Quantum description of the high-order harmonic generation in multiphoton and tunneling regimes

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We employ a recently developed *S*-matrix approach [L. Plaja and J. A. Pérez-Hernández, Opt. Express **15**, 3629 (2007)] to investigate the process of harmonic generation in tunnel and multiphoton ionization regimes. In contrast with most of the previous approaches, this model is developed without the stationary phase approximation and including the relevant continuum-continuum transitions. Therefore, it provides a full quantum description of the harmonic generation process in these two ionization regimes, with a good quantitative accuracy with the exact results of the time-dependent Schrödinger equation. We show how this model can be used to investigate the contribution of the electronic population ionized at different times, thus giving a time-resolved description that, up to now, was reserved only to semiclassical models. In addition, we will show some aspects of harmonic generation beyond the semiclassical predictions as, for instance, the emission of radiation while the electron is leaving the parent ion and the generation of harmonics in semiclassically forbidden situations.

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I. INTRODUCTION

The interaction of intense laser fields with atoms constitutes a paradigmatic example of a quantum process beyond the perturbative limit. This particular case, however, results are especially interesting since theory can be contrasted with experiments on a regular basis. This key fact results from the extraordinary development of intense laser during the last two decades. Besides its fundamental theoretical interest, the study of intense-field processes has also lead to the emergence of new technological applications as, for instance, coherent x-ray generation [2] and the production of attosecond pulses of electromagnetic radiation [3]. These particular developments are based specifically on the properties of the harmonic generation in the strong interaction regime. For instance, soft x rays can be generated with some efficiency since the dipole radiation spectrum shows a plateau in which the harmonic intensities are at the same level, until a high-frequency (soft x-ray) cutoff is reached [4,5]. Also, for ultrashort laser pulses, it has been demonstrated that the highest-order harmonics are phase matched, allowing for the reconstruction of trains of high-frequency pulses with durations below the femtosecond scale [6–8].

The basic concepts relevant for understanding the harmonic generation have been established a decade ago in the frame of a three-step model [9]. According to this view, the higher frequency harmonics are generated by the recollision of the electrons previously detached from the atom. The energy acquired by the electron during this excursion in the continuum is released in the form of harmonic radiation, in the event of recombination with the parent ion. The rigorous theoretical support to this view was established in [10], within the strong field approximation (SFA) (neglecting the effect of the Coulomb potential in the dynamics of the electrons in the continuum). This model becomes especially fruitful when tackled with the stationary phase approximation (SPA), since then the theory becomes semiclassical and extraordinarily intuitive, in terms of electronic trajectories in free space. However, the use of the stationary phase approximation imposes some conditions to the harmonic processes: the electron should recombine at the same location where it was previously ionized, and the velocity right after ionization is zero (actually negative kinetic energy, but it is often considered as a zero velocity condition [11]). It is generally accepted that these conditions are only applicable in the intensity regime of tunneling ionization.

Despite the success of the semiclassical approach, the lack of quantitative agreement has limited the applicability of the theory as a rigorous description. It is known that it requires profound *ad hoc* corrections in order to approach the exact results, computed numerically [11,12] or found experimentally [13]. It should be noted, however, that even for those cases, the corrections are not tested quantitatively for a range of intensities, and in some cases require the numerical solution of the Schödinger equation to include Coulomb corrections [13]. Also, the description in terms of semiclassical trajectories neglects some important quantum aspects, as the spreading of the wave packet and the possibility of recombination in neighbor potential wells, as in the case of elongated molecules. Recently an *S* matrix theory has been demonstrated to provide a quantitative description for a wide range of situations (multiphoton, tunnel, and soft over-the-barrier ionization) in the realistic case of Coulomb binding potentials and for electrons exposed to electromagnetic fields of arbitrary pulse shape. This theory does not employ the stationary phase approximation and includes the relevant part of the continuum-continuum transitions, while still being very efficient in computational terms (about two orders of magnitude faster than the exact integration of the time-dependent Schrödinger equation, TDSE). The aim of this paper is to use this tool to perform a detailed analysis of the harmonic generation in tunneling and multiphoton regimes, which may provide some insight on the role of these quantum aspects beyond the SPA.

The paper is organized in two sections. We shall begin with a detailed derivation of the model, and a brief discus-

sion on its quantitative accuracy, complementary to that of [1]. In the second section, we will exploit the possibilities of the SFA formalism to develop a time-resolved description of the harmonic generation process, in a similar fashion as the semiclassical SPA models do, but now including the full quantum aspect of the dynamics.

II. THEORETICAL APPROACH

This section complements our previous work [1] in two aspects. First we will provide a detailed discussion on the derivation and approximations of the model and, second, we will provide some more comparisons with the exact results of the TDSE to demonstrate the accuracy of our description against variations of the carrier phase, field envelope, and wavelength.

Our derivation begins with the standard definition of the evolution of the wave packet in terms of the propagator

$$|\psi(t)\rangle = iG^+(t,t_0)|\psi(t_0)\rangle \tag{1}$$

(t_0 being an initial time before the interaction starts), associated with the time-dependent Hamiltonian $H(t)=H_a+V_i(t)$. Here we will consider $H_a=p^2/2m+V_c(r)$, the atomic Hamiltonian $[V_c(r)=-Zq^2/r]$, and $V_i(t)=-(q/mc)\mathbf{A}(t)\cdot\mathbf{p}+q^2/(2mc^2)\mathbf{A}^2(t)$ the interaction with the electromagnetic field in the velocity gauge and in the dipole approximation. Note that the SFA S-matrix theories are known to be gauge dependent, therefore the accuracy of this description in the length gauge remains as an open question for a future work. We can consider the following *exact* identity, useful for developing recursive approximations to the full evolution of the wave function,

$$G^+(t,t_0) = G_a^+(t,t_0) + \hbar^{-1}\int_{t_0}^{t} dt' G^+(t,t')V_i(t')G_a^+(t',t_0), \tag{2}$$

where G_a^+ is the propagator for the field free case, i.e., associated with H_a.

Assuming a classical description of the electromagnetic field, the harmonic radiation is proportional to the electron's acceleration $a(t)=\langle\psi(t)|\hat{a}(t)|\psi(t)\rangle$ with $\hat{a}(t)\equiv(1/m)[-\nabla V_c+q\mathbf{E}(t)]$. Note that, if the full quantum nature of the field is to be computed, then this mean value has to be replaced by the dipole matrix element [14]. Using Eqs. (1) and (2), together with the definition of the acceleration, we may write

$$\begin{aligned} a(t) = {} & \langle\psi(t_0)|G_a^-(t_0,t)\hat{a}(t)G_a^+(t,t_0)|\psi(t_0)\rangle + \hbar^{-1}\int_{t_0}^{t} dt_1\langle\psi(t_0)|G_a^-(t_0,t)\hat{a}(t)G^+(t,t_1)V_i(t_1)G_a^+(t_1,t_0)|\psi(t_0)\rangle \\ & + \hbar^{-1}\int_{t_0}^{t} dt_1\langle\psi(t_0)|G^-(t_1,t)V_i(t_1)G_a^-(t_0,t_1)\hat{a}(t)G_a^+(t,t_0)|\psi(t_0)\rangle \\ & + \hbar^{-2}\int_{t_0}^{t} dt_2\int_{t_0}^{t} dt_1\langle\psi(t_0)|G_a^-(t_0,t_2)V_i(t_2)G^-(t_2,t)\hat{a}(t)G^+(t,t_1)V_i(t_1)G_a^+(t_1,t_0)|\psi(t_0)\rangle. \end{aligned} \tag{3}$$

Note that the third term in the right-hand side of Eq. (3) is the complex conjugate of the second. On the other hand, we may use the following identity

$$\int_{t_0}^{t}\int_{t_0}^{t} dt'dt''f(t')g(t'') \equiv \int_{t_0}^{t} dt'\int_{t_0}^{t'} dt''f(t')g(t'') + \int_{t_0}^{t} dt''\int_{t_0}^{t''} dt'f(t')g(t'') \tag{4}$$

to rewrite Eq. (3) as

$$\begin{aligned} a(t) = a_{dr}(t) + a_{ar}(t) + \text{c.c.} = {} & \hbar^{-1}\int_{t_0}^{t} dt_1\langle\psi(t_0)|G_a^-(t_0,t)\hat{a}G^+(t,t_1)V_i(t_1)G_a^+(t_1,t_0)|\psi(t_0)\rangle \\ & + \hbar^{-2}\int_{t_0}^{t} dt_2\int_{t_0}^{t_2} dt_1\langle\psi(t_0)|G_a^-(t_0,t_2)V_i(t_2)G^-(t_2,t)\hat{a}G^+(t,t_1)V_i(t_1)G_a^+(t_1,t_0)|\psi(t_0)\rangle + \text{c.c.} \end{aligned} \tag{5}$$

As discussed in [1], the first term (a_{dr}, direct recombination) corresponds to a *direct* process, in which the electron is first ionized by the field and, afterwards, recombined to the ground state. The second term (a_{ar}, assisted recombination), takes into account the continuum-continuum transitions and refers to the possibility of the electron being recombined not to the ground but to an excited state (see Fig. 1 in [1]). For reasons given below, we shall consider this process as a recombination *assisted* by the field. This second path is usually neglected, and has been demonstrated to be small for the

case of zero-range atomic potentials [15]. However, it is also demonstrated to be relevant in the Coulombic case, especially in the multiphoton regime [1].

To proceed, let us first consider the direct term a_{dr}. In the strong field approximation (SFA), we neglect the atomic potential in the dynamics of the ionized states, so we may write

$$iG^{+}(t,t_1)|\phi_{\mathbf{k}}(t_1)\rangle \simeq e^{-(i/\hbar)\int_{t_1}^{t} d\tau \epsilon_{\mathbf{k}}(\tau)}|\phi_{\mathbf{k}}(t_1)\rangle, \tag{6}$$

with $\epsilon_{\mathbf{k}}(\tau)=[\hbar\mathbf{k}-(q/c)\mathbf{A}(\tau)]^2/2m$. Thus, inserting the completeness relation $\int d\mathbf{k}|\phi_{\mathbf{k}}(t)\rangle\langle\phi_{\mathbf{k}}(t)|=1$ in the direct recombination term of Eq. (5) we may decompose

$$a_{dr}(t) = \int d\mathbf{k} a(\mathbf{k},t) \tag{7}$$

with

$$a(\mathbf{k},t) \simeq -\frac{i}{\hbar}\int_{t_0}^{t} dt_1 e^{i\epsilon_0(t-t_1)/\hbar} e^{-(i/\hbar)\int_{t_1}^{t}\epsilon_{\mathbf{k}}(\tau)d\tau} \times \alpha_{0,\mathbf{k}} V_{i,\mathbf{k}}(t_1)\phi(\mathbf{k}) \tag{8}$$

with $|\psi(t_0)\rangle=|\phi_0\rangle$, the initial atomic eigenstate, and $|\mathbf{k}\rangle$ a plane wave, and $V_{i,\mathbf{k}}(t_1)=\langle\mathbf{k}|V_i(t_1)|\mathbf{k}\rangle$, $\phi(\mathbf{k})=\langle\mathbf{k}|\phi_0\rangle$,

$$\langle\phi_{\mathbf{k}}|\phi_0\rangle = \frac{1}{\pi}\left(\frac{2a_0}{Z}\right)^{3/2}\frac{1}{[1+(a_0/Z)^2k^2]^2} \tag{9}$$

and $\alpha_{0,\mathbf{k}}=\langle\phi_0|\hat{a}|\mathbf{k}\rangle$,

$$\langle\phi_0|\left(-\frac{1}{m}\frac{\partial V_c}{\partial z}\right)|\phi_{\mathbf{k}}(t)\rangle = -\sqrt{2}ik_z\frac{Zq^2}{\pi m}\left(\frac{Z}{a_0}\right)^{3/2}\frac{1}{k^2}\left[1-\frac{\arctan(ka_0/Z)}{(ka_0/Z)}\right]. \tag{10}$$

By definition $iG_a^{+}(t_1,t_0)|\psi(t_0)\rangle=\exp[-i\epsilon_0(t_1-t_0)/\hbar]|\phi_0\rangle$. In fact, the computation of $a(\mathbf{k},t)$ involves a single integral which can be carried on very effectively. This can be easily noticed by considering its differential form

$$\frac{d}{dt}a(\mathbf{k},t) = \frac{i}{\hbar}[\epsilon_0-\epsilon_{\mathbf{k}}(t)]a(\mathbf{k},t) - \frac{i}{\hbar}\alpha_{0,\mathbf{k}}V_{i,\mathbf{k}}(t)\phi(\mathbf{k}). \tag{11}$$

Since this is an uncoupled system its computation can be carried on very efficiently, for instance using a Runge-Kutta algorithm. Note that the uncoupling is inherent to the SFA-S-matrix approach, but it has not been generally exploited. Instead, most authors seem to consider unavoidable the use of the stationary phase approximation to make the computation of Eqs. (7) and (8) tractable.

Now we turn our attention to the acceleration due to the assisted recombination, a_{ar}. After employing the SFA and using the completeness relation twice (before and after the acceleration operator), we find

$$a_{ar}(t) = \int d\mathbf{k}\int d\mathbf{k}'\int_{t_0}^{t}\frac{dt_2}{\hbar}e^{i(\epsilon_0/\hbar)t_2}e^{(i/\hbar)\int_{t_2}^{t}d\tau\epsilon_{\mathbf{k}'}(\tau)}\langle\phi_0|V_i(t_2)|\mathbf{k}'\rangle \times\langle\mathbf{k}'|\int_{t_0}^{t_2}\frac{dt_1}{\hbar}e^{-i(\epsilon_0/\hbar)t_1}e^{-(i/\hbar)\int_{t_1}^{t}d\tau\epsilon_{\mathbf{k}}(\tau)}\hat{a}|\mathbf{k}\rangle V_{i,\mathbf{k}}(t_1)\phi(\mathbf{k}). \tag{12}$$

As noted above, this term describes the recombination to a state excited by the field. Since the operator $\hat{a}$ is relevant at short distances from the atom, the contribution to the integral is non-negligible only for times $t_2\simeq t$. Otherwise, the final excited state would have been driven by the field far from the potential origin, and the overlap with $\hat{a}$ would be almost zero. We shall, therefore, replace the lowest bound (t_0) of the t_2 integral by $t-\Delta t$, Δt being a small quantity that represents the time lapse during which the overlap is not negligible. In this situation, the only relevant term in this integral is the phase $\frac{i}{\hbar}\int_{t_2}^{t}d\tau\epsilon_{\mathbf{k}'}(\tau)\simeq\frac{i}{\hbar}\epsilon_{\mathbf{k}'}(t)(t-t_2)$, while t_2 can be replaced by t in the arguments of the rest of the functions (including the upper limit of the integral over t_1). According to our interpretation of the time interval Δt, we consider the integral to vanish in its lower limit. In this form, the integral over t_2 can be computed easily as

$$a_{ar}(t) \simeq \frac{i}{\hbar}\int d\mathbf{k}\int d\mathbf{k}' e^{i(\epsilon_0/\hbar)t}\frac{1}{\epsilon_{\mathbf{k}'}(t)-\epsilon_0}\langle\phi_0|\left[\epsilon_{\mathbf{k}'}(t)-\frac{\hbar^2k'^2}{2m}\right] \times|\mathbf{k}'\rangle\langle\mathbf{k}'|\int_{t_0}^{t}dt_1 e^{-i(\epsilon_0/\hbar)t_1}e^{-(i/\hbar)\int_{t_1}^{t}d\tau\epsilon_{\mathbf{k}}(\tau)}\hat{a}|\mathbf{k}\rangle \times V_{i,\mathbf{k}}(t_1)\phi(\mathbf{k}), \tag{13}$$

where we have expressed $V_i(t)$ in terms of the Volkov energies $\epsilon_{\mathbf{k}'}(t)$. The computation of the integral in momentum space requires some further approximations. On one hand, the wave packet expressed in the $\mathbf{k}'$ space corresponds to a ionized wave function close to the nucleus, therefore it is reasonable to approximate its energy by the time-averaged $\epsilon_{\mathbf{k}'}(t)\simeq\epsilon_0+U_p$, U_p being the ponderomotive energy at the field maximum. Note that this is a rough approximation, and it can only be fully justified by the results of our theory. On the other hand, the matrix element can be processed as

$$\langle\phi_0|\frac{\hbar^2k'^2}{2m}|\mathbf{k}'\rangle \equiv \left\langle -\frac{\hbar^2\nabla^2}{2m}\phi_0|\mathbf{k}'\right\rangle \tag{14}$$

and then substituted in Eq. (13) together with the time-averaged energies. Now the integral in $\mathbf{k}'$ is unity, and we are left with the following matrix element

$$\left\langle -\frac{\hbar^2\nabla^2}{2m}\phi_0\middle|\hat{a}|\mathbf{k}\right\rangle = \langle(H_a-V_c)\phi_0|\hat{a}|\mathbf{k}\rangle = \langle\phi_0|\hat{a}|(\epsilon_0-V_c)\mathbf{k}\rangle = \langle\phi_0|\hat{a}|(\epsilon_0-H+H_f)|\mathbf{k}\rangle \simeq \langle\phi_0|\hat{a}\frac{\hbar^2k^2}{2m}|\mathbf{k}\rangle, \tag{15}$$

H_f being the atom-free Hamiltonian, that has also been approximated by the average $\frac{\hbar^2k^2}{2m}+U_p$, and H the total energy,

approximately ϵ_0+U_p. After these manipulations, Eq. (13) can be written as

$$a_{ar}(t) \simeq -\int d\mathbf{k}\left(1+\frac{\epsilon_0-\hbar^2k^2/2m}{U_p}\right)a(\mathbf{k},t)+\text{c.c.} \quad (16)$$

Thus the total acceleration defined in Eq. (5) can be, therefore, written as

$$a(t) \simeq -C_F\int d\mathbf{k}\frac{\epsilon_0-\hbar^2k^2/2m}{U_p}a(\mathbf{k},t)+\text{c.c.}, \quad (17)$$

where we have introduced a Coulomb factor in the amplitude of the Volkov waves $C_F=(2k_B|\epsilon_0|/E_0)^{Z/k_B}$, with $k_B=\sqrt{2m|\epsilon_0|}$ and E_0 the maximum field amplitude. This correction is derived as an approximation of the prefactor found in [16] and demonstrated to give accurate quantitative results for the computation of ionization rates in the tunneling regime [17]. Figures 1(a)–1(c) shows the comparison of the results of the formulation above with the exact integration of the Schrödinger equation for the hydrogen atom interacting with a linearly polarized electromagnetic field of the form $E(t)=E_0\epsilon(t)\sin(2\pi ct/\lambda+\phi_0)$. The spectra have been calculated according to the definition of the Fourier series $a(t)=\Sigma_j a(\omega_j)\exp(-i\omega_j t)$, where $\omega_j=j\Delta\omega$, $\Delta\omega=2\pi/t_{int}$, with t_{int} the total time lapse of the computation. The subplots 1(a) and 1(b) correspond to a field envelope $\epsilon(t)=\sin^2(\pi ct/\lambda/8)$ with $\lambda=800$ nm, $\phi_0=0$, and computation time t_{int} of eight cycles. These two cases complement those presented in [1], that where computed with another carrier phase, $\phi_0=\pi/2$. Therefore, together with the previous results of [1], these figures demonstrate the ability of our formulation to reproduce quantitatively the changes in the high-frequency part of the spectra caused by the variation of the carrier phase. Also, Fig. 1(b) demonstrates the accuracy of our S-matrix calculation in situations where the ionization is non-negligible (see inset of the figure). A further check of the accuracy of our approach can be found in Fig. 1(c), where the comparison has been carried out for a completely different situation: envelope $\epsilon(t)=\cosh^{-2}[(ct-8\lambda)/2\lambda]$, wavelength $\lambda=1064$ nm, and interaction time of 16 cycles. Note that Fig. 1(a) corresponds to a multiphoton situation (Keldysh parameter 1.5), while Figs. 1(b) and 1(c) correspond to a tunnel (Keldysh parameters 0.57 and 0.47, respectively).

As commented above, traditionally the efficient evaluation of the S-matrix acceleration given by Eqs. (7) and (8) has been tackled with the use of the stationary phase approximation, instead of Eq. (11). The semiclassical picture emerging from this approximation is closely connected with the so-called three-step model. Although this is a quite successful model that describes several aspects of the harmonic spectra (cutoff frequency, harmonic chirping, etc.), it is known to be limited to the tunneling ionization regime and neglects some quantum effects as the wave function spreading. Furthermore, it needs additional corrections to describe the harmonic generation with elliptically polarized laser beams, or to describe the same process for a potential with various scattering centers (as elongated molecules) which give rise to interfering paths from harmonic emission.

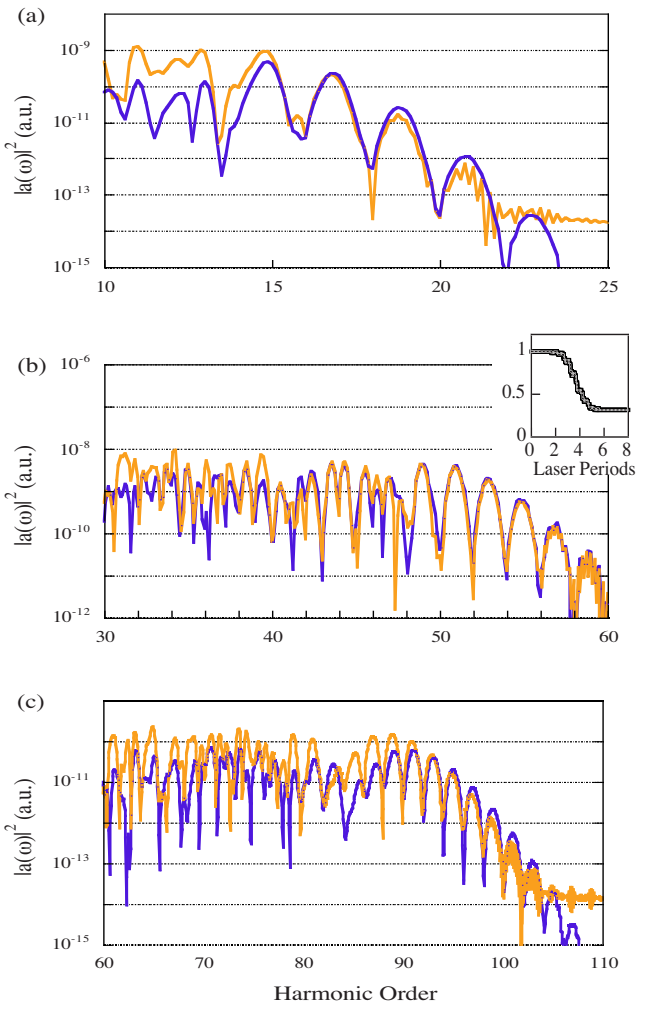

FIG. 1. (Color online) Compared harmonic spectra of hydrogen atom for (a), (b) $\sin^2$ and (c) $\cosh^{-2}$ envelopes [orange (grey) lines: exact results from the TDSE; blue (black) lines: results of our formulation, no rescaling of the data has been done]. (a) and (b) correspond to an eight-cycle pulse of $\lambda=800$ nm with intensities $\simeq 5\times10^{13}$ and $\simeq 3.5\times10^{14}$ W/cm^2, respectively. (c) corresponds to $\lambda=1064$ nm, intensity $\simeq 3\times10^{14}$, and interaction time of 16 cycles. The inset in (b) shows the evolution of the population of the ground state during the interaction.

As a main advantage, our method preserves the quantum character of the interaction dynamics, while still offering a substantial increase in computing speed. Therefore, it offers the possibility of analyzing the process of harmonic generation from a fully quantum perspective and in situations not restricted to the tunnel ionization regime. On the other hand, as we will see below, the nature of the S-matrix approach also allows for a time-resolved analysis of the process which is unavailable from the numerical integration of the TDSE. In our case, this analysis is the quantum counterpart of the interpretation based on classical trajectories that come up if the stationary phase approximation is employed. In the following, therefore, we will use our derivation to perform an analysis of the harmonic generation process beyond the semiclassical description in tunnel and multiphoton regimes.

III. SEMICLASSICAL AND QUANTUM ASPECTS OF HARMONIC GENERATION

The fundamental understanding of the high-order harmonic generation process can be drawn directly from the S-matrix expression of the a_{dr} term in Eq. (5): the electron remains in the ground state of the atom until it is perturbed by the field, at some time instant t_1. Afterwards, it evolves under the influence of the field and the Coulomb potential, until it recombines with the ground state emitting radiation through the $\hat{a}$ operator. The strong field approximation (SFA) simplifies this picture by assuming that the electron, once detached from the atom, evolves in the continuum with the solely influence of the electromagnetic field. If the stationary phase approximation (SPA) is also employed, a semiclassical picture emerges in which the electron ionizes with zero velocity and localized at the potential center, evolves as a classical particle, and recombines when its trajectory crosses again the electrons birth coordinate. The maximum energy of the harmonics radiated is, then, directly related with the electron's kinetic energy at the moment of the recollision. In this picture, therefore, the whole process of harmonic generation is the sum of the contributions of each possible electron trajectory associated to a particular ionization event during the interaction. Two of these classical trajectories are plotted as orange (grey) lines in Figs. 2(b) and 3(b), corresponding to the electron ionized when the electric field is close to its maximum and when it is zero, respectively. As can be easily seen, the nature of the process of harmonic generation differs substantially between these two cases: when the electron is released near the field maximum it rescatters with the nucleus after less than a half laser period, while if released at a zero of the field no rescattering occurs after ionization. Therefore, according to the semiclassical picture, harmonics are only generated in the first case. The kinetic energy associated to the classical trajectories is plotted as a orange (grey) line in Figs. 2(a) and 3(a). As can be noticed, this quantity oscillates strongly with the electron quiver during its excursion after ionization. According to the semiclassical picture, the harmonics associated with the case of Fig. 2 will have a maximum frequency equal to the instantaneous kinetic energy at the recollision, plus the energy of the ground state. By inspection it is easy to notice that this value is around $|\epsilon_0|+3.2U_p$.

Up to now, this time-resolved description of the harmonic generation process has been generally associated to the ensemble of classical trajectories resulting from the SPA of the SFA S-matrix formulas. However, the structure of the S-matrix integral (5) also allows to isolate the contribution of a particular ionization instant in a natural way, by simply replacing $V_i(t_1)$ by $V_i'(t_1) \equiv W(t_1)V_i(t_1)$, $W(t_1)$ being a Gaussian time window defined as $W(t_1)=\exp[-(t-t_c)^2/\tau^2]$. Note that $W(t_1)$ only restricts the time interval allowed for the ionization process, i.e., the electron dynamics in the continuum is unaffected. In correspondence with the semiclassical view expressed above, Figs. 2(b), 2(c), 3(b), and 3(c) show the time dependence of the total acceleration (17) when the time window $W(t_1)$ is centered near the maximum (t_c =3.29 field periods) and the zero of the field (t_c=3.5 field periods), respectively. In both cases the width of the window is defined as $\tau=1/16$ field periods. A close inspection of both cases reveal some features unexpected with the semiclassical analysis.

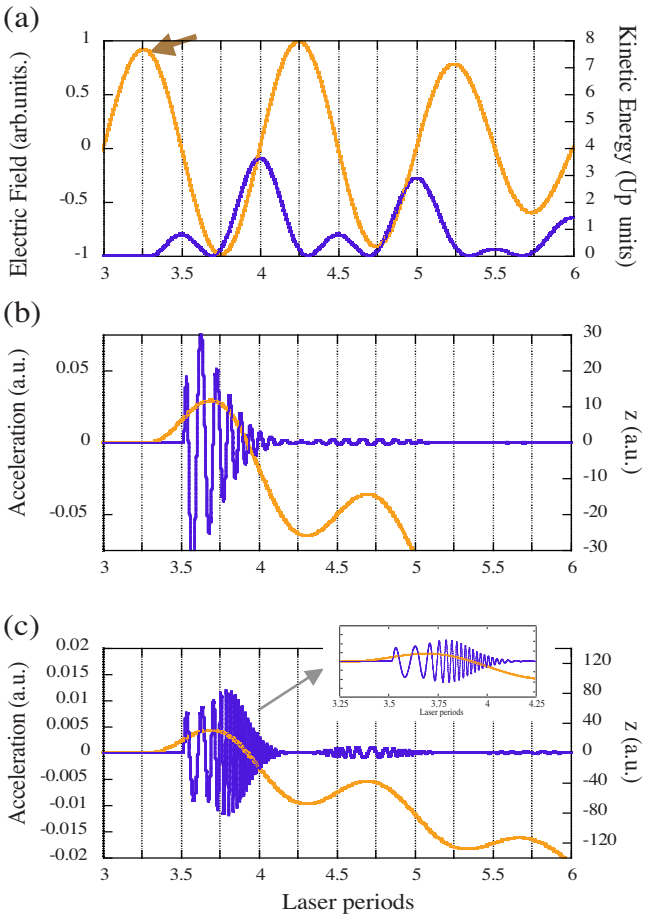

FIG. 2. (Color online) (a) Electric field [orange (grey) line] and kinetic energy [blue (black) line] associated with the classical trajectory of an electron assumed to be ionized near the field's maximum (arrow). (b) Electron trajectory associated to this situation [orange (grey) line] and dipole's acceleration computed from our S-matrix formulation for the corresponding time window for the multiphoton case of Fig. 1(a). (c) The same as for the tunnel case of Fig. 1(b). The inset shows a magnification of the acceleration near the classical time for recollision.

Let us begin considering the data shown in Fig. 2. Figure 2(b) corresponds to the multiphoton case whose spectrum is shown in Fig. 1(a), and Fig. 1(c) to the tunneling case of Fig. 1(b). In both situations, the orange (grey) line corresponds to the classical trajectory of an electron born near the field maximum, at the time where the window $W(t_1)$ is centered. There are two interesting features, in both ionization regimes, which depart from the ideas drawn by the semiclassical approach. On one hand, a strong acceleration (and therefore harmonic radiation) is generated *before* the rescattering event. Therefore, this demonstrates that harmonics are also generated in the *way out* of the ionization process, when the electron departures from the atom. In the tunneling case, however, the acceleration is strongly chirped, so that the higher spectral frequencies are generated near the classical

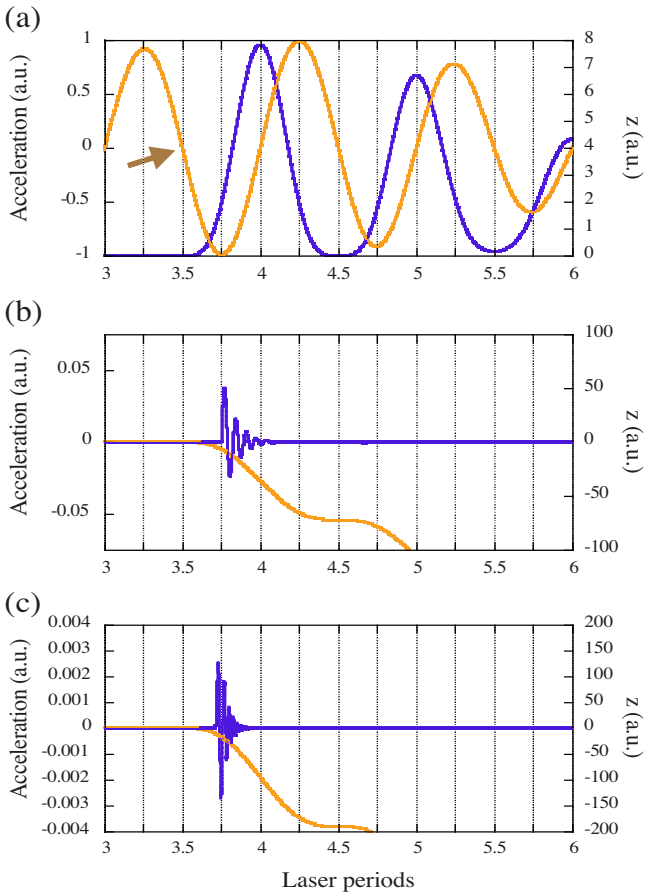

FIG. 3. (Color online) (a) Electric field [orange (grey) line] and kinetic energy [blue (black) line] associated with the classical trajectory of an electron assumed to be ionized at the field's zero (arrow). (b) Electron trajectory associated to this situation [orange (grey) line] and dipole's acceleration computed from our *S*-matrix formulation for the corresponding time window for the multiphoton case of Fig. 1(a). (c) The same for the tunnel case of Fig. 1(b).

times of the rescattering. This explains the success of the semiclassical theory to describe the nature of the harmonic radiation at the higher frequency part of the spectrum. Note that the up chirping near the time of rescattering can be interpreted classically as due to the contribution of short trajectories. However, in the multiphoton regime the chirp is reduced and the frequencies of the harmonics generated during the way out are similar to those generated at the rescattering.

The second interesting feature is the presence of secondary bursts of radiation associated with times in which the distance of the quivering electron with the parent atom reaches a minimum. Note that, as the electron trajectory does not cross the coordinate origin, this mechanism of *close up* radiation is forbidden in the semiclassical theory. These bursts are found to be down chirped in their first half and up chirped in the second, in correspondence to the kinetic energy of the electron around the time instant of maximal approach to the atom. In the tunneling regime, the maximum frequency of these burst falls well into the plateau region, so the semiclassical theory is safe when neglecting its contribution to describe the higher spectral frequencies. In the multiphoton case, the frequency of the closeup radiation is again similar to the those at the end of the plateau, and cannot be neglected to explain the source of the higher-order harmonics.

To our understanding, the physical mechanisms underlying way-out and closeup harmonic radiation are associated with the delocalization of the wave function in the continuum: first, the electron's wave packet is not perfectly localized at the instant of ionization, second, the wave packet spreads during the excursion through the continuum and, finally, the recombination does not take place at the coordinate origin, as the acceleration operator has some spatial extension. As a result, the overlap of the ionized wave function with the ground state and the acceleration operator remains operative in situations where the classical trajectories are far from the nucleus.

The same analysis can also be applied to the case of Figs. 2(b) and 2(c) where we show the acceleration associated to the ionization of a wave packet near the zero of the field. In this case, the classical trajectory never comes back to the nucleus, therefore the harmonic generation is forbidden semiclassically. Our results show, however, that harmonics are generated right after the ionization, when the electron leaves the parent ion. This *way-out* radiation is typically decreasing shortly after ionization and has frequencies also in the middle of the plateau. Note that although the electron's kinetic energy can reach $8U_p$ in its way out (for instance at the beginning of the four cycle), this possibility is not mapped into the harmonic frequencies, since they are dominated by the slower part of the wave function, which is the one remaining closer to the parent ion. Note that, in the tunneling case, the amplitude of the acceleration during this way-out process is substantially smaller than when the electron departs near a maximum of the field. This later fact is consistent with the known dramatic decrease of the tunnel ionization probabilities when the field amplitude decreases.

IV. CONCLUSION

We have developed and used an *S*-matrix theory for the computation of the harmonic generation in a wide variety of situations. The theory is proven to be quantitative accurate in the description of the higher frequency part of the spectra, and to give also physical insights on the time evolution of the generation process. In particular, we have shown two radiative processes in addition to the conventional electron rescattering. This *way out* or the *closeup* mechanisms of radiation are demonstrated to be irrelevant in the qualitative understanding of the generation of the higher harmonics in the tunneling case, but not in the multiphoton regime. Further investigation on the applicability of our model to species different from hydrogen and to molecules is currently being done.

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