Divergence gating towards far-field isolated attosecond pulses

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Abstract

Divergence gating, a novel method to generate far-field isolated attosecond pulses (IAPs) through controlling divergences of different pulses, is proposed and realized by relativistic chirped laser–plasma interactions. Utilizing various wavefronts for different cycles of incident chirped lasers, reflected harmonics with minimum divergences are obtained only at the peak cycle when plasma targets are adjusted to proper distances from foci of lasers. Therefore, the corresponding attosecond pulse is isolated in far field due to much slower decay during propagation than others. Confirmed by three-dimensional numerical simulations, millijoule-level sub-50 as IAPs with intensity approaching $10^{16}$ W cm$^{-2}$ ($10^{17} – 10^{18}$ W sr$^{-1}$) are obtained by our scheme, where low-order harmonics can be preserved.

1. Introduction

Isolated attosecond pulses (IAPs) have attracted broad interests during decades [1–3] for their rich applications among the resolution and control of ultrafast electron movement in atoms [4], molecules [5], materials [6, 7], and high-energy-density plasma [8, 9]. Getting rid of intensity limitations from the ionization threshold, relativistic high-order harmonic generation (RHHG) [10–15] has been regarded as the most promising way towards ultraintense attosecond pulses (APs) for attosecond-pump-attosecond-probe experiments [16], which are challenging for those produced from gaseous high-order harmonic generation [17] due to low photon flux as well as pulse energy in the magnitude of $10^{-9}$ J [16]. However, such APs are obtained generally in form of trains as the harmonic emission is driven by periodical interactions between lasers and overdense plasmas [13].

Series of methods [18–28] have been proposed to generate IAPs through RHHG, which can be classified as ‘temporal gating’ and ‘spatial gating’. Different from temporal gating where APs can only be generated efficiently once because of few-cycle incident lasers [18], well-controlled laser polarization [19–21] or specially designed targets [22] as well as their transparency [23, 24], spatial gating separates APs in space through different reflected directions of them due to attosecond lighthouses [25–27] or tightly focused lasers [28]. For temporal gating, not only do some of methods require ultrathin targets [22–24], but also most of them have to filter out low-order harmonics (photon energy $E_{ph} \lesssim 30$ eV typically) which are usually produced efficiently for several optical cycles. While attosecond lighthouse effects used in spatial gating can relax the requirement of filters to $E_{ph} \gtrsim 20$ eV, experimentally challenging preplasma scales $L \lesssim 10^{-2} \lambda_0$ is required [25], which greatly limits the generation efficiency at the same time [29, 30]. Here, $\lambda_0$ represents the central wavelength of incident lasers. The introduction of high-pass spectral filters greatly suppresses the intensity of IAPs due to fast decay of harmonic intensities [13]. What’s more, the removed spectra ranging from near ultraviolet (NUV) to vacuum ultraviolet (VUV) are critical for attosecond science in valence-shell phenomena [31, 32], molecule excitation [33] and nonlinear cluster dynamics [9, 34].

To generate IAPs with spectra and energy preserved as much as possible, the idea of divergence gating is proposed in this paper. Divergence gating separates APs through the modulation of divergences, where APs
emitted in the peak optical cycles of incident lasers have minimum divergences and experience much lower energy density decay during propagation than other pulses. As all harmonics including fundamental frequency waves are influenced, divergence gating has the potential to generate near-axis IAPs in the far field with lower requirements of filters, even preserving all harmonics. Recently, the optically-curved plasma surfaces due to laser–plasma interactions have been proved to act like plasma mirrors (PMs) which significantly affect the divergences of harmonics from RHHG and can be controlled by the convex wavefronts of defocused quasi-monochromatic incident lasers. However, as the wavefronts are identical, significantly affect the divergences of harmonics from RHHG and can be controlled by the convex surfaces due to laser–plasma interactions have been proved to act like plasma mirrors (PMs) which determine the bandwidth of Gaussian envelope by 0.

The scheme of divergence gating is illustrated in figure 1(a). A chirped Gaussian incident laser (orange plane of incident laser), the transverse incident electric field $E$ acting on the target is given as

$$E = E_0 \frac{w_0}{w_s} \exp \left(-2 \ln \frac{\xi^2}{\tau_L^2} - \frac{r^2}{w_s^2}\right) \sin \left[\frac{\omega_0 \left(\xi + \frac{\Delta \xi^2}{2}\right)}{2} - \frac{kr^2}{2R_L} + \phi_0\right],$$

where $r = \sqrt{\rho^2 + s^2}$ and $\xi \equiv t - x_t/c$. Here, $w_s = w_0\sqrt{1 + (\xi/\Delta \xi)^2}$, $R_L = x_t + \Delta \xi/\epsilon_0$, $\Delta \xi = \pi w_0^2/\lambda$, $k = 2\pi/\lambda$, $\lambda = \lambda_0/(1 + \xi)$, $\omega_0 = 2\pi c/\lambda$, and $s = \pm \sqrt{(\Delta \omega/\omega_0 \tau_l)^2 - (4 \ln 2/\omega_0^2 \tau_l^2)^2}$ [41] represent the beam radius at which the field amplitude is 1/e times of the axial value, radius of curvature, Rayleigh length, wave number, wavelength, central angular frequency and chirp parameter (>0 for positive chirp and <0 for negative chirp) of the incident laser on plasma surface, respectively. Besides, $w_0$, $\Delta \omega$, $\tau_l$, $c$, $\gamma$ and $z$ represent the beam waist, band width, pulse duration (full-width at half-maximum), speed of light in vacuum and the positions in two orthogonal transverse directions, respectively. The phase $\phi_0 = \phi_C + \phi_G$ includes the contribution of carrier envelope phase $\phi_C$ (CEP) and Gouy phase $\phi_G \equiv \arctan(x/\Delta \xi)$. We note that $\xi = 0$ when the temporal envelope of the laser field achieves its maximum. Besides, as illustrated in figure 2(b), values of $s$ increase quickly with the increase of $\Delta \omega$ or decrease of $\tau_l$ for most of situations ($\tau_l > 4\sqrt{2 \ln 2}/\Delta \omega$ in specifically), while the minimum of the second one is limited by the first due to the transform limit (white area). The relationship between $s$ and $\tau_l$ for a given $\Delta \omega$ is non-monotonical as both the Gaussian envelope and frequency chirping contribute to the bandwidth. Therefore, for fixed $\Delta \omega$, the chirp parameter $s$, which characterizes the changing rate of frequency, changes nonlinearly with different $\tau_l$ which determines the bandwidth of Gaussian envelope by $0.44/\tau_L [46]$ and equals 0 for transform-limited (TL) and infinite-duration beams.

Reflected along X-axis, the reflected laser (purple hollow column) is focused by the optically-curved dense electron layer, which is compressed by the driving laser and acts like a PM. Due to the spatial distribution of the light pressure $\propto E^2$, the displacement of surface electrons varies in transverse directions, resulting in parabolic PMs formed naturally, whose focal length $f_p \approx w_0^2(1 + e^{-1})/4L \cos^2 \alpha$ and profile illustrated in figure 1(b) (solid). Here, $E \equiv a_L \lambda_0(1 - \sin \alpha)/\pi L$ accounts for the electron dynamics of

2. Theoretical analysis

The scheme of divergence gating is illustrated in figure 1(a). A chirped Gaussian incident laser (orange hollow column), which is linear-polarized in the y direction, propagates along X-axis and irradiates on the overdense plasma target (grey block) obliquely with angle $\alpha$ and central wavelength $\lambda_0$, where exponential preplasma density profile with scale $L$ is assumed. Keeping a distance $x_t$ between the plasma bulk and focal plane of incident laser, the transverse incident electric field $E$ acting on the target is given as

where $r \equiv \sqrt{\rho^2 + s^2}$ and $\xi \equiv t - x_t/c$. Here, $w_s = w_0\sqrt{1 + (\xi/\Delta \xi)^2}$, $R_L = x_t + \Delta \xi/\epsilon_0$, $\Delta \xi = \pi w_0^2/\lambda$, $k = 2\pi/\lambda$, $\lambda = \lambda_0/(1 + \xi)$, $\omega_0 = 2\pi c/\lambda$, and $s \equiv \pm \sqrt{(\Delta \omega/\omega_0 \tau_l)^2 - (4 \ln 2/\omega_0^2 \tau_l^2)^2}$ [41] represent the beam radius at which the field amplitude is 1/e times of the axial value, radius of curvature, Rayleigh length, wave number, wavelength, central angular frequency and chirp parameter (>0 for positive chirp and <0 for negative chirp) of the incident laser on plasma surface, respectively. Besides, $w_0$, $\Delta \omega$, $\tau_l$, $c$, $\gamma$ and $z$ represent the beam waist, band width, pulse duration (full-width at half-maximum), speed of light in vacuum and the positions in two orthogonal transverse directions, respectively. The phase $\phi_0 \equiv \phi_C + \phi_G$ includes the contribution of carrier envelope phase $\phi_C$ (CEP) and Gouy phase $\phi_G \equiv \arctan(x/\Delta \xi)$. We note that $\xi = 0$ when the temporal envelope of the laser field achieves its maximum. Besides, as illustrated in figure 2(b), values of $s$ increase quickly with the increase of $\Delta \omega$ or decrease of $\tau_l$ for most of situations ($\tau_l > 4\sqrt{2 \ln 2}/\Delta \omega$ in specifically), while the minimum of the second one is limited by the first due to the transform limit (white area). The relationship between $s$ and $\tau_l$ for a given $\Delta \omega$ is non-monotonical as both the Gaussian envelope and frequency chirping contribute to the bandwidth. Therefore, for fixed $\Delta \omega$, the chirp parameter $s$, which characterizes the changing rate of frequency, changes nonlinearly with different $\tau_l$ which determines the bandwidth of Gaussian envelope by $0.44/\tau_L [46]$ and equals 0 for transform-limited (TL) and infinite-duration beams.

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plasma [35]. Its reciprocal $e^{-1}$ describes the influence of laser intensity on the optical properties of curved PMs, which is negligible when $a_1 \gg 1$ as $e^{-1} \sim 0$. The dimensionless parameter $a_{1s} \equiv e\bar{E}/m_ee\psi_0$ is the normalized amplitude of electric field strength with $m_e$ and $e$ representing the mass and absolute charge of electrons respectively. Assuming the reflected harmonics have Gaussian spatial distributions either, their radii of curvature $R_i$ on PMs satisfy $1/R_i - 1/R_f = 1/\rho_\pi \cos \alpha$ on the meridional plane [47] and are given as

$$R_i = \frac{x_f^2 + (1 + s_\xi)^2 x_{00}}{x_f - b_0(1 + s_\xi)^2 x_{00}}$$  \hspace{1cm} (2)$$

where $b_0 \equiv 4\pi L \cos \alpha / \lambda_0(1 + e^{-1})$ is actually the ratio of the optimal $x_f$ to $x_{00}$ to realize divergence gating as discussed later and $x_{00} \equiv \pi \epsilon \omega_0^2 / \lambda_0$ is the Rayleigh length of incident lasers for the central wavelength. It is clear that the curvature $R_i^{-1}$ on $\xi$ is introduced by various wavefronts of chirped lasers (dashed) and profiles of parabolic PMs (solid) as shown in figure 1(b). Then, the propagation of reflected harmonics is considered with their radii on PMs $w_{n\beta} = \beta_n w_0 (\beta_n \leq 1)$ assumed, where the intensity is $1/e^2$ times of the axial value. In this paper, symbols with subscript $n$ represent the corresponding physical quantities for harmonics whose photon energy $E_{\beta n} = nc_0 \epsilon_0$ and $c_0$ is the energy of single photon at central wavelength $\lambda_0$. Utilizing the well-known $q$-parameters of Gaussian beams [46], we have $q_{\beta n} = R_i^{-1} - i\lambda_0 / n\pi \epsilon_0 w_0^2$ and $q_{\beta n} = -i\beta_n \lambda_0 \epsilon_0 w_0^2$ for harmonics on the PM and at their focal planes respectively, where $x_{0n}$ represents the Rayleigh length of the corresponding harmonic. Considering an equivalent definition of $q$-parameter as $q \equiv x + ix_0 \epsilon_0$ [46], $q_{0n}$ and $q_{\beta n}$ satisfy $q_{i\beta n} = (q_{0n} + x_{0n})^{-1}$ for the free propagation distance $x_{0n}$ in vacuum between the two positions. After derivation, the expressions of $x_{0n}$ and $x_{R\beta n}$ are given as

$$x_{0n} = n/\beta_n^2 \left[ x_f / (1 + s_\xi)^2 x_{00} - b_0 \right] x_{R\beta n},$$  \hspace{1cm} (3)$$

![Figure 1. 3D PIC simulations and far-field results. (a) 3D schematic for the generation of far-field IAPs through divergence gating, where a $\gamma$-polarized Gaussian chirped laser (orange hollow column) obliquely irradiates an overdense target (grey block).](image-url)
Intensity on PMs take full advantage of divergence gating. Specifically, for optimal field condition $x_f o c u s$ represents the incident laser exactly offset the focusing effect of concave PMs within the central optical period.

\[ I_f o c u s \approx I_n a r e a / [1 + (x_{n a r e a}/x_{f o c u s})^2] \]

where $I_{n a r e a}$ represents the intensity of the reflected harmonic at its focus. Similarly, the near-field harmonic intensity on PMs $I_{n a r e a} = I_{f o c u s} / [1 + (x_{n a r e a}/x_{f o c u s})^2]$. Therefore, $I_{n a r e a} \approx I_n a r e a / [1 + (x_{n a r e a}/x_{f o c u s})^2]$. Assuming $I_{n a r e a} = I_n a r e a / [1 + (x_{n a r e a}/x_{f o c u s})^2]$ and considering equations (3)–(5) as well as the far field condition $x_{f o c u s} \gg x_{f o c u s}$, we obtain

\[ I_{n a r e a} \approx I_n a r e a / [1 + (x_{n a r e a}/x_{f o c u s})^2] \]

where $I_n a r e a$ and $\eta_n(\xi)$ represent the focal intensity of incident lasers and generation efficiency of harmonics, respectively. As $I_{n a r e a} \propto \theta_n^{-2}$, the minimum $\theta_n$ should be achieved exactly at the peak laser cycle ($\xi = 0$) to take full advantage of divergence gating. Specifically, for optimal $x_f = b_0 x_{f o c u s}$, we get $R_{t o p}$ and $\theta_{t o p}$ as

\[ R_{t o p} = b_0 + (1 + \xi)^2 b_0^{-1} x_{f o c u s} \]

\[ \theta_{t o p} = \theta_0 \sqrt{(n \beta_\alpha(\xi))^{-2} + \beta_\alpha^2(\xi) b_0^2 (1 - (1 + \xi)^{-2})^2} / (1 + b_0 (1 + \xi)^{-2}) \]

In this situation, foci of harmonics generated at the peak laser cycle ($\xi = 0$) are on the PMs as $x_{f o c u s} = 0$ according to equation (3), indicating that the convex wavefront of the incident laser exactly offset the focusing effect of concave PMs within the central optical period.
As shown in equation (7), the flat near-field wavefronts \( R_{0}^{\text{ph}} = \infty \) of reflected pulses are realized when \( \xi = 0 \), which indicates the minimum divergence and lowest decay during propagation comparing to harmonics generated at other cycles with either concave \( (R_{0}^{\text{ph}} < 0) \) or convex \( (R_{0}^{\text{ph}} > 0) \) wavefronts on PMs. Considering APs are generated efficiently once a cycle \( \sim T_0 \equiv \lambda_0/c \) for oblique incidence, we have \( \theta_0^{\text{ph}}(\pm T_0) = \theta_0\sqrt{n - 1}j_1^n(\pm T_0) + 4j_0^n(\pm T_0)\beta_0^2(1 + I_0)/I_0 \) for small \( sT_0 \ll 1 \), which is much bigger than the divergence of harmonics at the peak cycle \( \theta_0^{\text{ph}}(0) = \theta_0/n\beta_0(0)\sqrt{1 + I_0} \). Defining \( \Gamma_0(\xi) \equiv \beta_0^2(\xi)\theta_0^{-2} \), we get the effect of divergence gating as

\[
\frac{\Gamma_0(0)}{\Gamma_0(\pm T_0)} = 1 + 4n^2\beta_0(0)\beta_0^2(0)T_0^2 \tag{9}
\]

which is independent on \( \tau_1 \) for a fixed chirp parameter.

Finally, as the satellite harmonics can be ignored compared with the one generated at the peak laser cycle when \( I_0(0)/I_0(\pm T_0) \gg 1 \), we get the critical photon energy of filters \( E_{\text{ph}}^{\text{min}} \sim 1.26\varepsilon_0/sb_0 T_0 \beta_0^2(0) \) to produce far-field IAPs through divergence gating. Here, \( e \) is the base of the natural logarithm. Considering that influences of temporal envelope \( \exp(-4 \ln 2 \xi^2/\tau_1^2) \) and generation efficiency \( \eta_0(\xi) \) on \( I_0(0)/I_0(\pm T_0) \) have been neglected during theoretical analysis for simplicity, the requirements of \( E_{\text{ph}}^{\text{min}} \) can be further relaxed as shown in simulations. We note that there is astigmatism for reflected pulses from parabolic PMs when \( \alpha \neq 0 \) as harmonics satisfy \( 1/R_0 - 1/R_0 s = \cos \alpha /\xi f \) on the sagittal plane \([48]\). As a result, the corresponding optimal defocused distance \( x_1 = b_0 x_{ph} \cos \alpha \), whose relative error comparing to the one on the meridional plane is smaller than 0.1 when \( \alpha < 18^\circ \) and can be neglected.

Besides, the chirp induced by plasma surface deformation hardly affects our scheme as it does not influence the focusing properties of PMs.

3. Simulation results

3D numerical simulations are carried out with time-domain near-to-far-field transformation (TDNFFT) method \([49, 50]\) to verify our theory where particle-in-cell (PIC) code 'EPOCH' \([51]\) is used to provide near-field data for it. In simulations, except noted in text, the Gaussian-like linear-polarized chirped laser (orange hollow column) with \( \lambda_0 = 800 \text{ nm} (\varepsilon_0 \approx 1.55 \text{ eV}), w_0 = 2 \mu \text{m}, \tau_1 = 8 \text{ fs} \sim 3T_0, \Delta \omega = 0.5\omega_0 \) \((s \approx 0.162T_0^{-1})\), \( \phi_0 = 0 \) and peak intensity \( I_0 \approx 1.92 \times 10^3 \text{ W cm}^{-2} (\alpha_0 = 30) \) at focus is incident along \( X \)-axis from the right boundary as shown in figure 1(a). The quadratic polynomial phase factor about \( \xi \) in equation (1) is introduced to PIC simulations to initialize the chirp. According to reference \([45]\), the influence of temporal envelope on the formula of incident beams considered in simulations can be neglected.

Irradiated by lasers with incident angle \( \alpha = 15^\circ \) and defocused distance \( x_1 \), the bulk density \( n_0 \) and thickness \( d \) of the thin foil (grey block) are 100n_0 \((n_0 = m_0\omega_0^2/4\pi\varepsilon_0^2)\) and 0.5\lambda_0 respectively. Clearly, for the peak cycle \( (\xi = 0) \), the laser intensity acting on plasma surfaces \( I_0 = I_0 / (1 + x_1^2/x_{\Sigma 0}^2) \). Before the bulk, the preplasma with exponential density scale \( L = 0.1\lambda_0 \) is set. As the interaction between the beam and preplasma is dominant and the width of interaction region is about the skin depth, the small thickness of plasma bulk is chosen to save computational resources whose rationality has been verified \([51]\). In addition to computationally expensive 3D PIC simulations which are performed with limited numerical resolution (illustrated in figure 1 with simulation time step \( \Delta t \approx T_0/300 \)), high-precision two-dimensional (2D) PIC simulations are also carried out \((\Delta t \approx T_0/2263)\) for figures 3–5, where the 3D far-field results are calculated through TDNFFT method based on axisymmetric approximations. We note that all far-field results are calculated in 3D geometries except for figure 3(f) where the result in 2D simulation is shown for the comparison. The size of 3D (2D) PIC simulation box \( x \times y \times z \) \((x \times y) = 8.5\lambda_0 \times 25\lambda_0 \times 20\lambda_0 \) \((10\lambda_0 \times 35.5\lambda_0)\), containing 850 \(\times 2500 \times 2000 \) \((8000 \times 28400)\) cells with \( (2 \times 16) \) and \( (1 \times 8) \) superparticles per cell for electrons and immobile ions, respectively. The hole-boring effect is negligible in our scheme. All of far-field results are calculated after reflected pulses propagating a distance \( x_{ph} = 10^5\lambda_0 \).

In figure 1(a) where \( x_1 = b_0 x_{ph} \sim 23.31\lambda_0, I_0 \approx 7.97 \times 10^{20} \text{ W cm}^{-2} \), curvatures of chirped incident laser change for different optical cycles due to the time-varying wavelengths (colorful solid), which is indicated by its spatial–temporal distribution of intensities (orange shadow). Such various spatial distributions cause variation in the focal length \( f_0 \) of the optically-curved PM, resulting in wavefronts (green solid) of near-field reflected laser change significantly for different optical cycles. As shown in the intensity distribution (purple shadow), the flat reflected wavefront is obtained only at the peak cycle \( \xi = 0 \), which meets our expectation and indicates the realization of divergence gating. Clearly, the IAP (red solid) is obtained in the far field after passing through a proper filter (brown cylinder, \( E_{\text{ph}} = 10 \text{ eV} \) while the
divergence gating is considered in figure 2(c). To obtain far-field IAPs, the requirement of $x_0$ for the near-field reflected pulses (purple solid in figure 1(a)). Quantitatively, as illustrated in figure 1(c), (an attosecond pulse train (APT, purple solid in figure 1(d)) is obtained in near field with the same filter used for the near-field reflected pulses (purple solid in figure 1(a)). The numerical results (red cycles) of curvature $1/R_{\text{opt}}$ are consistent with the theoretical predictions (black solid) for different cycles, calculated from wavefronts (green solid in figure 1(a) and equation (7) respectively. We note the varying distances between circles are introduced by the chirp.

Figure 2(a) illustrates the theoretical $\theta_n$ for the same case as figure 1(a), where $\beta_n$ is assumed to be 0.9 for $n = 5, 10$ and 20 (black, blue and red). Because the approximate values of $\theta_n^\delta(\xi) \approx \theta_n \sqrt{[n \cdot \beta_n \xi + 4\beta_n(\xi) - \beta_n \xi^2 + 4\beta_n \xi^2]/(1 + \beta_n^2)}$ represented by crosses are close to the corresponding precise theoretical values calculated from equation (8) (solids) when $|\xi| \leq T_0$, equation (9) is valid and capable to evaluate $\xi_{\text{min}}^\delta$ required by divergence gating. Besides, the lines and crosses confirm that the minimum divergences are obtained at the peak cycle ($\xi = 0$) as expected. Here, the chirped parameter $s \approx 0.162T_0^{-1}$ is determined by $\Delta \omega$ and $T_1$ as shown in figure 2(b). The influence of incident angle $\alpha$ on divergence gating is considered in figure 2(c). To obtain far-field IAPs, the requirement of $\xi_{\text{min}}^\delta$ is increased for bigger $\alpha$ which means more narrow spectra as well as less energy are preserved. Therefore, although the most efficient RHHG is achieved when $\alpha \sim 55^\circ$ [53], $\alpha = 15^\circ$ is chosen to balance the generation efficiency and isolation of AEs with astigmatism considered at the same time. Further, figure 2(d) illustrates the dependence of $T_n(0)/T_n(\pm T_0)$ on $E_{\text{ph}}$ for $\alpha = 15^\circ$. Clearly, IAPs are predicted to be produced when $E_{\text{ph}}^\delta \sim 10$ eV for $s \approx 0.162T_0^{-1}$ and $\beta_n = 0.9$. The same $\beta_n$ is set for all harmonics to show the effects of divergence gating briefly in theory and the value 0.9 is chosen based on the simulation result of harmonics contained in the peak AP.

As illustrated in figures 3(a) and (b) where $\alpha = 0^\circ$ ($I_\text{th} \approx 7.60 \times 10^{20}$ W cm$^{-2}$) and $15^\circ$ ($I_\text{th} \approx 7.97 \times 10^{20}$ W cm$^{-2}$) respectively, the unchirped incident laser with the same intensity as the chirped one can only generate ATPs (blue solids) on the axis when harmonics whose $E_{\text{ph}} < 10$ eV are filtered out. Corresponding to our theoretical predictions, once divergence gating is realized by chirped lasers, the intensity distributions of reflected AEs (red solids, $E_{\text{ph}}^\delta = 10$ eV) show that satellite pulses can be ignored if their emission intervals with the peak AP approach one cycle as their intensities are lower than $I_{\text{AP}}^\delta/e^2$ (red dashed). Here, $I_{\text{AP}}^\delta$ represents intensity of the peak AP and all far-field results are obtained when $x_{\text{tar}} = 10^3 \lambda_0$. However, as AEs are generated each half cycle for normal incidence, intense far-field IAP is only obtained for $\alpha = 15^\circ$ with intensity and duration about $10^{19}$ W cm$^{-2}$ and 40 as respectively, as shown in figure 3(b). The little higher $I_{\text{AP}}^\delta$ for cases using chirped lasers is caused by slightly smaller divergences. Comparisons with cases using TL pulses with the same bandwidth $\Delta \omega = 0.5\omega_0$ are discussed in the appendix. Besides, for the case shown in the top half of figure 3(b), its spatial–temporal distribution on z–t plane ($y = 0$) in the far field is illustrated in figure 3(c), where near-axis IAP is obtained with total angular spread reaching 12 mrad ($|z| \lesssim 600 \lambda_0$ when $x_{\text{tar}} = 10^3 \lambda_0$). Therefore, the energy of it is about 1.5 mJ and its peak intensity can be boosted to $10^{20} - 10^{21}$ W cm$^{-2}$ after refocused to micrometer scales. Besides, the curved...
wavefronts of APs indicate their arriving time varies with transverse positions of the observation plane, caused by the natural spherical far-field wavefront of the Gaussian beam whose radius approximates to \( x_{\text{far}} \).

The time-frequency analysis of APs obtained from chirped (the top half) and non-chirped driving lasers (the bottom half) are presented in figure 3(d) and (e), which verify that divergence gating greatly suppresses intensities of satellite APs while hardly affect the peak one as expected. What’s more, to compare with 3D far-field results shown in figure 3(b), the 2D result (red solid) for chirped laser–plasma interactions (same parameters as the case represented by the red solid in figure 3(b)) is illustrated in figure 3(f). As expected, a weaker effect of divergence gating is observed in the 2D situation where the APT results for unchirped driving pulses (blue solid in figure 3(b)).

Our scheme works for a broad range of parameters as illustrated in figures 4(a)–(c) where filters with \( E_{\text{ph}}^{\text{min}} = 10 \) eV are used. Figure 4(a) shows that with the increase of band width \( \Delta \omega \), the intensity rate between the strongest and second strong APs \( I_{\text{rate}} \) (red circle) improves while the duration \( \tau_{\text{AP}} \) of peak APs (blue triangle) keeps constant \( \approx 40 \) as, consistent with our theoretical predictions shown in figure 2(d). Besides, the smallest bandwidth \( \sim 0.35\omega_0 \) is required to obtain IAP in the far field. Figure 4(b) shows the relations of \( L \) with \( I_{\text{rate}} \) and IAPs can be obtained for \( 0.075 \lesssim L/\lambda_0 \lesssim 0.125 \) which is feasible in experiments nowadays [54]. We note that the decline of the performance of divergence gating for \( L > 0.1\lambda_0 \) stems from the decreased harmonic generation efficiency as well as worse optical properties of PMs because of instability [55]. Furthermore, there is about 50% probability to generate IAPs for random \( \phi_0 \), i.e. random CEP, as illustrated in figure 4(c), which is capable to perform pump-probe experiments requiring IAPs without the stabilization of CEP with the help of CE tagging technology [18, 56, 57]. We note that such result is three times of those obtained from reference [18] where unchirped three-cycle lasers and filters with \( E_{\text{ph}}^{\text{min}} \) as high as 35 eV are used.

More interesting, the far-field IAP (blue solid) is obtained without filters when \( \phi_0 = -0.1\pi \), \( \tau_L = 8 \) fs and \( \Delta \omega = 0.5\omega_0 \) (\( s \approx 0.162T_0^{-1} \)) as shown in figure 4(d). Due to all harmonics are preserved, the intensity and energy of the IAP (\( \tau_{\text{AP}} = 49 \) as) reach \( 1.8 \times 10^{16} \) W cm\(^{-2}\) (1.1 \( \times 10^{14} \) W cm\(^{-2}\)) and mJ, respectively. Finally, in the bottom half of the same figure, we present that the divergence gating is valid for lasers with long pulse duration as \( \tau_L \approx 16 \) fs whose bandwidth \( \Delta \omega = 0.5\omega_0 \) (\( s \approx 0.084T_0^{-1} \)). Through increasing the \( E_{\text{ph}}^{\text{min}} \) to 20 eV based on the prediction in figure 2(d), the far-field IAP (red solid, \( \sim 4 \times 10^{14} \) W cm\(^{-2}\) or \( 2.5 \times 10^{16} \) W cm\(^{-2}\)) is generated with little longer \( \tau_{\text{AP}} \approx 72 \) as. Here, intensities of incident beams are the same as the one used in figure 1 and \( \phi_0 = 0 \). Further simulations (not shown) confirm that the divergence gating also works for incident laser intensities in \( 10^{20} \) W cm\(^{-2}\) which is achievable for 100 TW class facilities. What’s more, according to the theoretical prediction of equation (9) and the estimation based on high-precision 1D simulations, IAPs in the water-window regime (282–533 eV) can be obtained through divergence gating with such laser intensities irradiated on the PM.
Figure 5. The far-field temporal intensity distributions of the reflected on-axis APs (after transformed to 3D geometry) obtained from 2D PIC simulations for cases where TL pulses with bandwidth $\Delta \omega = 0.5 \omega_0$ are used. (a) and (b) are the results when $x_f = b_0 x_R \approx 23.68 \lambda_0$ and $x_f = 0$ are respectively taken. The other parameters are $\alpha = 15^\circ$, $\tau_L = 2.3 \text{ fs}$, $I_0 = 6.53 \times 10^{21} \text{ W cm}^{-2}$ ($I_0 \approx 2.66 \times 10^{21} \text{ W cm}^{-2}$ for the defocused case) and $\phi_0 = 0$. Note that the dashed line represents the value of $I_{\text{peak}} / e^2$.

4. Conclusions

In summary, we have proposed a scheme called divergence gating realized by defocused chirped lasers to obtain IAPs in far field. Theoretical analysis and numerical simulations show that the divergence of the reflected peak AP is much smaller than others when solid targets are placed at a position behind the focal plane of the incident laser with distance $x_f = \left[4 \pi L \cos \alpha / \lambda_0 (1 + \epsilon - 1) \right] x_R_0$. As the much lower decrease of on-axis intensity, far-field IAPs can be obtained after propagation, if filters satisfy $E_{\text{min}} \sim 1.26 \epsilon_0 / s b_0 T_0 s_0^2(0)$. 3D simulation results show that our scheme is robust and bright sub-50 as mJ-level IAPs are obtained in the far field without the requirement of stable CEP by chirped lasers with intensities of $1.92 \times 10^{21} \text{ W cm}^{-2}$. Besides, our scheme can produce intense IAPs even without filters, whose intensities reach $1.8 \times 10^{16} \text{ W cm}^{-2}$ $(1.1 \times 10^{18} \text{ W sr}^{-1})$. Through divergence gating, generated mJ-level IAPs with broad spectrum and intense photon flux benefit the metrology and utilization of IAPs [58] greatly, especially for attosecond-pump-attosecond-probe experiments.

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Data availability statement

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

Appendix. Comparison with cases using transform-limited driving lasers

For the typical chirped incident laser used in this paper ($I_0 \approx 1.92 \times 10^{21} \text{ W cm}^{-2}$, $\tau_L = 8 \text{ fs}$, $\Delta \omega = 0.5 \omega_0$), its intensity can be increased to $6.53 \times 10^{21} \text{ W cm}^{-2}$ with duration decreased to $2.3 \text{ fs}$ after compressed to the TL beam [46]. Utilizing such TL pulses, 2D PIC simulations are carried out with far-field results calculated in 3D geometries. The far-field on-axis intensity distributions of cases where TL pulses interact with targets away from their focal planes ($x_f = b_0 x_R \approx 23.68 \lambda_0$, $I_0 \approx 2.66 \times 10^{21} \text{ W cm}^{-2}$) or at foci are illustrated in figures 5(a) and (b), respectively. Other simulation parameters are the same as those used in the case illustrated by the red line of figure 3(b).

Clearly, because of larger divergences caused by optically-curved PMs, the case using the focused TL pulse (figure 5(b)) generates much weaker far-field APs than those using proper defocused pulses no matter...
they are TL beams (figure 5(a)) or not (figure 3(b)) [36]. Besides, for defocused situations, although the IAP generated by the TL beam (figure 5(a)) is intenser due to higher incident intensity, its isolation property \( I_{\text{iso}} \approx 12 \) is worse than the one obtained through the divergence gating, illustrated by red solids \( I_{\text{iso}} \approx 16 \) in figure 3(b) and it is very challenging to compress such incident lasers to the TL situation (as short as 2.3 fs) in experiments.

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**References**

[1] Sansone G et al 2006 Science 314 443–6
[2] Chini M, Zhao K and Chang Z 2014 Nat. Photon. 8 178–86
[3] Chang Z, Corkum P B and Leone S R 2016 J. Opt. Soc. Am. B 33 1081–97
[4] Hu S X and Collins L A 2006 Phys. Rev. Lett. 96 073004
[5] Lépine F, Ivanov M Y and Vrakking M J J 2014 Nat. Photon. 8 195–204
[6] Cavaliere A L et al 2007 Nature 449 1029–32
[7] Li J, Lu J, Chen A, Han S, Li J, Wu Y, Wang H, Ghimire S and Chang Z 2020 Nat. Commun. 11 2748
[8] Meyer-ter-Vehn J, Honrubia J, Geissler M, Karsch S, Krausz F, Tsakiris G and Witte K 2005 Plasma Phys. Control. Fusion 47 B07–13
[9] Saalmann U, Georgescu I and Rost J M 2008 New J. Phys. 10 025014
[10] Bulanov S V, Naumova N M and Pegoraro F 1994 Phys. Plasmas 1 745–57
[11] Gibson P 1996 Phys. Rev. Lett. 76 50–3
[12] Lichters R, Meyer-ter-Vehn J and Pukhov A 1996 Phys. Plasmas 3 3425–37
[13] Baeva T, Gordionko S and Pukhov A 2006 Phys. Rev. E 74 046404
[14] an der Brügge D and Pukhov A 2010 Phys. Plasmas 17 033110
[15] Edwards M R and Mikhailova J M 2020 Sci. Rep. 10 5154
[16] Sansone G, Palotto L and Núñez M 2011 Nat. Photon. 5 656–64
[17] Winterfeldt C, Spießma C and Gerber G 2008 Rev. Mod. Phys. 80 117–40
[18] Heissler P et al 2012 Phys. Rev. Lett. 108 235003
[19] Baeva T, Gordionko S and Pukhov A 2006 Phys. Rev. E 74 066401
[20] Rykovanov S G, Geissler M, Meyer-Ter-Vehn J and Tsakiris G D 2008 New J. Phys. 10 025025
[21] Yeung M et al 2015 Phys. Rev. Lett. 115 193903
[22] Xu X et al 2020 Optica 7 355–8
[23] Zhang Y X, Rykovanov S, Shi M Y, Zhong C L, He X T, Qiao B and Zepf M 2020 Phys. Rev. Lett. 124 114802
[24] Zhong C, Qiao B, Zhang Y, Zhong L, Li X, Wang J, Zhou C, Zhu S and He X 2021 New J. Phys. 23 063080
[25] Vincenti H and Quéré F 2012 Phys. Rev. Lett. 108 113904
[26] Wheeler J A, Borot A, Monchoce S, Vincenti H, Ricci A, Malvache A, Lopez-Martens R and Quéré F 2012 Nat. Photon. 6 829–33
[27] Kallala H, Quéré F and Vincenti H 2020 Phys. Rev. Lett. 124 043007
[28] Naumova N M, Nees J A, Sokolov I V, Zhong C L, He X T, Zepf M and Qiao B 2017 Phys. Rev. Lett. 92 063902
[29] Rödel C et al 2012 Phys. Rev. Lett. 109 125002
[30] Dollar F et al 2013 Phys. Rev. Lett. 110 175002
[31] Reiter F et al 2010 Phys. Rev. Lett. 105 243902
[32] Hassan M T et al 2016 Nature 530 66–70
[33] Cheng Y, Chini M, Wang X, González-Castrillo A, Palacios A, Argenti L, Martin F and Chang Z 2016 Phys. Rev. A 94 023403
[34] Wabnitz H et al 2002 Nature 420 482–5
[35] Vincenti H, Monchoce S, Kahaly S, Bonnaud G, Martin F and Quéré F 2014 Nat. Commun. 5 3403
[36] Zhang Y, Zhong C L, Zhu S P, He X T, Zepf M and Qiao B 2021 Phys. Rev. Appl. 16 024042
[37] Major Z et al 2009 Rev. Laser Eng. 37 431–6
[38] Kessel A 2018 Generation and Parametric Amplification of Few-Cycle Light Pulses at Relativistic Intensities (Cham: Springer)
[39] Danson C N et al 2019 High Power Laser Sci. Eng. 7 E54
[40] Galván B J, Salamin Y I, Liseykina T V, Harman Z and Keitel C H 2011 Phys. Rev. Lett. 107 185002
[41] Mackenroth F, Gnonoskov A and Marklund M 2018 Phys. Rev. Lett. 117 104801
[42] Dumlu C K 2010 Phys. Rev. D 82 045007
[43] Du H, Luo L, Wang X and Hu B 2012 Opt. Express 20 9713–25
[44] Yang Y-Y et al 2013 Opt. Express 21 2195–205
[45] Kumar S, Gupta D N, Malik H K, Singh D, Lee J and Yoon M 2020 Phys. Plasmas 27 043105
[46] BEA S and MCT 2019 Fundamentals of Photonics (New York: Wiley)
[47] Marcuse D 1982 Light Transmission Optics (Boston, MA: Artech House Publishers)
[48] Jari T 1986 Appl. Opt. 25 2908–11
[49] Taflove A and Hagness S 2005 Computational Electrodynamics: The Finite-Difference Time-Domain Method (Boston, MA: Artech House Publishers)
[50] García S G, Olmedo B G and Martín R G 2000 Microwave. Opt. Technol. Lett. 27 427–32
[51] Arber T D et al 2015 Plasma Phys. Control. Fusion 57 113001
[52] Xu X, Qiao B, Yu T, Yin Y, Zhuo H, Liu K, Xie D, Zou D and Wang W 2019 New J. Phys. 21 103013
[53] Thaury C and Quéré F 2010 J. Phys. B: At. Mol. Opt. Phys. 43 213001
[54] Chopineau L et al 2019 Phys. Rev. X 9 011050
[55] Vincenti H 2019 Phys. Rev. Lett. 123 105001
[56] Wittmann T, Horvath B, Helml W, Schätzel M G, Gu X, Cavalieri A L, Paulus G G and Kienberger R 2009 Nat. Phys. 5 357–62
[57] Ren X et al 2017 J. Opt. 19 124017
[58] Orfanos I, Makos I, Liontos I, Skantzakis E, För B, Charalambidis D and Tzallas P 2019 APL Photon. 4 080901