Generation of isolated attosecond pulses using a plasmonic funnel-waveguide

Joonhee Choi¹, Seungchul Kim¹,², In-Yong Park¹,³, Dong-Hyub Lee¹, Seunghwoi Han¹ and Seung-Woo Kim¹,⁴

¹ Ultrafast Optics for Ultraprecision Group, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 305-701, Korea
² Max Planck Center for Attosecond Science (MPC-AS), San 31 Hyoja-Dong, Namku, Pohang, Kyungbuk 790-784, Korea
³ Center for Nano-Imaging Technology, Division of Industrial Metrology, Korea Research Institute of Standards and Science (KRISS), Daejeon 305-340, Korea
E-mail: swk@kaist.ac.kr

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Abstract. We theoretically investigated the possibility of generating attosecond pulses by means of plasmonic field enhancement induced in a nanostructured metallic funnel-waveguide. This study was motivated by our recent experimental demonstration of ultrashort extreme-ultraviolet (EUV) pulses using the same type of three-dimensional waveguides. Here, with emphasis on generation of isolated attosecond pulses, the finite-domain time-difference method was used to analyze the funnel-waveguide with respect to the geometry-dependent plasmonic features such as the field enhancement factor, enhanced plasmonic field profile and hot-spot location. Then an extended semi-classical model of high-order harmonic generation was adopted to predict the EUV spectra generated from the funnel-waveguide in consideration of the spatial inhomogeneity of the plasmonic field within the hot-spot volume. Our simulation finally proved that isolated attosecond pulses can be produced at fast repetition rates directly from a few-cycle femtosecond laser or by synthesizing a two-color laser consisting of two multi-cycle pulses of cross-polarized configuration.

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1. Introduction

Attosecond light pulses are a promising tool to resolve the electron motion in an atom or molecule at the attosecond time scale (10^{-18} s) [1, 2]. For the purpose, attosecond pulses should be produced in the form of isolated pulses with well controlled burst timing. At the same time, the pulse repetition rate needs to be at the MHz level to avoid the space charge effect in pump–probe experiments through fast data acquisition [3]. It is well known that attosecond pulses are generated through nonlinear frequency up-conversion when an intense laser field is focused onto noble gaseous atoms. This process of high-order harmonic generation (HHG) requires laser intensities stronger than 10 TW cm^{-2}, for which femtosecond pulses amplified by means of chirped pulse amplification (CPA) are preferably used as the driving laser. However, the CPA process based on a Ti : sapphire crystal restricts the pulse repetition rate to ∼1 MHz level or below [4, 5], so attention is being paid to alternative methods such as pulse amplification using an external passive cavity [6–8] or single-pass strong amplification [9, 10] with the aim of achieving pulse power amplification at high repetition rates in the MHz range or faster.

Another alternative way of producing attosecond pulses at high repetition rates is to exploit the plasmonic field enhancement occurring in metallic nanostructures [11, 12]. The plasmonic resonance of free electrons boosts the intensity level of a modest femtosecond laser pulse readily by a factor of more than 100, leading to generation of high-order harmonics without reducing the repetition rate. Since the first experimental demonstration of the plasmon-driven HHG using bow-tie nanostructures, several theoretical investigations were performed to validate the possibility of generating isolated attosecond pulses using various metallic nano-antennas [13–15]. These theoretical studies also revealed some distinctive characteristics of the plasmonic HHG spectra which are mostly attributed to the spatial inhomogeneity of the plasmonic field in the vicinity of the hot spot formed within nano-antennas [16, 17].

Recently we proposed a three-dimensional (3D) design of a plasmonic funnel-waveguide as an improved means for efficient implementation of the plasmon-driven HHG [12]. The funnel-waveguide leads to nanofocusing of the propagating surface plasmon polaritons (SPPs) so that the incident laser field becomes concentrated on a hot spot beyond the diffraction limit. Unlike bow-ties fabricated on metallic thin films, the funnel-waveguide can be built on a bulk metal substrate to provide high immunity to thermal damage. Further, the peak intensity enhanced by the funnel-waveguide is found less sensitive to the geometrical inaccuracy that is inevitably encountered in fabrication of nanostructures. Consequently, high-order harmonics can be produced with improved conversion efficiency and thermal stability, extending the
harmonic cut-off up to the 43rd order that corresponds to a 18.6 nm wavelength in the EUV region [12].

In this paper, we conducted a theoretical analysis to investigate the feasibility of generating isolated attosecond pulses from the same funnel-waveguide. Our work proceeded in two steps: firstly, we solved the Maxwell equations to clarify how the plasmonic near-field intensity inside the funnel-waveguide is affected by its cross-section ellipticity. In doing that, a commercially available finite-domain time-difference (FDTD) software program (FDTD Solutions, LUMERICAL Inc., version 7.5) was used. Secondly, the high-order harmonic EUV spectra generated from the waveguide were numerically calculated by adopting an extended semi-classical HHG model that takes into account the spatial inhomogeneity of the enhanced plasmonic field. This two-step simulation enabled us to better understand the underlying nano-scale physics of strong plasmonics occurring within the funnel-waveguide and also clearly examine the possibility of producing isolated attosecond pulses by selectively filtering the resulting high-order harmonics, albeit not yet substantiated by experiment.

2. Design of the funnel-waveguide by finite-domain time-difference simulation

The funnel-waveguide takes the shape of a 3D plasmonic nanostructure that is made of silver with a hollow tapered hole of elliptical cross-section inside as depicted in figure 1(a). The geometrical shape of the funnel-waveguide is characterized by four design parameters; the major-axis diameter of the inlet aperture, the major-axis diameter of the exit aperture, the waveguide length and the ellipticity of the cross-section. In our simulation, the first three parameters were fixed as 4.4, 0.2 and 9 µm, respectively, as already optimized by Park et al [12]. Only the cross-section ellipticity ξ, which is defined as the ratio of the minor-axis diameter to the major-axis diameter, was taken as the variable design parameter since it was found to critically affect plasmonic behavior such as the intensity-enhancement factor, plasmonic pulse duration, hot-spot position and spatial field inhomogeneity.

The driving laser was assumed to be precisely focused on the inlet aperture along the optical axis of the funnel-waveguide. Three distinct temporal profiles were selected for simulation as the pulse shape of the driving laser, which are named Pulse I, Pulse II-1 and Pulse II-2, respectively. Specifically, Pulse I was a few-cycle pulse of 4 fs duration with 800 nm carrier wavelength. Pulse II-1 was a multi-cycle pulse of 10 fs duration with the same 800 nm carrier wavelength, whereas Pulse II-2 was a long pulse of 60 fs duration with 1560 nm carrier wavelength. In reality, Pulse I represented a carrier-envelope-phase (CEP) stabilized few-cycle femtosecond laser pulse produced at a MHz-level repetition rate [18] and Pulse II-1 and Pulse II-2 were long pulses of different carrier wavelengths, which were to be combined together in order to synthesize a two-color pulse capable of producing attosecond pulses by spatiotemporal symmetry-breaking despite their long pulse duration.

The three pulse profiles were tested separately by FDTD simulation in the first instance. They were assumed as linearly polarized plane waves with uniform intensity across the wavefront. The polarization direction was aligned parallel with the minor axis of the elliptical inlet aperture of the waveguide. As each pulse propagates through the hollow core of the waveguide, it was anticipated that the photonic mode of the incident electric wave was adiabatically converted to the plasmonic mode [19–21]. Our simulation confirmed the adiabatic conversion which guides the plasmonic mode to build up sharply in the hot spot formed near the exit aperture.
Figure 1. FDTD simulation of the funnel-waveguide. (a) Geometry of the funnel-waveguide, (b) maximum intensity enhancement factor versus ellipticity, (c) plasmonic pulse duration versus ellipticity, (d) hot-spot position versus ellipticity and (e) spatial field inhomogeneity at the hot-spot position. In (b)–(d), min. (maj.) denotes the polarization direction of the incident plane wave parallel to the minor- (major-) axis of the elliptical cross-section.

Figure 1(b) shows how the peak intensity-enhancement factor, $|E_{\text{loc}}/E_0|^2$, defined as the square of the ratio of the peak amplitude of the plasmonic field ($E_{\text{loc}}$) to that of the incident electric field ($E_0$), varies with the ellipticity $\xi$ of the cross-section. The simulation result predicts that $|E_{\text{loc}}/E_0|^2$ has a decreasing tendency as $\xi$ increases. Nonetheless, for all three pulse profiles, $|E_{\text{loc}}/E_0|^2$ builds up strongly enough to reach the HHG-threshold intensity of $10\,\text{TW cm}^{-2}$ from moderate input intensities of $0.1$–$1\,\text{TW cm}^{-2}$. It is also worthy of note that, when the input field
polarization is rotated by 90° to be in parallel with the major-axis direction, \( |E_{\text{loc}}/E_0|^2 \) decreases significantly with a reversed dependence on \( \xi \) for all three pulse profiles. This indicates that the adiabatic nanofocusing of the plasmonic mode occurs more efficiently when the hollow core of the funnel-waveguide has a smaller taper angle [19], which is the case of aligning the polarization direction to be parallel with the minor-axis direction along with decreasing \( \xi \).

The plasmonic pulse formed inside the funnel-waveguide has a temporal profile that is broader than that of the input pulse. This pulse broadening is caused by the wavelength-dependent behavior of the guided plasmonic mode within the hollow core with its cross-section continuously reducing from the inlet to the exit aperture. Figure 1(c) shows how the temporal duration of Pulse I and Pulse II-1 varies with \( \xi \). The pulse broadening becomes severe for small \( \xi \). Figure 1(d) summarizes the simulation result of the hot-spot location that is measured from the exit aperture. It is important to note that Pulse II-1 and Pulse II-2 share the same hot-spot location for \( \xi = 0.5 \) when the two pulses propagate in the cross-polarized configuration. The ellipticity \( \xi \) also affects the near-field intensity distribution as illustrated in figure 1(e). The spatial field inhomogeneity \( d_{\text{inh}} \) is quantitatively defined as the length over which the plasmonic field amplitude remains within 50% of its maximum within the hot-spot volume. For \( \xi = 0.5 \), \( d_{\text{inh}} \) is worked out to be \( \sim 1000 \) nm but it reduces to \( \sim 200 \) nm when \( \xi = 1.0 \) due to the increased curvature and arc-like distribution of conduction electrons at the inner wall of the circular-shaped cross-section.

Figure 2(a) presents the temporal profile of Pulse I whose plasmonic pulse is broadened from 4 to 6.0 fs duration in the hot spot for the case of \( \xi = 0.5 \). The leading and trailing edges of the plasmonic pulse are measured to be 2.7 and 3.3 fs, respectively. The CEP of the plasmonic pulse has a shift of \( \sim \pi/2 \), so the sine-shaped input pulse gradually evolves into a cosine-shaped field inside the waveguide. Figure 2(b) shows the spectral distribution of the plasmonic pulse in comparison to that of the incident pulse. The spectral bandwidth of the driving pulse of 4 fs duration is 110 THz, while the plasmonic pulse of 6.0 fs duration has a spectral bandwidth of 77 THz. Considering that the FT-limited pulse duration of a 77 THz spectral bandwidth centered at 375 THz is 5.9 fs, the plasmonic field is found to experience no significant group-delay dispersion within the waveguide.

The reduced spectral bandwidth of the plasmonic pulse is attributed to the spatial separation of the spectral power, as depicted in figure 2(c), leading to a prolonged inhomogeneous dephasing time [22]. The funnel-waveguide supports an anti-symmetric SPP mode propagating along its hollow core of decreasing effective radius [23]. The maximum wavelength that can be confined in the anti-symmetric SPP mode is restricted by the radius, so the spectral bandwidth continuously reduces as the SPP mode propagates along the hollow core of decreasing radius [24]. For the funnel-waveguide optimized with \( \xi = 0.5 \), our simulation shows that the spectral power separation can be squeezed to a short spot of \( \sim 200 \) nm length, thereby producing a few-cycle ultrashort plasmonic pulse that is adequate for the generation of isolated attosecond pulses. The two-color HHG process can also be considered in generating attosecond pulses as it offers some advantages based on temporal symmetry breaking [25–27].

3. Plasmon-driven high-harmonic spectra of the funnel-waveguide

Our simulation on the high-order EUV spectrum to be generated from the plasmon-driven HHG was conducted in the framework of the extended Lewenstein’s model based on the strong-field approximation [28]. Specifically, we adopted the approach of Husakou et al [17] that
Figure 2. Temporal and spectral characteristics of the plasmonic pulse produced from Pulse I (4 fs duration and 800 nm central wavelength) in the funnel-waveguide optimized with $\xi = 0.5$. (a) Normalized temporal profiles of the input pulse and the plasmonic pulse, (b) Fourier-transformed spectra of the input pulse and the plasmonic pulse and (c) spectral power distribution of three frequency components (320, 375, 430 THz) in the plasmonic pulse near the exit aperture.

enables incorporation of the spatial field inhomogeneity of the plasmonic pulse by use of first-order perturbation theory. Accordingly, the two-dimensional nonlinear dipole moment $\vec{d}(t)$ is expressed in the form

$$\vec{d}(t) = 2\text{Re} \left\{ \int_{-\infty}^{t} dt' \left( \frac{\pi}{\epsilon + i(t-t')/2} \right)^{3/2} \vec{d}^* \left[ \vec{p}_{st} - e\vec{A}(t) \right] \exp \left[ -i \frac{S(t, t')}{\hbar} \right] \left[ \vec{E}(t') \cdot \vec{d} \left[ \vec{p}_{st} - e\vec{A}(t') \right] \exp \left[ -i \int_{-\infty}^{t} w(t') dt' \right] \right] \right\}. \tag{1}$$

In equation (1), $e$ is the electric charge, $\vec{E}(t)$ is the electric field of the plasmonic pulse obtained by FDTD simulation and $\vec{A}(t)$ is the associated vector potential. In addition, $\epsilon$ is a positive regularization constant, $w(t)$ is the tunnel field ionization rate calculated by the Ammosov–Delone–Krainov (ADK) theory [29]. Further, $d$ is the dipole matrix element for bound–free transitions, and $p_{st}$ and $S$ are the modified stationary momentum and quasi-classical...
action, respectively:

\[
d(\vec{p}) = \frac{2^{7/2}(2\pi m_e I_p)^{5/4}}{\pi} \frac{\vec{p}}{(\vec{p}^2 + 2m_e I_p)^{3/2}},
\]

(2)

\[
\tilde{p}_a(t, t') = e \left[ \tilde{A}(t') + \frac{\Delta B - \tilde{A}(t') \Delta t + \beta [0.5(\Delta \tilde{B})^2 - \Delta \tilde{D} + \tilde{C}(t') \Delta t]}{\Delta t - \beta [2 \Delta \tilde{G} + \Delta t (B(t) + \tilde{B}(t'))]} \right],
\]

(3)

\[
S(t, t') = I_p \Delta t - \sum_{i=x,y} 0.5e^2m_e^{-1}[(\Delta B_i)^2/\Delta t - \Delta C_i]
+ \sum_{i=x,y} \beta e^2m_e^{-1}(p_{a,i}/e)^2[2\Delta G_i + [B_i(t) + B_i(t')]\Delta t]
+ \sum_{i=x,y} \beta e^2m_e^{-1}(p_{a,i}/e)(-\Delta C_i \Delta t + 2\Delta D_i)
+ \sum_{i=x,y} \beta e^2m_e^{-1}[(C_i(t) + C_i(t'))\Delta B_i - 2\Delta F_i \Delta t],
\]

(4)

with

\[
\begin{align*}
\tilde{A}_i(t) &= -E_i(t), \quad \tilde{B}_i(t) = A_i(t), \quad \tilde{C}_i(t) = A_i^2(t), \\
\tilde{D}_i(t) &= C_i(t), \quad \tilde{F}_i(t) = C_i(t)E_i(t), \quad \tilde{G}_i(t) = B_i(t),
\end{align*}
\]

where \(i = x, y\) and \(\beta = e/m_e d_{inh}\). For any function \(f\), we define \(\Delta f \equiv f(t) - f(t')\). In this simulation, with the focus on the effect of spatial field inhomogeneity, we neglected the freed electron absorption near the metallic surface. The reason is that, unlike planar nanostructures, the 3D funnel-waveguide yields a strong plasmonic electric field along the central axis of the hollow core, not on the metallic interface. Consequently, EUV emission is presumed to occur without significant electron absorption by the metallic wall surrounding the hollow core. Besides, our calculation revealed that the spatial inhomogeneity of \(d_{inh} = 200\)nm gives rise to a small correction to the unmodified three-step HHG model, which consequently justifies adopting the first-order perturbation theory. Specifically, the amount of first-order correction made by the introduction of \(\beta\) accounts for about 1% of the unmodified terms in the quasi-classical momentum and action given in (3) and (4). An extra ADK rate was considered in order to take into account the ground state depletion of a target gas atom.

Figure 3 presents the calculation result of the harmonic spectra obtained from Pulse I with Ar gas. The induced plasmonic pulse was assumed to have an enhanced peak intensity of 200 TW cm\(^{-2}\) with a zero CEP. The EUV spectra plotted in figure 3(a) with two different values of \(d_{inh}\) indicate how the plasmon-driven HHG process is affected by the spatial inhomogeneity of the induced plasmonic pulse. The value of \(d_{inh}\) reduces from 1000 to 200 nm when the ellipticity \(\xi\) varies from 0.5 to 1.0 as discussed in figure 1(e). For \(d_{inh} = 200\) nm, which is the case of the circular cross-section with \(\xi\) set to 1.0, the harmonic cut-off extends considerably to beyond the fortieth harmonic (H40). At the same time, even-order harmonics are produced together with odd-order harmonics. These HHG spectrum-broadening phenomena near the cut-off region that are desirable in view of the generation of attosecond pulses result from the inversion symmetry breaking by the spatial inhomogeneity of the plasmonic field. The broken spatial symmetry leads freed electrons to select the short quantum paths more efficiently. At
Figure 3. Simulation for the plasmon-driven HHG from Pulse I-induced plasmonic pulse (6 fs duration, 2.7 fs in the leading edge, 3.3 fs in the trailing edge, 200 TW cm$^{-2}$ intensity and 800 nm central wavelength).  
(a) High-harmonic spectra for different field inhomogeneity lengths with CEP $= 0$.  
(b) High-harmonic spectra for different CEP values with $d_{inh} = 200$ nm. The downward arrows indicate the harmonic cut-off positions for the corresponding CEP values.  
(c) Attosecond pulses generated with different harmonics filtering ranges with CEP $= 0$ and $d_{inh} = 200$ nm.  
(d) Comparison of single attosecond pulses generated with two different values of field inhomogeneity.

The same time, ponderomotive energy in short path trajectories is presumed to increase due to the stronger acceleration of freed electrons in the continuum state prior to recollision onto the parent ions [16]. This physical interpretation is backed up by monitoring the variation of the HHG spectrum with different CEP values. Since the inversion symmetry is broken by the increased plasmonic field inhomogeneity, the high-harmonic spectrum for CEP $= 0$ is no longer the same as that for CEP $= \pi$ as calculated in figure 3(b) for the case of $d_{inh} = 200$ nm. When CEP $= \pi$, freed electrons experience the plasmonic field on the decreasing side that decelerates the electron motion in the continuum state, shortening the harmonic cut-off.
Figure 3(c) presents the attosecond pulses computed with different harmonic filtering ranges. Three cases of (15–19th), (10–45th), and (40–60th) for harmonics filtering were tested in consideration of available EUV-region metal filters such as Ge, Al and Zr. It is noted that, when the plasmonic field is almost homogeneous with $d_{\text{inh}} = 1000 \, \text{nm}$, isolated attosecond bursts of $\sim 360 \, \text{as}$ duration can be produced only from (35–60th) filtering. On the other hand, the plasmonic field inhomogeneity of $d_{\text{inh}} = 200 \, \text{nm}$ compresses the pulse duration to 210 as. At the same time, the pedestal pulses adjacent to the main burst are effectively suppressed as shown in figure 3(d). This indicates that the plasmonic field inhomogeneity of the circular cross-section is useful in producing isolated, sharp attosecond pulses.

4. Time–frequency distribution of plasmon-driven high-order harmonic generation spectra

In order to clarify in more detail how the presence of spatial field inhomogeneity affects the plasmon-driven HHG spectrum, the so called time–frequency distribution was calculated as shown in figure 4. For this simulation, with the plasmonic-driven HHG conditions of figure 3, we first solved Newton’s law of motion for the given inhomogeneous plasmonic field so as to obtain the generated EUV photon energy as a function of either the ionization time or recombination time, i.e.,

$$m_e \frac{d^2 x}{d t^2} = - \frac{\partial H_I(x, t)}{\partial x},$$

$$H_I(x, t) = ex E(t) = ex (1 + x/d_{\text{inh}}) E_0 f(t) \cos(\omega_0 t + \phi_{\text{CEP}}).$$

In the above equation of electron motion, the variable $x$ is the instantaneous position of a freed electron in its trajectory, $m_e$ is the electron mass and $H_I(x, t)$ is the interaction Hamiltonian. Other variables $E_0$, $f(t)$, $\omega_0$ and $\phi_{\text{CEP}}$ are the peak amplitude, envelope function, center frequency and CEP of an enhanced few-cycle plasmonic pulse, respectively. The time-dependent electron trajectory $x(t)$ for a given inhomogeneous plasmonic field, which is the solution of equation (5), can be calculated using the Verlet algorithm [30]. The boundary conditions are as given; the electron becomes ionized at $t_i$ so that $x(t_i) = 0$ and $v(t_i) = dx(t_i)/dt = 0$, and terminates its laser-driven journey by recombination onto the parent ion at $t_r$, i.e., $x(t_r) = 0$. Once $x(t)$ and $v(t)$ are obtained, the EUV photon energy emitted from the electron upon return is finally determined as $E_{\text{EUV}} = m_e v(t_r)^2/2 + I_p$ with $I_p$ being the ionization potential of the gas atom.

Figure 4(a) plots the produced EUV photon energies in units of the harmonic order as a function of the ionization time $t_i$ (magenta circle) and also the recombination time $t_r$ (cyan rectangle). Since the funnel-waveguide has a relatively small degree of spatial field inhomogeneity compared to that of bow-ties [11, 17], its effect is apparent when the electric field of the driving plasmonic pulse reaches its maximum, i.e. during a temporal period of $\sim 4.4$ to $\sim 5.4$ laser cycles (1 cycle $= 2.67 \, \text{fs}$). The simulation result of figure 4(a) clearly indicates that, for the spatial field inhomogeneity given by $d_{\text{inh}} = 200 \, \text{nm}$ and CEP $= 0$, the harmonic cutoff is extended most extensively beyond the 35th harmonic compared with other combinations of $d_{\text{inh}}$ and CEP. In addition, the field inhomogeneity tends to suppress electrons of low ponderomotive energies driven by previous laser cycles, allowing for more high energy electrons triggered by the very peak of the plasmonic field to participate in generating higher-order harmonics near the cutoff. Between the two paths, short and long, for returning electrons, the latter contributes less
Figure 4. Time–frequency distribution of the plasmon-driven HHG spectrum with different inhomogeneity and CEP values. (a) Generated EUV photon energies as a function of the ionization time (magenta circle) and the recombination time (cyan rectangle) for an enhanced few-cycle plasmonic pulse (6 fs, 200 TW cm\(^{-2}\)). The yellow solid box indicates the time–frequency data during a temporal period of 4.4–5.4 laser cycles and its enlarged view is compared with those of two different conditions of field inhomogeneity or CEP. (b) Time–frequency distribution of the plasmon-driven HHG spectrum within a temporal period of 4.5–6.5 laser cycles (green dashed box) calculated by full quantum mechanical analysis using the dipole moment obtained from equation (1).

to HHG due to the wave-packet diffusion effect [28]. As a consequence, the HHG spectrum near the harmonic cut-off is produced mostly by the short-path electrons of high energies, resulting in a broadened, continuous EUV spectrum in which protruding harmonic peaks are not seen at integer multiples of the fundamental frequency of the driving laser.

Figure 4(b) shows the time–frequency distribution directly attained through windowed Fourier transform of the dipole moment of equation (1). This quantum mechanical calculation reveals that for the case of \(d_{\text{inh}} = 200\) nm and CEP = 0, the short-path electron produced by the peak of the plasmonic pulse overwhelms other electrons of the previous laser pulses, particularly for harmonic orders above \(\sim 30\). This implies that high energy electron wave-packets being free from quantum interference over a wide spectral span can lead to effective generation of single isolated attosecond pulses. For other cases of \(d_{\text{inh}} = 1000\) nm and CEP = \(\pi\), wave-packet interference occurs among several electrons of different quantum paths, bringing
Figure 5. Simulation for plasmon-driven HHG using the two-color pulse combining Pulse II-1 with Pulse II-2. Panels (a), (c) and (e) are for the case of combining Pulse II-1 (12 fs duration, 100 TW cm$^{-2}$ intensity and 800 nm central wavelength) and Pulse II-2 (60 fs duration, 80 TW cm$^{-2}$ intensity and 1560 nm central wavelength. Panels (b), (d) and (f) are for another case in which the duration of Pulse II-1 and II-2 are reduced to 6 and 15 fs, respectively, with other pulse parameters remaining the same. Panels (a) and (b) show synthesized temporal profiles of the produced plasmonic pulses, panels (c) and (d) are the corresponding two-color HHG spectra and (e) and (f) are single attosecond pulses generated by harmonics filtering with $\delta = 0$.

about clearly modulated harmonic peaks but with shortened harmonic cut-off. In addition, as inferred in figure 4(b), it is noteworthy that the presence of spatial inhomogeneity compels returning electrons to emit attosecond EUV bursts repeatedly at every full cycle, not half cycle, of the driving plasmonic field. This permits generation of even-order harmonics, which results
from the broken inversion symmetry. This time–frequency analysis based on either classical or quantum mechanical approaches gives a good exploration for the HHG spectra of figure 3 obtained by the semi-classical three-step model.

5. Plasmon-driven two-color high-harmonic spectra of the funnel-waveguide

Figure 5 presents the simulation result for the two-color input pulse synthesized by combining Pulse II-1 and Pulse II-2. The temporal profile of the combined pulse is given in figure 5(a). The corresponding high-harmonic spectrum was calculated following the same procedure as in figure 3, with the phase delay $\delta$ between Pulse II-1 and Pulse II-2 being set to zero. The pulse intensity and $d_{inh}$ were assumed to be 100 TW cm$^{-2}$ and 200 nm for Pulse II-1 and 80 TW cm$^{-2}$ and infinite for Pulse II-2 by reflecting the near-field intensity distribution obtained by FDTD simulation.

As shown in figure 5(c), the HHG spectrum resulting from the combined plasmonic field yields a stretched harmonic cut-off in comparison to that from Pulse II-1 alone. Specifically, high-order harmonics are generated at $(2N)\omega_0$, $(2N + 1/2)\omega_0$, $(2N + 1)\omega_0$ and $(2N + 3/2)\omega_0$, where $N$ is an integer and $\omega_0$ is the center frequency of Pulse II-1; even-order harmonics at $(2N)\omega_0$ result from the spatial inhomogeneity caused by Pulse II-1, and those harmonics at $(2N + 1/2)\omega_0$ and $(2N + 3/2)\omega_0$ have their basis in the inversion symmetry breaking added by Pulse II-2. Although the high harmonic spectra are sensitive to the relative phase delay between the orthogonal two color laser fields, harmonic filtering of the plane cut-off region of (50–60th) yields a single attosecond burst with a duration of $\sim$370 as (see figure 5(e)).

Another simulation result is shown in figures 5(b), (d) and (f) in which the pulse duration of Pulse II-1 and Pulse II-2 was simply shortened to 6 and 15 fs, respectively, while other pulse parameters including CEP remained the same. As expected, the HHG spectrum in figure 5(d) exhibits a more flattened spectral profile as well as strengthened harmonic emission near the cut-off region as compared to that of figure 5(c). This is because a more efficient temporal gate can be created on the HHG process by the added ultrashort infrared pulse, leading to a further reduction in the attosecond pulse duration to $\sim$330 as (see figure 5(f)).

In the filtering process of attaining isolated attosecond pulses, the idea of utilizing the polarization gating technique may be considered, as previously investigated for nano-antennas [14, 31], so as to minimize the photon flux loss encountered in the spectral range selection using metal filters. Besides, the exit aperture of the funnel-waveguide can act as a spectral high-pass filter with long wavelengths being blocked by the diffraction limit, so controlling the exit aperture size would be a convenient means of spectral range selection for generating isolated attosecond pulses.

6. Conclusions

To conclude, our theoretical analysis elaborated in this study proved the feasibility of generating isolated attosecond pulses at high repetition rates by means of plasmonic field enhancement using the nano-structured funnel-waveguide of Park et al [12]. Two distinct input lasers were considered in our simulation; one is a few-cycle 4 fs laser of 800 nm carrier wavelength and the other is a two-color laser synthesized by combining a multi-cycle 10 fs pulse of 800 nm carrier wavelength with a perturbed long 60 fs pulse of 1560 nm carrier wavelength. The pulse duration of the generated attosecond pulses was calculated to be $\sim$210 as for the few-cycle laser
and \( \sim 370 \) as for the two-color laser, respectively. This simulation result suggests that the funnel-waveguide could be a useful tool to produce isolated attosecond pulses flashing at high repetition rates with the high spatiotemporal resolution needed for precise attosecond-scale spectroscopic studies such as an EUV pump–probe experiment.

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