Quantum theory of radiation by nonstationary systems with application to high-order harmonic generation

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In quantum theory, the description of radiation by a system of charged particles in stationary and nonstationary cases requires different approaches. While radiative processes in stationary systems are covered in textbooks, the emission of radiation by nonstationary systems driven by an external time-dependent force has not received due attention. In this paper, we develop a quantum theory describing the spectrum of photons emitted by a generic nonstationary atomic system and show how to implement it in practical calculations. We also discuss the approximation in which the exact quantum formula for the spectrum reduces to the well-known classical formula with the classical dipole moment replaced by the expectation value of the dipole moment calculated quantum mechanically. This approximation underlies the classical ansatz commonly used in the theory of high-order harmonic generation (HHG). To illustrate the theory, we apply it to calculating HHG spectrum for a one-dimensional zero-range-potential model. The spectra obtained from the quantum formula and from the ansatz are different and the difference grows as the probability of ionization grows. This difference is an observable effect predicted by the present theory.

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I. INTRODUCTION

In classical theory, the radiation of electromagnetic waves by a system of charged particles under certain approximations is completely described by the time-dependent electric dipole moment $\mathbf{d}_{\rm cl}(t)$ of the system. In particular, the spectral density of the radiation at frequency $\omega > 0$ is given by [1,2]

$$I_{\rm cl}(\omega) = \frac{2\omega^4}{3\pi c^3}|\mathbf{d}_{\rm cl}(\omega)|^2. \tag{1}$$

Here and in the following, for any function of time $f(t)$ its Fourier transform is defined by

$$f(\omega) = \int f(t)e^{i\omega t}\,dt. \tag{2}$$

Equation (1) holds for stationary systems, whose Hamiltonians do not depend on time, as well as for nonstationary systems interacting with some external time-dependent forces. This classical formula is derived under the following two approximations. First, the particles are nonrelativistic, which amounts to the dipole approximation. Second, the particles are assumed to perform a given motion driven by their interactions with each other and with external forces, but not affected by the emitted radiation. In other words, they move as if there were no radiation, and this motion defines the dipole moment $\mathbf{d}_{\rm cl}(t)$.

Consider the radiation by charged particles under the same two approximations, but in the framework of quantum theory. Stationary and nonstationary systems in this case require different treatments. Quantum theory of radiation by stationary systems is presented in textbooks [3,4]. An elementary process considered in this theory is the spontaneous emission of a photon in transitions of an atom from one stationary state to another stationary state with lower energy in the discrete or continuous spectrum. The second of the approximations indicated above means that the interaction with the electromagnetic field causing the emission should be taken into account in the leading order of perturbation theory, so the treatment is based on Fermi's golden rule. Because of the first approximation, the emission rate is expressed in terms of a matrix element of the dipole operator $\hat{\mathbf{d}}$. However, this standard approach is not applicable to nonstationary systems. This can be seen already from the fact that nonstationary systems do not have stationary states; therefore, radiative processes cannot be described by rates. Instead, one should consider the total probability of emitting a photon during the action of an external time-dependent force. Furthermore, there exists radiation induced by the external force, which would not appear without it. The description of this radiation requires a different approach and, except for the case of a weak external force which can be treated perturbatively (as, e.g., in the problem of scattering of a weak electromagnetic field by an atom [4]), is not discussed in textbooks. The goal of this paper is to develop a quantum theory which would enable one to calculate the spectrum of photons emitted by a nonstationary system under the same approximations as in Eq. (1).

To be more specific, we mention that our interest to this general problem is motivated by its relation to the theory of high-order harmonic generation (HHG). The point is that in all calculations in the theory of HHG, beginning with the pioneering papers [5–10], it is tacitly postulated that the spectrum of harmonics generated by an atom in a strong laser field is described by Eq. (1) with the classical dipole moment replaced by the expectation value of the dipole moment calculated quantum mechanically,

$$\mathbf{d}_{\rm cl}(t) \to \mathbf{d}(t) = \langle\psi(t)|\hat{\mathbf{d}}|\psi(t)\rangle, \tag{3}$$

where $|\psi(t)\rangle$ is the state vector of the atom interacting with the laser field, i.e., of the nonstationary system which emits the HHG radiation. We call this replacement the classical ansatz. In earlier studies on the quantum theory of HHG [11,12], it was recognized that the replacement involves an *additional* approximation not used in the derivation of the classical formula (1). From the point of view of the general theory, it is desirable to clarify the dynamical origin of this approximation. From the point of view of applications, it is important to realize its consequences in the theory of HHG. In the present paper we address both these issues.

The paper is organized as follows. In Sec. II, a quantum theory of radiation for a generic nonstationary system is developed. We derive the quantum analog of Eq. (1) and show how to implement this formula in practical calculations. We also compare the present theory with the standard treatment of spontaneous emission in the stationary case (Appendix) and with a theory for the nonstationary case proposed in Ref. [12] (Sec. II G). In Sec. III, we discuss how the classical ansatz (3) emerges from the present theory and analyze the difference between the exact quantum formula and the ansatz in two analytically tractable cases. In Sec. IV, the theory is illustrated by calculating HHG spectrum for a model one-dimensional atom. Section V concludes the paper.

II. QUANTUM THEORY

A. Time-dependent Schrödinger equation

We consider a one-electron atom described by a time-independent potential $\hat{V}_{\rm a}$ interacting with a time-dependent potential $\hat{V}_{\rm ext}(t)$ representing an external force exerted on the electron and a quantized electromagnetic field. The time-dependent Schrödinger equation for the combined atom-field system reads (atomic units are used throughout)

$$i\frac{\partial|\Psi(t)\rangle}{\partial t} = [\hat{H} + \hat{V}_{\rm ext}(t)]|\Psi(t)\rangle, \tag{4}$$

where $|\Psi(t)\rangle$ is the state vector of the system and $\hat{H}$ is its Hamiltonian in the absence of the external force. We have

$$\hat{H} = \hat{H}_0 + \hat{H}_{\rm int}, \tag{5}$$

where $\hat{H}_0$ describes the noninteracting atom and field,

$$\hat{H}_0 = \hat{H}_{\rm a} + \hat{H}_{\rm f}, \tag{6}$$

$\hat{H}_{\rm a}$ is the Hamiltonian of the unperturbed atom,

$$\hat{H}_{\rm a} = \frac{\hat{\mathbf{p}}^2}{2} + \hat{V}_{\rm a}, \tag{7}$$

$\hat{H}_{\rm f}$ is the field Hamiltonian, and $\hat{H}_{\rm int}$ represents the atom-field interaction. The electromagnetic field is quantized in a cubic box of volume L^3 with periodic boundary conditions [3,4]. The field modes in the box are enumerated by a multi-index $\nu = \mathbf{q}\lambda$, where $\mathbf{q} = 2\pi(l_x, l_y, l_z)/L = \omega\mathbf{n}/c$ is the discretized wave vector, l_x, l_y, and l_z are integers, $\omega > 0$ is the mode frequency, $\mathbf{n}^2 = 1$, and $\lambda = 1, 2$ labels real polarization vectors $\mathbf{e}_\nu$ satisfying $\mathbf{e}_\nu\mathbf{q} = 0$ and $\mathbf{e}_{\mathbf{q}\lambda}\mathbf{e}_{\mathbf{q}\lambda'} = \delta_{\lambda\lambda'}$. The field Hamiltonian is

$$\hat{H}_{\rm f} = \sum_\nu \omega\hat{a}_\nu^\dagger\hat{a}_\nu, \tag{8}$$

where $\hat{a}_\nu^\dagger$ and $\hat{a}_\nu$ are the creation and annihilation operators for photons in the mode ν obeying the commutation relation $[\hat{a}_\nu, \hat{a}_\nu^\dagger] = 1$. Following classical theory [1,2] leading to Eq. (1), the atom-field interaction is treated in the dipole approximation and within the first order of perturbation theory—this amounts to the two approximations formulated below Eq. (2). Under these approximations [3,4]

$$\hat{H}_{\rm int} = \alpha\hat{\mathbf{A}}\hat{\mathbf{p}}, \quad \alpha = 1/c, \tag{9}$$

where $\hat{\mathbf{A}}$ is the vector potential operator,

$$\hat{\mathbf{A}} = \sum_\nu \mathbf{A}_\nu(\hat{a}_\nu^\dagger + \hat{a}_\nu), \quad \mathbf{A}_\nu = \sqrt{\frac{2\pi c^2}{\omega L^3}}\,\mathbf{e}_\nu. \tag{10}$$

The notation α in Eq. (9) incorporating the electron charge (equal to -1 in atomic units) is introduced to explicitly indicate the perturbation theory parameter. The external potential $\hat{V}_{\rm ext}(t)$ can be arbitrarily strong and is treated without any approximations. It is assumed to act during a finite time interval,

$$\hat{V}_{\rm ext}(t \leqslant 0) = \hat{V}_{\rm ext}(t \geqslant T) = 0. \tag{11}$$

The present treatment based on the first order of perturbation theory in α is restricted to not too large values of T. Let $\Gamma = O(\alpha^2)$ denote the characteristic rate of spontaneous radiative decay for atomic states involved in the dynamics described by Eq. (4). The applicability of the treatment requires

$$\Gamma T \ll 1. \tag{12}$$

This condition ensures that radiative processes do not affect the electronic motion, which complies with the second of the approximations used in the derivation of Eq. (1). The atomic subsystem described by the Hamiltonian $\hat{H}_{\rm a} + \hat{V}_{\rm ext}(t)$ is a nonstationary system meant in the title. Our goal is to analyze the emission of photons by this system caused by its interaction with the quantized field.

B. Bare and dressed states

Let us introduce eigenstates of the different Hamiltonians. The eigenstates of the unperturbed atom in the discrete spectrum are defined by

$$\hat{H}_{\rm a}|\psi_n\rangle = E_n|\psi_n\rangle. \tag{13}$$

We need to consider two sets of eigenstates in the continuous spectrum,

$$\hat{H}_{\rm a}|\psi_{\mathbf{k}}^{\pm}\rangle = E_{\mathbf{k}}|\psi_{\mathbf{k}}^{\pm}\rangle, \quad E_{\mathbf{k}} = \mathbf{k}^2/2, \tag{14}$$

where $\mathbf{k}$ is the incident momentum and the superscripts $+$ and $-$ refer to states with outgoing and incoming scattering waves, respectively [13,14]. For brevity, in the following expansions over the complete set of atomic states, we retain only sums over the discrete spectrum, omitting integrals over the continuous spectrum, and employ the notation $|\psi_n^{\pm}\rangle$, to indicate which set of continuum states is meant. For states in the discrete spectrum the superscript is omitted.

The eigenstates of the field oscillators are defined by

$$\hat{a}_\nu^\dagger\hat{a}_\nu|N\rangle_\nu = N|N\rangle_\nu, \quad N = 0, 1, \ldots. \tag{15}$$

The eigenstates of the free field are given by products of such states for each oscillator. Thus we introduce the vacuum field state containing no photons,

$$|0\rangle = \prod_{\nu} |0\rangle_{\nu}, \quad \hat{H}_{\rm f}|0\rangle = 0. \tag{16}$$

A state with one photon in the mode ν is defined by

$$|\nu\rangle = \hat{a}_{\nu}^{\dagger}|0\rangle = |1\rangle_{\nu} \prod_{\nu' \neq \nu} |0\rangle_{\nu'}, \quad \hat{H}_{\rm f}|\nu\rangle = \omega|\nu\rangle. \tag{17}$$

One can similarly define two-photon field states, etc., but we do not need them for the analysis.

We now consider the combined atom-field system. The eigenstates of the noninteracting system are called *bare* states. We introduce zero-photon and one-photon bare states,

$$\hat{H}_0|\psi_n^{\pm}\rangle|0\rangle = E_n|\psi_n^{\pm}\rangle|0\rangle, \tag{18a}$$

$$\hat{H}_0|\psi_n^{\pm}\rangle|\nu\rangle = (E_n + \omega)|\psi_n^{\pm}\rangle|\nu\rangle. \tag{18b}$$

The operator $\hat{H}_{\rm int}$ couples the electronic and field degrees of freedom. The eigenstates of the interacting system are called *dressed* states. The corresponding zero-photon and one-photon dressed states are defined by

$$\hat{H}|\psi_n^{\pm}, 0\rangle = E_{n0}|\psi_n^{\pm}, 0\rangle, \tag{19a}$$

$$\hat{H}|\psi_n^{\pm}, \nu\rangle = E_{n\nu}|\psi_n^{\pm}, \nu\rangle. \tag{19b}$$

They can be found using stationary perturbation theory [13]. In the first order in α, we obtain $E_{n0} = E_n + O(\alpha^2)$, $E_{n\nu} = E_n + \omega + O(\alpha^2)$, and

$$|\psi_n^{\pm}, 0\rangle = |\psi_n^{\pm}\rangle|0\rangle + \alpha \sum_{\nu} \hat{G}_a^{\pm}(E_n - \omega)\mathbf{A}_{\nu}\hat{\mathbf{p}}|\psi_n^{\pm}\rangle|\nu\rangle + O(\alpha^2), \tag{20a}$$

$$|\psi_n^{\pm}, \nu\rangle = |\psi_n^{\pm}\rangle|\nu\rangle + \alpha\hat{G}_a^{\pm}(E_n + \omega)\mathbf{A}_{\nu}\hat{\mathbf{p}}|\psi_n^{\pm}\rangle|0\rangle + \alpha \times (\text{two-photon states}) + O(\alpha^2), \tag{20b}$$

where $\hat{G}_a^{\pm}(E)$ are the Green's operators for the unperturbed atom,

$$\hat{G}_a^{\pm}(E) = (E - \hat{H}_a \pm i\epsilon)^{-1} \tag{21a}$$

$$= \sum_n \frac{|\psi_n^-\rangle\langle\psi_n^-|}{E - E_n \pm i\epsilon}, \tag{21b}$$

satisfying outgoing (+) and incoming (−) wave asymptotic boundary conditions. Here and in the following, $\epsilon = +0$ denotes an infinitesimally small real positive number. Note that any of the sets $|\psi_n^{\pm}\rangle$ could be used in the spectral resolution (21b). If the subscript n corresponds to a discrete state of the atom, then the superscript $\pm$ in the notation of the state on the RHS of Eqs. (20) can be omitted (throughout the paper, LHS and RHS stand for the left- and right-hand side, respectively). However, this superscript also appears in the Green's operator, so it should be retained on the LHS of the equations. In this case, from Eq. (20a) we have

$$|\psi_n^+, 0\rangle = |\psi_n^-, 0\rangle - 2i\pi\alpha \sum_{\nu} \delta(E_n - \omega - \hat{H}_{\rm a}) \times \mathbf{A}_{\nu}\hat{\mathbf{p}}|\psi_n\rangle|\nu\rangle + O(\alpha^2), \tag{22}$$

where the second term on the RHS originates from the pole in Eq. (21a). In the time-dependent picture, the + (−) dressed states can be obtained from the corresponding bare states at $t \to -\infty$ ($t \to +\infty$) by adiabatically turning on (off) the interaction $\hat{H}_{\rm int}$. This important physical asymmetry between the + and − states will be taken into account in defining observables (see Sec. II D).

The following comment regarding the dressed states is in order here. The perturbation theory expansion in α is known to encounter a problem which appears already in the second-order correction to the eigenenergies in Eqs. (19). This correction for dressed states originating from discrete states of the unperturbed atom is generally complex. Its real part describes the Lamb shift of the atomic level [15]. This shift diverges and requires regularization [3,4]. Its imaginary part accounts for the decay of the atomic state by emitting or absorbing a photon; the characteristic decay rate appears in the condition (12). Because of the decay, complex-energy dressed states do not belong to the discrete spectrum of $\hat{H}$ but are diluted in its real-energy continuum, as in the case of autoionizing states of atoms [16]. Thus, strictly speaking, the structure of the set of dressed states essentially differs from that of bare states [3]. However, this problem does not reveal itself in the first order in α. In this approximation, dressed states are in one-to-one correspondence with bare states from which they originate according to Eqs. (20). The two sets bearing the same superscript are related by a unitary transformation. Let us discuss this *dressing* transformation in more detail.

For definiteness, we consider the − states, since this is what is needed for the following; the + states can be treated similarly. The dressing transformation is defined by

$$|\Psi_{\rm dressed}^-\rangle = \hat{\mathcal{U}}|\Psi_{\rm bare}^-\rangle, \tag{23}$$

where $|\Psi_{\rm bare}^-\rangle$ stands for any of the bare states, including multiphoton states not discussed above, and $|\Psi_{\rm dressed}^-\rangle$ denotes the corresponding dressed state. We show the unitarity of $\hat{\mathcal{U}}$ by explicitly constructing this operator. This can be done by solving Eq. (4) for $\hat{V}_{\rm ext}(t) = 0$. To obtain $|\Psi_{\rm dressed}^-\rangle$, one has to substitute $\hat{H}_{\rm int} \to \hat{H}_{\rm int}e^{-\epsilon t}$ and find the solution satisfying $|\Psi(t \to \infty)\rangle = e^{-i\hat{H}_0 t}|\Psi_{\rm bare}^-\rangle$. By comparing with Eq. (23) one obtains $\hat{\mathcal{U}}$. Thus, in the first order in α, we find

$$\hat{\mathcal{U}} = 1 + i\hat{\mathcal{R}} + O(\alpha^2), \tag{24}$$

where $\hat{\mathcal{R}} = O(\alpha)$ is given by

$$\hat{\mathcal{R}} = \int_0^{\infty} e^{i\hat{H}_0 t}\hat{H}_{\rm int}e^{-i\hat{H}_0 t}e^{-\epsilon t}dt \tag{25a}$$

$$= \alpha \sum_{\nu} (\hat{R}_{\nu}\hat{a}_{\nu}^{\dagger} + \hat{R}_{\nu}^{\dagger}\hat{a}_{\nu}) \tag{25b}$$

and

$$\hat{R}_{\nu} = \int_0^{\infty} e^{i\hat{H}_{\rm a}t}\mathbf{A}_{\nu}\hat{\mathbf{p}}\, e^{-i\hat{H}_{\rm a}t}e^{i\omega t - \epsilon t}dt \tag{26a}$$

$$= i \sum_{nm} \frac{|\psi_n^-\rangle\langle\psi_n^-|\mathbf{A}_{\nu}\hat{\mathbf{p}}|\psi_m^-\rangle\langle\psi_m^-|}{E_n - E_m + \omega + i\epsilon}. \tag{26b}$$

The operator $\hat{\mathcal{R}}$ is manifestly Hermitian, which ensures (to the first order in α) the condition of unitarity $\hat{\mathcal{U}}^\dagger\hat{\mathcal{U}} = 1$. For zero-photon and one-photon states Eq. (23) takes the form

$$|\psi_n^-, 0\rangle = \hat{\mathcal{U}}|\psi_n^-\rangle|0\rangle = |\psi_n^-\rangle|0\rangle + i\alpha\sum_\nu \hat{R}_\nu|\psi_n^-\rangle|\nu\rangle + O(\alpha^2), \tag{27a}$$

$$|\psi_n^-, \nu\rangle = \hat{\mathcal{U}}|\psi_n^-\rangle|\nu\rangle = |\psi_n^-\rangle|\nu\rangle - i\alpha\hat{R}_\nu^\dagger|\psi_n^-\rangle|0\rangle + \alpha \times (\text{two-photon states}) + O(\alpha^2). \tag{27b}$$

Taking into account Eqs. (21b) and (26b), it can be seen that Eqs. (27) agree with Eqs. (20).

From the unitarity of the dressing transformation it follows that zero-photon and one-photon dressed states with the same superscript are orthogonal, $\langle\psi_m^-, \nu|\psi_n^-, 0\rangle = \langle\psi_m^+, \nu|\psi_n^+, 0\rangle = 0$. However, such states with different subscripts are not orthogonal, which reflects the fact that dressing transformations for $+$ and $-$ states are different. Thus, for zero-photon states $|\psi_n^+, 0\rangle$ originating from discrete states of the unperturbed atom, using Eq. (22) we obtain

$$\langle\psi_m^-, \nu|\psi_n^+, 0\rangle = -2i\pi\alpha\langle\psi_m^-|\mathbf{A}_\nu\hat{\mathbf{p}}|\psi_n\rangle\delta(E_m + \omega - E_n). \tag{28}$$

We show below (see Sec. II D and the Appendix) that this projection describes spontaneous decay of the state $|\psi_n^+, 0\rangle$.

C. Virtual and real photons

The first-order correction to the eigenvector in Eq. (27a) dresses the unperturbed atomic state by a cloud of *virtual* photons [17]. These photons are bound to the atom; they cannot leave the system and therefore do not contribute to the emitted radiation. Accordingly, the dressed state on the LHS of Eq. (27a) is said to contain no *real* photons. Similarly, the dressed state on the LHS of Eq. (27b) contains one real photon, while the RHS of Eq. (27b) contains virtual zero-photon and two-photon field states. The central quantity in the following analysis is the number of photons in a given mode. To properly define this quantity, it is important to distinguish between virtual and real photons. Here we introduce the corresponding formalism.

The creation $\hat{a}_\nu^\dagger$ and annihilation $\hat{a}_\nu$ operators are defined by their action on bare states. In particular, bare states are the eigenstates of the operator $\hat{a}_\nu^\dagger\hat{a}_\nu$ giving the number of photons in the mode ν. For example, $\hat{a}_\nu^\dagger\hat{a}_\nu|\psi_n^-\rangle|0\rangle = 0$ and $\hat{a}_\nu^\dagger\hat{a}_\nu|\psi_n^-\rangle|\nu\rangle = |\psi_n^-\rangle|\nu\rangle$. However, these operators do not distinguish between virtual and real photons. To single out real photons, we need operators which would act on dressed states in the same way as $\hat{a}_\nu^\dagger$ and $\hat{a}_\nu$ act on bare states. We introduce such operators by a canonical transformation defined by Eq. (23),

$$\hat{d}_\nu = \hat{\mathcal{U}}\hat{a}_\nu\hat{\mathcal{U}}^\dagger, \quad \hat{d}_\nu^\dagger = \hat{\mathcal{U}}\hat{a}_\nu^\dagger\hat{\mathcal{U}}^\dagger. \tag{29}$$

This transformation preserves the commutation relation $[\hat{d}_\nu, \hat{d}_\nu^\dagger] = 1$. The dressed operators (29) have the same properties as $\hat{a}_\nu^\dagger$ and $\hat{a}_\nu$, but in the basis of dressed states. In particular, $\hat{d}_\nu^\dagger\hat{d}_\nu|\psi_n^-, 0\rangle = 0$ and $\hat{d}_\nu^\dagger\hat{d}_\nu|\psi_n^-, \nu\rangle = |\psi_n^-, \nu\rangle$. This enables us to interpret $\hat{d}_\nu^\dagger$ and $\hat{d}_\nu$ as the creation and annihilation operators for real photons in the mode ν. Using Eqs. (24) and (25), in the first order in α we obtain

$$\hat{d}_\nu = \hat{a}_\nu - i\alpha\hat{R}_\nu + O(\alpha^2), \tag{30a}$$

$$\hat{d}_\nu^\dagger = \hat{a}_\nu^\dagger + i\alpha\hat{R}_\nu^\dagger + O(\alpha^2). \tag{30b}$$

These equations clarify the meaning of the operators $\hat{R}_\nu$.

D. Initial condition and observables

We still have to specify the initial condition for Eq. (4) and define observables of interest here. These issues are related. Let us discuss them.

In standard treatments of radiative processes in stationary systems [3,4], the dynamics is described in terms of transitions between bare states. In the present case, radiative transitions are caused by the atom-field interaction $\hat{H}_{\text{int}}$. This interaction persists at all times; hence the transitions also occur at all times. The amplitudes of bare states continue to oscillate with time and a special limiting procedure is required to extract observables at $t \to \infty$. This approach is recalled in the Appendix.

We adopt an alternative approach in which both the initial condition for Eq. (4) and observables are defined in terms of dressed states. We assume that the atom is initially in a discrete ground or excited state indicated by the subscript 0 and there are no photons. This initial condition is specified by

$$|\Psi(0)\rangle = |\psi_0^+, 0\rangle. \tag{31}$$

The initial state is represented by a $+$ state because it is prepared in the past. Dressed states are the eigenstates of $\hat{H}$, so transitions between them are caused only by the external potential $\hat{V}_{\text{ext}}(t)$. Such transitions are localized in time in the interval $0 < t < T$, see Eq. (11), so observables can be extracted from the solution to Eqs. (4) and (31) at any time $t \geqslant T$ after the action of $\hat{V}_{\text{ext}}(t)$ is over. We are interested in two kinds of observables: the probability to find the system in a given zero-photon state,

$$P_n = |\langle\psi_n^-, 0|\Psi(t \geqslant T)\rangle|^2, \tag{32}$$

and the total probability to emit a photon in a given mode ν,

$$Q_\nu = \sum_n |\langle\psi_n^-, \nu|\Psi(t \geqslant T)\rangle|^2. \tag{33}$$

Note that the final states here are represented by $-$ states, which complies with the general rule for transitions to continuous spectrum [13]. The probability (32) describes the distribution of atomic states resulting from excitation and ionization processes not accompanied by emitting photons. The probability (33) defines the spectral, angular, and polarization distributions of the emitted photons. Of main interest here is the spectral density of radiation obtained from Q_ν in the limit $L \to \infty$ by integrating over the direction $\mathbf{n} = c\mathbf{q}/\omega$ of the wave vector of a photon and summing over its polarization λ,

$$I(\omega) = \frac{L^3\omega^3}{(2\pi c)^3}\sum_\lambda \int Q_\nu d\mathbf{n}. \tag{34}$$

To obtain the quantum analog of Eq. (1), one has to substitute here Q_ν calculated in the leading-order approximation in α.

In the stationary case, $\hat{V}_{\rm ext}(t) = 0$, the system spontaneously emits photons if the subscript 0 in Eq. (31) corresponds to an excited atomic state. We show in the Appendix that in the present approach based on Eqs. (31) and (33) spontaneous photons are an immanent property of the initial dressed state and this approach yields in the stationary case the same results as the approach used in textbooks [3,4].

The solution to Eq. (4) can be expanded in dressed states. Although the operator $\hat{V}_{\rm ext}(t)$ acts only on the electronic degrees of freedom, it couples dressed states with different numbers of photons, because of correlation between electronic and photonic degrees of freedom in the states. In the first order in α, only zero-photon and one-photon states are populated during the evolution; two-photon states are populated in the second order in α, etc. Thus the solution to Eq. (4) can be presented in the form

$$|\Psi(t)\rangle = \sum_n A_n(t) e^{-iE_n t} |\psi_n^-, 0\rangle + \alpha \sum_{n\nu} B_{n\nu}(t) e^{-i(E_n+\omega)t} |\psi_n^-, \nu\rangle, \tag{35}$$

where the coefficients $A_n(t)$ and $B_{n\nu}(t)$ are of order $O(\alpha^0)$. These coefficients cease to depend on time after the action of the external potential is over,

$$A_n(t \geqslant T) = A_n, \quad B_{n\nu}(t \geqslant T) = B_{n\nu}. \tag{36}$$

Thus the probabilities (32) and (33) can be expressed as

$$P_n = |A_n|^2, \quad Q_\nu = \alpha^2 \sum_n |B_{n\nu}|^2. \tag{37}$$

Using Eqs. (35)–(37) and the operators $\hat{d}_\nu^\dagger$ and $\hat{d}_\nu$ introduced in Sec. II C, we obtain yet another representation for Q_ν,

$$Q_\nu = \langle \Psi(t)|\hat{d}_\nu^\dagger \hat{d}_\nu|\Psi(t)\rangle|_{t\geqslant T}. \tag{38}$$

This formula shows that Q_ν defined by Eq. (33) gives the number of real photons emitted in the mode ν.

To calculate the observables P_n and Q_ν, one has to solve Eq. (4) with the initial condition (31). We next discuss two approaches to solving this problem.

E. Expansion in bare states

The solution to Eqs. (4) and (31) can be expanded in bare states. In the first order in α, it can be sought in the form

$$|\Psi(t)\rangle = |\psi(t)\rangle|0\rangle + \alpha \sum_\nu e^{-i\omega t} |\phi_\nu(t)\rangle|\nu\rangle, \tag{39}$$

where

$$|\psi(t)\rangle = \sum_n a_n(t) e^{-iE_n t} |\psi_n^-\rangle \tag{40}$$

and

$$|\phi_\nu(t)\rangle = \sum_n b_{n\nu}(t) e^{-iE_n t} |\psi_n^-\rangle \tag{41}$$

are atomic state vectors and the coefficients $a_n(t)$ and $b_{n\nu}(t)$ are of order $O(\alpha^0)$. Substituting Eq. (39) into Eqs. (4) and (31) and projecting from the left on $\langle 0|$ using Eq. (20a), we obtain

$$i\frac{\partial|\psi(t)\rangle}{\partial t} = [\hat{H}_{\rm a} + \hat{V}_{\rm ext}(t)]|\psi(t)\rangle, \tag{42a}$$

$$|\psi(0)\rangle = |\psi_0\rangle. \tag{42b}$$

Projecting on $\langle \nu|$ gives

$$i\frac{\partial|\phi_\nu(t)\rangle}{\partial t} = [\hat{H}_{\rm a} + \hat{V}_{\rm ext}(t)]|\phi_\nu(t)\rangle + e^{i\omega t}\mathbf{A}_\nu\hat{\mathbf{p}}|\psi(t)\rangle, \tag{43a}$$

$$|\phi_\nu(0)\rangle = \hat{G}_a^+(E_0-\omega)\mathbf{A}_\nu\hat{\mathbf{p}}|\psi_0\rangle. \tag{43b}$$

We have neglected terms of order $O(\alpha^2)$ and $O(\alpha)$ in Eqs. (42) and (43), respectively, which is consistent with neglecting terms of order $O(\alpha^2)$ in Eq. (39). These equations form the basis of the present approach and provide a means to implement it in practical calculations. Let us discuss them.

Equation (42) describes the atomic subsystem completely neglecting its interaction with the quantized field. The condition (11) means that the coefficients in Eq. (40) satisfy

$$a_n(t \geqslant T) = a_n, \tag{44}$$

where a_n are constants. The unitarity of the evolution described by Eq. (42) ensures that

$$\sum_n |a_n|^2 = 1. \tag{45}$$

Substituting Eqs. (27) into Eq. (35) and comparing the zeroth-order term with such a term in Eq. (39), we obtain $A_n(t) = a_n(t)$; hence $A_n = a_n$. Thus the distribution of atomic states (32) in the leading order in α is not affected by the atom-field interaction and given by

$$P_n = |a_n|^2. \tag{46}$$

Following classical theory leading to Eq. (1), we assume that the solution to Eq. (42), and hence the coefficients a_n, are known. In practice, for general potentials $\hat{V}_{\rm a}$ and $\hat{V}_{\rm ext}(t)$, the solution can be found only numerically. Note that the theory of HHG [5–10] is based on Eq. (42) with

$$\hat{V}_{\rm ext}(t) = \mathbf{F}(t)\hat{\mathbf{r}}, \tag{47}$$

where $\mathbf{F}(t)$ is the electric field of an intense laser pulse. Solving Eq. (42) numerically is also the only option in this case.

Equation (43) describes the emission of photons. This equation is inhomogeneous: the last term on the RHS of Eq. (43a), containing the solution to Eq. (42), serves as a source of photons. Because of this term, the norm of $|\phi_\nu(t)\rangle$ is not conserved. The initial condition (43b) describes photons present in the initial state (31). Using Eqs. (22) and (27a), we can rewrite it in the form

$$|\phi_\nu(0)\rangle = i[\hat{R}_\nu - 2\pi\delta(E_0-\omega-\hat{H}_{\rm a})\mathbf{A}_\nu\hat{\mathbf{p}}]|\psi_0\rangle, \tag{48}$$

where the two terms on the RHS describe virtual and real (spontaneous, see the Appendix) photons, respectively. Note that the second term vanishes if the atom is initially in the ground state. Because of the inhomogeneous term in Eq. (43a), the coefficients in Eq. (41) do not become constant at $t \geqslant T$. Substituting Eqs. (40) and (41) into Eq. (43a) and using Eq. (26b), we obtain

$$b_{n\nu}(t \geqslant T) = b_{n\nu} + i\, e^{i(E_n+\omega)t}\langle\psi_n^-|\hat{R}_\nu|\psi(t)\rangle, \tag{49}$$

where $b_{n\nu}$ are constants. Comparing the first-order terms in Eqs. (35) and (39) and using (49) we find $B_{n\nu} = b_{n\nu}$. Thus the distribution of photons (33) in the leading order in α is

given by

$$Q_\nu = \alpha^2 \sum_n |b_{n\nu}|^2. \tag{50}$$

The coefficients $b_{n\nu}$ defining this distribution are to be found from Eq. (49) by solving Eq. (43). This problem is technically similar to solving Eq. (42) and, in the general case, it also can be treated only numerically. Note, however, that it should be solved for each photon mode ν, which presents a much more laborious computational task. Equation (34) with Q_ν substituted from Eq. (50) is the quantum analog of Eq. (1) for the present system.

There exists an interesting relation connecting $|\psi(t)\rangle$ and $|\phi_\nu(t)\rangle$. Using Eqs. (42a) and (43a), we obtain

$$i\frac{\partial}{\partial t}\langle\psi(t)|\phi_\nu(t)\rangle = e^{i\omega t}\mathbf{A}_\nu \mathbf{p}(t), \tag{51}$$

where

$$\mathbf{p}(t) = \langle\psi(t)|\hat{\mathbf{p}}|\psi(t)\rangle, \quad \mathbf{p}(t \leqslant 0) = \mathbf{0} \tag{52}$$

is the expectation value of the electron momentum in the state $|\psi(t)\rangle$. From the initial conditions (42b) and (43b) we have $\langle\psi(0)|\phi_\nu(0)\rangle = 0$. Thus

$$\langle\psi(t)|\phi_\nu(t)\rangle = -i\int_0^t \mathbf{A}_\nu \mathbf{p}(t')e^{i\omega t'}dt'. \tag{53}$$

The LHS of this equation for $t \geqslant T$ can be calculated using Eqs. (44) and (49). The result of substituting Eq. (49) into Eq. (41) can be presented in the form $|\phi_\nu(t \geqslant T)\rangle = |\phi'_\nu(t)\rangle + |\phi''_\nu(t)\rangle$, where the two terms correspond to the two terms in Eq. (49). We have $\langle\psi(t)|\phi'_\nu(t)\rangle|_{t\geqslant T} = \sum_n a_n^* b_{n\nu}$ and $\langle\psi(t)|\phi''_\nu(t)\rangle|_{t\geqslant T} = e^{i\omega t}\langle\psi(t)|\hat{R}_\nu|\psi(t)\rangle$. On the other hand, using Eq. (26a) we find

$$e^{i\omega t}\langle\psi(t)|\hat{R}_\nu|\psi(t)\rangle|_{t\geqslant T} = \int_t^\infty \mathbf{A}_\nu \mathbf{p}(t')e^{i\omega t'}dt'. \tag{54}$$

Substituting all this into Eq. (53) gives [we recall Eq. (2) defining the notation]

$$\sum_n a_n^* b_{n\nu} = -i\mathbf{A}_\nu \mathbf{p}(\omega). \tag{55}$$

This relation is useful for testing consistency of the solutions to Eqs. (42) and (43).

It is instructive to consider the above equations in the stationary case, $\hat{V}_{\rm ext}(t) = 0$. In this case, Eqs. (44) and (49) hold at all times $t \geqslant 0$. From Eq. (42b) we obtain $a_n = \delta_{n0}$. Then from Eq. (43b) using Eq. (28) we find that $b_{n\nu}$ is given by Eq. (A5). Thus both sides of Eq. (55) turn to zero, so the relation holds.

F. Expansion in dressed states

The solution to Eqs. (4) and (31) can be also expanded in dressed states, as in Eq. (35). In the first order in α, the difference between dressed and bare one-photon states in the second term in Eq. (35) can be neglected, so we can rewrite this expansion in the form

$$|\Psi(t)\rangle = \sum_n A_n(t)e^{-iE_n t}|\psi_n^-, 0\rangle + \alpha\sum_\nu e^{-i\omega t}|\varphi_\nu(t)\rangle|\nu\rangle, \tag{56}$$

where

$$|\varphi_\nu(t)\rangle = \sum_n B_{n\nu}(t)e^{-iE_n t}|\psi_n^-\rangle. \tag{57}$$

We have already shown that $A_n(t) = a_n(t)$. Thus the procedure of finding the atomic distribution (46) remains the same as in the previous approach: one has to solve Eq. (42). Here we discuss an alternative procedure of finding the distribution of photons (50).

The state $|\varphi_\nu(t)\rangle$ differs from the corresponding state $|\phi_\nu(t)\rangle$ in Eq. (39) because the first term in Eq. (56) also contains terms of order $O(\alpha)$. Substituting Eq. (27a) into Eq. (56) and comparing with Eq. (39), we obtain

$$|\varphi_\nu(t)\rangle = |\phi_\nu(t)\rangle - i\,e^{i\omega t}\hat{R}_\nu|\psi(t)\rangle. \tag{58}$$

The operator $\hat{R}_\nu$ satisfies the commutation relation

$$[\hat{H}_{\rm a}, \hat{R}_\nu] = -\omega\hat{R}_\nu + i\mathbf{A}_\nu\hat{\mathbf{p}}. \tag{59}$$

Differentiating Eq. (58) in time and using Eqs. (42), (43), (48), and (59), we obtain an equation describing the evolution of $|\varphi_\nu(t)\rangle$,

$$i\frac{\partial|\varphi_\nu(t)\rangle}{\partial t} = [\hat{H}_{\rm a} + \hat{V}_{\rm ext}(t)]|\varphi_\nu(t)\rangle + i\,e^{i\omega t}[\hat{V}_{\rm ext}(t), \hat{R}_\nu]|\psi(t)\rangle, \tag{60a}$$

$$|\varphi_\nu(0)\rangle = -2i\pi\delta(E_0 - \omega - \hat{H}_{\rm a})\mathbf{A}_\nu\hat{\mathbf{p}}|\psi_0\rangle. \tag{60b}$$

This equation replaces Eq. (43) in the dressed-state expansion approach. It is also inhomogeneous; however, in contrast to Eq. (43a), the source term in Eq. (60a) vanishes after the action of $\hat{V}_{\rm ext}(t)$ is over, so the norm of $|\varphi_\nu(t)\rangle$ is conserved at $t \geqslant T$. From the second of Eqs. (37) and Eq. (57) we obtain

$$Q_\nu = \alpha^2\langle\varphi_\nu(t)|\varphi_\nu(t)\rangle|_{t\geqslant T}. \tag{61}$$

Thus the distribution of photons up to a factor α^2 coincides with the norm of $|\varphi_\nu(t)\rangle$. The possibility to characterize the radiation dynamics by the norm of the solution to Eq. (60) is an advantage of the present approach. For $\hat{V}_{\rm ext}(t) = 0$, the norm does not depend on time and remains equal to that of the RHS of Eq. (60b). Substituting this into Eq. (61) gives the probability of spontaneous decay of the initial state [see Eq. (A4) in the Appendix]. For $\hat{V}_{\rm ext}(t) \neq 0$, the last term in Eq. (60a) causes a variation of the norm, and this describes the radiation induced by the external potential. In terms of the state $|\varphi_\nu(t)\rangle$, the relation (55) takes the form

$$\langle\psi(t)|\varphi_\nu(t)\rangle|_{t\geqslant T} = -i\mathbf{A}_\nu\mathbf{p}(\omega). \tag{62}$$

We will see in Sec. III that this relation is a bridge from the quantum theory to the classical formula (1).

G. Heisenberg's picture and comparison with earlier theories

The approach used in textbook treatments of radiative processes [3,4] is applicable only to stationary systems. To treat the emission of radiation by nonstationary systems, this approach must be generalized. The theory developed above presents one of the possible generalizations. There exists another generalization initially proposed for a two-state model [11] and then developed for the general system of charged particles [12]. In both papers, the authors were motivated by applications to the theory of HHG; however, their approach

can be extended to an arbitrary form of the external time-dependent potential. Here, we discuss a relation between the present theory and the theory proposed in these earlier studies.

To compare the theories, we apply the approach of Ref. [12] to the present system. The approach employs Heisenberg's picture, so we need to introduce some additional notation. Let $\hat{U}(t)$ and $\hat{u}(t)$ denote the evolution operators for Eqs. (4) and (42a) defined by $|\Psi(t)\rangle = \hat{U}(t)|\Psi(0)\rangle$ and $|\psi(t)\rangle = \hat{u}(t)|\psi(0)\rangle$, respectively. We obtain

$$\hat{U}(t) = \hat{u}(t)e^{-i\hat{H}_{\rm ft}t}\Bigg[1 - i\alpha\sum_{\nu}\int_0^t \mathbf{A}_\nu\hat{\mathbf{p}}(t')(\hat{a}_\nu^\dagger e^{i\omega t'} + \hat{a}_\nu e^{-i\omega t'})dt' + O(\alpha^2)\Bigg], \tag{63}$$

where $\hat{\mathbf{p}}(t) = \hat{u}^\dagger(t)\hat{\mathbf{p}}\hat{u}(t)$. Let $|\Psi^{\rm D}(t)\rangle$ be the solution to Eq. (4) satisfying

$$|\Psi^{\rm D}(0)\rangle = |\psi_0\rangle|0\rangle. \tag{64}$$

In Ref. [12], the number of photons in the mode ν is defined by

$$Q_\nu^{\rm D}(t) = \langle\Psi^{\rm D}(t)|\hat{a}_\nu^\dagger\hat{a}_\nu|\Psi^{\rm D}(t)\rangle \tag{65a}$$

$$= \langle\Psi^{\rm D}(0)|\hat{a}_\nu^\dagger(t)\hat{a}_\nu(t)|\Psi^{\rm D}(0)\rangle, \tag{65b}$$

where $\hat{a}_\nu(t) = \hat{U}^\dagger(t)\hat{a}_\nu\hat{U}(t)$. Using Eq. (63), we obtain

$$\hat{a}_\nu(t) = e^{-i\omega t}\Bigg[\hat{a}_\nu - i\alpha\int_0^t \mathbf{A}_\nu\hat{\mathbf{p}}(t')e^{i\omega t'}dt' + O(\alpha^2)\Bigg]. \tag{66}$$

Substituting this into Eq. (65b) gives

$$Q_\nu^{\rm D}(t) = \alpha^2\int_0^t\int_0^t \langle\psi_0|\mathbf{A}_\nu\hat{\mathbf{p}}(t'')\mathbf{A}_\nu\hat{\mathbf{p}}(t')|\psi_0\rangle e^{i\omega(t'-t'')}dt'dt''. \tag{67}$$

This formula is the result of the application of the theory of Ref. [12] to the present problem. It coincides (up to notation) with Eq. (16) from Ref. [12] which, in turn, corresponds to Eq. (7) from Ref. [11]. Note that it yields the number of photons as a function of time. The observable photon distribution should probably be obtained from Eq. (67) in the limit $t \to \infty$. However, neither the limiting procedure was discussed nor the final result for $Q_\nu^{\rm D} = Q_\nu^{\rm D}(t \to \infty)$ was given in Ref. [12].

As can be seen from the above derivation, the theory of Ref. [12] is based on Eqs. (64) and (65a). In the present theory, these equations are replaced by Eqs. (31) and (38), respectively. The use of a dressed state (31) instead of a bare state (64) in the initial condition is justified by the fact that the Lamb shift [15] is experimentally observable. The counting of only real photons in Eq. (38) instead of all photons, as in Eq. (65a), is justified by the observation that virtual photons dress the atomic subsystem [17], but do not contribute to the emitted radiation. Thus we believe that the present theory is in closer correspondence to physical reality than the theory based on Eqs. (64) and (65a).

We can further elucidate the difference between the theories by presenting our result for Q_ν in a form which can be more directly compared with Eq. (67). Similar to Eq. (65b), we can rewrite Eq. (38) as

$$Q_\nu = \langle\Psi(0)|\hat{d}_\nu^\dagger(t)\hat{d}_\nu(t)|\Psi(0)\rangle|_{t\geqslant T}, \tag{68}$$

where $\hat{d}_\nu(t) = \hat{U}^\dagger(t)\hat{d}_\nu\hat{U}(t)$. For simplicity, we assume that the atom is initially in the ground state, so that $|\psi_0^+, 0\rangle = |\psi_0^-, 0\rangle$ and hence $|\Psi(0)\rangle = \hat{\mathcal{U}}|\Psi^{\rm D}(0)\rangle$. Then it can be easily seen that Eq. (68) differs from Eq. (65b), because $\hat{\mathcal{U}}^\dagger\hat{U}(t)\hat{\mathcal{U}} \neq \hat{U}(t)$. From Eqs. (30) we have

$$\hat{d}_\nu(t) = \hat{a}_\nu(t) - i\alpha\hat{R}_\nu(t) + O(\alpha^2), \tag{69}$$

where $\hat{R}_\nu(t) = \hat{u}^\dagger(t)\hat{R}_\nu\hat{u}(t)$. Using Eqs. (24), (25), and (66) we obtain

$$\hat{d}_\nu(t)|\Psi(0)\rangle = -i\alpha\, e^{-i\omega t}\Bigg[\int_0^t \mathbf{A}_\nu\hat{\mathbf{p}}(t')e^{i\omega t'}dt' + e^{i\omega t}\hat{R}_\nu(t) - \hat{R}_\nu + O(\alpha)\Bigg]|\Psi^{\rm D}(0)\rangle. \tag{70}$$

By solving the Heisenberg equation for $\hat{R}_\nu(t)$ using Eq. (59), we find

$$\hat{R}_\nu(t) = e^{-i\omega t}\Bigg[\hat{R}_\nu - \int_0^t \mathbf{A}_\nu\hat{\mathbf{p}}(t')e^{i\omega t'}dt' + i\int_0^t [\hat{V}_{\rm H}(t'), \hat{R}_\nu(t')]e^{i\omega t'}dt'\Bigg], \tag{71}$$

where $\hat{V}_{\rm H}(t) = \hat{u}^\dagger(t)\hat{V}_{\rm ext}(t)\hat{u}(t)$. Substituting this into Eq. (70) gives

$$Q_\nu = \alpha^2\int_0^T\int_0^T \langle\psi_0|[\hat{V}_{\rm H}(t'), \hat{R}_\nu(t')][\hat{V}_{\rm H}(t), \hat{R}_\nu(t)]|\psi_0\rangle \times e^{i\omega(t-t')}dt\,dt'. \tag{72}$$

This formula has the structure of Eq. (67), but the operators involved are different. Note that in the present case the RHS of Eq. (60b) vanishes and the solution can be expressed as

$$|\varphi_\nu(t)\rangle = \hat{u}(t)\int_0^t [\hat{V}_{\rm H}(t'), \hat{R}_\nu(t')]e^{i\omega t'}dt'|\psi_0\rangle. \tag{73}$$

Substituting this into Eq. (61) also leads to Eq. (72). We can obtain another representation for Q_ν by noting that $\hat{u}(t \geqslant T) = e^{-i\hat{H}_{\rm a}(t-T)}\hat{u}(T)$; hence

$$e^{i\omega t}\hat{R}_\nu(t) = \int_t^\infty \mathbf{A}_\nu\hat{\mathbf{p}}(t')e^{i\omega t' - \epsilon t'}dt'. \tag{74}$$

Substituting this into Eq. (70) gives

$$Q_\nu = \alpha^2\int_0^\infty\int_0^\infty \langle\psi_0|\mathbf{A}_\nu\delta\hat{\mathbf{p}}(t')\mathbf{A}_\nu\delta\hat{\mathbf{p}}(t)|\psi_0\rangle \times e^{i\omega(t-t') - \epsilon(t+t')}dt\,dt', \tag{75}$$

where

$$\delta\hat{\mathbf{p}}(t) = \hat{\mathbf{p}}(t) - e^{i\hat{H}_{\rm a}t}\hat{\mathbf{p}}\,e^{-i\hat{H}_{\rm a}t}. \tag{76}$$

The latter formula, Eq. (75), can be directly compared with Eq. (67). In this formula, the way of taking the limit $t \to \infty$ in Eq. (67) is specified. In addition, the Heisenberg momentum operator $\hat{\mathbf{p}}(t)$ for an electron moving in the total potential $\hat{V}_{\rm a} + \hat{V}_{\rm ext}(t)$ is replaced by $\delta\hat{\mathbf{p}}(t)$, where the electron momentum in the atomic potential $\hat{V}_{\rm a}$ is subtracted. We can further rewrite Eq. (75) in the form

$$Q_\nu = \alpha^2\sum_n\left|\int_0^\infty \langle\psi_n^-|\mathbf{A}_\nu\delta\hat{\mathbf{p}}(t)|\psi_0\rangle e^{i\omega t - \epsilon t}dt\right|^2, \tag{77}$$

where the summation runs over all final states of the atom, as in Eqs. (33), (37), and (50).

Summarizing the comparison, we note that although Eqs. (65b) and (67) obviously differ from Eqs. (68) and (75), respectively, we cannot state that the observable distribution $Q_\nu^{\rm D}$ following from Eq. (67) in the limit $t \to \infty$ necessarily differs from Q_ν. One can easily see that in the stationary case, $\hat{V}_{\rm ext}(t) = 0$, $Q_\nu^{\rm D}$ reduces to Q'_ν given by Eq. (A4), and thus coincides with Q_ν, if the limit is understood as explained in the Appendix. We must admit that the coincidence may hold also in the nonstationary case, $\hat{V}_{\rm ext}(t) \neq 0$, provided that the limit is properly defined. In this sense, Eq. (75) specifies the proper definition.

To close the theory section, we emphasize that Eqs. (50), (61), (72), (75), and (77) give different representations for the same quantity—the number of photons emitted in the mode ν obtained in the leading order in α. Any of them substituted into Eq. (34) gives the quantum analog of Eq. (1). Although the expressions on the RHS of the equations are shown to be formally equivalent, they are quite different from the point of view of implementation. While Eqs. (50) and (61) can be readily implemented by solving Eqs. (43) and (60), respectively, Eqs. (72), (75), and (77) [as well as Eq. (67)] are formal and not suitable for practical calculations. We are going to illustrate our theory by calculations (see Sec. IV), so the difference is important. This explains our priorities in the presentation.

III. CLASSICAL ANSATZ

A. Classical approximation

The quantum theory developed above can be used to shed some light on the dynamical origin of the classical ansatz. It enables us to locate the approximation underlying the replacement (3) at a more fundamental level. Let us try to find an approximate solution to Eq. (60) in the form

$$|\varphi_\nu(t)\rangle \approx f_\nu(t)|\psi(t)\rangle, \tag{78}$$

where $f_\nu(t)$ is a function of time. Projecting this equation from the left on $\langle\psi(t)|$ and taking into account that $\langle\psi(t)|\psi(t)\rangle = 1$, we find $f_\nu(t) = \langle\psi(t)|\varphi_\nu(t)\rangle$. Substituting this into Eq. (61) and using Eq. (62), we obtain

$$Q_\nu \approx Q_\nu^{(\rm c)} = \alpha^2 |\langle\psi(T)|\varphi_\nu(T)\rangle|^2 = \alpha^2 |\mathbf{A}_\nu \mathbf{p}(\omega)|^2. \tag{79}$$

For the present system $\mathbf{p}(t) = -\dot{\mathbf{d}}(t)$, where $\mathbf{d}(t)$ is the expectation value of the dipole moment defined by Eq. (3). Thus

$$Q_\nu^{(\rm c)} = \alpha^2 \omega^2 |\mathbf{A}_\nu \mathbf{d}(\omega)|^2. \tag{80}$$

Substituting this into Eq. (34) leads to Eq. (1) with the classical dipole $\mathbf{d}_{\rm cl}(t)$ replaced by $\mathbf{d}(t)$. This is the classical ansatz. Accordingly, we refer to Eq. (78) as the classical approximation.

Comparing Eqs. (61) and (79) and using the Cauchy-Bunyakovsky-Schwarz inequality, we obtain

$$Q_\nu \geqslant Q_\nu^{(\rm c)}. \tag{81}$$

Thus the exact quantum result Q_ν for the number of photons in the mode ν always exceeds the result $Q_\nu^{(\rm c)}$ obtained in the classical approximation. This inequality is a rigorous consequence of the theory. The equality is achieved if and only if the state $|\varphi_\nu(T)\rangle$ up to a constant factor coincides with the state $|\psi(T)\rangle$. The approximation in Eq. (78) assumes that this is the case for any t.

Equation (79) could be also obtained from Eq. (75) by substituting

$$\langle\psi_0|\mathbf{A}_\nu \delta\hat{\mathbf{p}}(t'')\mathbf{A}_\nu \delta\hat{\mathbf{p}}(t')|\psi_0\rangle \to \langle\psi_0|\mathbf{A}_\nu \delta\hat{\mathbf{p}}(t'')|\psi_0\rangle\langle\psi_0|\mathbf{A}_\nu \delta\hat{\mathbf{p}}(t')|\psi_0\rangle. \tag{82}$$

This replacement amounts to retaining only the term with $n = 0$ in the sum in Eq. (77). In other words, only radiative (accompanied by emitting a photon) processes in which the atom returns to the initial state are taken into account in the classical approximation. This explains the inequality (81). A replacement similar to Eq. (82) termed decorrelation ansatz was considered in Ref. [11]. It was pointed out that it is implicit in calculations of HHG spectra [5,6]. In Ref. [12], the decorrelation was justified for a system consisting of a large number of atoms. This, however, is not relevant for the present single-atom theory.

We suspect that the classical approximation (78) can be justified under certain conditions satisfied by the atomic system, external potential, and photon mode. Such a justification, together with a small parameter controlling the accuracy of the approximation, should emerge from the analysis of Eq. (60). The problem turns out to be nontrivial and we leave it for future studies. To illustrate the difference between the exact quantum result for Q_ν and the approximation (79) based on Eq. (78), in the rest of this section we compare them in two situations which can be treated analytically. For simplicity, we assume that the atom is initially in the ground state, so the RHS of Eq. (60b) turns to zero.

B. Weak external potential: Perturbation theory

We first consider a weak external potential. In this case, the solutions to Eqs. (42) and (60) can be found using time-dependent perturbation theory [13]. In the first order in $\hat{V}_{\rm ext}(t)$, we obtain

$$Q_\nu = \alpha^2 \sum_n \left| \mathbf{A}_\nu \sum_m \left[\frac{\mathbf{p}_{nm} V_{m0}(\omega + E_n - E_0)}{E_m - E_n - \omega - i0} + \frac{V_{nm}(\omega + E_n - E_0)\mathbf{p}_{m0}}{E_m - E_0 + \omega} \right] \right|^2, \tag{83}$$

where $\mathbf{p}_{nm} = \langle\psi_n^-|\hat{\mathbf{p}}|\psi_m^-\rangle$ and $V_{nm}(t) = \langle\psi_n^-|\hat{V}_{\rm ext}(t)|\psi_m^-\rangle$. On the other hand, from Eq. (79) we obtain

$$Q_\nu^{(\rm c)} = \alpha^2 \left| \mathbf{A}_\nu \sum_m \left[\frac{\mathbf{p}_{0m} V_{m0}(\omega)}{E_m - E_0 - \omega - i0} + \frac{V_{0m}(\omega)\mathbf{p}_{m0}}{E_m - E_0 + \omega} \right] \right|^2. \tag{84}$$

One can see that $Q_\nu^{(\rm c)}$ coincides with the term with $n = 0$ in the sum over n in Eq. (83), so the inequality (81) holds. Let us consider, for definiteness, the external potential of the form (47). In this case, the distribution (83) describes photons generated in scattering of a laser field $\mathbf{F}(t)$ by the atom [4]. The summation over n in Eq. (83) corresponds to the

summation over the final states of the atom. This quantum formula accounts for both elastic (Rayleigh) scattering processes, in which the atom is left in the initial state ($n = 0$), and inelastic (Raman) scattering processes, which result in excitation or ionization of the atom ($n \neq 0$). The classical approximation (84) accounts only for elastic scattering and neglects inelastic scattering. The relative contribution of Raman processes depends on the spectral contents of the field $\mathbf{F}(t)$ and the frequency ω of scattered photons. As far as we know, in the general case there is no parameter justifying the neglect of electronic Raman processes in atoms. Thus the classical approximation (78) is not expected to hold in the perturbative regime.

C. Pulsed external potential: Sudden approximation

We now consider a strong external potential acting during a very short time. To treat this case, it is convenient to use the coordinate representation in which the potential is represented by $V_{\rm ext}(\mathbf{r}, t)$, etc. Let $V_{\rm ext}$ characterize the magnitude of $V_{\rm ext}(\mathbf{r}, t)$ and $t_{\rm a}$ denote the characteristic atomic time. We assume that $T \ll t_{\rm a}$ and $V_{\rm ext}T \gg 1$, which ensures that $V_{\rm ext} \gg \Delta E_{\rm a}$, where $\Delta E_{\rm a} \sim 1/t_{\rm a}$ is the characteristic energy distance between atomic states. Under these conditions Eqs. (42) and (60) can be solved using the sudden approximation. From Eq. (42) we obtain

$$\psi(\mathbf{r}, t \leqslant T) = e^{iS(\mathbf{r},t)}\psi_0(\mathbf{r}), \tag{85}$$

where

$$S(\mathbf{r}, t) = -\int_0^t V_{\rm ext}(\mathbf{r}, t')\, dt'. \tag{86}$$

In the same approximation

$$\hat{\mathbf{p}}\psi(\mathbf{r}, t) = \mathbf{p}(\mathbf{r}, t)\psi(\mathbf{r}, t), \quad \mathbf{p}(\mathbf{r}, t) = \nabla S(\mathbf{r}, t). \tag{87}$$

The characteristic frequency of the emitted photons in this case is $\omega \sim 1/T \gg \Delta E_{\rm a}$. At such high frequencies the operator (26) can be approximated by $\hat{R}_\nu = i\mathbf{A}_\nu\hat{\mathbf{p}}/\omega$. Thus the solution to Eq. (60) is

$$\varphi_\nu(\mathbf{r}, T) = -i\mathbf{A}_\nu\mathbf{p}(\mathbf{r}, \omega)\psi(\mathbf{r}, t). \tag{88}$$

Substituting this into Eq. (61), we find

$$Q_\nu = \alpha^2 \int \psi_0^*(\mathbf{r})|\mathbf{A}_\nu\mathbf{p}(\mathbf{r}, \omega)|^2\psi_0(\mathbf{r})d\mathbf{r}. \tag{89}$$

On the other hand, from Eq. (79) we obtain

$$Q_\nu^{(\rm c)} = \alpha^2\left|\int \psi_0^*(\mathbf{r})\mathbf{A}_\nu\mathbf{p}(\mathbf{r}, \omega)\psi_0(\mathbf{r})d\mathbf{r}\right|^2. \tag{90}$$

The classical approximation (90) again differs from the quantum formula (89). The integral in Eq. (90) is a coherent sum of the emission amplitudes of the different volume elements, while that in Eq. (89) is an incoherent sum of the emission intensities. It can be seen that the inequality (81) holds. The difference between Eqs. (89) and (90) strongly depends on the shape of the momentum $\mathbf{p}(\mathbf{r}, \omega)$ as a function of $\mathbf{r}$. In particular, for the external potential of the form (47), the momentum does not depend on $\mathbf{r}$ and the difference disappears. This example supports our suspicion that, under certain conditions on the system, the classical approximation (78) can be justified by the analysis of Eq. (60).

IV. APPLICATION TO HIGH-ORDER HARMONIC GENERATION

To illustrate the theory, we apply it to the calculation of the HHG spectrum in a one-dimensional model. Before we turn to the model, let us comment on the applicability of the present theory to the HHG process. First, in a fully quantum theory of HHG, the strong infrared laser field should be treated quantum mechanically, on an equal footing with the HHG field. However, the high intensity of the laser field means large occupation numbers of photonic states. In this case, the field can be treated classically [4] and its interaction with the atom can be described by the potential (47). Note that in Refs. [11,12] the laser field was also treated classically. Second, a quantitative theory of HHG should take into account medium effects which can modify experimentally observable HHG spectra compared to theoretical predictions for a single atom. However, the spectra are still based on a single-atom response. This justifies the interest to single-atom treatments of HHG in Refs. [5–9] and below.

It is convenient to use the coordinate representation. We consider a one-dimensional atom described by a zero-range potential,

$$\hat{H}_{\rm a} = -\frac{1}{2}\frac{d^2}{dx^2} - \varkappa\delta(x). \tag{91}$$

This model was used previously for developing the adiabatic theory of strong-field ionization [18] and HHG [19]. The unperturbed atom has only one bound state,

$$E_0 = -\varkappa^2/2, \quad \psi_0(x) = \varkappa^{1/2}e^{-\varkappa|x|}. \tag{92}$$

Its scattering states meant in Eq. (14) are given by

$$E_k = k^2/2, \quad \psi_k^{(\pm)}(x) = e^{ikx} + R(\pm|k|)e^{\pm i|kx|}, \tag{93a}$$

$$R(k) = \frac{i\varkappa}{k - i\varkappa}, \quad -\infty < k < \infty. \tag{93b}$$

The external potential is $\hat{V}_{\rm ext}(t) = F(t)x$, where $F(t)$ is the electric field of a laser pulse. Let us modify the notation $\nu \to \omega$ and $\mathbf{A}_\nu \to A_\omega$ for the present one-dimensional case and omit an unessential factor in the spectrum of photons by setting $\alpha A_\omega = 1$. Then Eqs. (42) and (43) take the form

$$i\frac{\partial\psi(x, t)}{\partial t} = [\hat{H}_{\rm a} + F(t)x]\psi(x, t), \tag{94a}$$

$$\psi(x, 0) = \psi_0(x), \tag{94b}$$

and

$$i\frac{\partial\phi_\omega(x, t)}{\partial t} = [\hat{H}_{\rm a} + F(t)x]\phi_\omega(x, t) + e^{i\omega t}\hat{p}\psi(x, t), \tag{95a}$$

$$\phi_\omega(x, 0) = \frac{-i\varkappa^{3/2}}{\omega}\mathrm{sgn}(x)(e^{-\varkappa|x|} - e^{-\varkappa(\omega)|x|}), \tag{95b}$$

where $\hat{p} = -id/dx$ and

$$\varkappa(\omega) = \sqrt{\varkappa^2 + 2\omega}. \tag{96}$$

Equations (94) and (95) are solved numerically using a method described in Ref. [18]. This yields the solutions at the

end of the pulse, $\psi(x,T)$ and $\phi_\omega(x,T)$. The observables are then calculated by projecting these functions onto the atomic states. Using Eqs. (40) and (44), we obtain

$$a_0 = e^{iE_0T}\int \psi_0(x)\psi(x,T)dx, \tag{97a}$$

$$a_k = e^{iE_kT}\int \psi_k^{(-)*}(x)\psi(x,T)dx. \tag{97b}$$

The probability for the atom to remain in the initial state and the photoelectron momentum distribution (PEMD) are given by

$$P_0 = |a_0|^2, \quad P(k) = |a_k|^2. \tag{98}$$

The unitarity condition (45) takes the form

$$P_0 + P_{\rm ion} = 1, \quad P_{\rm ion} = \int P(k)\frac{dk}{2\pi}. \tag{99}$$

Similarly, using Eq. (41), we obtain

$$b_{0\omega}(T) = e^{iE_0T}\int \psi_0(x)\phi_\omega(x,T)dx, \tag{100a}$$

$$b_{k\omega}(T) = e^{iE_kT}\int \psi_k^{(-)*}(x)\phi_\omega(x,T)dx. \tag{100b}$$

Then from Eq. (49) we find

$$b_{0\omega} = b_{0\omega}(T) + \int \frac{e^{i(E_0+\omega-E_{k'})T}\,p_{0k'}a_{k'}}{E_0+\omega-E_{k'}+i0}\frac{dk'}{2\pi}, \tag{101a}$$

$$b_{k\omega} = b_{k\omega}(T) + \frac{e^{i(E_k+\omega-E_0)T}\,p_{k0}a_0}{E_k+\omega-E_0} + \int \frac{e^{i(E_k+\omega-E_{k'})T}\,p_{kk'}a_{k'}}{E_k+\omega-E_{k'}+i0}\frac{dk'}{2\pi}, \tag{101b}$$

where

$$p_{k0} = \int \psi_k^{(-)*}(x)\hat{p}\,\psi_0(x)\,dx = \frac{2k\varkappa^{3/2}}{\varkappa^2+k^2}, \tag{102a}$$

$$p_{kk'} = \int \psi_k^{(-)*}(x)\hat{p}\,\psi_{k'}^{(-)}(x)\,dx = 2\pi k\delta(k-k') - \frac{2\varkappa}{k^2-k'^2+i0}\left(\frac{k'|k|}{|k|-i\varkappa} - \frac{k|k'|}{|k'|+i\varkappa}\right). \tag{102b}$$

According to Eq. (50), the quantum spectrum of HHG photons is given by

$$Q(\omega) = |b_{0\omega}|^2 + \int |b_{k\omega}|^2\frac{dk}{2\pi}. \tag{103}$$

In the classical approximation, the spectrum is obtained from Eq. (79),

$$Q^{(c)}(\omega) = |p(\omega)|^2, \tag{104}$$

where $p(\omega)$ is the Fourier transform of

$$p(t) = \int \psi^*(x,t)\hat{p}\psi(x,t)dx \tag{105}$$

calculated as described in Ref. [19]. Equation (104) amounts to the classical ansatz commonly used in calculations of HHG [5–9].

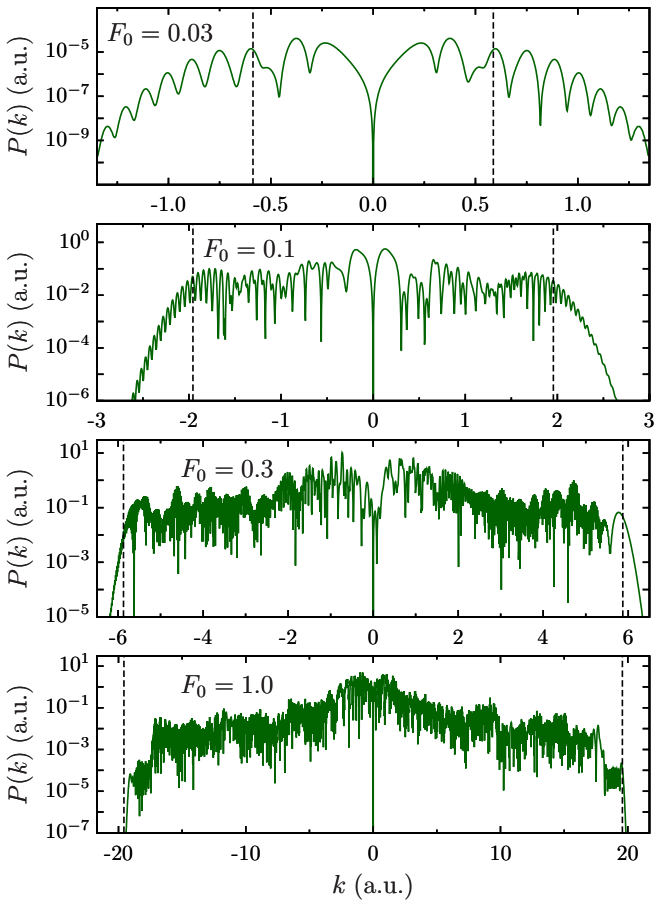

FIG. 1. Photoelectron momentum distributions $P(k)$, Eq. (98), generated by pulses with frequency $\omega_0 = 0.114$ ($\lambda \approx 400$ nm) and amplitudes indicated in the figure. The survival probability for $F_0 =$ 0.03, 0.1, 0.3, and 1 is $P_0 = 1 - 0.324\times10^{-5}$, 0.948, 0.456×10^{-1}, and 0.204×10^{-2}, respectively. The vertical dashed lines show the classical cutoffs at $k^2/2 = 10.007\times U_p$ [22].

We consider pulses with the Gaussian envelope,

$$F(t) = F_0 e^{-(2t'/\tau)^2}\cos(\omega_0 t'), \quad t' = t - T/2, \tag{106}$$

where F_0, ω_0, and $\tau = 2\pi n_{\rm oc}/\omega_0$ are the pulse amplitude, frequency, and width, respectively, and $n_{\rm oc}$ is the number of optical cycles in the pulse. To comply with Eq. (11), we solve Eqs. (94) and (95) in a finite interval $0 \leqslant t \leqslant T$, where T is chosen to be sufficiently large so that the observables do not depend on this parameter. All the calculations reported below are performed with $n_{\rm oc} = 5$ and $T = 8\tau$. We are mainly interested in the adiabatic regime relevant to strong-field physics [20], which corresponds to sufficiently low frequencies and high intensities of the laser field [18,21]. Accordingly, we consider pulses with two frequencies $\omega_0 = 0.114$ and 0.057 corresponding to the wavelengths $\lambda \approx 400$ nm and 800 nm, respectively. For each of the frequencies, we performed calculations for four pulses with amplitudes $F_0 = 0.03$, 0.1, 0.3, and 1 corresponding to intensities from 3.2×10^{13} W/cm^2 to 3.5×10^{16} W/cm^2. In the calculations we set $\varkappa = 1$, so $E_0 = -0.5$. For completeness of the discussion, we consider observables of both kinds, the PEMDs and the HHG spectra. The results obtained are shown in Figs. 1–4.

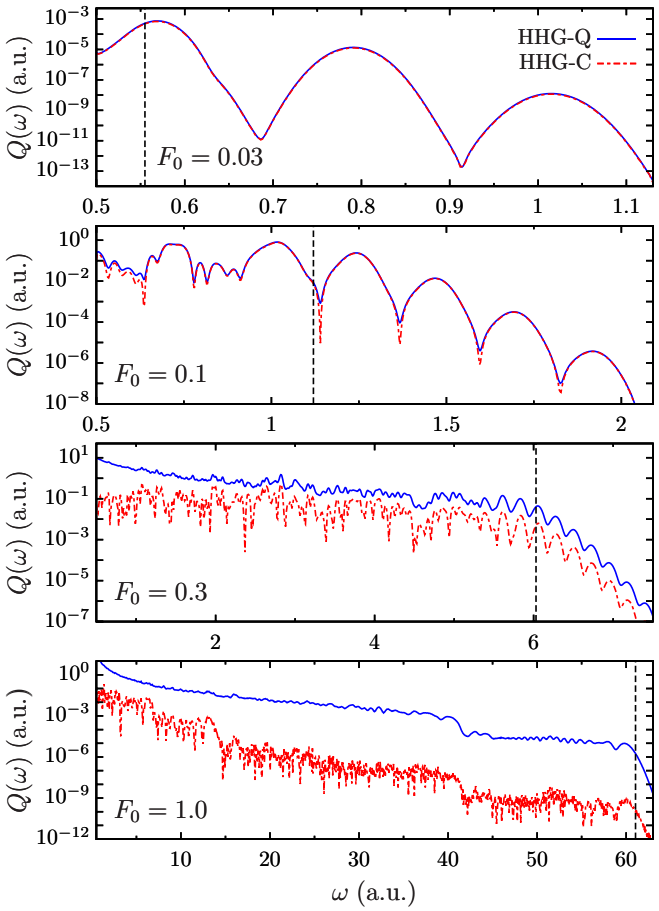

FIG. 2. HHG spectra for the same pulses as in Fig. 1. Solid (blue, HHG-Q) lines show results of the present quantum theory obtained from Eq. (103). Dashed (red, HHG-C) lines show results of the classical approximation from Eq. (104). The vertical dashed lines show the classical cutoff at $\omega = |E_0| + 3.173\times U_p$ [23].

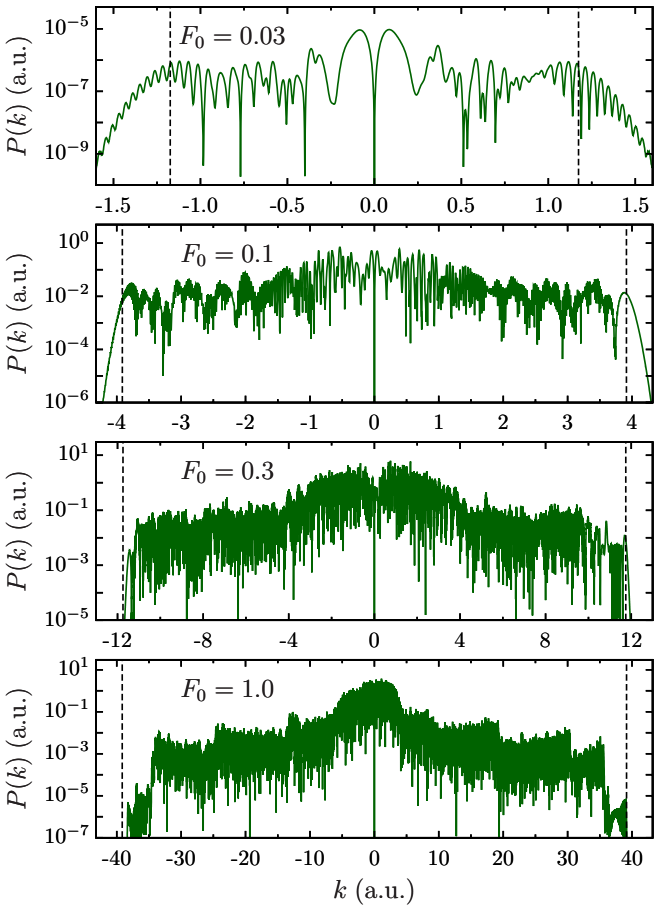

FIG. 3. Similar to Fig. 1, but for pulses with frequency $\omega_0 = 0.057$ ($\lambda \approx 800$ nm). The survival probability for $F_0 = 0.03$, 0.1, 0.3, and 1 is $P_0 = 1 - 0.227\times10^{-5}$, 0.935, 0.397×10^{-4}, and 0.112×10^{-3}, respectively.

Figures 1 and 3 present survival probabilities P_0 (given in the captions) and PEMDs $P(k)$ for pulses with $\omega_0 = 0.114$ and 0.057, respectively, obtained from Eqs. (98). For both frequencies, P_0 rapidly decreases as the pulse amplitude grows and becomes negligibly small [which means almost complete ionization, see Eq. (99)] for the largest value of F_0 considered. Note that for the lower frequency the decrease of P_0 is not monotonic. This is explained by adiabatic recombination which is the inverse process to tunneling ionization [18]. In the adiabatic regime, the PEMD $P(k)$ is localized in k between classical cutoffs at $k^2/2 = 10.007\times U_p$ [22] shown by vertical dashed lines, where $U_p = F_0^2/4\omega_0^2$ is the ponderomotive potential. The cutoffs are determined by backward rescattering trajectories born near the main positive and negative maxima of the field $F(t)$. The secondary cutoffs seen as abrupt jumps in the bottom panels of Figs. 1 and 3 at smaller $|k|$ correspond to backward rescattering trajectories born near secondary maxima of $F(t)$. More details on the behavior of $P(k)$ for the present model in the adiabatic regime can be found in Ref. [18].

Figures 2 and 4 present HHG spectra for the same pulses as in Figs. 1 and 3, respectively. Solid (blue) lines denoted by HHG-Q show quantum spectra obtained from Eq. (103). Dashed (red) lines denoted by HHG-C show the corresponding spectra obtained in the classical approximation from Eq. (104). The HHG-C spectra for the present model in the adiabatic regime were analyzed in Ref. [19]. They are localized in ω below the classical cutoff at $\omega = |E_0| + 3.173 \times U_p$ [23] shown by vertical dashed lines. Similar to the PEMD case, this cutoff is determined by a rescattering trajectory born near the main maximum of $|F(t)|$. The secondary cutoffs seen in the bottom panels of Figs. 2 and 4 at smaller ω correspond to rescattering trajectories born near secondary maxima of $|F(t)|$. For both frequencies, the HHG-Q spectra obtained from the present quantum theory differ from the HHG-C spectra obtained in the classical approximation, and this is the main conclusion to be drawn from the calculations. For the weakest field considered, $F_0 = 0.03$, the difference is marginal. However, it grows with the field and becomes dramatic for $F_0 = 1$. In all the cases, the inequality (81) is seen to hold. Note that the HHG-Q and HHG-C spectra have more or less similar shapes; in particular, they both vanish beyond the classical cutoff. The difference manifests itself in their magnitudes, especially at higher frequencies near the cutoff.

This difference can be explained as follows. In the adiabatic regime, the solution to Eq. (94) can be divided into adiabatic and rescattering parts representing electrons which

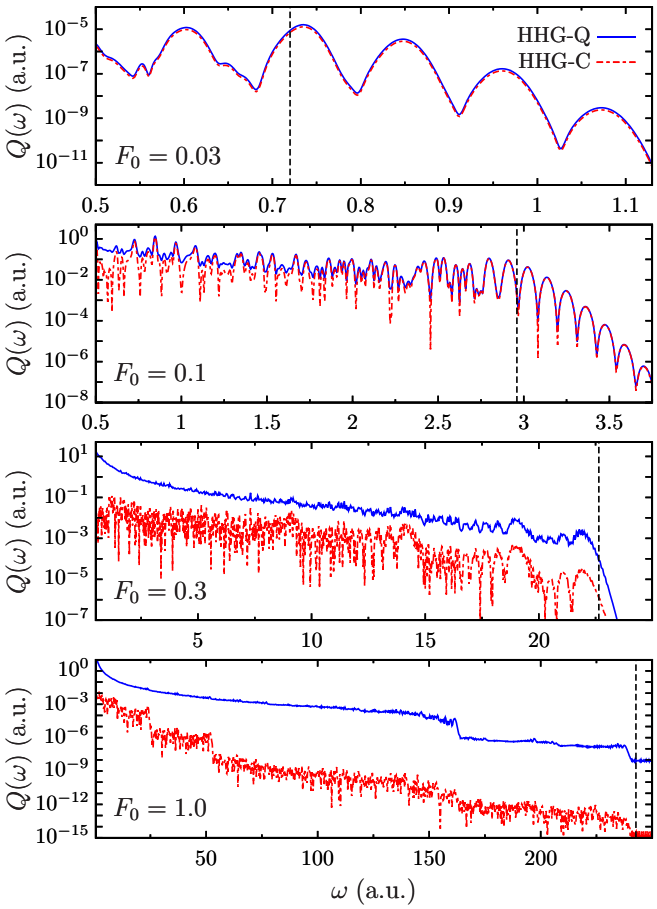

FIG. 4. Similar to Fig. 2, but for the same pulses as in Fig. 3. The continuation of both spectra beyond the cutoff in the bottom panel is an artifact caused by the lack of convergence of our calculations in this region.

remain bound and are liberated from the atom by tunneling ionization in the strong laser field, respectively [18,21]. The HHG-C spectrum (104) is determined by the cross term involving the adiabatic and rescattering parts in the integral (105); the contribution of the rescattering-rescattering term to the HHG-C spectrum in the adiabatic regime is small [19]. To contribute to the integral, the adiabatic and rescattering parts must overlap in space. To this end, the liberated electrons must return to the parent ion. This happens for the first time within one optical cycle after tunneling ionization. The maximum energy of electrons arriving for rescattering determines the cutoff frequency of the emitted HHG-C radiation [23]. If a part of the initial state survives until the maximum of the pulse envelope, the cutoff is determined by the pulse amplitude F_0. However, if the field is so strong that almost complete ionization occurs within one optical cycle or so in the rising part of the pulse envelope, the maximum energy of electrons arriving for rescattering before the initial state is completely depleted is determined by the *instantaneous* value of the pulse envelope at the moment of ionization, which is smaller than F_0. This results in a lower effective cutoff frequency. Thus the magnitude of the HHG-C spectrum near the cutoff determined by F_0 should start to decrease at sufficiently high intensities. This behavior is confirmed in Figs. 2 and 4. On the other hand, the HHG-Q spectrum contains contributions from processes in which returning electrons recombine to continuum states represented by terms with $n \neq 0$ in Eq. (77). Such radiative processes yield photons with frequency up to the classical cutoff, as in the HHG-C case. In contrast to the HHG-C case, they occur even if the initial state is completely depleted before the pulse envelope attains its maximum. A more detailed analysis of mechanisms defining the magnitude and shape of the HHG-Q spectrum is certainly required. This, however, goes beyond the scope of the present paper and is left for future studies.

Summarizing the results shown in Figs. 2 and 4, the present quantum theory predicts saturation of the HHG yield near the classical cutoff as the laser intensity grows and the ionization probability approaches unity, while the classical ansatz commonly used in the theory of HHG [5–9] predicts that the yield decreases. This behavior should be confirmed by further calculations involving the full dimensionality of the HHG problem.

V. CONCLUSION

We have developed a quantum theory describing the emission of electromagnetic radiation by a generic nonstationary atomic system under the same approximations as used in the derivation of the classical formula (1). The quantum spectrum of radiation is given by Eq. (34), where the distribution of photons Q_v is equivalently represented by Eqs. (50), (61), (72), (75), and (77). In practical calculations, this distribution can be obtained from Eq. (50) or Eq. (61) by solving Eq. (43) or Eq. (60), respectively. We have shown that under an additional approximation specified by Eq. (78) the exact quantum formula for the spectrum reduces to Eq. (1), where one should substitute Eq. (3). This classical ansatz is commonly used in the theory of HHG [5–10]. The approximation (78) sheds some light on the dynamical origin of the ansatz. We have shown that this approximation can be justified by the analysis of Eq. (60) under certain conditions on the system (see Sec. III C). To illustrate the theory, we have calculated HHG spectrum for a model one-dimensional atom. The results obtained from the quantum formula and from the classical ansatz are different. In particular, the quantum yield of HHG photons always exceeds that predicted by the ansatz. The difference grows and becomes dramatic as the probability of ionization approaches unity. Thus the present theory may have important implications for the theory of HHG.

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APPENDIX: SPONTANEOUS EMISSION IN THE STATIONARY CASE

In the absence of the external potential, $\hat{V}_{\rm ext}(t) = 0$, the system described by Eq. (4) becomes stationary. Spontaneous emission of photons by such a system is treated in textbooks [3,4]. Here, we reconsider this problem in order to elucidate

the difference between the standard approach used in textbooks and the present approach introduced in Sec. II D.

In the standard approach, the initial condition and observables are defined in terms of bare states. The initial condition formulated above Eq. (31) is specified by

$$|\Psi'(0)\rangle = |\psi_0\rangle|0\rangle. \tag{A1}$$

The solution to Eq. (4) satisfying Eq. (A1) is denoted by $|\Psi'(t)\rangle$; the prime distinguishes it from the solution to Eqs. (4) and (31). Similar to Eq. (35), the solution $|\Psi'(t)\rangle$ can be expanded in bare states. The amplitudes $\alpha B'_{n\nu}(t) = e^{i(E_n+\omega)t}\langle\nu|\langle\psi_n^-|\Psi'(t)\rangle$ of one-photon bare states populated during the evolution can be found by solving Eqs. (4) and (A1) using time-dependent perturbation theory [13]. In the first order in α one obtains

$$\begin{aligned}\alpha B'_{n\nu}(t) &= -i\int_0^t e^{i(E_n+\omega-E_0)t'}\langle\nu|\langle\psi_n^-|\hat{H}_{\rm int}|\psi_0\rangle|0\rangle dt' \\ &= -\alpha\langle\psi_n^-|\mathbf{A}_\nu\hat{\mathbf{p}}|\psi_0\rangle\frac{e^{i(E_n+\omega-E_0)t}-1}{E_n+\omega-E_0}.\end{aligned} \tag{A2}$$

The total probability to emit a photon in the mode ν is given by

$$Q'_\nu = \alpha^2\sum_n |B'_{n\nu}(t\to\infty)|^2. \tag{A3}$$

Primes in the notation $B'_{n\nu}(t)$ and Q'_ν indicate that these quantities are defined in terms of bare states. The amplitudes (A2) oscillate with time and do not have a definite limit at $t\to\infty$. However, the limit in (A3) exists if one considers Q'_ν as a generalized function of ω. The limiting procedure is based on the relation $\omega^{-2}\sin^2\omega t|_{t\to\infty} = \pi t\delta(\omega)$ also used in the derivation of Fermi's golden rule [13]. The result is

$$Q'_\nu = 2\pi\alpha^2 t\sum_n |\langle\psi_n^-|\mathbf{A}_\nu\hat{\mathbf{p}}|\psi_0\rangle|^2\delta(E_n+\omega-E_0). \tag{A4}$$

Note that, because of the δ function, the summation here runs only over discrete atomic states with energies below E_0. Hence spontaneous photons do not appear, $Q'_\nu = 0$, if the atom is initially in the ground state.

The present approach to the same problem is based on Eqs. (31) and (33). For $\hat{V}_{\rm ext}(t) = 0$, the solution to Eqs. (4) and (31) is $|\Psi(t)\rangle = e^{-iE_0t}|\psi_0^+, 0\rangle$. The amplitudes $\alpha B_{n\nu} = e^{i(E_n+\omega)t}\langle\nu, \psi_n^-|\Psi(t)\rangle$ of one-photon dressed states in Eq. (35) can be found using Eq. (28),

$$B_{n\nu} = -2i\pi\langle\psi_n^-|\mathbf{A}_\nu\hat{\mathbf{p}}|\psi_0\rangle\delta(E_n+\omega-E_0). \tag{A5}$$

These amplitudes do not depend on time. Substituting them into the second of Eqs. (37) and replacing one of the two δ functions by $t/2\pi$ (we use this common trick to shorten the discussion), we obtain Q_ν. The result coincides with Q'_ν given by Eq. (A4).

The above consideration illustrates the difference between formalisms of the two approaches. While in the standard approach the number of photons oscillates with time and the observable distribution (A4) emerges only at $t\to\infty$, in the present approach spontaneous photons are an immanent property of the initial dressed state (31) present in the system at all times $t > 0$. However, in spite of the difference, the results for the probability to spontaneously emit a photon in a given mode are identical, $Q_\nu = Q'_\nu$.

As follows from Eq. (A4), the probability Q_ν linearly grows with time, so one usually considers the partial emission rate $\Gamma_\nu = Q_\nu/t$. The total rate of emitting a photon is $\Gamma = \sum_\nu \Gamma_\nu$. The linear growth of Q_ν continues as long as $\Gamma t \ll 1$; at larger t, the depletion of the initial state due to its radiative decay cannot be neglected. The decay rate Γ is also a property of the initial dressed state (31) given by $\Gamma = -2\,\mathrm{Im}\,E_{n0}$, where E_{n0} is the complex energy of the state defined by Eq. (19a). We have $\Gamma = O(\alpha^2)$, so the decay is not accounted for by the present treatment restricted to the first-order approximation in α. As a result, the spectrum of spontaneous photons obtained by substituting Eq. (A4) into Eq. (34) consists of discrete lines located at frequencies $\omega = E_0 - E_n$ with $E_n < E_0$. Because of the assumption $\Gamma t \ll 1$, the lines have a δ-function shape. At larger times satisfying the opposite condition, $\Gamma t \gg 1$, their shape becomes Lorentzian [3,4]; this, however, goes beyond the present treatment.

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