theoretical front [1]. The HHG and consequently the production of attosecond pulses (ASPs) have enabled researchers to probe the fundamental processes of atomic and molecular phenomena with unprecedented resolution. The ASP is particularly important for investigating electron correlation effects and observing characteristic temporal delays in photoemission from different atomic orbitals [2, 3]. The ASP can also probe the detailed microscopic motion of electrons in atoms, molecules, or any other nanoscale structures, and those effectively bridge the gap between various fields of basic sciences [4–8].

The HHG originates from the interaction of an intense laser with gas atoms, which leads to the generation of coherent radiation at higher harmonics of the laser frequency. Experimentally, it has been observed that higher harmonic spectra consist of a plateau where the harmonic intensity is nearly constant over many orders of magnitude followed by a sharp cutoff [9, 10]. Ever since the inception of the idea of the generation of higher harmonics by laser–atom interaction, research objectives around the globe have been aimed toward the enhancement of the harmonic cutoff of the HHG and also to increasing the corresponding intensity of the emitted harmonics. There are various studies devoted to the achievement of these goals. The effect of the pulse chirp [11–16], the pulse duration [17–19], the synthesis of a laser pulse using two or more color laser fields [20–24], and plasmonic fields [25–28] on the harmonic cutoff and the intensity of the emitted harmonics has been reported in the past. Moreover, some
studies show that the phase of emitted harmonics is greatly influenced by the harmonic emission time and the carrier-envelope phase of the driving laser pulse [29, 30]. The fundamental motivation behind the enhancement of the harmonics (in both the cutoff and the intensity) is the generation of an intense single attosecond pulse (ASP) instead of an ASP train [2, 31–36].

The harmonics at the cutoff of the HHG spectrum are emitted in a short time with a relatively constant phase. The superposition of these harmonics can lead to the generation of an isolated ASP [37–40]. On the other hand, the harmonics lying in the plateau region are generated by two primary electron paths (the so-called short and long paths) having two clearly distinguished ionization and recombination times. The superposition of these plateau harmonics leads to a short light burst of sub-femtosecond or attosecond duration separated by twice the laser frequency, and for a multi-cycle driving pulse, the superposition of plateau harmonics is observed to produce an ASP train [7, 41].

The typical shape of the sinc function makes it very interesting from the perspective of the generation of higher-order harmonics. The sinc pulse has a flat top spectral distribution, and also has a single relatively extreme field amplitude which can assist in accelerating the electron to achieve higher kinetic energies. If the electron recombines with the parent ion, then the excess energy will be emitted in the form of energetic photons. Pulse-shaping techniques such as optical frequency combs have been around for approximately three decades [42, 43]. However, after the advent of optical arbitrary waveform generation, pulse shaping has mainly been achieved by trains of identical optical pulses produced by mode-locked lasers [44]. These mode-locked lasers can have a pulse duration of a few femtoseconds with a repetition rate of a few gigahertz. In order to produce an arbitrary optical waveform, one needs a pulse shaper which can be updated for each pulse. This facet of pulse shaping is being actively worked upon by various researchers in the laser fraternity. We believe that sinc pulses can be generated by state of the art pulse-shaping techniques such as deformable mirrors, spatial light modulators, acousto-optic modulators, and many more. By directly synthesizing rectangular shaped, phase-locked frequency combs, researchers have reported the generation of sinc-shaped pulses of exceptional quality [45]. Contemporary technological advances have further increased the feasibility aspects of high-power sinc-shaped pulses for the near future.

In this work, we discuss a simple setup using sinc laser pulse(s), which promises a highly tunable harmonic cutoff in similar intensity ranges. This, in turn, translates to the realization of tunable radiation sources having photon energies ranging from XUV to soft x-rays of the electromagnetic spectrum. The theoretical and simulation aspects of the work are discussed in section 2, followed by the results and discussion in section 3. The self-contained theoretical formulation using the strong-field approximation along with the numerical implementation is discussed in section 4, and finally concluding remarks are made in section 5.

2. Theory and simulation details

The schematic diagram representing the setup is presented in figure 1. At present, there are no oscillators that can directly generate a sinc-shaped laser pulse. In view of this, a pulse shaper is introduced which transforms the incoming standard Gaussian laser into a sinc-pulse. The iris is placed in front of the pulse shaper to regulate the pulse energy. The shaped output pulse is then passed through the beam splitter. One of the pulses acquires a phase of 180° due to the odd number of reflections from the mirrors, and then interferes with the other pulse [46]. The distance d, as shown in the figure (mirror assembly in the bottom arm), can easily be tweaked on an optical bench, which in turn induces a path difference between the two pulses that results in a delay between them. This indicates that the delay parameter can be tuned easily. With this, we will see that the delay between two pulses does indeed cause a shift in the harmonic cutoffs. The temporal profile of the electric field of the synthesized pulse is then represented as:

$$E(t) = E_0(\tau) \left[ \frac{\sin(\omega_0(t - t_0 - \tau))}{\omega_0(t - t_0 - \tau)} - \frac{\sin(\omega_0(t - t_0))}{\omega_0(t - t_0)} \right] ,$$

where the laser frequency \( \omega_0 = 0.057 \text{ a.u.} \), \( \tau \) is the delay between the pulses, \( E_0(\tau) \) is a delay-dependent field amplitude, and \( t_0 \) introduces some constant phase. Note that \( \tau = 0 \) corresponds to the out-of-phase addition of the pulses which results in \( E(t) = 0 \) (refer to figure 1). Irrespective of the delay parameter \( \tau \), the pulse energy (\( \int |E(t)|^2 \, dt \)) can be fixed at some constant value by tweaks the amplitude \( E_0(\tau) \) using the iris (please refer to appendix A for further details). The use of an iris to control the pulse energy has been reported recently in a couple of experimental works by Nefedova et al [47, 48] and Boltel et al [49]. It should be noted that in figure 1 we have placed the iris before the pulse shaper, so that any deformations in the waveform of the incoming Gaussian pulse (introduced by the iris) can be compensated within the pulse shaper, and eventually the desired sinc-shaped waveform can be obtained. Moreover, the delay \( \tau \) can easily be tuned by varying the distance \( d \) on an optical bench. Throughout the manuscript, we will be using atomic units unless otherwise stated: this implies \( e = \hbar = m_e = 1 \).
We study the interaction of the synthesized laser pulse with a He atom by numerically solving the one-dimensional time-dependent Schrödinger equation (TDSE) based on the single-active electron approximation [50]. The TDSE in the length gauge is written as [51]:

$$i \frac{\partial \psi(x,t)}{\partial t} = \left[ -\frac{1}{2} \frac{\partial^2}{\partial x^2} + V(x) - xE(t) \right] \psi(x,t),$$

(2)

where $E(t)$ is the laser field as given by equation (1) and

$$V(x) = -\frac{1}{\sqrt{x^2 + \xi}}$$

(3)

is the ionic soft-core potential where the constant $\xi$ is dependent on the ionization potential of the atom under study. For a He atom $\xi = 0.484$ is considered such that the ground state energy (ionization potential) is found to be $\sim 0.9$ a.u. ($\sim 24.6$ eV), which is close to the experimental value of the ionization potential of helium. The general solution of TDSE is obtained by employing the time evolution operator $U(t_0 + \Delta t, t_0)$ on the initial state wavefunction of the electron $\psi_0(x, t_0)$,

$$\psi(x, t_0 + \Delta t) = U(t_0 + \Delta t, t_0) \psi_0(x, t_0),$$

(4)

and the TDSE is solved numerically by adopting the split-operator method [52] in which the evolution operator is factored as a product of the kinetic and potential energy propagators, i.e.,

$$U(t_0 + \Delta t, t_0) = e^{-i\gamma^{2}\Delta t/4} e^{-iv^{2}\Delta t/2} e^{-i\xi^2 \Delta t/4},$$

(5)

where $v(t) = V(x) - xE(t)$ is the effective potential in the length gauge.

The initial ground state is calculated by the imaginary time propagation method [53]. Starting from this initial state, the time-dependent wavefunction is obtained by solving the TDSE numerically. The time-dependent dipole acceleration $\ddot{d}(t)$ is evaluated following the Ehrenfest theorem as [54]:

$$\ddot{d}(t) = -(\psi(x,t)) \frac{\partial V(x)}{\partial x} + E(t)\psi(x,t).$$

(6)

The harmonic spectrum is then finally obtained by the Fourier transformation of $\ddot{d}(t)$, i.e.,

$$S(\omega) = \frac{1}{\sqrt{2\pi}} \int \ddot{d}(t)e^{i\omega t} dt.$$  

(7)

In our calculation, the simulation domain is confined in a finite space of $L \sim 1700$ a.u., where the grid spacing $\Delta x = 0.05$ a.u. and the simulation time step is decided from the relation $\delta t \sim (\Delta x)^2/2$. In order to avoid the reflection of the electron wave packet from the boundaries, an absorbing boundary of thickness 150 a.u. was placed at $x = \pm 700$ a.u. The absorption is routinely implemented by either the exterior complex scaling method [55] or the standard $\cos^{1/8}$ mask function [56]. The choice of the masking function does not have any effect on the actual results if the simulation domain is sufficiently large. The convergence is checked by varying the grid parameters, and for $L \gtrsim 1500$ a.u. the results are found to converge. We utilized the widely used Armadillo library for linear algebra purposes in our simulations [57].

The ASP is obtained by superposing several harmonics as [58]:

$$I(t) = \sum_{q} a_{q} \exp[iq\omega t]^{2},$$

(8)

where $q$ is the harmonic order and $a_{q}$ represents the inverse Fourier transformation given as:

$$a_{q} = \frac{1}{\sqrt{2\pi}} \int d(t)e^{-iqt} dt.$$  

(9)

A time-frequency analysis has been done to get an insight into the quantum recollision processes. The wavelet transform was performed using the standard Gabor time-frequency analysis [59, 60].

3. Results and discussions

In the following, we compare the harmonic spectrum of a Gaussian laser pulse and the synthesized pulse given by equation (1), followed by the effect of the delay $\tau$ on the harmonic cutoffs. Finally, we ascertain its impact on the generation of optimal ASPs.

3.1. Comparison with Gaussian pulse

Initially, in order to show the preeminence of our synthesized pulse for the generation of higher-order harmonics, we compared the HHG spectra generated by a synthesized laser pulse defined in equation (1) and the 800 nm/5 fs Gaussian-enveloped laser pulse. The temporal profile of the Gaussian laser pulse is given as:

$$E_{G}(t) = E_{0G}\ exp\left[ -4 \ln \frac{2\tau_{G}^{2} }{\tau_{c}^{2}} \ cos(\omega_{G} t) \right],$$

(10)

where $E_{0G}$ is the amplitude of the laser field, $\omega_{G}$ ($= 0.575$ a.u.) is the same laser frequency as the one used in the synthesized pulse, and $\tau_{G} = 5$ fs is considered. It should be noted that the pulse energy of both pulses is fixed at the value $\sim 1.91$ a.u., thus, the peak field amplitude for the Gaussian pulse is estimated to be $E_{0G} \sim 0.1567$ a.u. However, the field amplitude of the synthesized pulse is considered to be $E_{0} \sim 0.1453$ a.u.

The temporal electric field profile of the Gaussian-envelope and the synthesized pulse are presented in figures 2(a) and (b), respectively. For the synthesized pulse, the delay parameter $\tau$ is chosen to be 0.71 T, where $T = 2\pi/\omega_{0}$ is the time corresponding to one optical cycle (o.c.). Compared to the case of the Gaussian pulse, we can see that the laser cycle of the synthesized pulse is expanded. The electron ionized around the negative maximum of the synthesized laser field can gain higher energies in a relatively longer acceleration process. As a consequence, harmonics with a larger cutoff energy will then be achieved when they recombine with the parent ion. A comparison of the HHG power spectra for the Gaussian and the synthesized pulse is presented in figure 2(c). The differences in harmonic intensities between the two curves in the plateau
region are actually small, so for the purpose of clarity we have shifted the harmonic signal for the Gaussian pulse along the y-axis. These harmonic spectra have the characteristic structure of a typical HHG spectrum, i.e., irregular behavior toward the lower harmonics region. They then gradually become regular within the plateau region, with a final sudden drop in the harmonic intensity at the cutoff. The results show that the harmonic cutoff is impressively extended from $140\omega_0$ for the Gaussian field to $235\omega_0$ for the synthesized field. The harmonic structure in the supercontinuum region of the synthesized field is less modulated than the case of the Gaussian field. This condition is favorable for the generation of an isolated ASP. The cutoff rule [61] $I_p + 3.17U_p$ (where $U_p = E^2/4\omega_0^2$ is the quiver energy, and $I_p = 0.904$ a.u. is the ionization potential for the He atom) for the maximum possible harmonic photon energy predicts the cutoff at $106\omega_0$ (164 eV), while the harmonic cutoff for the case of the synthesized pulse is observed at $235\omega_0$ (364 eV). The inability of the three step model to predict the harmonic cutoff is due to the classical consideration of the fact that the laser pulse intensity should be constant during the quiver motion of electrons [17, 39]. Since the laser intensity in our case changes significantly in one laser cycle, we cannot expect that the celebrated three step model will predict the harmonic cutoff correctly.

Both the classical theory [9] and quantum time-frequency analysis are adopted to gain a deeper understanding of the generation of higher-order harmonics. In figures 2(c) and (d), we show the classical electron trajectories along with the time-frequency distribution of the HHG spectra for the above two cases. In the case of the synthesized field, the electron trajectory map shows two paths with different emission times (solid black circles) contributing to each harmonic in the HHG spectra (refer to figure 2(d)). The two branches of the emission time trajectory with positive and negative slopes correspond to the short and long paths, respectively. Likewise, in the time-frequency profile, there are two quantum paths contributing to each of the harmonics within half of the optical cycle of the synthesized laser field. It should be noted that the maximum harmonic order corresponding to the Gaussian pulse, marked by the point $P$ in figure 2(c), is approximately equal to $140\omega_0$, whereas the maximum harmonic order corresponding to the synthesized pulse, marked as $P'$ in figure 2(d), is observed at the value $235\omega_0$. Even though the field energy content of both the driving pulses is the same, the order of the cutoff harmonic corresponding to the synthesized pulse is about $95\omega_0$ (or 147 eV) more, corresponding to the Gaussian pulse. The harmonic spectrum for these cases is illustrated in figure 2(e), which complements the results presented using the time-frequency analysis.

Based on the classical trajectory maps, we can state that the peak $P$ originates from the ABC process marked in figure 2(a). The electron gets ionized by tunneling when the magnitude of the laser field amplitude reaches its maxima around the point $A$, then it gets accelerated by the next maxima at $B$ which results in it gaining high kinetic energy. Finally, the electron recombinesthe parent ion core in between the peak at $B$ and the dip at $C$, which results in the emission of harmonic photons up to the order of 135. The maximum energy at the peak $P$ is determined by the kinetic energy gained by the electron during the acceleration process. In contrast, in the case of the synthesized field (refer to figure 2(b)), the laser cycle is expanded, and therefore the electrons get ionized around $A'$. The free electron then accelerates for a relatively long time till peak $B'$. Then it returns to the core ion with higher kinetic energy in between $B'$ and $C'$ and recombinesthe with the ion. The recombination process leads to the emission of harmonics up to the order of 235.

### 3.2. Effect of delay on HHG and scaling law

We now show how the synthesized pulse waveform resulting from the time delay ($\tau$) between the two-component sinc pulses affects the resulting HHG spectra. The temporal profiles of the synthesized pulse for different time delays $\tau = 0.41$ T, $0.81$ T, and $1.21$ T are respectively presented in figures 3(a)–(c). The effect of the increasing delay on field strength can be seen as an extension of the laser cycle. The respective harmonic spectra (refer to equation (7)) for these delay parameters are presented in figures 3(d)–(f). The harmonic cutoff energies for these delays are observed respectively at $202\omega_0$, $245\omega_0$, and $350\omega_0$. Furthermore, one important feature observed in the harmonic spectrum presented in figures 3(e) and (f) is the enhancement of the HHG yield of the harmonics around the peaks $p_1$, $p_2$, $p_3$ marked in figure 3(e) and the peaks marked as $p_4$, $p_5$, $p_6$ in figure 3(f). The underlying mechanism behind the extension of the HHG cutoff

![Figure 2](attachment:figure2.png)
energy and the selective harmonic yield can be understood from the classical trajectories and the quantum time-frequency analysis of the quantum paths of the electron, as shown in figures 3(g)–(i). Here, classical trajectory analysis shows the effect of the time dependence of electron kinetic energy on the ionization (yellow circles) and also on the recombination (solid black circles) times.

The classical electron trajectories are consistent with the results of the time-frequency analysis of the electron quantum paths. The increasing delay time ($\tau$) results in the broadening of both the negative and positive cycle of the synthesized laser field. This eventually modifies the ionization and recombination time, and also the kinetic energy of the returning electron. The increased electron kinetic energy will cause an enhancement in the emitted harmonic photon energy as shown in figures 3(g)–(i).

In order to validate the numerical accuracy of the results obtained, we have also calculated the harmonic spectrum using the velocity gauge while solving the TDSE. For the case of the spatially homogeneous fields, the time evolution operator in equation (5) will have the following equivalent form in the velocity gauge [51]:

$$U(t_0 + \Delta t, t_0) = e^{-i(p' + A(t_0 + \Delta t/2)^2)\Delta t/4} e^{-iV(t)\Delta t} \times e^{-i(p + A(t_0 + \Delta t/2)^2)\Delta t/4},$$

where $A(t) = -\int_{t'_0}^t E(t') \, dt'$ is the vector potential of the laser field. Further, the dipole acceleration expectation value and eventually the spectral intensity of harmonic emission can be calculated by following a similar prescription as mentioned for the case of the length gauge in section 2.

In figure 4, we present the harmonic spectrum using the length (top panel) and velocity gauge (bottom panel) while numerically solving the TDSE. For the purpose of clarity, the harmonic intensities of $\tau = 0.31 \, \text{T}, 0.91 \, \text{T},$ and $1.11 \, \text{T}$ pulses are multiplied by factors of $10^{16}, 10^{11}$ and $10^5$, respectively.
Moreover, figure 4 presents the harmonic spectrum for different delay parameters. We can notice that it is not only the harmonic cutoff that changes with the delay parameter \( \tau \), but that some harmonic components are also observed to be enhanced as compared to the neighboring harmonics in the plateau region. The position of these enhanced harmonics is found to change for different time delays. The intensity of these harmonics is about one order of magnitude higher than the surrounding harmonics. The mechanism behind this selective enhancement has already been discussed above. These enhanced harmonics can be used further as a monochromatic source for many important applications, which include the seeding of an XUV free-electron laser or laser-plasma amplifiers [62]. In table 1, we have presented some typical harmonic cutoffs and their intensities for different values of the delay \( \tau \). Here we have also presented some enhanced harmonics and their intensities as a multiple of the intensity observed at the harmonic cutoff.

| \( \tau \) [T] | \( \omega_c \) | \( \log_{10} |S(\omega_c)| \) | \( \omega_{0}(\eta) \) |
|-------------|-------------|-----------------|-----------------|
| 0.81        | 248         | −3.18           | 26(8), 32(14), 44(24), 77(35) |
| 0.91        | 271         | −3.33           | 29(4), 36(7), 51(11), 90(10) |
| 1.01        | 291         | −3.58           | 32(2), 40(3), 57(5), 100(5) |
| 1.11        | 320         | −4.00           | 36(5), 45(14), 64(21), 112(17) |
| 1.21        | 350         | −4.55           | 39(33), 50(112), 72(132), 125(85) |

In figure 5(a), the observed harmonic cutoff \( \omega_c/\omega_0 \) for different delay parameters \( \tau \) is presented using the soft-core potential (equation (3)). Based on these observations, we infer that the HHG cutoff in the current setup scales as \( \sim \tau^{5/2} \) for a fixed driver pulse energy irrespective of atomic species. The scaling law is found to be consistent up to some large time delay such as 1.31 T. Moreover, a further increase in delay deforms the synthesized pulse by inducing a local field maximum between the two central peaks of the field. The intensities of the respective harmonic cutoff for different delay parameters are also illustrated in figure 5(a). It is observed that the cutoff intensities (\( \log_{10} |S(\omega_c)| \)) follow a \( \sim -\tau^3 \) scaling. The decrease in HHG yield can be explained by taking into account the spreading of the electronic wave-packet during propagation in the continuum. As we have mentioned earlier, increasing the delay in pulse synthesis would result in elongation in the laser cycle. Since the wave-packet propagation time in the continuum is proportional to the laser cycle, the wave-packet has more time to spread with increasing delay, thus scaling down the efficiency of the recombination process in harmonic emission.

Furthermore, to analyze the validity of these scaling laws in the three-dimensional environment, we have used a 1D modified potential referred to as a pliant-core potential [63]. This modified potential ensures a good agreement with the 3D TDSE calculations as reported in [63] and has the following form:

\[
V_{pc}(x) = -\frac{1}{(|x|^2 + \beta^2)^3/3},
\]

where the parameter \( \beta \) is used to avoid the Coulomb singularity. Also, the value of parameter \( \beta \) varies according to the ionization potential of the atom under study. For a He atom \( \beta = 0.49 \) is used [63].

In figure 5(b), we have presented the observed harmonic cutoff \( \omega_c/\omega_0 \) and the corresponding harmonic intensities obtained using the pliant-core potential (equation (12)). It is observed that the HHG cutoffs for different delay parameters are also scaled as \( \sim \tau^{5/2} \), showing an excellent agreement with the results obtained in the case of the soft-core potential. The intensities of the cutoff harmonics also follow a definite scaling of \( \sim -\tau^6 \) for using the pliant-core potential. As it was reported in [64, 65], the wavelength scaling of the harmonic intensity can vary significantly for different atoms. In order to see the universality of the scaling law, we have also obtained the scaling laws for the hydrogen atom (using \( \xi = 2 \) in equation (3)) with a reduced laser intensity of \( 10^{14} \) W cm\(^{-2} \), while keeping the field profile the same as the one used in figure 5. The harmonic cutoff and intensity scaling for the hydrogen atom are presented in figure 6. It has been observed that the harmonic cutoff scales as \( \sim \tau^{5/2} \) and the cutoff intensity scales as \( \sim -\tau^4 \).

The scaling laws for the harmonic cutoff and their respective intensities clearly demonstrate the utility of this setup. We not only predict the harmonic cutoff, but also its intensity in...
Figure 6. The scaling of the harmonic cutoff and the cutoff intensity with delay parameters for hydrogen atoms using a soft-core potential (refer to equation (3)) with $\xi = 2$. The laser intensity is considered to be $10^{14}$ W cm$^{-2}$ with the same field profile as used in figure 5. The pulse energy is kept fixed. The field amplitude and the pulse energy are normalized with respect to the values associated with the delay parameter $\tau = 0.71$ T. The peak field amplitude and the pulse energy for $\tau = 0.71$ T are respectively $\sim 0.065$ a.u. and $\sim 0.379$ a.u.

Figure 7. Classical trajectories are presented for different delay parameters and again the $\omega_2 \propto \tau^{1/2}$ scaling is also projected. All other laser parameters are the same as those presented in figure 5.

terms of the time delay between two oppositely polarized sinc pulses. The above-mentioned scaling law of the harmonic cutoff energy is also validated using classical trajectory analysis and the corresponding returning electron trajectory maps are presented in figure 7. Even though there is no way to comment on the cutoff intensities from classical analysis, the scaling property of the maximum radiated photon energy can be predicted promisingly. It is clear from figure 7 that for smaller time delays (i.e., $\tau < 0.41$ T) the cutoff harmonics are emitted due to the ionization around the peak marked as $O'$ in figure 2(b) of the synthesized laser pulse. The electrons ionize around the peak at $O'$ and later recombine between the dip at $A'$ and the peak at $B'$. These electrons accelerate for relatively longer times as compared to the electrons ionized around the dip at $A'$ and recombine between the peak at $B'$ and the dip at $C'$. Therefore, the energy of the emitted photons is higher for the former case.

Moreover, with the increasing delay, the width of the dip at $A'$ and the peak at $B'$ increases, and concurrently the amplitude of the peak at $O'$ decreases (refer to figures 3(a)–(c)). As a result, in cases for which $\tau$ is greater than 0.41 T, the quiver energy of the electrons ionized around the peak at $O'$ decreases, while the electrons ionized around the dip at $A'$ will return with higher kinetic energies as observed in figure 7. The typical nature of the sinc pulse yields the previously mentioned scaling of the cutoff energy and its intensity in the harmonic spectrum. The delay parameter actually controls the interference between the two pulses of opposite polarization, which eventually translates into the modification of the electron quantum path as well as the recombination energy of the returning electron.

3.3. Attosecond pulse generation

We now discuss the role of the higher-order harmonics in the generation of a single ASP. In figure 5, we observe nice scaling behavior of the harmonic cutoff and their respective intensity in terms of the delay parameter $\tau$. We now study how to generate an ASP by filtering several harmonics just before the cutoff but in the plateau region. The temporal profile of the ASP for a given delay parameter is obtained by superposing the contribution of the different harmonics and then performing the inverse Fourier transform (refer to equation (8)). In figure 8(a), we present isolated ASPs generated without any phase compensation, but just by superposing the last 100 harmonics up to the cutoff of the harmonic spectrum corresponding to a bandwidth of $\sim 155$ eV for different delay parameters. The pulses have been emitted within the same optical cycle, but just for the sake of clear presentation, we have shifted the pulses along the time scale. The maximum harmonic photon energy for different ASPs corresponding to the delay $\tau = 0.41$ T, 0.51 T, 1.21 T and 1.31 T are 314 eV, 323 eV, 543 eV and 574 eV respectively (refer to figure 5). The generated ASPs become shorter with increasing values of the delay parameter. Here we show that as we increase the delay from $\tau = 0.41$ T to $\tau = 1.31$ T, the pulse width of the generated ASPs decreases from 295 as to 99 as. We explain this behavior using figures 3(g)–(i). Here we see that as we increase the delay, the intensity of the short trajectory diminishes. This indicates that the contribution of the harmonics due to the shorter trajectories is much less in the HHG spectrum. Therefore, in the formation of ASPs, harmonics corresponding to the longer trajectories are present, which results in the shorter ASP. It should be noted that by the nature of the synthesized pulse (refer to equation (1)), harmonic emission takes place only for a single cycle of the driving pulse. Therefore, we will have at most two ASPs instead of a pulse train upon the superposition of a large range of harmonics in the plateau region of the HHG spectrum. These paired ASPs are a potential tool in pump-probe measurements. As we already discussed, with increasing delay the contribution of only a single trajectory (i.e., long trajectory) is dominant in the HHG spectrum. Hence, the intensity of one of the ASPs in the pair diminishes with increasing delay. The single ASP is more promising for applications in time-resolved spectroscopy. This method reduces the need to adopt various gating techniques to obtain the single ASP [66].

The harmonics in the HHG process are emitted over a range of time, which means the phase of each harmonic is arbitrary.
As a result, the superposition of all the plateau harmonics cannot generate the shortest ASP. However, the phase dispersion can be compensated by propagating the ASP through either a gas medium or a thin metal foil [36, 67]. In our simulations, the harmonic phase has been compensated by taking a constant phase difference between two consecutive harmonics. The resultant ASPs after the phase compensation for different delay parameters are presented in figure 8(b). After the phase compensation, intense isolated ASPs with durations (photon energy) of 26 as (314 eV), 27 as (323 eV), 29 as (543 eV) and 27 as (574 eV) can be achieved for time delays $\tau = 0.41 \, \text{T}$, 0.51 T, 1.21 T and 1.31 T, respectively. Moreover, their intensity has been increased up to one order of magnitude compared to the scenario without any phase compensation.

One can notice that despite having a large harmonic continuum in the case of the synthesized field, the generated ASP is not as short as it is in the case of a Gaussian pulse [20]. This can be understood by taking into account the variation of the phases of the emitted harmonics. The phase of the emitted harmonic is calculated as:

$$\theta_n = \arctan 2 \left( \text{Im}[D_n], \text{Re}[D_n] \right),$$

where $D_n$ is the Fourier transform of the dipole acceleration as calculated in equation (6). A comparison between the emitted harmonic phases for a synthesized and Gaussian pulse is depicted in figures 9(a) and (b), respectively. We see that for the synthesized pulse, the previous 15 harmonics from the cutoff (between the 360th–375th order) have constant phase variation between consecutive harmonics, while for the Gaussian pulse, nearly 50 harmonics (between the 90th–140th order) are separated by a constant phase. The superposition of these phase-locked harmonics can generate an ASP of shorter duration without applying any phase compensation techniques. However, if the phase dispersion is properly compensated, then the obtained large HHG continuum in the case of the synthesized pulse can be used to produce a shorter ASP. The discussion related to the ASP generation by Gaussian laser pulse is presented in appendix B.

### 4. Strong field approximation

So far we have discussed how the harmonic cutoffs can be regulated by simply changing the delay between the two oppositely polarized laser pulses. The cutoff scaling is found to be a property of the laser pulse envelope, and how it changes with the delay parameter. In order to further validate the scaling laws, in this section we study the high harmonic generation process under the framework of the strong field approximation (SFA). The main assumption made in SFA is that the dynamics of the electron after ionization is solely controlled by the strong laser field, and the effect of the core potential is small enough to be ignored. Within the regime of tunnel ionization, the Keldysh parameter $\gamma \ll 1$ [68] and the time-dependent wavefunction $|\psi(x, t)\rangle$ of the electron can be written as (dropping the $x$-dependence):

$$|\psi(t)\rangle = a(t)|g\rangle + |\phi(t)\rangle$$

where $|g\rangle$ is the ground state which is calculated by the imaginary-time propagation method [53], $|\phi(t)\rangle$ represents the continuum part of the wavefunction and $a(t)$ is the amplitude of the ground state and can be computed using the quasistatic approximation $|a(t)|^2 = \exp[- \int_0^t w(t') \, dt']$, where $w(t)$ is the instantaneous ionization rate calculated by the Ammosov–Delone–Krainov formula [69].

The continuum part $|\phi(t)\rangle$ under SFA satisfies the equation [10]:

$$i \frac{d}{dt} \phi = H_v(t) \phi - E(t) x a(t) |g\rangle,$$  

Figure 8. The temporal profiles of the ASPs generated by properly selecting the filtering window of 100 harmonics before the cutoff for time delays $\tau = 0.41 \, \text{T}$, 0.51 T, 1.21 T, and 1.31 T without (a) and with (b) phase compensation is presented. For the purpose of clarity, the ASPs are shifted in time scale and multiplied by some factors as shown. The cutoff energy ($E_c$) is also mentioned for each ASP.

Figure 9. The phase of the emitted harmonics in the case of: (a) a synthesized pulse with delay $\tau = 1.31 \, \text{T}$ and (b) a Gaussian-envelope pulse with laser parameters the same as those taken in figure 2(a).
where \( H_V \equiv -\frac{1}{2} \frac{\partial^2}{\partial \mathbf{r}^2} - E(t) \mathbf{r} + I_p \) is the Volkov Hamiltonian, with \( I_p \) being the ionization potential of the atomic species under study. The exact solution of equation (15) can be given as [10]:

\[
\langle p + A(t) | \phi \rangle = -i \int_0^t d't a'(t')E(t') \langle p + A(t') | x | g \rangle e^{-iS(p_g,t,t')} ,
\]

where the phase factor

\[
S(p_g,t,t') \equiv \int_t^{t'} \frac{1}{2} (p_g + A(t'))^2 dt'' + I_p (t - t')
\]

is the quasi-classical action and \( |p_g\rangle \) denotes the plane wave state

\[
|g\rangle = \frac{1}{\sqrt{2\pi}} e^{ip_g \mathbf{x}} .
\]

Also, \( A(t) = -\int_0^t E(t') dt' \) is the vector potential of the laser field \( E(t) \).

The expectation value of the time-dependent dipole acceleration \( d(t) \) defined in equation (6) is:

\[
\tilde{d}_{SFA}(t) = -\langle \psi(t)|V'(x)|\psi(t)\rangle + E(t)
\]

\[
= -[a'(t) \langle g| + \langle \phi(t) | |V'(x)| a(t) \langle g| + \langle \phi(t) |] + E(t) .
\]

Considering the continuum–bound transitions while dropping the \( E(t) \) and higher order continuum–continuum transitions, the dipole acceleration can be written as:

\[
\tilde{d}_{SFA}(t) = \tilde{\zeta}(t) + \tilde{\zeta}'(t) ,
\]

where,

\[
\tilde{\zeta}(t) = -a'(t) \langle g|V'(x)|\phi(t)\rangle .
\]

Now in order to calculate \( \tilde{\zeta} \), we insert the momentum space completeness relation \( \int |p + A(t)| \langle p + A(t)|dp_g \rangle \) in equation (21) and using equation (16), we get:

\[
\tilde{\zeta}(t) = \int_0^{t'} dt' \int dp_g \langle g|V'(x)|p + A(t)\rangle a'(t) a(t')
\]

\[
\times E(t') \langle p + A(t') | x | g \rangle e^{-iS(p_g,t,t')} .
\]

Applying the saddle point approximation to the momentum integration as in [70], we obtain the dipole acceleration as:

\[
\tilde{\zeta}(t) = \int_0^{t'} dt' \left( \frac{2\pi i}{1 - t' - \epsilon}\right)^{3/2} \langle g|V'(x)|p + A(t)\rangle a'(t)
\]

\[
\times a(t') E(t') \langle p + A(t') | x | g \rangle e^{-iS(p_g,t,t')} .
\]

Here the infinitesimal \( \epsilon \) comes from the regularized Gaussian integral around the stationary phase point, and the value of saddle point \( p_{ss} \) is given by:

\[
p_{ss} = -\frac{1}{t - t'} \int_0^{t'} A(t') \ dt''.
\]

Figure 10 depicts the main results of our SFA-based simulations for the He atom. To compare the results with the scaling

| Approach          | \( \mu \) | \( \eta \) | \( \delta \) | \( \kappa \) | \( \chi \) |
|-------------------|----------|----------|----------|----------|----------|
| Classical trajectories | 94.4     | 194.3    | —        | —        | —        |
| He, soft-core potential | 94.4     | 194.3    | 0.18     | 5.0      | -0.97    |
| He, pliant-core potential | 94.4     | 194.3    | 0.14     | 6.0      | -0.99    |
| He, soft-core potential, SFA | 94.0     | 194.8    | 0.08     | 5.0      | -0.98    |
| H, lower intensity | 19.3     | 42.09    | 0.17     | 4.0      | -0.95    |
laws suggested by the 1D TDSE simulations, the SFA-based harmonic spectrum was calculated for the same laser pulse as in the case of 1D TDSE, i.e., for a synthesized laser pulse with varying \( \tau \) values and a fixed pulse energy. The harmonic spectrum for different time delays is presented in figure 10(a). The HHG spectra corresponding to \( \tau = 0.41 \) T, 0.71 T and 1.01 T are shifted vertically for clarity purposes. In comparison with the HHG spectra obtained from the 1D TDSE solution and shown in figure 4, one can see that the harmonic cutoffs are identical for the corresponding delay cases. Moreover, selective enhancement in the intensity of harmonics at similar positions can also be seen. In figure 10(b), we have presented the harmonic cutoffs (\( \omega_c/\omega_0 \)) as obtained from the SFA-based simulations along with the fitted curve (solid line), showing the same scaling of \( \sim \tau^{3/2} \) as in the case of 1D TDSE (see figure 5(a)). The intensities of the respective harmonic cutoff for different time delays are also shown in figure 10(b), along with a reference curve (dashed line) which follows a scaling of \( \sim -\tau^5 \). It can be seen that for \( \tau > 0.71 \) T cases, the cutoff intensities (\( \log_{10}|S(\omega_0)| \)) follow the \( \sim -\tau^5 \) scaling, supporting the results obtained from 1D TDSE simulations (refer to figure 5(a)). However, for the \( \tau < 0.71 \) cases, the observed intensities of the cutoff harmonic start differing from the scaling law with decreasing values of delay \( \tau \). This can be understood by recalling the fact that we have taken the quasi-classical approximation in the SFA-based calculations, and as mentioned earlier, decreasing the value of delay parameter will shorten the laser cycle, thus lowering the effective wavelength of the synthesized pulse.

5. Summary and conclusions

We have theoretically investigated the HHG and the generation of a single ASP from a He atom. The driving field is sculpted from a sinc-shaped pulse by a simple experimentally realizable setup, though technological limitations currently restrict the generation of the sinc-shaped pulse at the mentioned intensities. This synthesized field can be considered as a single cycle pulse which can favorably control the electron quantum path. It has been observed that in comparison with few-cycle Gaussian pulses of the same energy, the bandwidth of an XUV supercontinuum spectrum is significantly broadened for the synthesized pulse. We find that the energy of the cutoff harmonics increases with the increasing delay parameter (\( \tau \)). Specifically, it nicely scales as \( \sim \tau^{3/2} \) for a fixed driver pulse energy. Furthermore, the intensity of the harmonic cutoff is also seen to follow a \( \sim -\tau^{4-6} \) scaling, depending on the atomic species or model potential under study. These well-defined scaling laws for the harmonic cutoff and its intensity indicate a realizable experimental setup, wherein radiation from XUV to soft x-rays can be generated by simply changing the distance \( d \) on an optical bench (refer to figure 1). The mentioned scaling laws for the harmonic cutoffs and the cutoff intensities are validated by (a) classical trajectory analysis (refer to figure 7), (b) gauge invariant HHG spectra (refer to figure 4), (c) soft-core and planar-core potentials (refer to figure 5), and (d) strong-field approximations (refer to figure 10). In table 2 we summarize the scaling parameters obtained through different approaches.

We have used these higher harmonics to generate ASPs with a central frequency in the XUV to the soft x-ray regime of the electromagnetic spectrum. This has been achieved by filtering the harmonics of bandwidth 155 eV from the respective harmonic cutoff for a given delay parameter. The quantum time-frequency analysis shows that the contribution of the harmonics corresponding to the shorter quantum path in the HHG supercontinuum spectrum decreases with the increasing delay of the pulse synthesis. As a result, an isolated \( \sim 100 \) as pulse for \( \tau = 1.31 \) T is straightforwardly obtained without any phase compensation. If the phase is compensated correctly, then an intense ultrashort \( \sim 27 \) as pulse can be generated with a photon energy of \( \sim 570 \) eV.

The advantage of the scheme proposed here lies in the tuning of the broadband XUV supercontinuum by merely adjusting the time delay on an optical bench, which seems feasible from an experimental point of view. The harmonic cutoff scaling is found to be a property of the laser pulse, however, the intensity scaling would depend on the atomic species or model potential under study. The enhancement in some satellite harmonics is also studied, and its dependence on the delay parameter \( \tau \) has been summarized. The exploration of higher dimensional effects on the electron quantum path and the corresponding ASP generation using this synthesized sinc pulse is reserved for a future project.

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Appendix A. Laser peak field and pulse energy

In our work, we have optimized the intensity of the component sinc pulse to keep the pulse energy of the synthesized pulse constant for different delay parameters (\( \tau \)). The pulse energy (\( E_0 \)) can be calculated by numerically solving the integral \( \int |E(t)|^2 dt \). The pulse amplitude is then tweaked to have the constant \( E_0 \). Figure A1 depicts the variation in the peak field amplitude \( E_0(\tau) \) of the component sinc pulse which is defined in equation (1). In figure A1, the energy of the synthesized pulse is also presented for different time delays. The curves have been normalized with respect to the values for the case of \( \tau = 0.71 \) T. One can see that the pulse energy is fixed for all the delay cases, while the peak field amplitude increases on either side of the delay \( \tau = 0.71 \) T case. The increase in the pulse amplitude on either side of the \( \tau = 0.71 \) T is related to the interference (constructive/destructive) of the central peaks of two component sinc pulses.

Appendix B. Attosecond pulse by Gaussian pulse

In the following, we studied the generation of attosecond pulses by the interaction of the He atom with a Gaussian pulse. Figure B1 presents the generated ASPs by superposing
the last 100 harmonics up to the harmonic cutoff \( E_c \sim 217 \text{ eV} \). The Gaussian-enveloped laser pulse of wavelength 800 nm and pulse length (FWHM) 5 fs with similar pulse energy \( \sim 1.91 \text{ a.u.} \) to the synthesized pulse is shown in figure 2(a) along with the respective harmonic spectra in figure 2(e) (red curve). In figure B1, the ASPs plotted with a dashed (solid) curve have been generated by simply superposing the harmonics of bandwidth \( \sim 155 \text{ eV (}\omega/\omega_0 = 41–140\text{)} \) without (with) phase compensation. For the case when phase compensation is not used, we observe two ASPs with a higher intensity pulse having a duration of \( \sim 377 \text{ as} \). The two ASPs in this case are because of the fact that the process of harmonic generation takes place for multiple cycles of Gaussian pulse, as can be seen in the time-frequency distribution of the corresponding HHG spectra (refer to figure 2(c)). Similarly, after compensating the phase we get an ASP (solid curve) of duration \( \sim 47 \text{ as} \) along with the well-separated lower intensity ASPs. Compared to the ASPs observed for sinc-shaped pulses, the generation of a single ASP (refer to figure 8) is not possible through pure Gaussian pulses without any additional mechanisms, be they chirp or multi-color laser pulses.

Moreover, the presented sinc-shaped synthesized pulse provides very good control over the tuning of harmonic cutoff energy by simply varying the distance \( d \) (refer to figure 1) on an optical bench. Such fine control would be difficult to achieve for conventional Gaussian pulses unless any additional mechanism is in place. In the case of the Gaussian pulse, the superposition of two such pulses with some time delay or with a difference in their carrier-envelope phases will not result in a nearly single-cycle driving pulse, which we observed in the proposed setup.

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