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Non-Perturbative Probing Atomic Ionization by Attosecond Pulse Trains

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Abstract

We present a theoretical study focused on the photoelectron spectrum of near-infrared (NIR) laser-driven ionization of hydrogen atoms by attosecond pulse trains composed of several HHs of the former. We analyze the effects of increasing the intensity of the NIR probe laser to account for the interference of multiple quantum pathways arising from mainbands formed in ionization by the attosecond pulse train within the strong-field approximation (SFA) beyond the commonly used first-order perturbative (in the NIR laser intensity) reconstruction of attosecond beating by interference of two-photon transitions (RABBIT). The structure of the energy bands formed in the photoelectron spectrum is governed by quantum interferences of the photoelectron wave packet released within one optical cycle of the NIR probe laser field—intracycle interference—and by the number of active high harmonic components, leading to higher-order Fourier contributions as a function of the NIR–XUV relative phase delay. We show that Fourier terms can be interpreted in terms of well-defined semiclassical trajectories. Our results demonstrate a significant departure from the standard two-path quantum-interference RABBIT picture, showing that both the phase-dependent oscillations of mainbands and sidebands and the extracted phase delays depend strongly on the probing laser intensity. The predictions of the SFA reveal that the above-threshold ionization bands exhibit systematic splitting and oscillation patterns as a function of the NIR intensity. SFA predictions are compared with results obtained within *ab initio* solutions of the time-dependent Schrödinger equation (TDSE), showing an excellent agreement, which evidences the minor effect of the Coulomb potential of the remaining ion on the escaping photoelectron for high energy above-threshold ionization. The precise study of the SFA reference phases is essential for the determination of the effect of the Coulomb potential on the escaping photoelectron for what these findings provide new insights into attosecond chronoscopy in the strong-field regime.

Keywords: atomic photoionization; RABBIT; non-perturbative theory; attosecond chronoscopy; phase delays; attosecond pulse train



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1. Introduction

The development of attosecond light sources and phase-controlled femtosecond laser pulses has enabled direct access to ultrafast electron dynamics in atoms and molecules [1–5].

The combination of extreme-ultraviolet (XUV) radiation with near-infrared (NIR) or visible laser fields in a pump-probe configuration constitutes the foundation of attosecond chronoscopy. The two main techniques developed are the attosecond streaking [6,7] and the reconstruction of attosecond beating by interference of two-photon transitions (RABBIT) [8,9].

In attosecond streaking, a single short isolated attosecond pulse ionizes the target and the released electron subsequently undergoes continuum–continuum transitions driven by the NIR probe field. In contrast, the RABBIT technique employs an attosecond pulse train (APT) as the pump, composed of odd harmonics of a concomitant fundamental laser, which probes the XUV one-photon photoionization process. Both techniques allow one to infer temporal information associated with the dynamics of the electron wavepacket during the photoionization process. Moreover, the phase of the photoelectron during the photoionization can be retrieved indirectly by disentangling the phase accumulated by the electron wavepacket released during the continuum–continuum transitions by calculating the photoionization yield as a function of the relative phase between the APT and the probing laser field [10–14].

The standard theoretical description of RABBIT relies on the first-order perturbative treatment of the probe field, assuming absorption and emission of a single NIR photon after the absorption of consecutive odd XUV harmonics [11,15]. In this regime, perturbation theory comprising two-photon transitions (one XUV and one NIR) accurately describes the process and the observed interference pattern, which can be interpreted in terms of a two-path quantum-interference model [11,16–18]. However, as the probe field intensity escalates, higher-order multiphoton transitions in the continuum become relevant, and the assumption underlying the perturbative description progressively breaks down [19,20].

The process of single ionization of atoms by short laser pulses can be assessed by numerically solving the TDSE. Under certain conditions ($\beta_0 = U_p/2\omega c \ll 1$, where $U_p = (F_0/2\omega)^2$ is the ponderomotive energy of a free electron in a monochromatic laser of frequency ω), the dipole approximation can be regarded as valid and the solution of the TDSE is considered “exact” [21]. On the other hand, in addition to the computational costs of solving the TDSE, the underlying mechanisms that take place in the ionization process are hard to extract from the propagated wavepackets. To provide simple interpretations, several models have been developed depending on the parameters of the laser and emission energies. The most used models are those based on the strong-field approximation (SFA). To address these high-intensity probe fields, non-perturbative approaches based on the SFA and semiclassical methods have proven to be particularly valuable [19,22–25]. By expressing the ionization amplitude as a coherent superposition of semiclassical trajectories associated with different ionization times, these approaches provide a simple physical interpretation of laser assisted photoionization, especially RABBIT processes [19,25–27]. In particular, semiclassical models naturally allow interpretations in terms of time-domain interferences, namely, intracycle and intercycle interferences [19,25]. Expressions for RABBIT signals accounting for ionization paths up to a specific order were recently derived [20] and a quantum path-integral model was developed to tailor attosecond time-resolved photoemission spectroscopy [28].

Fourth-order dipole transitions were considered to explain the oscillations in multi-sideband schemes for RABBIT [29,30]. Non-perturbative effects have been studied from a theoretical point of view in the strong field regime, where a multitude of quantum pathways contribute to the ionization process [19,25]. These works have primarily focused on the case of an APT composed of only two odd harmonics, showing that the characteristic *checkerboard* structure in the energy-relative phase for above-threshold ionization (ATI) can be modified by increasing the probe intensity. In particular, the lowest-order two-path interference

picture no longer applies when multiple continuum pathways become relevant. In a recent work that considered only two consecutive odd extreme ultraviolet (XUV) harmonics [25], we showed how electron emission can be interpreted as a result of quantum interferences in the time domain for non-perturbative intensities of the NIR probing laser and found that phase delays depend strongly on the fundamental laser intensity.

In the present work, we study the relevant case of atomic photoionization by an APT composed of several successive odd harmonics assisted by an NIR probe laser treated by means of a strong-field non-perturbative description. We include several odd XUV harmonics to explore their influence on above-threshold ionization as the laser intensity increases. The inclusion of multiple XUV frequencies substantially increases the number of interfering quantum paths, leading to complex patterns in the final high harmonic mainbands (MB) and sidebands (SB) as a function of the relative phase between the APT pump and the NIR probe. We investigate how the interplay between the active number of harmonics, the probe field intensity, and the relative XUV–NIR phase shape the photoelectron spectra (PES) and their phase-dependent oscillations. Therefore, the aim of this work is the study of the modification of the structure of MBs and SBs as a function of the relative phase between the probing laser and the attosecond pulse train when the number for multiple harmonics and the laser intensity is increased. The most notorious change is the splitting of MBs and SBs, which stems from the interference of different electron quantum paths originating from ionization by several high harmonics (HH) for atomic targets.

Using the SFA, we analyze the emergence of multi-path intracycle interference and perform a Fourier decomposition of the PES to extract the lowest phase of non-trivial order, related to the Eisenbud–Wigner–Smith photoionization time delay. We compute higher-order Fourier components allowing for the characterization of the transition from the perturbative two-photon regime to a strongly non-perturbative multiphotonic interference regime. Comparisons with *ab initio* calculations of the TDSE confirm the reliability of the SFA for energetic photoelectrons and provide physical insight into the mechanisms governing non-perturbative RABBIT interferometry. Therefore, we think that by considering quantum paths involving the exchange of more than two (XUV and NIR) photons our results within the SFA could explain experimental measurements of time delays in photoionization processes, unlike the first-order perturbative RABBIT theory.

The paper is organized as follows: In Section 2, we describe the non-perturbative theory based on the SFA used to evaluate the PES along the polarization direction in the RABBIT protocol and a semiclassical model relying on well-defined semiclassical electron trajectories. In Section 3, we present the results of the SFA and compare them to the *ab initio* calculations of the TDSE. Concluding remarks are presented in Section 4. Atomic units are used throughout the paper, unless otherwise stated.

2. Materials and Methods

We study the atomic ionization by a linearly polarized APT consisting of a superposition of odd high-order harmonics (HH) with frequencies $(2m + 1)\omega$, (where m is a positive integer number) accompanied by a near-infrared (NIR) laser of frequency ω . In the length gauge within the dipole approximation, the laser pulse can be completely defined by its electric field

$$\vec{F}(t) = f(t) \left\{ F_0 \cos(\omega t + \phi) + \sum_m F_m \cos[(2m + 1)\omega t + \phi_m] \right\} \hat{z}, \quad (1)$$

where $f(t)$ is the pulse envelope, F_0 corresponds to the field strength of the NIR component, ϕ its phase, and F_m and ϕ_m are the respective field strength and phases of the $(2m + 1)$ -th

high-order harmonic component $\text{HH}(2m + 1)$. In a region of a smooth envelope $f(t)$, the vector potential can be approximated as

$$\vec{A}(t) \simeq -\frac{f(t)}{\omega} F_0 \sin(\omega t + \phi) \hat{z} \quad (2)$$

when $m \gg 1$ or the harmonic intensities are very small compared to the fundamental component. Here, we explore further limits of the RABBIT protocol for an APT of several HHs when the NIR probe intensity cannot be considered perturbative and the atomic potential can be neglected in the continuum.

The photoelectron emission spectrum can be calculated from the transition matrix T as

$$\frac{dP}{dE \sin \theta d\theta} = 2\pi \sqrt{2E} |T|^2, \quad (3)$$

where $E = \vec{k}^2/2$ is the electron emission energy, $\vec{k}$ the corresponding momentum, and θ the polar emission angle relative to the laser field polarization axis.

We solve the TDSE numerically in the length gauge using the generalized pseudospectral method [31]. The wave function is decomposed in spherical harmonics and we use a radial box of 2400 a.u. (including absorbing walls to avoid reflections) and a maximum of 200 angular momenta. The time evolution of the wave function is performed with the split-operator method with a time step of $\Delta t \simeq 0.1$. We perform the computation of the two-dimensional momentum distribution by projecting the post-pulse wave function onto outgoing continuum states. The atom is assumed to be initially prepared in its ground state. Owing to the cylindrical symmetry of a linearly polarized laser field, the magnetic quantum number is conserved throughout the time evolution.

Since we want to model regions where the energy of the emitted electron is far enough from the threshold, the ionization by the NIR and the core–electron interaction can be neglected. Within the SFA and the rotating wave approximation, the transition matrix T can be written as [19,25]

$$T = \sum_m T_m, \quad (4)$$

where each high-harmonic transition term can be calculated as

$$T_m = F_m \int_{-\infty}^{\infty} \ell(t) e^{i[S_m(t) - \phi_m]} dt, \quad (5)$$

with

$$\ell(t) = -\frac{i}{2} f(t) \hat{z} \cdot \vec{d}[\vec{k} + \vec{A}(t)], \quad (6)$$

and the generalized Volkov phase is given by

$$S_m(t) = - \int_t^{\infty} dt' \left\{ \frac{[\vec{k} + \vec{A}(t')]^2}{2} + I_p - (2m + 1)\omega \right\}. \quad (7)$$

In Equation (6), $\vec{d}(\vec{v}) = \langle \vec{v} | \vec{r} | \varphi(r) \rangle$ is the dipole transition matrix element and I_p stands for the atomic ionization potential energy in Equation (7).

If we consider the temporal translational symmetry for the vector potential, i.e., $\vec{A}(t + 2\pi j/\omega) = \vec{A}(t)$, and for the generalized Volkov phase, i.e., $S_m(t + 2\pi j/\omega) = S_m(t) + 2\pi a j/\omega$, we can factorize T_m in Equation (5) as

$$T_m = F_m e^{-i\phi_m} \frac{\sin(a\pi N/\omega)}{\sin(a\pi/\omega)} e^{ia\pi(N-1)/\omega} I_m(\vec{k}), \quad (8)$$

where $a = \frac{k^2}{2} + I_p + U_p - (2m + 1)\omega$ and

$$I_m(\vec{k}) = \int_0^{2\pi/\omega} \ell(t) e^{i[S_m(t) - \phi_m]} dt \quad (9)$$

corresponds to the intracycle amplitude factor, which is responsible for the phase modulation of the mainbands (MB) and sidebands (SB) as a function of ϕ and F_0 . For a full proof of Equation (8), the reader can see ref. [27]. The prefactors of Equation (8) other than $I_m(\vec{k})$ correspond to the so-called intercycle amplitude factor, which gives rise to the formation of MBs and SBs for $a = 2n\pi$ at energies

$$E_m^n = (2m + 1 + n)\omega - I_p - U_p. \quad (10)$$

for even and odd $|n|$, respectively. We want to highlight that in our theory we have ruled out laser-assisted dynamic interference [32–34] since photoionization during the rising and falling flanks of the XUV pulse is negligible in our case.

In strong-field physics, semiclassical electron-trajectory models provide a more intuitive physical picture. To analyze the intracycle interference and the effects on the phase of the electron wavepacket, we briefly follow the semiclassical model (SCM) posed in ref. [25]. We make use the saddle point approximation to evaluate the integral of Equation (9). For linear polarized lasers, the main electron emission yield occurs along the polarization direction. For these reason, we decided to focus on the emission parallel to the polarization axis since it is the direction of major yield. Our study can be generalized to other different directions and angle-integrated observables like the total probability. For simplicity, we consider forward emission, i.e., $\vec{k} = k\hat{z}$, where we get two different solutions to the saddle equation for each electron final momentum given by

$$A(t_s) = \beta_+(k) \equiv v_{0,m} - k, \quad (11)$$

with $v_{0,m}^2/2 = (2m + 1)\omega - I_p$ corresponding to the energy of photoelectrons ionized by the high-frequency pump pulse in absence of the probe field of fundamental frequency. Within the SCM, the intracycle factor reads

$$I_m(k) = 2g_m(k, t_s) e^{i\tilde{S}_m} \cos\left(\frac{\Delta S}{2} - \frac{\pi}{4} \text{sgn}[\beta_+(k)]\right), \quad (12)$$

where t_s are the saddle times (t_1 and t_2), $\tilde{S}_m = (S_m(t_1) + S_m(t_2))/2$, $\Delta S = S_m(t_2) - S_m(t_1)$, and

$$g_m(k, t_s) = \frac{-i\sqrt{2\pi}d_z[k + A(t_s)]}{2v_{0,m}^{1/2}|F_0 \cos(\omega t_s + \phi)|^{1/2}}. \quad (13)$$

The prefactors g_m in Equation (12) are equal for both saddle times t_1 and t_2 . If we consider transitions to a particular MB or SB of order n starting from an HH($2m + 1$), the intracycle factor can be written as

$$I_m^{(n)} = 2g_m(k_{m,n}, t_s) e^{-in\phi} \tilde{I}_m^{(n)}, \quad (14)$$

where $k_{m,n} = \sqrt{2E_m^n}$ is the forward momentum at the ATI peak, t_s is any of the two saddle times t_1 or t_2 , and

$$\tilde{I}_m^{(n)} = e^{inf_m(F_0)} \cos\left(\frac{\Delta S}{2} - \frac{\pi}{4} \text{sgn}[\beta_+(k_n)]\right). \quad (15)$$

The average action $\tilde{S}_m$ can be decomposed as $\tilde{S}_m = -n\phi + nf_m(F_0)$ (see Appendix B of [25]) allowing to analyze the PES in terms of ϕ separately from the intensity effects.

When the APT is composed by a number ν of HHs in the range $\{m, m+1, \dots, m+\nu-1\}$, the interferences in the PES [Equation (3)] can be obtained spanning $|T|^2$ [see Equation (4)] as

$$\begin{aligned} |T|^2 &= \sum_j |T_j|^2 + 2 \sum_{j < k} |T_j| |T_k| \cos(\varphi_j - \varphi_k) \\ &= \sum_j |T_j|^2 + 2 \sum_{\substack{j,k=1 \\ j < k}}^{\nu} |T_j| |T_k| \cos[(n_k - n_j)\phi + (\phi_j - \phi_k) + \lambda_{j,k}^{n_j, n_k}], \end{aligned} \quad (16)$$

where $j, k \in \{m, m+1, \dots, m+\nu-1\}$, ϕ_m is the absolute phase of the HH($2m+1$), and we have used $\lambda_{j,k}^{n_j, n_k} = \arg(\tilde{I}_k^{(n_k)}) - \arg(\tilde{I}_j^{(n_j)})$. It can be noted that Equation (16) explicitly show that interference terms oscillate with frequency $|n_k - n_j|$ as a function of ϕ . Considering that n is the number of energy steps of size ω to reach a particular MB or SB from the corresponding HH peak, we can observe that, for the particular case of a number ν of HHs, the phase oscillations are given by harmonic combinations of $(n_j - n_k) = \pm 2, \pm 4, \dots, \pm 2(\nu-1)$ since the lowest energy difference between two neighboring HHs is 2ω , and between the lowest HH($2m+1$) and the highest HH($2(m+\nu)-1$) the energy difference is $\pm 2(\nu-1)\omega$, as shown in Figure 1.

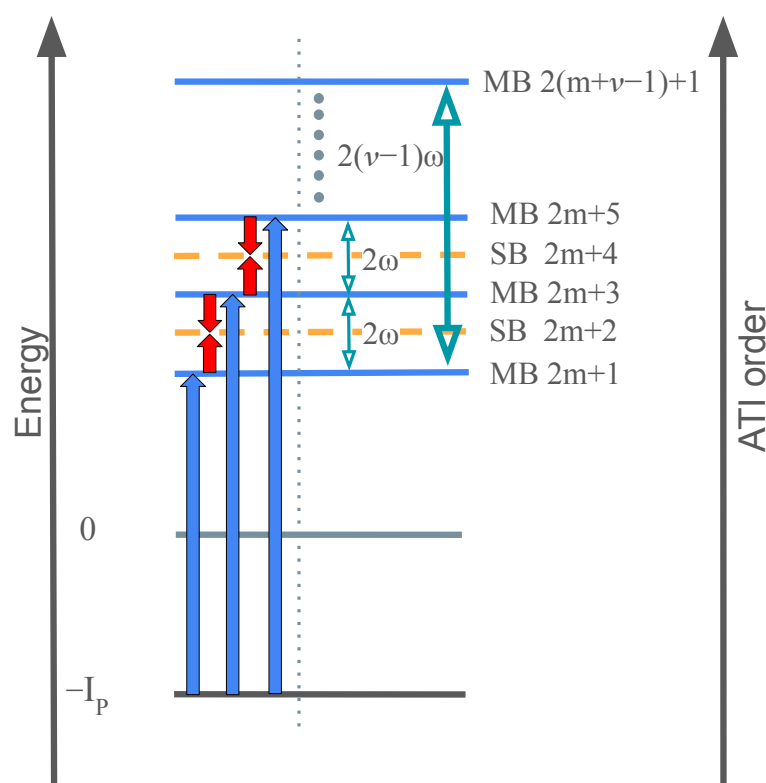


Figure 1. Scheme of transitions involved in the APT involving a number ν of HHs. **Left:** typical RABBIT protocol to reach a particular SB. **Right:** minimum and maximum energy differences between HHs.

From Equation (16), the inherent interference structure of HHs and SBs arises as a sum of several terms of different order. To get more insight into the behavior of the ATI peaks, we perform a Fourier analysis of the HHs and SBs signals as a function of ϕ as was done for

the $\omega - 2\omega$ atomic photoionization in [22,23]. Therefore, the Fourier series decomposition will be performed as a function of F_0 , for fixed MBs and SBs, i.e.,

$$P(F_0, \phi) = \frac{a_0(F_0)}{2} + \sum_{\mu=1}^{\infty} a_{\mu}(F_0) \cos(\mu\phi + \delta_{\mu}(F_0)), \quad (17)$$

where the coefficients $a_{\mu}(F_0)$ measure the amplitude of each term in the Fourier series and $\delta_{\mu}(F_0)$ is the phase delay of each term. On the other hand, there are several $(\nu - 1)$ terms in Equation (16) contributing to the oscillatory pattern with frequency $|n_j - n_k| = 2$ as a function of ϕ . Particularly, the Eisenbud–Wigner–Smith delay can be obtained for $\mu = 2$, i.e., $\delta_2(F_0 \rightarrow 0)$ in the typical finite differences interpretation provided by RABBIT in the standard form for perturbative probe laser intensities, i.e., $A + B \cos(2\phi + \delta)$. At this point, it is important to highlight that $\delta_{\mu}(F_0)$ factors in Equation (17) are in general different from $\lambda_{j,k}^{n_j, n_k}$ factors in Equation (16). For example, if we consider a particular SB, with the increase of probe intensity, besides the noticeable influence of higher orders $\mu = 4, \dots, 2(\nu - 1)$, there are more interference terms in Equation (16) oscillating with frequency 2 as a function of ϕ , arising from non-neighbor HHs. These extra terms “contaminates” the perturbative RABBIT protocol for extracting the Eisenbud–Wigner–Smith delays. In Appendix B, we pose two examples for clarification of the relationship between Equations (16) and (17). In the next section, we analyze these facts using the SFA theory to understand the PES as a function of F_0 and the phase ϕ . Finally, it is very important to recall that our theory considers a multitude of quantum paths (in fact, infinite paths), since it is not a perturbative treatment for the laser pulse as in the case of the typical RABBIT setup [11,16,17,35] where only two paths interfere as shown in the left side of Figure 1.

3. Results and Discussion

As we show strong-field interference effects, the atomic target potential is non-essential, since the only signature on its action on the escaping photoelectron within the SFA is the ionization potential energy (IP) in Equation (7) and the dipole moment in Equation (6) of the corrected manuscript. Our analysis can be easily extended from the hydrogen atom to deal with multi-electron targets on the single-active-electron (SAE) approximation, which is widely employed in strong-field and attosecond physics. Within this framework, the influence of the remaining electrons is incorporated through an effective potential and the corresponding ionization potential. emission energies. We show in Supplementary Materials calculations within TDSE and SFA for emission of electrons from the ground state $3p0$ orbital in Ar. However, we restrict our analysis to hydrogen in the following.

In the previous section, we have shown that the intracycle interference is the main responsible for the oscillations in the PES as we vary the phase ϕ [see Equations (8) and (9)]. We can retrieve the intracycle interference through the photoionization emission by an APT with a duration of only one cycle of the fundamental NIR laser, i.e., $\tau_X = 2\pi/\omega$, (~ 3 fs) where $\omega = 0.05$ a.u. corresponding to $\lambda = 912$ nm (NIR) and for each XUV and NIR pulse we consider a trapezoidal flat-top envelope with a linear ramp on and ramp off consisting of only one cycle. The peak fields of all the harmonics considered are $F_m = 0.02$ or zero. The relative phases for each harmonic were chosen to be $\phi_m = 0$. We show this case in Figure 2 for an APT composed of the three harmonics HH27, HH29, and HH31. For $\tau_X = 2\pi/\omega$, there is no formation of MBs or SBs due to the absence of intercycle interference. The intracycle interference exhibits a 2π periodic oscillation as a function of the relative phase ϕ that follows the classical energy streaking of the photoelectrons with $F_0 = 0.003, 0.005$, and 0.008 in (a,b), (c,d), and (e,f), respectively. In dashed lines, the classical streaking energy of photoelectrons ionized by the HH27, HH29, and HH31 are displayed [see Equation (A1) in

Appendix A and Supplementary Materials], showing a contact point between RABBIT and attosecond streaking techniques [28]. The interference structures become more complex as the probe laser intensity grows. It is worth mentioning the excellent agreement between the SFA results (left column) and the TDSE results (right column), which demonstrates the reliability of the SFA due to the minor effect of the ionic core potential on the escaping photoelectron for the energy ranges in our study.

For the case of a longer APT, MBs and SBs arise as a consequence of the intercycle interference among the electron wavepackets released at different NIR optical cycles [see the intercycle factor of Equation (8)]. In Figure 3, we show calculations for APT with a duration of three optical cycles of the probe NIR laser, i.e., $\tau_X = 6\pi/\omega$ (~ 9 fs) for the same intensity values as in Figure 2. In this case, the intercycle interference allows for emission mainly in the MBs (denoted by vertical dashed lines) and SBs according to Equation (10). Whereas the intracycle interference pattern in Figure 2 displays a 2π periodicity as a function of ϕ , the total (intra- and intercycle) ionization spectrograms in Figure 3 are π -periodic. The horizontal bars show the classical energy ranges for each HH [see Equation (A2)] and give a rough idea of the range of the effect of each HH on neighboring MBs and SBs. In Figure 3a,b, we observe a typical two-lobe interference pattern as a function of ϕ since the probe laser intensity ($F_0 = 0.003$ a.u.) allows the exchange of only one probe photon, which is compatible with the first-order perturbative RABBIT theory. In other words, only two classical energy ranges overlap on SB28 and SB30 allowing for emission and absorption of only one ω photon. While SBs peak at $\phi = 0$ and π , MBs do it at $\phi = \pi/2$ and $3\pi/2$, giving rise to phase delays $\delta_{SB} = 0$ and $\delta_{HH} = \pi$, respectively [see Equation (17) with $\mu = 2$]. As the probe laser intensity grows, the number of visible marginal SBs with energies higher than E_{15}^0 (HH31) and lower than E_{13}^0 (HH27) increases. More importantly, as the probe laser becomes more intense, the total interference pattern in the MBs and SBs in Figure 3 present a higher number of lobes. For $F_0 = 0.005$ a.u. in Figure 3c,d, the HH29 peak is affected by exchange of two probe photons after ionization produced by HH27 and HH31. In fact, all the MBs and SBs between HH27 and HH29 are affected by the exchange of two probe photons from all HHs. Nonetheless, the number of lobes in the marginal SBs in Figure 3c,d is two, since not all the HHs are affecting them. For example, neither does HH31 affect the low-energy marginal SBs, nor does HH27 the high-energy marginal SBs, as their effective ranges do not cover them (horizontal bars). We observe a splitting of the lobes going from two lobes in Figure 3a,b to four lobes in Figure 3c,d. It is clear that the influence of the (classically) allowed energies (shown in the Figure as horizontal bars) for each HH, is directly related to the increasing of the number of lobes. However, this number in MBs and SBs does not increase from the second to third row of Figure 3, despite the increase in the intensity of the probe laser. This stems from the fact that the number of HHs is $\nu = 3$ and the energy gap between the highest HH31 and the lowest HH27 in the APT is $2(\nu - 1)\omega = 4\omega$, activating the $\mu = 2$ and $\mu = 4$ terms in Equation (17).

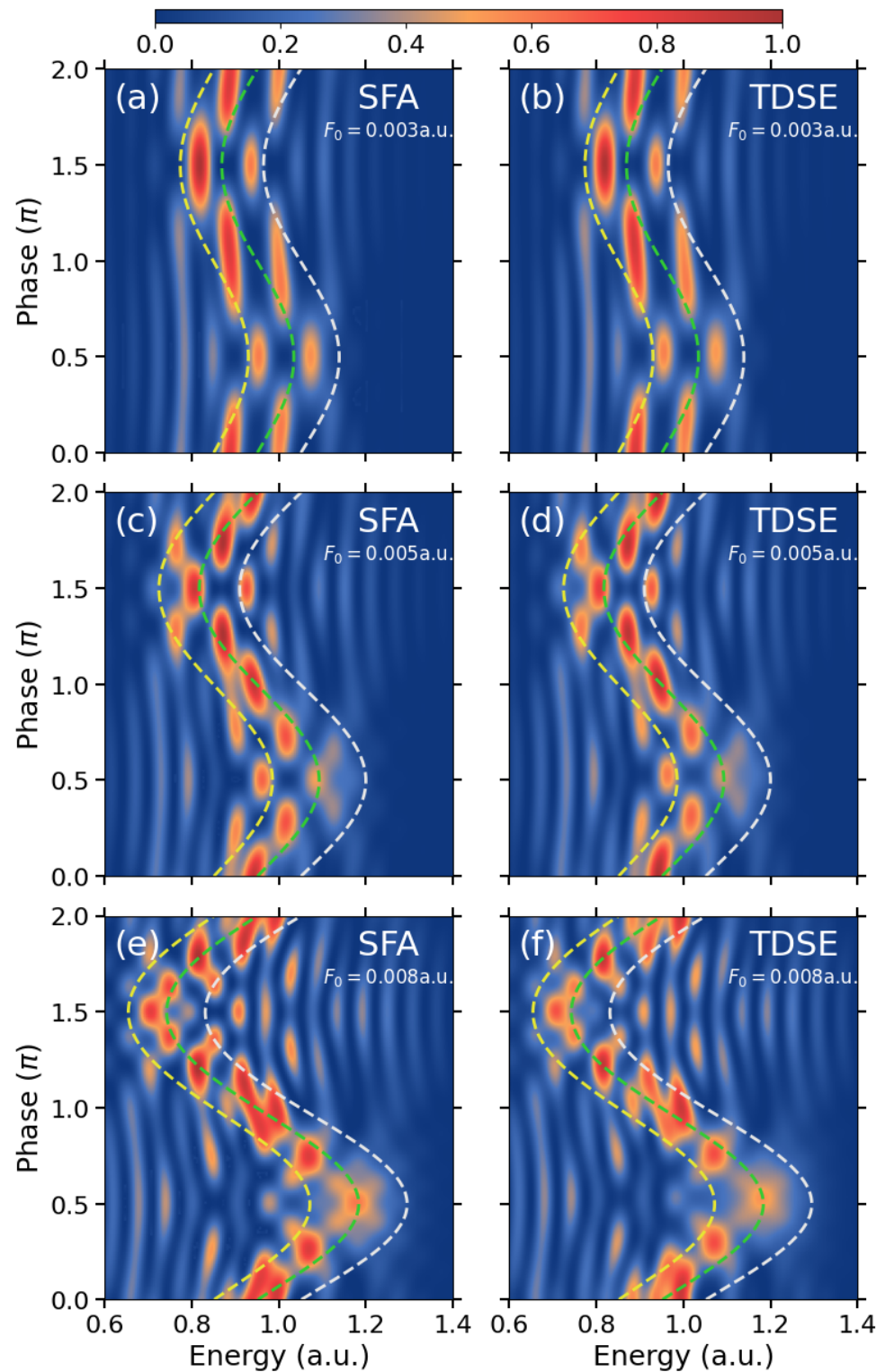


Figure 2. PES the forward direction as a function of the emission energy and the relative phase ϕ between the APT and the probing laser. The duration of the APT comprises one optical cycle of the probing laser ($\tau = 2\pi/\omega$). PES are calculated for three XUV components HH27, HH29 and HH31. The SFA calculations are shown on the left column (a,c,e), meanwhile TDSE calculations are shown in the right column (b,d,f). Calculations are performed for laser intensities $F_0 = 0.003$ a.u. in (a,b), $F_0 = 0.005$ a.u. in (c,d), and $F_0 = 0.008$ a.u. in (e,f). Dashed lines correspond to classical streaking energies of the three HHs [Equation (A1) in the Appendix A].

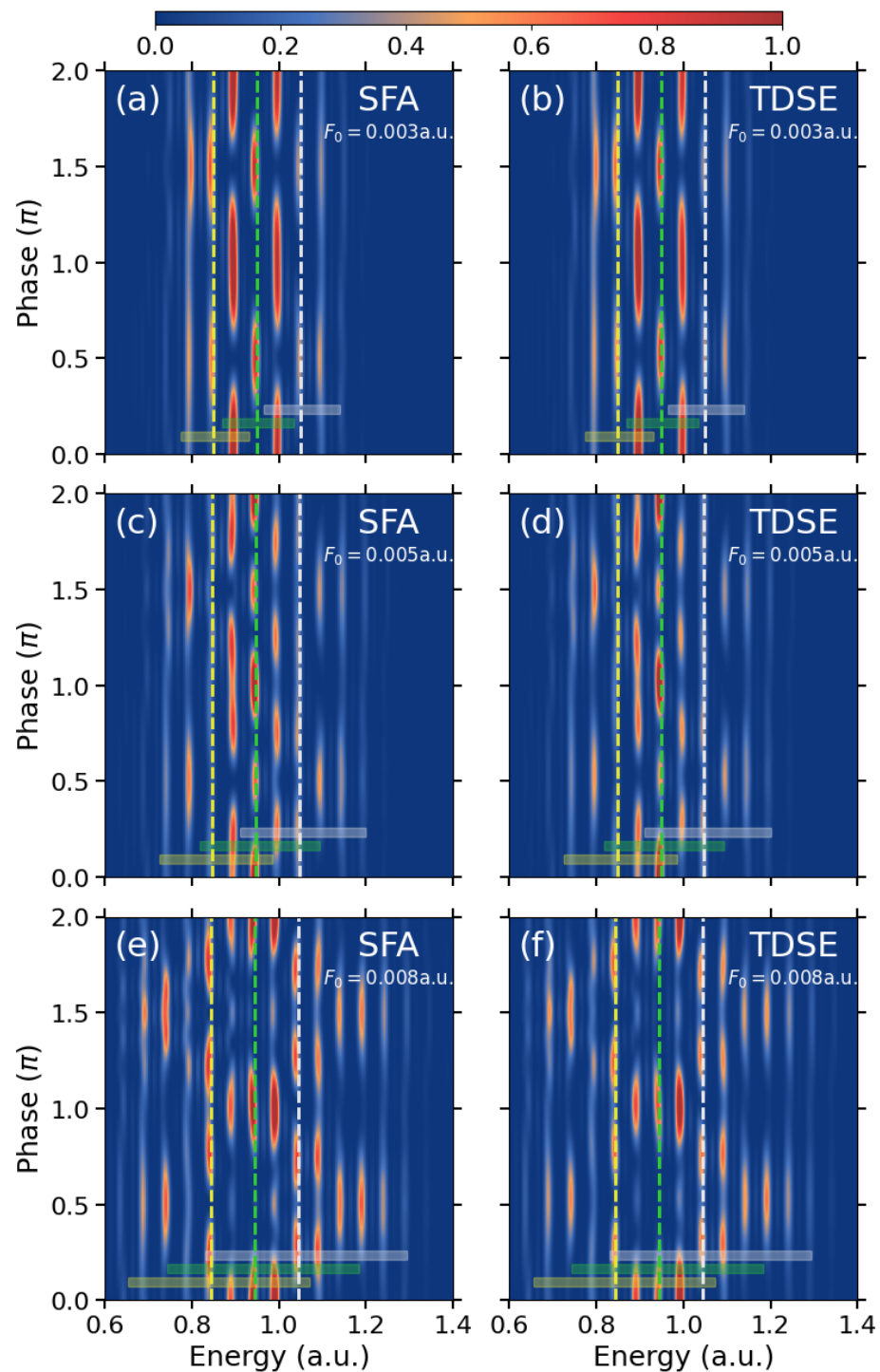


Figure 3. PES for ionization of hydrogen in the forward direction as a function of the emission energy and the relative phase ϕ between the APT and the probing laser. The duration of the APT comprises three optical cycles of the probing laser ($\tau = 6\pi/\omega$). PES are calculated for three XUV components HH27, HH29 and HH31. The SFA calculations are shown on the left column (a,c,e), meanwhile TDSE calculations are shown in the right column (b,d,f). Calculations are performed for laser intensities $F_0 = 0.003$ a.u. in (a,b), $F_0 = 0.005$ a.u. in (c,d), and $F_0 = 0.008$ a.u. in (e,f). The peak positions of harmonics are shown with colored dashed vertical lines: HH27 (yellow), HH29 (green), HH31 (white). Light colored horizontal bars represent the classical energy ranges of each HH according to Equation (A2) in the Appendix A. Each bar is colored as its corresponding HH peak.

In general, laser-assisted photoionization can be divided into two distinct regimes: (i) attosecond streaking when the duration of the XUV pulse is much less than an optical

period of the NIR laser, i.e., for $\tau_x \ll 2\pi/\omega$ and the (ii) sideband regime when the XUV pulse is much longer than an optical period of the NIR laser, i.e., $\tau_x \gg 2\pi/\omega$. For the former, a streaking undulation of the electron yield as a function of the phase delay between the two pulses is observed, whereas for the latter, MBs and SBs are formed due to the multiple exchange of NIR photons, making explicit the transition from the classical electron picture as a particle to the quantum electron picture as a wave [36]. The intracycle interference pattern is formed in just the limiting case between the two aforementioned scenarios, i.e., $\tau_x = 2\pi/\omega$. In this case, there is no formation of sidebands since intercycle interference is not possible but a clear reminiscence of the streaking oscillation following the negative of the vector potential [See Equation (A1)] is inherited. The classical streaking trajectories discussed in Appendix A determine the overall energy modulation and the corresponding classical energy ranges. However, from a semiclassical standpoint, the observed intracycle interference patterns arise from the coherent superposition of two quantum pathways released at t_s satisfying Equation (11) within the same optical cycle. Thus, while classical streaking establishes the general energy framework, the detailed lobe structures originate from quantum intracycle interference effects.

We explore the increase in the number of HH components accompanied by a high-intense NIR probe to observe the contributions to the many oscillations of the ATI bands. In Figure 4, we plot SFA results as a function of the electron emission energy and the relative phase ϕ between the APT and the probe laser, extending the above results for the case of for four ($\nu = 4$), five ($\nu = 5$), and six ($\nu = 6$) HH components in the panels (a), (b), and (c), respectively. In the cases where the probe laser intensity allows the overlap of classical energy ranges for the paths starting from all the HHs involved, we observe the interference modulations as $\cos[2(\nu - 1)\phi + \delta_{\nu-1}]$. For example, MB33 in Figure 4a with six lobes and nodes, SB32 with eight lobes and nodes in Figure 4b, and SB32 in Figure 4c with ten lobes and nodes. It is important to state that the interplay of different orders varies at different ATI peaks, making the visual inspection a very fuzzy tool for assessing its importance.

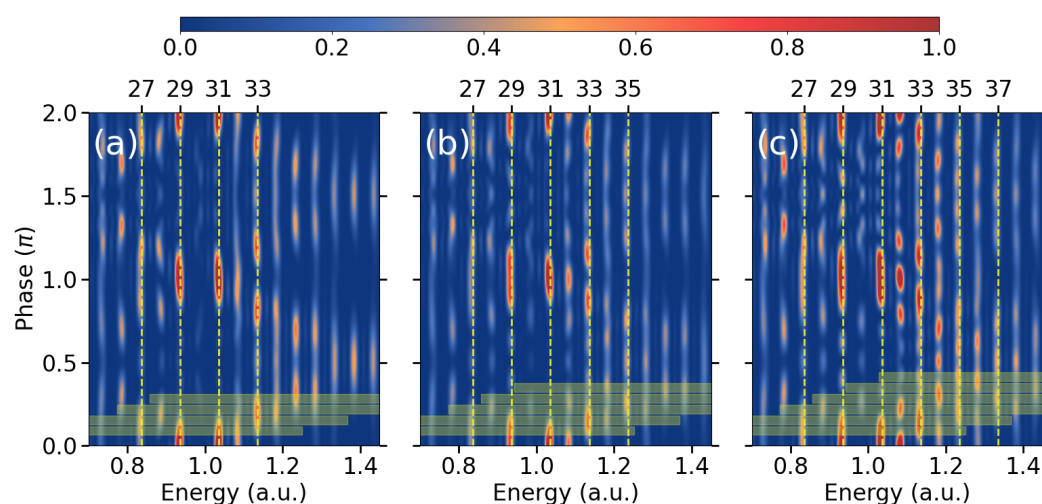


Figure 4. PES for ionization of hydrogen in the forward direction as a function of the emission energy and the relative phase ϕ between the APT and the probing laser. In all the cases, $F_0 = 0.012$ a.u. and the APT has duration of three optical cycles composed by the odd harmonics of order (a) HH27 to HH33, (b) HH27 to HH35, and (c) HH27 to HH37. The XUV field strengths is $F_X = 0.01$ a.u. Light colored horizontal bars represent the classical energy ranges of each HH according to Equation (A2) in the Appendix A. Each bar is colored as its corresponding HH peak.

In order to show the above mentioned behaviors, we explore the effect of the laser intensity and the phase of each ATI band. In Figure 5, we show the effect of the probe laser intensity on the interference patterns of MB29, SB30, and MB31 as a function of the

relative phase ϕ for the case of an APT formed with four HHs—from HH27 to HH33. We note that the structure of MBs and SBs becomes more complex as F_0 increases. For very low intensities, MB29 in Figure 5a and MB31 in Figure 5c have noticeable emission probabilities, with phases peaking at $\phi = \pi/2$ and $3\pi/2$. On the other hand, for SB30, the emission probability rises from zero at $F_0 = 0$, as expected, since SBs can be reached only by absorbing NIR photons, and the probability distributions peak at $\phi = 0$ and $\phi = \pi$. These behaviors at low intensities were described in detail in ref. [25]. As intensity increases (3×10^{-3} a.u. $\lesssim F_0 \lesssim 7 \times 10^{-3}$ a.u.), the lobe structures split to give rise to four lobes. For $F_0 \gtrsim 0.007$ a.u., a new splitting of the lobes gives rise to a total number of six lobes, reaching its maximum $2(\nu - 1)$ for $\nu = 4$ HHs.

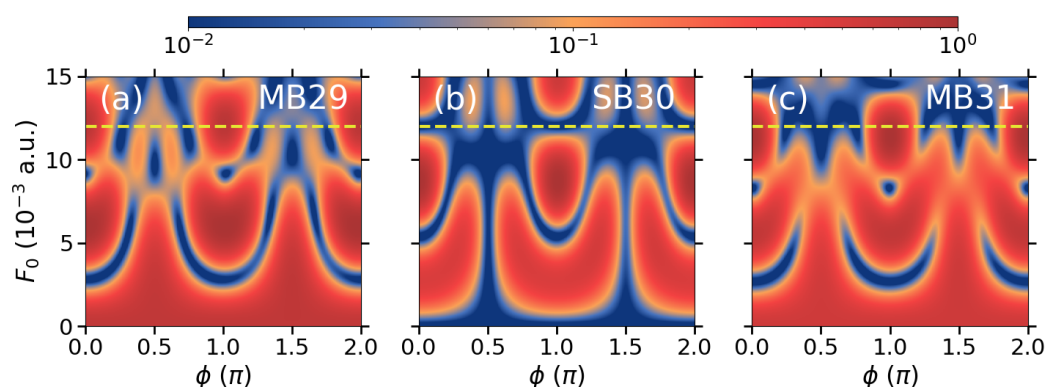


Figure 5. PES of forward emission as a function of the field strength F_0 and the relative phase ϕ in log scale for three different ATI bands: (a) MB29, (b) SB30, and (c) MB31. The APT is composed of four high-order harmonic components from HH27 to HH33. The yellow dashed line correspond to $F_0 = 0.012$ a.u. used in Figure 4a.

In Figure 6, we show the first even orders $\{0, 2, 4, 6, 8\}$ of the Fourier decomposition [Equation (17)] for the same three particular ATI peaks of the calculations shown in Figure 5, namely, MB29, SB30, and MB31 as a function of the increasing probe laser intensity. Odd orders are not shown since they are close to zero within the numerical error, as expected. We observe that Fourier components turn on gradually as the electric field increases for the first even orders. We can see that the last relevant order for this configuration is the 6th one $[2(\nu - 1)]$ for the $\nu = 4$ components considered in the APT].

The amplitude a_4 can be considered as a measure of the departure from the finite differences interpretation for the derivative of the phase respect to the energy, namely, the Wigner time delay. Besides the effects of the fourth order, the second-order contribution stemming from electrons emitted by adjacent HHs becomes present in the same way. For example, at $F_0 \sim 2.5 \times 10^{-3}$ a.u. for SB30 shown in Figure 6b the blue curve corresponding to $\mu = 2$ represents the amplitude of the coherent sum of electrons originated by absorption of single photons of the HH27, HH29, and HH31.

In the lower row of Figure 6, we show the phase delays δ_μ as a function of the field strength. For perturbative intensities, where the term $\mu = 2$ dominates, $\delta_2 = 0$ for SBs and $\delta_2 = \pi$ for MBs, as expected in the typical perturbative RABBIT setup where the Coulomb interaction in the final channel is neglected [25,33]. As the field strength increases, δ_2 presents discrete jumps of π at particular values of F_0 where the coefficients $a_2(F_0)$ become null, as observed when comparing to the coefficients $a_\mu(F_0)$ in the first row of Figure 6. We observe an analogous behavior for the phases of higher orders, i.e., δ_4 and δ_6 . These values of F_0 are denoted as vertical dashed lines in Figure 6 with the corresponding colors to guide the eye.

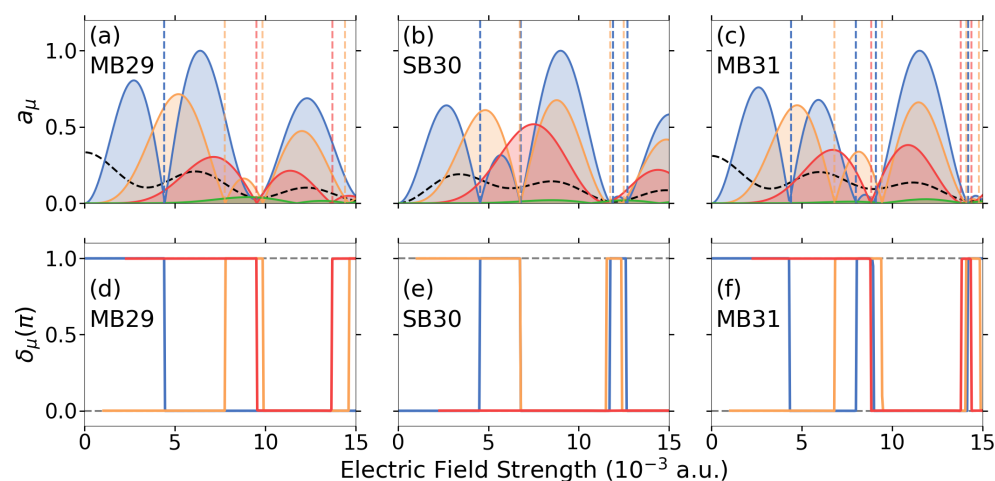


Figure 6. Even coefficients of Fourier decomposition in Equation (17) of the PES as a function of the intensity for (a) MB29, (b) SB30 and (c) MB31, and their respective phase delays in panels (d–f). Results for $\mu = \{0, 2, 4, 6, 8\}$ are displayed with colors black, blue, orange, red, and green, respectively. In the lower row, we only show relevant phase delays corresponding to $\mu = \{2, 4, 6\}$. The APT is composed of HH27, HH29, HH31, and HH33.

4. Conclusions

We have analyzed the PES in the RABBIT setup using the SFA non-perturbatively, including all infinite quantum paths. We have scrutinized the structure of MBs and SBs with increasing intensity of the probe laser beyond the first-order perturbative limit when multiple harmonics are present in the APT.

The presence of many harmonics enables a much larger number of quantum pathways and ATI interference channels, giving rise to effects that cannot be observed in simpler configurations. In particular, we have shown that contributions from non-neighboring ATI channels generate additional frequency components in the photoelectron signal, which are responsible for the splitting and complex structures observed in the MB and SB lobes.

Due to intracycle interference, the presence of many HHs leads to a splitting of the number of oscillating structures (lobes) of MBs and SBs as a function of the relative phase ϕ as the intensity of the probe laser grows. We have demonstrated that the number of lobes as a function of ϕ is related to the number of active HHs affecting the MB or SB under study, that is, the number ν of HHs whose energy ranges cover the studied HH or SB energy. We have also shown that the complexity of ATI structures increases with the number ν of HHs that form the APT.

A central aspect of the present work is the quantitative analysis of these effects through a Fourier decomposition of the transition amplitudes. This approach allows us to identify the physical origin of the different oscillatory components and to establish a direct connection between specific Fourier orders and the structures observed in the spectra. As a result, the manuscript not only reports these phenomena but also provides a detailed physical interpretation of the mechanisms responsible for their appearance.

In this sense, we have verified that the highest relevant order of MB or SB oscillations is $2(\nu - 1)$ at sufficient intensity of the probe laser. We have found that the phase delays of MBs and SBs experience discrete jumps of π for values of the intensity of the probe laser, corresponding to the zeros of their corresponding Fourier amplitudes. These jumps imply phase shifts of $\pi/2$ in the phase ϕ between the fundamental and its harmonics breaking down the typical checkerboard arrangement of PES as a function of ϕ found in the first-order perturbative RABBIT setup. Our results based on the SFA agree with *ab initio* calculations of the TDSE within the resolution threshold results for high emission energy. At lower photoelectron energies, particularly near threshold, Coulomb effects

may become relevant for phase-sensitive observables and could introduce deviations from SFA-based predictions. Non-perturbative studies of angle-resolved emission within the SFA and on conditions with appreciable effects of the Coulomb potential of the parent ion on the photoelectron are highly needed by the community.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/atoms14070047/s1>

This document provides additional theoretical and computational details that support the main findings reported in the article. Specifically, it contains:

- Specific details of the Fourier decomposition of the transition amplitudes, clarifying the physical origin of higher-order oscillatory components and their role in sideband and mainband structures and the connection with the saddle times [25,26].
- An analysis of interference between photoelectrons originated by absorption of non-neighboring high harmonics, showing how they generate additional frequency components and complex lobe patterns in the photoelectron spectra.
- A discussion on the validity of perturbation theory in RABBIT, including its breakdown at non-perturbative probe intensities.
- Computational details comparing SFA and TDSE simulations.
- Additional results for angle-resolved spectra applied to Ar atoms [31,37] to show the validity of results shown for hydrogen targets.

Author Contributions: Conceptualization, D.G.A. and S.D.L.; methodology, M.L.O., S.D.L., and D.G.A.; software, M.L.O., M.B., S.D.L., and D.G.A.; validation, M.L.O., S.D.L., and D.G.A.; formal analysis, M.L.O. and D.G.A.; investigation, M.L.O., S.D.L., and D.G.A.; resources, S.D.L. and D.G.A.; data curation, M.L.O., M.B., and S.D.L.; writing—original draft preparation, D.G.A. and S.D.L.; writing—review and editing, M.L.O., S.D.L., and D.G.A.; visualization, M.L.O.; supervision, S.D.L. and D.G.A.; project administration, S.D.L. and D.G.A.; funding acquisition, S.D.L. and D.G.A. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: Data are contained in this article and can be solicited to the corresponding authors.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

RABBIT	Reconstruction of attosecond beating by interference of two-photon transitions
APT	Attosecond pulse train
SFA	Strong-field approximation
TDSE	Time-dependent Schrödinger equation
PES	Photoelectron spectrum
HH n	High n -th order harmonic
NIR	Near infrared
XUV	Extreme ultraviolet
MB	mainband
SB	sideband
SCM	semiclassical model

Appendix A. Streaking Trajectories in RABBIT and Classical Ranges

Intracycle interference in RABBIT patterns is connected to classical streaking trajectories as can be inferred from Figure 2. Laser-assisted photoionization can be divided into two distinct regimes: (i) attosecond streaking, when the duration of the XUV pulse is much shorter than an optical period of the NIR laser, i.e., $\tau_X \ll 2\pi/\omega$, and (ii) the sideband regime, when the XUV pulse is much longer than an optical period of the NIR laser, i.e., $\tau_X \gg 2\pi/\omega$. For the former, a streaking undulation of the electron yield as a function of the phase delay between the two pulses is observed in Figure 2, whereas for the latter, sidebands are formed due to the multiple exchange of NIR photons as observed in Figure 3 of the paper, explicitly demonstrating the transition from the classical electron picture as a particle to the quantum electron picture as a wave [28,36].

In Figure 2, the intracycle interference exhibits a 2π -periodic oscillatory pattern as a function of the relative phase ϕ between the fundamental laser and the APT connecting the RABBIT and attosecond streaking methods to infer the phase delays in photoionization. In dashed line, we plot the streaking energy of final MB photoelectron energy as a function of ϕ given by [7]

$$\begin{aligned} E_{\text{str},j}(\phi) &= \left(\sqrt{2((2j+1)\omega - I_p)} + \frac{F_0}{\omega} \sin \phi \right)^2 / 2 \\ &\simeq (2j+1)\omega - I_p + 2\frac{F_0}{\omega} \sqrt{2((2j+1)\omega - I_p)} \sin \phi. \end{aligned} \quad (\text{A1})$$

The last line of Equation (A1) was obtained by neglecting the $F_0^2 \sin^2 \phi / \omega^2$ term for low intensity of the probe field.

The intracycle interference pattern is formed in the limiting case between the two aforementioned scenarios, i.e., $\tau_X = 2\pi/\omega$. In this case, there is no formation of sidebands since intercycle interference is not possible, but a clear reminiscence of the streaking oscillation following the negative of the vector potential [see Equations (A1) and (A2)] is inherited. The classical streaking trajectories discussed in Appendix A of the paper determine the overall energy modulation and the corresponding classical energy ranges. However, the observed intracycle interference patterns arise from the coherent superposition of multiple quantum pathways within the same optical cycle. Thus, while classical streaking establishes the general energy framework, the detailed lobe structures originate from quantum interference effects.

As has been mentioned in Section 3, the horizontal bars in Figures 3 and 4 demark the classical energy ranges for each MB (see ref. [25]), which are given by the extremes of $E_{\text{str},m}(\phi)$ in Equation (A1), that is, when $\sin \phi = 1$ for the upper boundary E_m^+ and $\sin \phi = -1$ for the lower boundary E_m^- , i.e.,

$$E_j^\pm = \frac{\left(\sqrt{2((2j+1)\omega - I_p)} \pm \frac{F_0}{\omega} \right)^2}{2}, \quad (\text{A2})$$

and $j = m, \dots, m + \nu - 1$ corresponds to the harmonic $\text{HH}(2j+1)$ for $m = 13$ and $\nu = 3$ in Figure 3.

Appendix B. Semiclassical Trajectories Interpretation

In the RABBIT protocol for the case of only two ($\nu = 2$) HHs present in the APT, from Equation (16), the photoelectron yield restricts to only a DC term and an ϕ -oscillating term

$$|T|^2 = A + B \cos(2\phi + \lambda_{m,m+1}^{n+2,n}), \quad (\text{A3})$$

where A is the DC term, B is the amplitude of first order, and $\lambda_{m,m+1}^{n+2,n}$ is the phase delay, namely

$$A = N^2 \left[F_m^2 |I_m^{(n+2)}|^2 + F_{m+1}^2 |I_{m+1}^{(n)}|^2 \right], \quad (\text{A4a})$$

$$B = 2N^2 F_m F_{m+1} |I_m^{(n+2)}| |I_{m+1}^{(n)}|, \quad (\text{A4b})$$

$$\lambda_{m,m+1}^{n+2,n} = \phi_{m+1} - \phi_m + \arg\left(\tilde{I}_m^{(n+2)}\right) - \arg\left(\tilde{I}_{m+1}^{(n)}\right). \quad (\text{A4c})$$

When the only two HHs considered are HH29 and HH31, i.e., $\nu = 2$, the difference $n_{29} - n_{31}$ is equal to ± 2 no matter which final MB or SB is analyzed. For example, to reach SB30, $n_{29} = 1$ and $n_{31} = -1$ leading to a $\cos(2\phi + \lambda_{29,31}^{1,-1})$ interference term in Equation (16), as expected [25]. In this simple case with only two HHs, the phases in the Fourier series and there is a straightforward identification between δ_2 in Equation (17) and $\lambda_{29,31}^{1,-1}$ in Equation (16).

For the case of three ($\nu = 3$) HHs present in the APT, from Equation (16) the transition probability can be written as

$$|T|^2 = A + B_1 \cos(2\phi + \lambda_{m,m+1}^{n+2,n}) + B_2 \cos(2\phi + \lambda_{m+1,m+2}^{n,n-2}) + C \cos(4\phi + \lambda_{m,m+2}^{n+2,n-2}), \quad (\text{A5})$$

where

$$A = N^2 \left(F_m^2 |I_m^{(n+2)}|^2 + F_{m+1}^2 |I_{m+1}^{(n)}|^2 + F_{m+2}^2 |I_{m+2}^{(n-2)}|^2 \right), \quad (\text{A6a})$$

$$B_1 = 2N^2 F_m F_{m+1} |I_m^{(n+2)}| |I_{m+1}^{(n)}|, \quad (\text{A6b})$$

$$B_2 = 2N^2 F_{m+1} F_{m+2} |I_{m+1}^{(n)}| |I_{m+2}^{(n-2)}|, \quad (\text{A6c})$$

$$C = 2N^2 F_m F_{m+2} |I_m^{(n+2)}| |I_{m+2}^{(n-2)}|, \quad (\text{A6d})$$

$$\lambda_{m,m+1}^{n+2,n} = \phi_{m+1} - \phi_m + \arg\left(\tilde{I}_m^{(n+2)}\right) - \arg\left(\tilde{I}_{m+1}^{(n)}\right), \quad (\text{A6e})$$

$$\lambda_{m+1,m+2}^{n,n-2} = \phi_{m+2} - \phi_{m+1} + \arg\left(\tilde{I}_{m+1}^{(n)}\right) - \arg\left(\tilde{I}_{m+2}^{(n-2)}\right), \quad (\text{A6f})$$

$$\lambda_{m,m+2}^{n+2,n-2} = \phi_{m+2} - \phi_m + \arg\left(\tilde{I}_m^{(n+2)}\right) - \arg\left(\tilde{I}_{m+2}^{(n-2)}\right). \quad (\text{A6g})$$

The second and third terms of Equation (A5) account for the lowest-order contribution ($\mu = 2$), while the last term is associated to the next-order contribution ($\mu = 4$) in Equation (17).

For example, for the HH27, HH29, and HH31, we have three interference terms in Equation (16), one oscillates as $\cos(2\phi + \lambda_{29,31}^{1,-1})$, other does it as $\cos(2\phi + \lambda_{27,29}^{3,1})$, and the last one as $\cos(4\phi + \lambda_{27,31}^{3,-1})$. Therefore, we can identify $\lambda_{27,29}^{3,1}$ in Equation (16) to δ_4 in Equation (17). However, we cannot directly identify $\delta_2 = \lambda_{29,31}^{1,-1}$ to δ_2 because of the “contaminating” term $\cos(2\phi + \lambda_{27,29}^{3,1})$. Only within the perturbative assumption of very low probe intensities, the amplitude of the contaminating term is negligible and the identification of $\delta_2 = \lambda_{29,31}^{1,-1}$ to δ_2 is valid. This relates the electron trajectories associated with the different quantum pathways to the observed interference patterns.

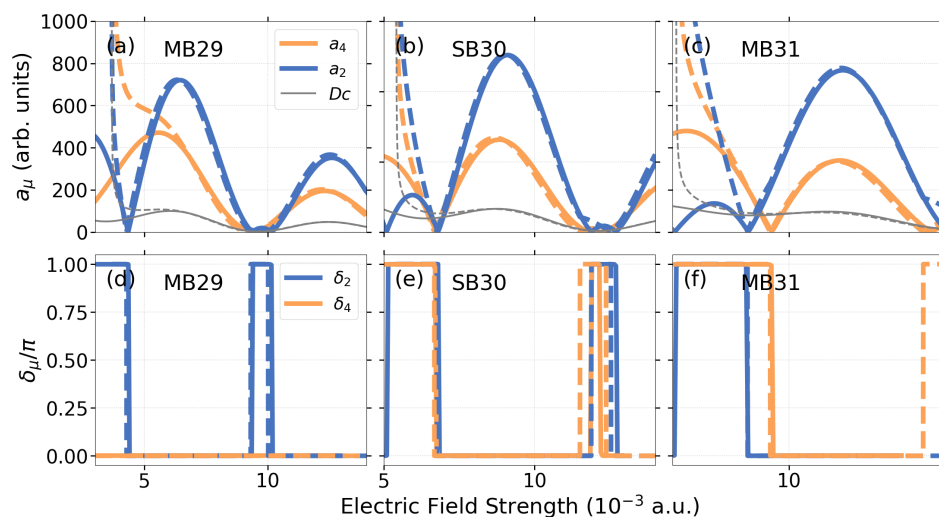


Figure A1. (a–c) are the Fourier amplitudes a_μ for $\mu = \{2, 4\}$ comparing the models SFA (solid line) and SCM (dashed line), step-on the ATI peaks MB29 (first column), SB30 (second column), and MB31 (third column). Corresponding Fourier phases δ_μ for the same values of μ are in panels (d–f). The results are obtained for a three-harmonic configuration consisting of HH27, HH29, and HH31, considering the energy region that is classically allowed for all three harmonics.

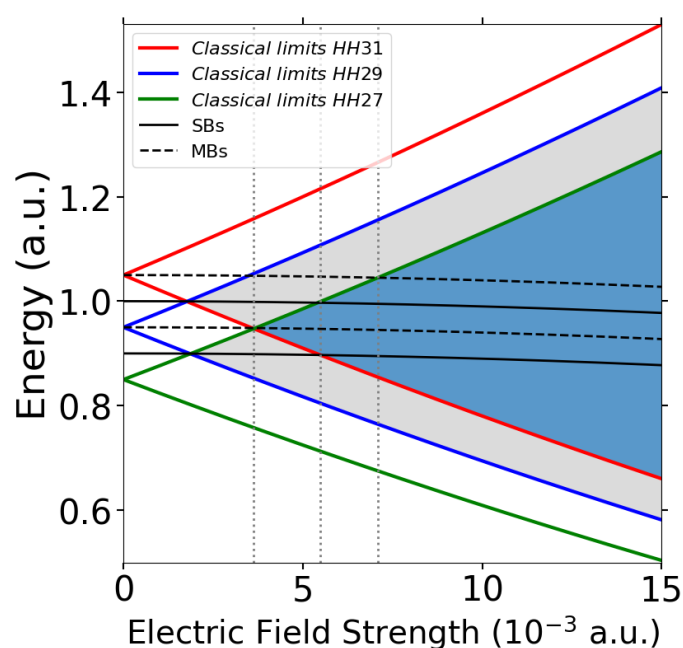


Figure A2. Classical regions for the harmonics HH31, HH29, and HH27 (red, blue, and green triangles, respectively). The white region corresponds to the non-classical domain, the gray region indicates the validity range for two HHs, and the blue region defines the validity range for all three HHs. The horizontal black lines represent the energies of the indicated ATI peaks, while the vertical gray lines denote the minimum values of F_0 above which the theory is valid for each of them.

Here, we establish the connection between the Fourier expansion terms in Equation (17) and the four terms of the SCM in Equation (A5). The first term is the DC term in the expansion. i.e., $a_0(F_0) = 2A$ and the term oscillating at frequency 4ϕ is directly identified with the corresponding Fourier component of the expansion,

$$C = a_4, \quad \lambda_{m,m+2}^{n+2,n-2} = \delta_4. \quad (\text{A7})$$

On the other hand, the identification of the Fourier component oscillating at frequency 2ϕ is not straightforward, since the corresponding contribution arises from the sum of the second and third term in Equation (A5). Hence, the identification is complete if a_2 and δ_2 in Equation (17) are related to the amplitudes in the second and third terms in Equation (A5) as

$$a_2 = \sqrt{B_1^2 + B_2^2 + 2B_1B_2 \cos(\lambda_{m,m+1}^{n+2,n} - \lambda_{m+1,m+2}^{n,n-2})}$$

$$\delta_2 = \tan^{-1} \left(\frac{B_1 \sin \lambda_{m,m+1}^{n+2,n} + B_2 \sin \lambda_{m+1,m+2}^{n,n-2}}{B_1 \cos \lambda_{m,m+1}^{n+2,n} + B_2 \cos \lambda_{m+1,m+2}^{n,n-2}} \right). \quad (\text{A8})$$

Having established the correspondence between the amplitudes and phases of the Fourier expansion and those of the semiclassical model, we now analyze this relation quantitatively. To this end, we consider a three-harmonic configuration with ($\nu = 3, m = 13$), corresponding to the harmonics HH27, HH29, and HH31. As can be seen in Figure A1, the SCM shows very good agreement with the Fourier coefficients obtained from the SFA calculations within the range of applicability of the semiclassical approximation, that is, far from the classical boundaries due to the well-known caustics, which are observed as divergencies in the SCM.

Figure A2 shows the classically allowed regions associated with the three harmonic components involved in the process (HH27, HH29, and HH31), corresponding to the interiors of the colored triangles indicated in the caption of Figure A1. In the region where all three harmonics are simultaneously classically allowed (blue region), all terms of Equation (A5) contribute to the transition probability, namely the DC term, the two terms oscillating at frequency 2ϕ , and the term oscillating at frequency 4ϕ . In contrast, in the regions where only two harmonics are classically allowed (gray regions), only the DC contribution and a single 2ϕ term survive. As a result, Equation (A5) reduces to Equation (A3).

This transition can be visualized by considering SB30, represented by the solid line originating at $E = 1$ a.u. in Figure A2. Moving along this line from right to left, one initially remains inside the blue region, where both first-order (2ϕ) contributions and the 4ϕ contribution are present (shown as a_2 and a_4 in Figure A1). At approximately $F_0 = 0.0055$ a.u., where the line intersects the upper boundary of the green triangle, a divergence appears, as shown in Figure A1b. For smaller values of F_0 , not shown in Figure A1 to avoid overcrowding the figure, the line of SB30 enters in a gray region where only two harmonics contribute, leaving a single 2ϕ term. Decreasing F_0 further, to values of F_0 such that the line reaches the intersection between the red and blue triangles at approximately $F_0 \simeq 0.0015$ a.u. (see Figure A2), we reach the minimum value of F_0 for that sideband, below which no classically allowed contributions exist within the SCM.

Therefore, Figure A2 provides a simple way to identify the parameter regions where the SCM is expected to be valid and to relate the changes in the contributing interference terms to the features observed in Figure A1.

This result demonstrates that the semiclassical trajectory interpretation provides a reliable framework for identifying and analyzing the different contributions involved in the interference process.

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