Interference in $\omega - 2\omega$ atomic ionization within the strong-field approximation: Beyond the perturbative regime

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We analyze interference processes in atomic ionization induced by a two-color laser with fundamental frequency $\omega$ and its second harmonic $2\omega$. The interplay between inter- and intracycle interference processes give rise to multiphoton peaks which can be named as main or ATI peaks and sidebands, in analogy to the well-known RABBIT (reconstruction of attosecond harmonic beating by interference of two-photon transitions). We use the saddle point approximation (SPA) to extract the complex ionization times of the interfering electron trajectories. Changing the relative phase between the two colors, the doubly differential momentum distribution of emitted electrons can be controlled. We study the dependence of the electron emission as a function of the relative phase between the $\omega$ and $2\omega$ fields within the strong field approximation (SFA) but beyond the perturbative regime. We focus on the extraction of the phase delays accounting the electron forward emission in the direction of the polarized electric fields. We characterize the time delays in the emission of electrons for visible frequency of the pump and its first harmonic as a probe [Ti:Sapphire laser (800 nm) together with the first harmonic (400 nm)] for a typical $\omega - 2\omega$ configuration for argon ionization. We find excellent agreement between our SPA results and the corresponding SFA (without any further approximation) and also with previous perturbative theories.

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I. INTRODUCTION

Photoionization is defined as the detachment of one or multiple electrons from a system, such as an atomic or molecular ion, a cluster, or a solid, due to the electromagnetic force exerted by external radiation. Two different pictures have been widely used to depict the photoionization process. On one hand, the exiting electron tunnels through the barrier formed by the parent structure and the external field when the time that the electron travels through the barrier is longer than the time variation of the external field (situation known as the tunneling regime). And on the other hand, when the variation of the external field is very rapid, tunneling is not possible and photoionization proceeds through the absorption of one or several photons allowing the target gain energy up to the continuum near threshold or even higher (situation known as the multiphoton regime) [1–8]. First experiments used rather weak lasers, for which the ionization processes were deep in the multiphoton regime [4]. Theories accompanying these experiments were firstly perturbative [5, 8] but, as irradiance of laser beams grew, these perturbation theories became obsolete and a new paradigm was necessary [7–9]. Based on rather simple pictures of the photoionization processes like the simpleman’s model (SMM) or the strong field approximation (SFA), interference structures in photoelectron spectra have been identified as a diffraction pattern from a time grating composed of inter- and intracycle interferences [10–13]. Whereas the intercycle interferences give rise to multiphoton peaks, intracycle interferences lead to a modulation of the ATI spectrum offering information on the subcycle ionization dynamics [11–13].

Metrology of atomic processes became accessible through pump-probe techniques such as attosecond streaking [16–18] and RABBIT (reconstruction of attosecond harmonic beating by interference of two-photon transitions) [19–21]. These techniques involve lasers of at least two very different frequencies: Electrons are emitted due to the absorption of an XUV field and probed by a moderately intense field in the near-infrared region of the electromagnetic spectrum. In this way, it was possible to measure phase shifts compatible with attosecond delays for noble gas atoms [22–25]. For attosecond streaking, the oscillating probe field moves classically the electron previously freed by the pump field, producing gains and losses of its final kinetic energy [16–20]. In RABBIT, ionization is given by two consecutive high harmonics fields of odd parity followed by absorption or emission of a photon with the fundamental frequency. By analyzing the interference between these two ionization paths, the intrinsic phase shifts in above threshold ionization (ATI) spectra could be extracted by means of second-order perturbative calculations [27] or by analyzing the asymptotic behavior of the scattered electron wave packet from numerically exact solutions of the time-dependent Schrödinger equation (TDSE) [28–31].

Strong-field ionization by laser fields with commensurate frequencies and well-defined relative phase permits the tune and control of the emission process [14, 15, 33–37]. ATI by laser pulses using the fundamental component and one of its harmonics were investigated [35, 37] and applied for controlled ionization [35, 41], dichroism [42, 43], orientation of molecules [44], and control of
interference fringes in the electron momentum distribution \[44, 45\]. The temporal shape of the two-color field is determined by the intensities of the two components and their relative phase. Coherent phase control refers to the manipulations of some physical processes through the relative phase \[37\]. The concept of phase shifts and time delays in RABBIT has been extended by Zipp et al. \[46\] for two-color (\(\omega - 2\omega\)) lasers with controlled relative phase. Very recently, we have theoretically explored the extraction of \(\omega - 2\omega\) phase delays by means of the ab initio solution of the TDSE and also through the development of a perturbation theory \[49\].

In this work, we developed a non-perturbation theory of the electronic photoemission process in atomic argon due to a two-color (\(\omega - 2\omega\)) linearly polarized short laser pulse in the multiphoton regime. In Sec. \[II\] we introduce the general theory based on the saddle point approximation (SPA) to calculate the ionization time of each interfering electron trajectory, firstly analyzing one-color ionization and then the \(\omega - 2\omega\) setup. Different interference structures of the doubly differential momentum distribution are analyzed. We focus on the extraction of the phase shifts using directional emission in the forward direction. We show that our SPA results are in excellent agreement with the results of the SFA (without any further approximation) and also the perturbation theories in the literature \[40, 41\]. We make our final remarks in Sec. \[III\]. Atomic units \((e = \hbar = m_e = 1\) a.u.\) are used throughout unless stated otherwise.

**II. NON-PERTURBATIVE STRONG-FIELD APPROXIMATION**

In general, ionization of an atomic system by a linearly polarized laser pulse can be considered in the single-active-electron approximation. The TDSE then reads

\[
i \frac{\partial}{\partial t} |\psi(t)\rangle = \left[ H_0 + H_{\text{int}}(t) \right] |\psi(t)\rangle,
\]

where \(H_0 = \hat{p}^2/2 + V(r)\) is the time-independent atomic Hamiltonian, whose first term corresponds to the electron kinetic energy and its second term to the electron-core Coulomb interaction. In Eq. (1), \(H_{\text{int}}(t)\) corresponds to the interaction hamiltonian between the atomic system and the external radiation field. Because of the presence of the external laser field, the electron initially bound in an atomic state |\(\phi_i\rangle\) can either remain in the same state, be excited to another atomic bound state, or be emitted to a final continuum state |\(\phi_f\rangle\) with final momentum \(\vec{k}\) and energy \(E = k^2/2\). In the latter case, we call the process photoionization and the transition amplitude within the time-dependent distorted wave theory in the prior form is expressed as \[48, 49\]

\[
T_{\text{ii}} = -i \int_{-\infty}^{+\infty} dt \langle \chi_f(r, t) | H_{\text{int}}(r, t) | \phi_i(r, t) \rangle,
\]

where \(\phi_i(r, t) = \phi_i(\vec{r}) e^{i \vec{k} \cdot \vec{r}}\) is the initial atomic state with ionization potential \(I_p\) and \(\chi_f(r, t)\) is the distorted final state. Eq. (2) is exact as far as the final channel \(\chi_f(r, t)\) is the exact solution of Eq. (1), within the dipole approximation. Throughout this paper, we will be considering linearly polarized laser fields (in the z direction).

Several degrees of approximation have been considered in the literature to solve Eq. (2). The widest known one is the strong field approximation (SFA), which neglects the Coulomb distortion (in the final channel) produced on the ejected-electron state due to its interaction with the residual ion and discard the influence of the laser field in the initial ground state and the depletion of the ground state \[8, 50\]. The SFA consists in approximating the distorted final state with the solution of the TDSE for a free electron in an electromagnetic field, namely, a Volkov function \[51\], i.e., \(\chi_f(r, t) = \chi_f^V(r, t)\), where

\[
\chi_f^V(r, t) = \frac{1}{(2\pi)^{3/2}} \exp \left\{ i \vec{k} + \vec{A}(t) \cdot \vec{r} \right\} \times \exp \left\{ \frac{i}{2} \int_t^{\infty} [\vec{k} + \vec{A}(t')]^2 dt' \right\}
\]

the vector potential due to the total external field is defined as \(\vec{A}(t) = -\int_{-\infty}^{t} dt' \vec{F}(t')\), and \(\vec{F}(t)\) denotes the external laser field. The final Volkov function in Eq. (3) is calculated within the length gauge, i.e., \(H_{\text{int}}(r, t) = \vec{F}(t) \cdot \vec{r}\).

Therefore, \(T\)-matrix in Eq. (2) can be written as

\[
T_{\text{ii}} = \int_{-\infty}^{+\infty} \ell(t) e^{i S(t)} dt,
\]

where

\[
\ell(t) = -i \vec{F}(t) \cdot \vec{d} \left[ \vec{k} + \vec{A}(t) \right]
\]

and

\[
S(t) = -\int_t^{\infty} dt' \left\{ \frac{[\vec{k} + \vec{A}(t')]^2}{2} + I_p \right\}
\]

with the dipole transition moment defined as \(\vec{d}(\vec{r}) = (2\pi)^{-3/2} (e^{i \vec{F}(\vec{r}) \vec{r}} \phi_i(\vec{r})\), and \(S(t)\) is the Volkov action.

We assume that the pump field is composed of \(2N\) optical cycles each of duration \(T = 2\pi/\omega\). Then,

\[
T_{\text{ii}} = \int_0^{NT} \ell(t) e^{i S(t)} dt = \sum_{j=0}^{N-1} \int_{jT}^{(j+1)T} \ell(t) e^{i S(t)} dt.
\]

We consider now a general electric field (and vector potential) with a smooth envelope with a central flat-top region where both \(\vec{F}(t)\) and \(\vec{A}(t)\) are oscillating with period \(T\). From Eq. (5), it is straightforward to realize that
$S(t) - at$ is a time-oscillating function with the same period of the laser field and vector potential,

$$S(t + jT) = S(t) + ajT,$$

where

$$a = \frac{k^2}{2} + I_p + U_p,$$

and $U_p = \int_0^{\alpha+T} dt' A(t')^2$. In light of the periodicity properties of the action in Eq. (7) and that $\ell(t + jT) = \ell(t)$, the transition matrix $T_{\text{if}}$ in Eq. (4) can be written in terms of the contribution of the first fundamental cycle or unit cell [13][52].

$$T_{\text{if}} = \sum_{j=0}^{N-1} \int_0^T \ell(t + jT)e^{iS(t+jT)}dt = \sum_{j=0}^{N-1} e^{ia_jT} \int_0^T \ell(t)e^{iS(t)}dt = \frac{\sin (aTN/2)}{\sin (aT/2)} e^{i(aT(2N-1)/2)} I(\vec{k}).$$

From the absolute value of the transition matrix we can extract probabilistic information, like the doubly differential momentum distribution or the angle resolved photoelectron spectrum. Because of the azimuthal symmetry, the electron distribution can be expressed in terms of only two physical magnitudes, i.e., the final electron momentum parallel $k_z$ and transversal $k_p$ to the field polarization direction or, alternatively, the final kinetic energy $E$ and the final polar emission angle $\theta$:

$$|T_{\text{if}}|^2 = \frac{dP}{2\pi k_p dk_z} = \frac{dP}{2\pi\sqrt{2E}dE d(cos \theta)},$$

The factor $I(\vec{k}) = \int_0^T \ell(t)e^{iS(t)}dt'$ in Eq. (9) corresponds to the contribution into one optical cycle of the $\omega$ field and $|I(\vec{k})|^2$ is known in the literature as the *intracycle* contribution to the ionization probability [19][52][53]. Thus, the photoelectron spectrum (PES) can be expressed as a product of the *intracycle* factor $|I(\vec{k})|^2$ and the *intercycle* factor $(\sin (aTN/2)/\sin (aT/2))^2$, being the latter the result of the phase interference arising from the $N$ different optical cycles of the field [11][13]. We want to point out that Eq. (9) is a mere consequence of the periodicity of the transition matrix with no further approximations, except for a flat-top pulse [52].

Finite maxima are reached at the zeroes of the denominator of the intercycle factor $(\sin (aTN/2)/\sin (aT/2))^2$, i.e., the energy values satisfying $aT/2 = n\pi$, since the numerator also cancels out at these points. Such maxima are recognized as the multiphoton peaks of the PES. They occur when

$$E_n = n\omega - I_p - U_p,$$

where we have used Eq. (8). In fact, when $N \to \infty$, the intercycle factor becomes a series of delta functions, i.e., $\sum_n \delta(E - E_n)$, satisfying the conservation of energy. Instead, for finite pulse durations $\tau = NT$ (composed of $N$ cycles), each multiphoton peak has a width $\Delta E \sim 2\pi/NT$, fulfilling the uncertainty relation $\Delta E \tau \sim 2\pi$.

The intracycle amplitude $I(\vec{k}) = \int_0^T \ell(t)e^{iS(t)}dt'$ in Eq. (9) can be calculated either numerically (SFA) or within the saddle point approximation (SPA). In the latter, the intracycle amplitude can be regarded as a superposition of all electron trajectories within any optical cycle or unit cell with final momentum $\vec{k}$

$$I(\vec{k}) \simeq \sum_{\beta} \ell(t_{\beta}) \frac{e^{iS(t_{\beta})}}{\left|\tilde{S}(t_{\beta})\right|^{1/2}},$$

each starting at a complex ionization times $t_{\beta}$ fulfilling the saddle equation $\tilde{S}(t_{\beta}) = 0$ (where the dot denotes the time derivative), i.e.,

$$\frac{[\vec{k} + \vec{A}(t_{\beta})]^2}{2} + I_p = 0.$$

In general, solutions of Eq. (13) come in pairs $(t_{\beta}, t_{\beta}^*)$, where the star means complex conjugate. From each couple, we select only the solution with positive imaginary part to avoid spurious exponential growth of probabilities and only keep exponential decays when calculating $\exp \{iS(t_{\beta})\} = \exp \{i \Re \{S(t_{\beta})\} \exp \left\{- \Im \{S(t_{\beta})\}\right\}$ in Eq. (12). The SMM considers real ionization times by neglecting $I_p$ and the perpendicular momentum reducing Eq. (13) to $k_z + A(t_{\beta}) = 0$.

### A. One color photoionization

For the case of atomic photoionization by a one color field $F(t) = F \cos(\omega t + \phi) \hat{z}$ with $f(t)$ a smooth function between 0 and 1 mimicking the pulse envelope, $\hat{z}$ the polarization direction, and $F$ the field strength, the action can be calculated from Eq. (5) as

$$S_0(t) = at + b \cos(\omega t) + c \sin(2\omega t),$$

where $a$ is given by Eq. (8), $b = F/\omega^2 \vec{k} \cdot \vec{A}$, $c = -U_p/2\omega$, and the ponderomotive energy is $U_p = (F/(2\omega))^2$. We denote $S_0$ the one-color action just to distinguish it from the two-color action in the next subsection.

From Eq. (13), two ionization times can be analytically calculated with the following expressions

$$t_1 = \frac{1}{\omega} \sin^{-1} \left[ \frac{\omega}{F} \left( k_z + i\sqrt{2I_p + k_p^2} \right) \right],$$

$$t_2 = \frac{1}{\omega} \sin^{-1} \left[ \frac{\omega}{F} \left( k_z + i\sqrt{2I_p + k_p^2} \right) \right],$$

for $k_z \geq 0$ and,

$$t_1 = \frac{1}{\omega} + \frac{1}{\omega} \sin^{-1} \left[ \frac{\omega}{F} \left( k_z + i\sqrt{2I_p + k_p^2} \right) \right],$$

$$t_2 = \frac{3\pi}{2\omega} - t_1^*,$$

(16)
for \( k_z \leq 0 \).

We show the real part of \( t_1 \) and \( t_2 \) in Fig. 1a and the imaginary parts in Fig. 1b for an electric field of strength \( F = 0.0469 \) a.u. and frequency \( \omega = 0.114 \), for the special case that \( k_\perp = 0 \) (forward and backward emission). The SMM times are drawn in dash line and were calculated by including a tiny ionization potential \( I_p = 10^{-6} \). The shaded region of longitudinal momentum \( |k_z| \leq 2 \sqrt{T_p} \) corresponds to the classical accessible region for the electron according to the SMM. Inside this region \( \text{Im}(t_1) = \text{Im}(t_2) = 0 \) while outside, \( \text{Im}(t_1) = \text{Im}(t_2) > 0 \). In turn, for the SPA, we have used an ionization potential \( I_p = 0.58 \) a.u. (corresponding to the ground state of atomic argon).

In Fig. 2a we show the intercycle factor \( \sin (aTN/2) / \sin (aT/2)^2 \) with \( N = 4 \) as a function of the longitudinal momentum \( k_z \) and the perpendicular momentum \( k_\perp \) (whose magnitude is equal to \( k_\perp \)). Isotropic rings are observed at radii \( k_\perp = \sqrt{2E_n} \), where \( E_n \) are given by Eq. (11), corresponding to the absorption of \( n \) photons. The intracycle factor is displayed in Fig. 2d exhibiting a strong angular dependence. The total doubly differential momentum distribution is proportional to the multiplication of the intercycle and intracycle factors and is displayed in Fig 2f. In this paper we omit in the calculations the factor \( l(t) \) containing the atomic dipole moment just to focus on interference aspects of the photoionization processes.

**B. Interference in \((\omega - 2\omega)\) photoionization**

The main goal of this work is to extend the well-known interference structures of electron photoemission in one-color atomic photoionization to the case of two colors, where one main frequency (harmonic) doubles the other (fundamental), i.e., \( \omega - 2\omega \) photoionization emission, within the SFA. If the fundamental intensity is very low compared to the intensity of its second harmonic, some connection between \( \omega - 2\omega \) ionization and RABBIT can be speculated and analyzed showing some similarities and some differences [45-47]. In the RABBIT jargon for even \( n \) in Eq. (11), the energy maxima are named ATI peaks and for odd \( n \) they are named sidebands, but although in our context that denomination...
of the multiphoton peaks is arbitrary, we will maintain it for clarity. In general \( n = n_{2\omega} + n_\omega \) denotes the absorption (emission) of a \( n_{2\omega} \) number of \( 2\nu \) photons and absorption (emission) and a \( n_\omega \) number of \( \omega \) photons for positive (negative) \( n_{2\omega} \) and \( n_\omega \) values. We consider the two-color electric field of the form

\[
\vec{F}(t) = f(t) \left[ F_{2\omega} \cos(2\omega t) + F_\omega \cos(\omega t + \phi) \right] \hat{\ell}, \tag{17}
\]

with \( \phi \) the relative phase of the second harmonic with respect to the fundamental laser field, \( f(t) \) is a smooth function between 0 and 1 mimicking the pulse envelope, \( \hat{\ell} \) is the polarization direction of both fields, and \( F_{2\omega} \) and \( F_\omega \) are the field strengths of the second harmonic and fundamental frequency, respectively. For a long pulse with adiabatic switch on and off the vector potential can be written in its central part \( f(t) \approx 1 \), as

\[
\vec{A}(t) = -f(t) \left[ \frac{F_{2\omega}}{2\omega} \sin(2\omega t) + \frac{F_\omega}{\omega} \sin(\omega t + \phi) \right] \hat{\ell}, \tag{18}
\]
giving rise to a periodicity property of the vector potential and the electric field, i.e., \( \vec{A}(t) = \vec{A}(t + 2j\pi/\omega) \) and \( \vec{F}(t) = \vec{F}(t + 2j\pi/\omega) \), with \( j \) any integer number provided that \( f(t + 2j\pi/\omega) = 1 \). For our calculations we use the same parameters as in Ref. [15] with \( F_{2\omega} = 0.0469 \) a.u. \( (I_{2\omega} = 8 \times 10^{13} \text{ W/cm}^2) \) and \( F_\omega = 0.00332 \) a.u. \( (I_\omega = 4 \times 10^{13} \text{ W/cm}^2) \). In Fig. 3 we show the electric field in black and the vector potential in red as a function of time. We have normalized both fields to the one-color case \( \vec{F}_0 = 0 \) displayed in dotted lines. As the one-color fields show obviously invariant under changes of the relative phase, there are small but appreciable changes in the two-color fields. For example, the one-color fields are \( \pi/\omega \)-periodic, whereas the two-color fields are \( 2\pi/\omega \)-periodic. As shown for the one-color case [11][12], the correct way to choose the unit cell corresponding to one optical cycle of the field is from a root of the vector potential. For the one-color case this corresponds to, for example, \( t = 0 \), whereas this value changes for the two-color case as a function of the relative phase \( \phi \). For example, considering the roots of the vector potential closest to the origin as the left border of the unit cells, they are determined by \( t \in [0, 2\pi/\omega] \) \( (\phi = 0 \text{ in Fig. 3a}) \), \( t \in [-1.243, -1.243 + 2\pi/\omega] \) \( (\text{in a.u. for } \phi = \pi/2 \text{ in Fig. 3b}) \), \( t \in [0, 2\pi/\omega] \) \( (\phi = \pi \text{ in Fig. 3b}) \), and \( t \in [1.243, 1.243 + 2\pi/\omega] \) \( (\text{in a.u. for } \phi = 3\pi/2 \text{ in Fig. 3b}) \).

Under the assumption of adiabatic switch on and off, the action in Eq. (15) can be analytically calculated (in the central region where \( f(t) = 1 \)) as

\[
S(t) = at + b(2\omega t) + c \sin(4\omega t) + d \cos(2\omega t + d) + e \sin(2\omega t + e) + f \sin(2\omega t + f) + g \sin(2\omega t + g), \tag{19}
\]

where \( a \) is given by Eq. (8), and

\[
b = \frac{F_{2\omega}}{4\omega^2} \hat{s} \cdot \hat{k}, \tag{20}
\]

\[
c = -\frac{U_{p,2}}{4\omega},
\]

\[
d = \frac{F_\omega}{\omega^2} \hat{s} \cdot \hat{k},
\]

\[
e = \frac{F_{2\omega} F_\omega}{4\omega^3},
\]

\[
f = -\frac{U_{p,1}}{2\omega},
\]

\[
g = -\frac{F_{2\omega} F_\omega}{12\omega^3},
\]

and \( U_p = U_{p,2} + U_{p,1} = (F_{2\omega}/(4\omega))^2 + (F_\omega/(2\omega))^2 \) defines the ponderomotive energy as the addition of the individual ponderomotive energies of each color. We have dropped diverging terms in Eq. (19) since only the accumulated action, computed as differences of phases, is relevant. The two-color action of Eq. (15) reduces to the one-color action of Eq. (14) when either \( F_{2\omega} = 0 \) (since \( d = e = f = g = 0 \)) or \( F_\omega = 0 \) (since \( b = e = e = 0 \)).
In Eq. (23) the intracycle amplitude stemming from the interference of the two electron trajectories released during half optical cycle of the \( k' \) field (or within one optical cycle of the 2\( \omega \) field) and (ii) the interhalfcycle interference between the contribution of the two half cycles of the \( k' \) field (or between the two optical cycles of the 2\( \omega \) field).

For the case of an \( \omega - 2\omega \) field with weak probe \( \omega \) field compared to the harmonic 2\( \omega \) field, there are four physical solutions of Eq. (13) per optical cycle (in the unit cell), i.e., \( t_1, t_2, t_3, \) and \( t_4 \). We have solved all the saddle times from Eq. (13) numerically by separating their real and imaginary parts obtaining two coupled equations:

\[
\begin{align*}
F_{2\omega} \sin (2\omega \text{Re}(t_\beta) \cosh(2\omega \text{Im}(t_\beta)) + F_{2\omega} \sin (2\omega \text{Re}(t_\beta + \phi) \cosh(2\omega \text{Im}(t_\beta)) &= k_z \\
F_{2\omega} \cos (2\omega \text{Re}(t_\beta) \sinh(2\omega \text{Im}(t_\beta)) + F_{2\omega} \cos (2\omega \text{Re}(t_\beta + \phi) \sinh(2\omega \text{Im}(t_\beta)) &= \sqrt{2}\sqrt{\rho} + k^2,
\end{align*}
\]

with \( \beta = 1, 2, 3, \) and 4. By neglecting either the first or the second terms of Eq. (22) and summing the two equations we recover the saddle times in Eqs. (16) and (15) for the one-color case.

From Eq. (12), the intracycle amplitude stemming from the electron trajectories with released times \( t_\beta \) \((\beta = 1, 2, 3, \) and 4) is proportional to

\[
\frac{4}{\beta=1} e^{iS(t_\beta)} = e^{iS_{1,2}} \cos \left( \frac{\Delta S_{1,2}}{2} \right) + e^{iS_{3,4}} \cos \left( \frac{\Delta S_{3,4}}{2} \right)
\]

where we have omitted the prefactors of each of the terms corresponding to the electron trajectories departing at \( t_1, t_2, t_3, \) and \( t_4 \), to highlight the interference patterns. In Eq. (23) \( \sum_{i,j} S_{i,j} = [S(t_i) + S(t_j)]/2 \) is the average action between \( t_i \) and \( t_j \) and \( \Delta S_{1,2} = S(t_j) - S(t_i) \) is the accumulated action between \( t_i \) and \( t_j \). The accumulated actions \( \Delta S_{1,2} \) and \( \Delta S_{3,4} \) in the last equation contribute to the intrahalfcycle interference of the first and second half cycles. In Fig. 4 we compare the intrahalfcycle factors of the first and second half cycles. They are similar but not equal since the vector potential in the first half cycle differs from the second half cycle. As the probe field is weak compared to the pump field, the intrahalfcycle distributions of Fig. 4 are also similar to the one color case exhibited in Fig. 2b. Taking the zeroth-order perturbation in the probe field \( (\Delta S_0)_{1,2} = (\Delta S_0)_{3,4} = \Delta S_0 \), where \( \Delta S_0 \) denotes the one-color accumulated action in Eq. (14). Therefore, the probability, calculated as the square of the absolute value of the coherent addition of the four different terms in Eq. (23), can be written as

\[
\begin{align*}
\left| \sum_{\beta=1}^4 e^{iS(t_\beta)} \right|^2 &\approx \left| e^{iS_{1,2}} + e^{iS_{3,4}} \right|^2 \cos^2 \left( \frac{\Delta S_0}{2} \right) \\
&\approx 4 \cos^2 \left( \frac{\Delta S}{2} \right) \cos^2 \left( \frac{\Delta S_0}{2} \right),
\end{align*}
\]

where \( \Delta S = S_{3,4} - S_{1,2} \). To get Eq. (24) we have considered the periodicity of \( S_0(t) \). Eq. (24) shows that the intracycle factor \( |\tilde{I}(k)|^2 \) can be approximately split as two factors: (i) the intrahalfcycle interference pattern \( \cos^2 \left( \frac{\Delta S_0}{2} \right) \) stemming from the interference of the two electron trajectories released during half optical cycle of the \( k' \) field (or within one optical cycle of the 2\( \omega \) field) and (ii) the interhalfcycle interference between the contribution of the two half cycles of the \( k' \) field (or between the two optical cycles of the 2\( \omega \) field).

If we go back to one-color ionization, i.e., \( S(t) = S_0(t) \), which fulfills the periodicity property \( S_0(t + jT/2) = S_0(t) + a\pi/\omega \), then, \( \sum_{i,j} S_{i,j} = S_{1,2} + a\pi \), and \( \Delta S = \Delta S_0 = a\pi/\omega = (E + I_p + U_p)\pi/\omega \). Replacing the energy for its value at the multiphoton peaks in Eq. (11), then \( \Delta S_0 = n\pi \). Therefore, the intracycle factor \( \cos(\Delta S_0/2) \) in Eq. (24) becomes equal to \( \pm 1 \) for even \( n \) (constructive interference for the ATI peaks) and 0 for odd \( n \) (destructive interference for the sidebands). It can be observed that the odd intercycle rings in Fig. 2a coincide with the minima of the intracycle factor in Fig. 2b. Thus, the sidebands are not formed in Fig. 2a and Fig. 2f, which simply means that sidebands are only present when an \( \omega \) field is applied.

More generally and beyond the last approximation, complex ionization (saddle) times depend on the longitudinal and transverse momenta, similar to the one color case. For \( \omega - 2\omega \) ionization, Fig. 5 displays the real parts of the saddle times \( t_1 \) (in black), \( t_2 \) (in green), \( t_3 \) (in cyan) and \( t_4 \) (in yellow) as a function of the longitudinal momentum \( k_z \) for \( k_z = 0 \) and for relative phases \( \phi = 0, \pi/2, \pi, \) and \( 3\pi/2 \). The same is displayed for the imaginary parts of the ionization times. For each value of \( k_z \), the different solutions of Eqs. (22) are very similar and cannot be distinguished in the figure. In Figs. 6 we show that the apparent degeneracy with respect of the relative phase is not such (close up of Fig. 4). Whereas for \( k_z \geq 0, \text{Re}(t_1) \) are the same for \( \phi = 0 \) and \( \pi \) and is zero for \( k_z = 0 \), it has smaller values for \( \phi = \pi/2 \) and higher values for \( \phi = 3\pi/2 \) being the difference of about
whereas it has smaller values for \( \phi = \pi \). The degeneracy of \( \text{Re}(t) \) is removed for \( k_z < 0 \) at expenses of a new degeneracy between \( \phi = \pi / 2 \) and \( 3\pi / 2 \). In Fig. 7b, we show that \( \text{Re}(t_2) \) lying in the second quarter cycle are the same for \( \phi = \pi / 2 \) and \( 3\pi / 2 \) for \( k_z \geq 0 \) whereas it has smaller values for \( \phi = \pi \) and higher values for \( \phi = 0 \). The degeneracy of \( \text{Re}(t_2) \) between \( \phi = \pi / 2 \) and \( 3\pi / 2 \) is removed for \( k_z < 0 \) at expenses of a new degeneracy between \( \phi = 0 \) and \( \pi \). \( \text{Re}(t_3) \) lying in the third quarter cycle is shown in Figs. 6c. The degeneracy is the same as for \( \text{Re}(t_1) \) whereas it has smaller values for \( \phi = 3\pi / 2 \) and higher values for \( \phi = \pi / 2 \) for \( k_z \geq 0 \) and \( \pi \) for \( k_z < 0 \). Finally, the same is observed for \( \text{Re}(t_4) \) with respect to \( \text{Re}(t_2) \) in Figs. 6d. We do not analyze the imaginary part of the ionization times in detail since it is not relevant for the interference patterns.

In Figs. 7 we show the SPA doubly differential pho-

![FIG. 5. Complex saddle times \( t_1, t_2, t_3, \) and \( t_4 \) as a function of the longitudinal momentum \( k_z \) for \( k_\perp = 0 \) for \( \omega - 2\omega \) ionization. In solid (dash) lines the results of the SPA real (imaginary) parts for different relative phases \( \phi = 0, \phi = \pi / 2, \phi = \pi, \) and \( \phi = 3\pi / 2 \). The variation of both real and imaginary parts of the saddle times with the relative phase is very small and cannot be discerned in this figure.

![FIG. 6. Close up of the real parts of the saddle times of Fig. 4 for different relative phases (a) \( \phi = 0 \), (b) \( \phi = \pi / 2 \), (c) \( \phi = \pi \), and (d) \( \phi = 3\pi / 2 \).

![FIG. 7. Doubly differential momentum distribution as a function of the longitudinal \( k_z \) and perpendicular momenta \( k_\perp \) for the \( \omega - 2\omega \) ionization within the SPA with relative phase \( \phi = 0 \). (a) Intercycle factor, (b) interhalfcycle factor, (c) multiplication of (a) and (b), (d) intrahalfcycle factor, (e) intracycle factor [multiplication of distributions in (d) and (b)\)], and (f) the total momentum distribution [multiplication of (c) and (d)]. All distributions are normalized.](image-url)
toelectron momentum distribution as a function of the longitudinal momentum $k_z$ and the perpendicular momentum $k_{\perp}$ for zero relative phase ($\phi = 0$) between the two colors. The intercycle interference pattern for $N = 4$ displays in Figs. 7a, a set of multiphoton (ATI and sidebands) peaks. The number of minima between consecutive multiphoton rings is $N - 1 = 3$. This factor is practically the same as the one-color intercycle factor in Fig. 2a with an almost imperceptible difference stemming from the inclusion of the ponderomotive energy of the $\omega$ field, which is $U_{p,1} = 8.5 \times 10^{-4}$ (2% of $U_{p,2}$). The intercycle factor is also independent of the electron emission angle and the relative phase $\phi$. The intracycle factor in Figs. 8a also consists in a set of concentric rings, but the isotropy is lost and the rings appear slightly stretched along the longitudinal momentum. Therefore, the minima of the intracycle rings do not perfectly match with the sidebands of the intercycle factor in Figs. 7a and thus, they survive when one multiply the inter- and intracycle factors as shown in Figs. 8a, unlike the one-color case. In Figs. 7b we show the intrahalfcycle factor calculated as the intrahalfcycle of the one color case, i.e., $\cos^2[(\Delta S_0)/2]$. Therefore, the intrahalfcycle pattern is independent of the relative phase $\phi$. The intracycle pattern (multiplication of the interhalf- and intrahalfcycle patterns) is shown in Figs. 8b. The quasi-isotropic intracycle factor appears modulated by the highly angle-dependent intrahalfcycle pattern (or viceversa). The total emission pattern is the multiplication of the inter- (Figs. 7b) and intracycle (Figs. 7c) (see Fig. 7f). ATI peaks and sidebands of Figs. 7c modulated by the intracycle interference pattern are observed in Figs. 7d.

The doubly differential photoelectron momentum distribution as a function of the longitudinal momentum $k_z$ and the perpendicular momentum $k_{\perp}$ for relative phase $\phi = \pi/2$ is shown in Figs. 8a. The intrahalfcycle factor in Figs. 8a also consists in a set of concentric rings with a discontinuity for $k_z = 0$ since the vector potential is not antisymmetric (with respect to the middle of the unit cell) as in the case of $\phi = 0$ (see Fig. 3b). Such discontinuities are an artifact of the SPA and also appears for laser assisted photoionization emission (two colors with one frequency much higher than the other) \cite{54, 55}. Again, the minima of the intrahalfcycle rings do not match with the sidebands of the intercycle factor in Figs. 7a and thus, they survive when one multiply the inter- and intrahalfcycle factors (see Fig. 8b). The intracycle factor in Fig 8c inherits the discontinuity of the intracycle factor of Figs. 8a. The total emission pattern is the multiplication of the inter- (Figs. 7b) and intracycle (Figs. 8c) patterns (see Fig. 8d).

Despite the case of $\phi = \pi/2$, for $\phi = \pi$ the two half cycles have the same duration and the intrahalfcycle distribution is continuous as displayed in Figs. 8a (as for $\phi = 0$). The multiplication of the intracycle factor of Figs. 7a and the intrahalfcycle factor of Figs. 8a is displayed in Figs. 9a, exhibiting all ATIs and sidebands. In Figs. 9b the intracycle pattern is displayed. The total

FIG. 8. Doubly differential momentum distribution as a function of the longitudinal momentum $k_z$ and the perpendicular momentum $k_{\perp}$ for the $\omega - 2\omega$ ionization within the SPA with relative phase $\phi = \pi/2$. (a) intrahalfcycle factor, (b) multiplication of the intracycle factor in Fig 7a and the intrahalfcycle factor in Figs. 8a, (c) intracycle factor (multiplication of the intrahalfcycle factor in Figs. 7a and the intrahalfcycle of Figs. 8a), and (d) total distribution calculated as the multiplication of the intercycle factor in Figs. 7d and the intracycle factor in Figs. 8a. All distributions are normalized.

FIG. 9. Doubly differential momentum distribution as a function of the longitudinal momentum $k_z$ and the perpendicular momentum $k_{\perp}$ for the $\omega - 2\omega$ ionization within the SPA with relative phase $\phi = \pi$. (a) intrahalfcycle factor, (b) multiplication of the intracycle factor in Fig 7a and the intrahalfcycle factor in Figs. 8a, (c) intracycle factor (multiplication of the intrahalfcycle factor in Figs. 7a and the intrahalfcycle of Figs. 8a), and (d) total distribution calculated as the multiplication of the intercycle factor in Figs. 7d and the intracycle factor in Figs. 8a. All distributions are normalized.
momentum distribution for $\phi = \pi$ is shown in Figs. 9. For $\phi = 3\pi/2$ the first half cycle of the vector potential is shorter than its second half cycle, thus, bigger intracycle rings yield for positive $k_z$, as shown in Fig. 10, instead of for negative $k_z$ in the case for $\phi = \pi/2$. The multiplication of the intercycle factor of Figs. 10a and the interhalfcycle factor of Figs. 10b is displayed in Fig. 10c, which shows ATIs and sidebands. In Fig. 10d the intracycle interference pattern exhibits again a discontinuity like the case of $\phi = \pi/2$ in Figs. 8. The total momentum distribution for $\phi = 3\pi/2$ is shown in Fig. 10d.

In order to test the validity of the SPA, we perform the time integral in Eq. (8) numerically within the SFA [49, 56, 57]. For the sake of simplicity, we model the atomic argon as a hydrogen-like atom with effective charge $Z_{\text{eff}} = \sqrt{2n^2I_p}$, where $n$ is the principal quantum number of the initial state, in this case $n = 3$ and the initial orbital quantum number is $l = 1$ ($p$-state) [58]. This effective charge ensures the ionization potential to be taken into account properly and, consequently, the intercycle fringes in the electron spectra to be situated at the energy values of Eq. (11). We consider an electric field with a ramp on and ramp off of duration $2\pi/\omega$ each and a flat-top region of duration $4\pi/\omega$. In Fig. 12 we show the doubly differential momentum distribution for relative phase $\phi = 0$ in (a), $\phi = \pi/2$ in (b), $\phi = \pi$ in (c), and $\phi = 3\pi/2$ in (d). In order to highlight the interference patterns we have multiplied the momentum distribution by $\exp(10E)$ and plotted in logarithmic scale to neutralize the exponential decay of the SFA as a function of the energy. The intercycle interference pattern appeared as concentric rings situated at $\sqrt{2E_n}$, whereas the intracycle interference pattern does it with the shape of waning and waxing moons (depending on the sign of $k_z$). Whereas distributions for $\phi = 0$ and $\pi$ in Figs 12a and 12c exhibit forward-backward symmetry, the momentum distribution for $\phi = \pi/2$ results in a small asymmetry enhancing forward emission and, contrarily, the momentum distribution for $\phi = 3\pi/2$ results in a small asymmetry enhancing backward emission. The agreement between the SPA distributions in Figs. 8, 9f, 10f, and 11f and the SFA distributions in Figs. 11 is very good. Some differences for the angular distribution near threshold stem from the effect of the dipole matrix element (from a $p$-state to the continuum) in the SFA, which has been disregarded within the SPA. We have checked this performing calculations for a hydrogenic atom from a fictitious $1s$ state with $I_p = 0.58$ (not shown).

C. Phase delays in $\omega - 2\omega$ ionization

In order to get a simple close form, and considering that ionization takes place at times near the extremes of the electric field [Eq. (17)], at zeroth-order perturbation, these ionization times are $t_j = (j - 1)\pi/(2\omega)$ with $j = 1, 2, 3, \text{and } 4$. After a bit of algebra the accumulated action becomes...
This shows that the phase delay of any sideband is expansion in terms of the perturbation parameter where in the last term we have performed a series maximization.

For an ATI peak, n is even then replacing Eq. 25 into Eq. (24) we demonstrate that the intracycle interference probability factor \( \cos^2 \left( \Delta S \right) \) is equal to

\[
\cos^2 \left[ \chi \cos \left( \phi + \pi/4 \right) \right] = 1 - \frac{\chi^2}{2} + \frac{\chi^2}{2} \cos \left[ 2 \left( \phi + 3\pi/4 \right) \right] + O(\chi^4),
\]

where in the last term we have performed a series expansion in terms of the perturbation parameter. This shows that the phase delay of an ATI peak is \( \phi_0 = -3\pi/4 \). In Fig. 12 we display the analytical expression \( \cos^2 \left[ \chi \cos \left( \phi + \pi/4 \right) \right] \) and its first order perturbative approximation \( 1 - \chi^2/2 + \chi^2 \cos \left[ 2 \left( \phi + 3\pi/4 \right) \right] /2 \) for the value \( \chi = 0.5 \) which reproduces the SFA in 45 and the perturbative theory in 46. The variation of the probabilities of the ATI peaks and sidebands as a function of the relative phase \( \phi \) is small if \( \chi \) is small, or equivalently, if the probe field is weak. It is worth to notice from Eqs. 26 and 27 that the probability of ATI peaks and sidebands as a function of the relative phase \( \phi \) have opposite phases (phase difference of \( \pi \)), as expected. The fact that the addition of the intracycle pattern of ATI and sidebands is unity assures the conservation of probability. We obtain similar results to the non-perturbative theory in Ref. 34 for RABBIT. Especially, Eqs. 26 and 27 are similar to Eqs. (14) and (15) in 31. The factor \( \chi \sim -\alpha_2 \left( 1 + F_{2\omega}/(3\omega E_2) \right) \) is \( \sqrt{2} = -\alpha_2 \cdot \vec{k} \left( 1 + \alpha_2 \cdot \vec{k}/(3E_2) \right) / \sqrt{2} \), where \( \alpha_2 = \vec{F}_2/\omega^2 \), \( \alpha_2 = F_{2\omega}/(2\omega)^2 \) represents the quiver vector for the two different \( \omega \) and \( 2\omega \) fields and \( E_2 = k_2^2/2 \). Therefore, it may be thought that the interhalfcycle interference pattern stem from two point sources

\[
\sin^2 \left[ \chi \cos \left( \phi + \pi/4 \right) \right] = \sin^2 \left[ \vec{k} \cdot \left( \vec{R}_+ - \vec{R}_- \right) / 2 \right] \quad \text{(ATI)}
\]

\[
\sin^2 \left[ \chi \cos \left( \phi + \pi/4 \right) \right] = \sin^2 \left[ \vec{k} \cdot \left( \vec{R}_+ - \vec{R}_- \right) / 2 \right] \quad \text{(SB)}
\]

at \( \vec{R}_{+/-} = \pm \alpha_2 \left( 1 + \alpha_2 \cdot \vec{k}/(3E_2) \right) \cos \left( \phi + \pi/4 \right) / \sqrt{2} \), similar to a diatomic molecule aligned along the polarization axis. For the case of the ATIs, these two point sources emit in phase and constructive interference is produced at perpendicular emission; instead, for sidebands, the two point sources emit in counterphase, leading to partial destructive interference in the perpendicular direction since in this case \( \chi = -F_{2\omega}/(3\sqrt{2}\omega^3) \) and not zero as for the emission from a diatomic molecule. In Figs. 13a, 8c, 9c, and 10c we observe a minimum as a function of the angle, whereas ATIs exhibit maxima values.

In Fig. 13 we show the energy spectrum in the forward direction as a function of the relative phase \( \phi \) between the two colors within the SPA. In Fig 13a, we show that the intercycle factor is independent \( \phi \). All multiphoton peaks (ATIs and sidebands) are present in the intercycle factor in Fig. 13a with separation of one \( \omega \) photon energy. The interhalfcycle factor can be observed as a \( 2\pi \)-periodic function in Fig. 12b. The separation between intercycle maxima corresponds to a \( 2\omega \) photon energy and the amplitude of the oscillation increases with energy, since the accumulated action \( \Delta S \) in Eq. (25) increases with \( k_z = \sqrt{2E} \) through the factor \( \chi \). The interplay between the inter- and interhalfcycle interferences is plotted in Fig. 13b, where both ATI peaks and sidebands arise. From Eq. (11), we see that the first multiphoton peak just above threshold \( E_\text{th} = 0.0038 \text{ a.u.} \) corresponds to \( n = 11 \) and, as it is an odd number, it is a sideband with \( \phi_0 = 3\pi/4 \) and \( 7\pi/4 \) (as all sidebands). In turn, ATI peaks maximize at \( \phi_0 = \pi/4 \) and \( 5\pi/4 \). This confirms our prediction of Eqs. 26 and 27 shown in Fig. 12.
of the argon atom.

the cooper minimum stemming from the $3p$ state. We have multiplied the momentum distribution by $\exp(15E)$ to neutralize the exponential decay of the SFA as a function of the energy. One observe a very good agreement between SFA (Fig. 14) and SPA results (Figs. 13c and 13f). In order to highlight the interference patterns we have also calculated the SFA forward emission spectrum as a function of the relative phase $\phi$ for the $\omega - 2\omega$ ionization within the SPA. In order to highlight the interference patterns we show the corresponding asymmetry parameter calculated within the SFA. At first sight, there are significant differences between the SPA and SFA results, however, when one inspect on the asymmetry at the position of the first ATI (in Fig. 29b) and second sideband (in Fig. 29f), similar oscillatory behaviors are found, maximizing the ATIs at $\phi = 0.25\pi$ and $1.25\pi$ and thee sidebands at $\phi = 0.75\pi$ and $1.75\pi$.

For a close comparison between our SPA and SFA results with the perturbative theory developed in Ref. 15 accompanying an experiment for the ionization of atomic argon by a $\omega - 2\omega$ and also our recent perturbative theory developed in Ref. 16, we perform the transformation $t = t' + \phi'/(2\omega) - \pi/(4\omega)$ and $\phi = -\phi'/2 - \pi/4$ in Eq. 17, becoming the electric field in Eq. 30 equivalent.
to the following expression [see Eq. (1) of Ref. [45] only differing in a factor 2 for the definition of the frequencies, and Eq. (1) of Ref. [46],

$$\tilde{F}(t) = f(t)[2\omega \sin(2\omega t + \phi') + F_\omega \sin(\omega t')] \hat{\xi}, \quad (30)$$

where we have supposed that the envelope $f(t)$ remains invariant due to its smoothness as a function of time. In Eq. (30) $\phi'$ is the relative phase of the second harmonic with respect to the fundamental laser field. Figs. 13c, 13f, and 15 show that the ATIs maximize at $\phi' = \pi/4$ and $5\pi/4$, which is equivalent to $\phi'_0 = -2(\phi_0 + \pi/4) = \pi$, and $3\pi$, whereas the sidebands maximize at $\phi'_0 = 3\pi/4$ and $7\pi/4$, which is equivalent to $\phi'_0 = -2(\phi_0 + \pi/4) = 0$, and $2\pi$ (modulo $2\pi$). Therefore, there are an agreement not only between our SFA and SPA calculations but also with our own perturbation theory [40] and the perturbation theory in Ref. [44].

III. CONCLUSIONS

We have developed a non-perturbative strong field theory for the atomic ionization by a linearly polarized $\omega - 2\omega$ laser pulse. We have derived the formation of sidebands as a result of the interplay between inter- and interhalfcycle interference patterns stemming from the effect of a $\omega$ field with respect to a stronger $2\omega$ component. We have individualized both interhalf- and intrahalfcycle interferences conforming the intracycle interference pattern. We show that phase delays calculated within our SPA agree not only with our SFA calculations but also with previous perturbation theories [45, 46] extending their validity to stronger pulses.

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