Derivation of coupled Maxwell-Schrödinger equations describing matter-laser interaction from first principles of quantum electrodynamics

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We study the general problem of matter-laser interaction within the framework of nonrelativistic quantum electrodynamics, with particular emphasis on strong laser field effects. Consequently, we formulate a well-defined approximation leading in a straightforward manner toward the conventional semiclassical mode of description. Namely, we arrive naturally to two coupled equations of motion: (i) the Schrödinger equation which governs the quantum dynamics of an atomic system driven by classically described radiation field (composed of an incoming laser pulse plus radiation emitted from the atom), and (ii) the classical Maxwell wave equation which describes the emission of radiation from the mentioned atomic source. Employing the formalism of adiabatic Floquet theory, we derive a simple criterion of validity of the just described semiclassical approach. It shows that the semiclassical treatment is justified in most situations. On the other hand, it turns out that the semiclassical approximation breaks down completely in certain special but realistic cases, regardless of the fact that the incoming laser pulse contains a huge number of photons. Under such special circumstances, we anticipate new effects arising due to the quantized nature of the radiation field, to be observable, for example, in harmonic generation spectra. Our considerations are illustrated more explicitly using a simple model of a two-level atom strongly driven by a laser. The quantum dynamics of this model problem is resolved within the framework of quantum electrodynamics while adopting well-defined and physically justifiable approximations. As an outcome, analytic formulas are found serving as a quantitative criterion of (non)applicability of the semiclassical approach and demonstrating the breakdown of semiclassical theory under well-defined conditions. An illustrative numerical calculation is provided.

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I. INTRODUCTION

The problem of the interaction between atoms and intense laser light possesses a basic physical importance. In spite of the remarkable attention devoted in the literature to the subject (see for example Refs. [1–7]), the vast majority of theoretical work was based upon simplifications whose adequacy is not always evident. Most often, a classical description of the radiation field is adopted and the treatment of the problem is divided into two steps:

- In the first step, the quantum dynamics of a laser-driven atom is described by solving the time-dependent Schrödinger equation for the atomic wave function. The associated semiclassical Hamiltonian contains the conventional momentum gauge coupling term, which accounts for an interaction between the quantum mechanical particles and the incoming classical electromagnetic wave.
- In the second step, the scattered and emitted light is obtained by solving the classical wave equation of Maxwell. Radiation occurs here due to an inhomogeneous source term defined as the time-dependent expectation value of the electric current operator. An expectation value is taken here over the previous constructed wave function of the atomic system.

A more advanced approach is to solve simultaneously the coupled system of Schrödinger plus Maxwell equations, as being studied theoretically in Ref. [8] and computationally in Ref. [9]. This allows us to account for eventual changes of the wave function of the atom due to the emission of radiation, and more importantly, also for an influence of the given atomic source by the radiation emitted by other atomic sources. The latter effect might be particularly significant when studying harmonic generation from a linear chain of atoms or quantum dots, where the harmonics emitted by different sources cumulate and may thus reach a considerable intensity.

The just outlined semiclassical viewpoint obviously requires a deeper justification, which can be found only by a consistent application of the first principles of the quantum theory. Namely, the radiation field needs necessarily to be treated within the framework of QED and the studied system "atom plus radiation field" must be regarded as one nonseparable quantum entity. Mutual coupling between the atomic sources and the quantized field must be properly incorporated into the total Hamiltonian of the whole system.

It is the purpose of the present article to employ the rigorous formalism sketched in the previous paragraph and to investigate what kind of approximations are hidden behind the semiclassical approach. It seems also worthy to search for a criterion of validity of these approximations and to evaluate explicitly the correction terms arising from the quantized nature of the radiation field. Moreover, it is challenging to explore whether and under what circumstances the semiclassical description may break down and what might be the consequences regarding the harmonic generation spectra in such a case.

To our best knowledge, theoretical analysis of this kind has not been adequately addressed in the literature so far, in spite of the fact that various important contributions were made toward better understanding of strong field phenomena using the quantum electrodynamics formalism, see Refs. [10–17] and citations therein.

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The article is organized as follows. Section II reviews the notoriously well-known formalism used for the description of matter-light interaction within the framework of nonrelativistic quantum electrodynamics and defines in particular the quantum mechanical Hamiltonian of the studied system "atom plus radiation field." Sections III and IV describe a canonical transformation which offers a formally exact physical picture of the matter-laser interaction process and which is suitable for further theoretical analysis from the semiclassical viewpoint. In Sec. V, we introduce a well-defined approximation leading to the semiclassical approach mentioned previously. In Sec. VI we derive consequently the semiclassical system of coupled Maxwell-Schrödinger equations. Applicability and limitations of the semiclassical approximation are critically examined within Sec. VII. Concluding remarks are given in Sec. VIII. In the Appendix, we consider a simple model problem of a two-level atom driven by laser. The general theory developed in the main text is illustrated more explicitly on this concrete system. More importantly, this model allows us to go beyond the semiclassical mode of description and to obtain a significant amount of physical insights within the quantum electrodynamics framework while adopting well-controlled approximations. As an outcome, simple analytic formulas are found serving as a quantitative criterion of (non)applicability of the semiclassical approach and demonstrating explicitly its breakdown under well-defined conditions. An illustrative numerical example is provided.

II. BASIC THEORETICAL BACKGROUND

The physical system of our interest is described by the conventional momentum gauge Hamiltonian (used, e.g., by Cohen-Tannoudji [18] and Milonni [19])

$$\mathbf{H} = \mathbf{H}_R + \sum_{j=1}^{N} \frac{1}{2\mu_j} \left(\vec{\mathbf{p}}_j - \frac{q_j}{c} \vec{\mathbf{A}}_R^{\perp}(\vec{\mathbf{r}}_j) \right)^2 + V(\vec{\mathbf{r}}^N). \quad (1)$$

Here, $\mathbf{H}_R$ represents the standard Hamiltonian of a free radiation field,

$$\mathbf{H}_R = \sum_{\nu\wp} \hbar\omega_\nu \mathbf{a}_{\nu\wp}^\dagger \mathbf{a}_{\nu p}; \quad (2)$$

whereas $\vec{\mathbf{A}}_R^{\perp}(\vec{r})$ is the associated Coulomb gauge vector potential operator, given by the usual modal expansion

$$\vec{\mathbf{A}}_R^{\perp}(\vec{r}) = \sum_{\nu\wp} \sqrt{\frac{2\pi\hbar c^2}{\omega_\nu V}} [e^{+i\vec{k}_\nu\cdot\vec{r}} \mathbf{a}_{\nu\wp} + e^{-i\vec{k}_\nu\cdot\vec{r}} \mathbf{a}_{\nu\wp}^\dagger] \vec{\varepsilon}_{\nu\wp}. \quad (3)$$

In the above formulas, each field mode is characterized by its wave vector $\vec{k}_\nu = (2\pi/L)(\nu_x, \nu_y, \nu_z)$ (whose values are discretized using a cubic quantization volume $V = L^3$) and by its polarization vector $\vec{\varepsilon}_{\nu\wp}$ (with $\wp = 1, 2$). An index j in summation (1) runs over all the charged particles forming the matter system (atom, molecule) under our study. The meaning of other symbols should be self evident. For the sake of clarity, let us point out here that the modal summations (2) and (3) are subjected to an implicit high frequency cutoff. Correspondingly, all the bare masses μ_j in Eq. (1) are cutoff dependent, as to provide a well-defined renormalized theory in the infinite cutoff limit. Keeping in mind this remark, we shall not comment more on matters of renormalization within the present text.

Dynamical quantum state of the entire system "atom plus quantized radiation field" is generally described by some state vector $|\Psi(t)\rangle$ whose time evolution obeys the time-dependent Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} |\Phi(t)\rangle = \mathbf{H} |\Phi(t)\rangle. \quad (4)$$

The choice of an adequate initial condition for $|\Phi(t)\rangle$ will be discussed later in the next section.

III. CANONICAL TRANSFORMATION USING DISPLACEMENT OPERATORS

In the case when matter interacts with a laser pulse, an analysis of the problem (4) is considerably simplified by switching into another equivalent representation to be defined in the following. Within the first step, we consider an unitary displacement operator

$$\mathbf{U}_R(t) = \mathbf{D}(\{\alpha_{\nu\wp}(t) e^{-i\omega_\nu t}\}); \quad (5)$$

introduced originally by Glauber [20] and Mollow [21]. Contrary to the works of Glauber and Mollow, we allowed here the complex amplitudes $\alpha_{\nu\wp}(t)$ to be explicitly time dependent in an as yet unspecified manner.

The displacement operator possesses basic property [21]

$$\mathbf{U}_R^\dagger(t) \vec{\mathbf{A}}_R^{\perp}(\vec{r}) \mathbf{U}_R(t) = \vec{\mathbf{A}}_R^{\perp}(\vec{r}) + \vec{A}_L^{\perp}(\vec{r}, t, \{\alpha_{\nu\wp}(t)\}); \quad (6)$$

where

$$\vec{A}_L^{\perp}(\vec{r}, t, \{\alpha_{\nu\wp}(t)\}) = \sum_{\nu\wp} \sqrt{\frac{2\pi\hbar c^2}{\omega_\nu V}} [e^{-i(\omega_\nu t - \vec{k}_\nu\cdot\vec{r})} \alpha_{\nu\wp}(t) + e^{+i(\omega_\nu t - \vec{k}_\nu\cdot\vec{r})} \alpha_{\nu\wp}^*(t)] \vec{\varepsilon}_{\nu\wp}, \quad (7)$$

is a c-number field characterized completely by the amplitudes $\{\alpha_{\nu\wp}(t)\}$. We shall interpret $\vec{A}_L^{\perp}(\vec{r}, t, \{\alpha_{\nu\wp}(t)\})$ as a classical electromagnetic wave, which contains both an incoming laser pulse and the light scattered or emitted as a consequence of an interaction with the atom. Consistently with the just mentioned interpretation, we shall require that the as yet unrestricted time-dependent amplitudes $\{\alpha_{\nu\wp}(t)\}$ must possess an infinite past limit

$$\lim_{t\to-\infty} \alpha_{\nu\wp}(t) = \alpha_{\nu\wp}^{\mathrm{inc}}; \quad (8)$$

with the asymptotic values $\{\alpha_{\nu\wp}^{\mathrm{inc}}\}$ being chosen to describe an incoming freely propagating light pulse

$$\vec{A}_L^{\perp}(\vec{r}, t, \{\alpha_{\nu\wp}^{\mathrm{inc}}\}) = \sum_{\nu\wp} \sqrt{\frac{2\pi\hbar c^2}{\omega_\nu V}} \left[e^{-i(\omega_\nu t - \vec{k}_\nu\cdot\vec{r})} \alpha_{\nu\wp}^{\mathrm{inc}} + e^{+i(\omega_\nu t - \vec{k}_\nu\cdot\vec{r})} \alpha_{\nu\wp}^{\mathrm{inc}*} \right] \vec{\varepsilon}_{\nu\wp}, \quad (9)$$

that vanishes in the atomic region for $t \to -\infty$.

Operator (5) transforms the Hamiltonian (1) into an equivalent form

$$\mathbf{H}(t) = \mathbf{U}_R^\dagger(t) \mathbf{H} \mathbf{U}_R(t) - i\hbar \mathbf{U}_R^\dagger(t) \frac{\partial}{\partial t} \mathbf{U}_R(t). \quad (10)$$

Straightforward manipulations yield a more explicit formula

$$\mathbf{H}(t) = \mathbf{H}_R + \sum_{j=1}^{N} \frac{1}{2\mu_j}\left(\vec{\mathbf{p}}_j - \frac{q_j}{c}\vec{\mathbf{A}}_R^{\perp}(\vec{\mathbf{r}}_j) - \frac{q_j}{c}\vec{A}_L^{\perp}(\vec{\mathbf{r}}_j, t, \{\alpha_{\nu\wp}(t)\})\right)^2 + V(\vec{\mathbf{r}}^N) - i\hbar\sum_{\nu\wp}\left(\dot{\alpha}_{\nu\wp}(t)e^{-i\omega_\nu t}\mathbf{a}_{\nu\wp}^{\dagger} - \dot{\alpha}_{\nu\wp}^{*}(t)e^{+i\omega_\nu t}\mathbf{a}_{\nu\wp}\right); \quad (11)$$

where the shorthand symbol $\dot{\alpha}_{\nu\wp}(t) = [\partial\alpha_{\nu\wp}(t)/\partial t]$. Clearly, an appropriate equation of motion is now

$$i\hbar\frac{\partial}{\partial t}|\Psi(t)\rangle = \mathbf{H}(t)|\Psi(t)\rangle; \quad (12)$$

where $|\Psi(t)\rangle = \mathbf{U}_R^{\dagger}(t)|\Phi(t)\rangle$.

Importantly, in an infinite past limit of $t \to -\infty$, the transformed Hamiltonian (11) coincides exactly with its untransformed and time-independent counterpart (1). This fact enables us to define now in a rigorous fashion an initial state $|\Psi(t_{\text{init}})\rangle$ of the system. We wish to associate $|\Psi(t_{\text{init}})\rangle$ (for $t_{\text{init}} \to -\infty$) with the ground atomic state, which is embedded in the quantum vacuum (and is thus entangled with the quantized field modes in a nontrivial way) while awaiting an approach of the incoming c-number light pulse (7). The choice of such an initial state is natural and unique: We identify $|\Psi(t_{\text{init}})\rangle$ with the ground state of the Hamiltonian (1). If so, the time-dependent Schrödinger equation (12) possesses a unique solution $|\Psi(t)\rangle$ whose properties are analyzed within the rest of this article.

IV. CANONICAL TRANSFORMATION USING A TIME-DEPENDENT ATOMIC BASIS SET

Proceeding further in our derivation, we consider another unitary operator

$$\mathbf{U}_A(t) = \sum_n |\varphi_n(t)\rangle_A\langle\varphi_n^0(t)|. \quad (13)$$

Here, vectors $|\varphi_n^0(t)\rangle_A = e^{-(i/\hbar)\mathcal{E}_n^0(t-t_0)}|\varphi_n^0\rangle_A$ represent the stationary states of a field free atom, that is,

$$\left(\sum_{j=1}^{N}\frac{\vec{\mathbf{p}}_j^2}{2\mu_j} + V(\vec{\mathbf{r}}^N)\right)|\varphi_n^0\rangle_A = \mathcal{E}_n^0|\varphi_n^0\rangle_A. \quad (14)$$

An index n is used above to formally label both discrete and continuous parts of the atomic energy spectrum. Vectors $|\varphi_n(t)\rangle_A$ stand for the dynamical states of an atom dressed by the c-number field (7). It holds

$$i\hbar\frac{\partial}{\partial t}|\varphi_n(t)\rangle_A = \mathbf{H}^{\text{sc}}(t)|\varphi_n(t)\rangle_A, \; |\varphi_n(t_0)\rangle_A = |\varphi_n^0\rangle_A, \quad t_0 \to -\infty; \quad (15)$$

where $\mathbf{H}^{\text{sc}}(t)$ is the semiclassical "atom-light" Hamiltonian,

$$\mathbf{H}^{\text{sc}}(t) = \sum_{j=1}^{N}\frac{1}{2\mu_j}\left(\vec{\mathbf{p}}_j - \frac{q_j}{c}\vec{A}_L^{\perp}(\vec{\mathbf{r}}_j, t, \{\alpha_{\nu\wp}(t)\})\right)^2 + V(\vec{\mathbf{r}}^N). \quad (16)$$

Operator (13) facilitates an unitary transformation of the Hamiltonian (11) into an equivalent form

$$\tilde{\mathbf{H}}(t) = \mathbf{U}_A^{\dagger}(t)\mathbf{H}(t)\mathbf{U}_A(t) - i\hbar\mathbf{U}_A^{\dagger}(t)\frac{\partial}{\partial t}\mathbf{U}_A(t). \quad (17)$$

After some manipulations we arrive toward a more explicit expression

$$\tilde{\mathbf{H}}(t) = -\sum_{nn'}\sum_{j=1}^{N}\frac{q_j}{c}|\varphi_n^0(t)\rangle_A\langle\varphi_n(t)|\vec{\mathbf{A}}_R^{\perp}(\vec{\mathbf{r}}_j)\cdot\vec{\mathbf{v}}_j^{\text{sc}}(t) \times |\varphi_{n'}(t)\rangle_A\langle\varphi_{n'}^0(t)| - i\hbar\sum_{\nu\wp}\left[\dot{\alpha}_{\nu\wp}(t)e^{-i\omega_\nu t}\mathbf{a}_{\nu\wp}^{\dagger} - \dot{\alpha}_{\nu\wp}^{*}(t)e^{+i\omega_\nu t}\mathbf{a}_{\nu\wp}\right] + \sum_n |\varphi_n^0(t)\rangle_A\left(\mathcal{E}_n^0 + \mathbf{H}_R\right)_A\langle\varphi_n^0(t)| + \sum_{nn'}\sum_{j=1}^{N}\frac{q_j^2}{2c^2\mu_j}|\varphi_n^0(t)\rangle_A\langle\varphi_n(t)| \times[\vec{\mathbf{A}}_R^{\perp}(\vec{\mathbf{r}}_j)]^2|\varphi_{n'}(t)\rangle_A\langle\varphi_{n'}^0(t)|; \quad (18)$$

where

$$\vec{\mathbf{v}}_j^{\text{sc}}(t) = \frac{1}{\mu_j}\left[\vec{\mathbf{p}}_j - \frac{q_j}{c}\vec{A}_L^{\perp}(\vec{\mathbf{r}}_j, t, \{\alpha_{\nu\wp}(t)\})\right]. \quad (19)$$

An appropriate equation of motion is now

$$i\hbar\frac{\partial}{\partial t}|\tilde{\Psi}(t)\rangle = \tilde{\mathbf{H}}(t)|\tilde{\Psi}(t)\rangle; \quad (20)$$

where $|\tilde{\Psi}(t)\rangle = \mathbf{U}_A^{\dagger}(t)|\Psi(t)\rangle$. An initial condition for $|\tilde{\Psi}(t)\rangle$ is again identified with the ground state of the Hamiltonian (1), as in the preceding section.

V. SEMICLASSICAL APPROXIMATION

All the manipulations carried out so far have been fully exact, such that $|\tilde{\Psi}(t)\rangle$ describes the exact light-induced quantum dynamics of the entangled system "atom plus quantized radiation field." The main purpose of the present article is to relate the previously discussed rigorous QED formalism with the semiclassical "quantized atom-classical light" treatment, as being motivated in the Introduction. Importantly, it turns out that the semiclassical approach is obtained from the exact formulations (18) through (20) by imposing two well-defined approximations:

(a) We erase all the off-diagonal ($n \neq n'$) matrix elements

$$_A\langle\varphi_n(t)|\vec{\mathbf{A}}_R^{\perp}(\vec{\mathbf{r}}_j)\cdot\vec{\mathbf{v}}_j^{\text{sc}}(t)|\varphi_{n'}(t)\rangle_A, {}_A\langle\varphi_n(t)| \times(\vec{\mathbf{A}}_R^{\perp}(\vec{\mathbf{r}}_j))^2|\varphi_{n'}(t)\rangle_A, \quad (21)$$

appearing in the formula (18). Index n becomes then a good quantum number.

(b) In addition, we neglect in Eq. (18) also the diagonal ($n = n'$) matrix elements

$$_A\langle\varphi_n(t)|(\vec{\mathbf{A}}_R^{\perp}(\vec{\mathbf{r}}_j))^2|\varphi_n(t)\rangle_A. \quad (22)$$

The implementation of the approximations (a) and (b) leads to a simplified Hamiltonian

$$\overline{\mathbf{H}}(t) = \sum_n |\varphi_n^0(t)\rangle_A\left(\mathcal{E}_n^0 + \mathbf{H}_R + \mathbf{h}_R^{nn}(t)\right)_A\langle\varphi_n^0(t)|; \quad (23)$$

where the field operator

$$\mathbf{h}_R^{nn}(t) = -\sum_{j=1}^{N} \frac{q_j}{c} {}_A\langle\varphi_n(t)|\vec{\mathbf{A}}_R^{\perp}(\vec{\mathbf{r}}_j)\cdot\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)|\varphi_n(t)\rangle_A - i\hbar\sum_{\nu\wp}\big(\dot{\alpha}_{\nu\wp}(t)e^{-i\omega_\nu t}\mathbf{a}_{\nu\wp}^{\dagger} - \dot{\alpha}_{\nu\wp}^{*}(t)e^{+i\omega_\nu t}\mathbf{a}_{\nu\wp}\big). \tag{24}$$

Within the previously introduced approximation, the "atom-light" interaction process is described by an initial value problem

$$i\hbar\frac{\partial}{\partial t}|\overline{\Psi}(t)\rangle = \overline{\mathbf{H}}(t)|\overline{\Psi}(t)\rangle,\ |\overline{\Psi}(t_0)\rangle = \big|\varphi_n^0\big\rangle_A|\{0\}\rangle_R,\quad t_0\to-\infty. \tag{25}$$

Symbol $|\{0\}\rangle_R$ stands here of course for the quantum vacuum (i.e., for the ground state of the free radiation field). Since n is a good quantum number, we are allowed to choose some concrete value $\overline{n}$ of index n and to identify the desired solution $|\overline{\Psi}(t)\rangle$ with a direct product state

$$|\overline{\Psi}(t)\rangle = e^{-(i/\hbar)\mathcal{E}_{\overline{n}}^0(t-t_0)}\big|\varphi_{\overline{n}}^0\big\rangle_A|\chi_{\overline{n}}(t)\rangle_R. \tag{26}$$

The field component $|\chi_{\overline{n}}(t)\rangle_R$ is obtained by solving an effective Schrödinger equation for the degrees of freedom of the radiation

$$i\hbar\frac{\partial}{\partial t}|\chi_{\overline{n}}(t)\rangle_R = \big[\mathbf{H}_R + \mathbf{h}_R^{\overline{n}\overline{n}}(t)\big]|\chi_{\overline{n}}(t)\rangle_R,\ |\chi_{\overline{n}}(t_0)\rangle_R = |\{0\}\rangle_R,\quad t_0\to-\infty. \tag{27}$$

It is reasonable to require that the field state $|\chi_{\overline{n}}(t)\rangle_R$ remains unpopulated by photons for all values of t. If so, an information about the light present in the system is carried solely by the c-number term (7) and not by the approximative quantum state (26). Requirement $|\chi_{\overline{n}}(t)\rangle_R \approx |\{0\}\rangle_R$ is fulfilled if, and only if, the Hamiltonian term $\mathbf{h}_R^{\overline{n}\overline{n}}(t) \equiv \mathbf{0}$. Cancellation of $\mathbf{h}_R^{\overline{n}\overline{n}}(t)$ is accomplished via a convenient choice of the as yet unspecified field amplitudes $\{\alpha_{\nu\wp}(t)\}$. Namely, we set

$$\dot{\alpha}_{\nu\wp}(t) = +ie^{+i\omega_\nu t}\sqrt{\frac{2\pi}{\hbar\omega_\nu V}}\sum_{j=1}^{N} {}_A\langle\varphi_{\overline{n}}(t)|\times e^{-i\vec{k}_\nu\cdot\vec{\mathbf{r}}_j}\big[q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\cdot\vec{\varepsilon}_{\nu\wp}\big]|\varphi_{\overline{n}}(t)\rangle_A; \tag{28}$$

while taking into account an initial condition (8). Solution (26) of an initial value problem (25) is then written down explicitly as follows

$$|\overline{\Psi}(t)\rangle = e^{-(i/\hbar)\mathcal{E}_{\overline{n}}^0(t-t_0)}e^{-(i/\hbar)\mathcal{E}_{ZPE}(t-t_0)}\big|\varphi_{\overline{n}}^0\big\rangle_A|\{0\}\rangle_R. \tag{29}$$

For the sake of clarity, we note in passing that initial value problems (8) and (28) fixes unambiguously also the amplitudes $\{\alpha_{\nu\wp}(t)\}$ entering into the exact nonapproximative Hamiltonian (18) of the previous section.

Let us extract now from the previously constructed approximative state vector $|\overline{\Psi}(t)\rangle$ some more concrete information about physical observables. Suppose that $\mathbf{Q}_A$ is an arbitrary Schrödinger picture operator acting on the atomic degrees of freedom and keeping the radiation field unaffected. The position and momentum of an electron may serve as concrete examples. An expectation value of $\mathbf{Q}_A$ over the quantum state $|\overline{\Psi}(t)\rangle$ is then evaluated as

$$\langle\mathbf{Q}_A\rangle(t) = {}_A\langle\varphi_{\overline{n}}(t)|\mathbf{Q}_A|\varphi_{\overline{n}}(t)\rangle_A. \tag{30}$$

In other words, all the measurable characteristics of the atomic system are encoded in the associated dynamical state $|\varphi_{\overline{n}}(t)\rangle_A$, whose time evolution is governed by the c-number field (7) via the Schrödinger equation (15).

Further, let $\mathbf{Q}_R$ be an arbitrary Schrödinger picture operator acting on the degrees of freedom of the radiation field and keeping the atomic variables unaffected. Electric and magnetic field strengths or the vector potential may serve as concrete examples. An expectation value of $\mathbf{Q}_R$ over the quantum state $|\overline{\Psi}(t)\rangle$ is then found to be

$$\langle\mathbf{Q}_R\rangle(t) = {}_R\langle\{\alpha_{\nu\wp}(t)e^{-i\omega_\nu t}\}|\mathbf{Q}_R|\{\alpha_{\nu\wp}(t)e^{-i\omega_\nu t}\}\rangle_R; \tag{31}$$

where the amplitudes of a coherent field state $|\{\alpha_{\nu\wp}(t)e^{-i\omega_\nu t}\}\rangle_R = \mathbf{D}(\{\alpha_{\nu\wp}(t)e^{-i\omega_\nu t}\})|\{0\}\rangle_R$ obey the differential Eq. (28). Of special importance is an expectation value

$$\langle\vec{\mathbf{A}}_R^{\perp}(\vec{r})\rangle(t) = \vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}); \tag{32}$$

which is identified immediately with the c-number light (7). Expression (32) provides an expected physical evidence of the "atom-light" interaction process. In the limit of $t\to-\infty$, only the incoming light pulse $\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}^{\mathrm{inc}}\})$ is present in the system. As soon as this light pulse reaches the atom, some radiation starts to be scattered and emitted and hence the originally constant field amplitudes $\{\alpha_{\nu\wp}\}$ vary accordingly in time, as determined by Eq. (28).

VI. THE MAXWELL WAVE EQUATION

The dynamical evolution of the c-number field (32) is determined by relations (7) and (28). Let us examine now this dynamical time evolution more explicitly. The total time derivative of the quantity (7) equals to

$$\frac{\partial}{\partial t}\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) = \sum_{\nu\wp}\sqrt{\frac{2\pi\hbar c^2}{\omega_\nu V}}[e^{-i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\dot{\alpha}_{\nu\wp}(t) + e^{+i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\dot{\alpha}_{\nu\wp}^{*}(t)]\times\vec{\varepsilon}_{\nu\wp} - i\sum_{\nu\wp}\omega_\nu\sqrt{\frac{2\pi\hbar c^2}{\omega_\nu V}}[e^{-i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\alpha_{\nu\wp}(t) - e^{+i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\alpha_{\nu\wp}^{*}(t)]\vec{\varepsilon}_{\nu\wp}. \tag{33}$$

A substitution of the formula (28) leads to a more explicit expression

$$-\frac{1}{c}\frac{\partial}{\partial t}\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) = \vec{E}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) + \frac{4\pi}{V}\sum_{j=1}^{N}\sum_{\nu\wp}\omega_\nu^{-1}{}_A\langle\varphi_{\overline{n}}(t)|\times\sin[\vec{k}_\nu\cdot(\vec{r}-\vec{\mathbf{r}}_j)]\big(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\cdot\vec{\varepsilon}_{\nu\wp}\big)\vec{\varepsilon}_{\nu\wp}|\varphi_{\overline{n}}(t)\rangle_A; \tag{34}$$

where $\vec{E}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\})$ is of course the transversal electric field derived from the Coulomb gauge vector potential (7). The second line of Eq. (34) consists of odd functions of variable $\vec{k}_\nu$ and therefore vanishes exactly after carrying out the summation over index ν. The contributions associated with the given value $(+\vec{k}_\nu)$ cancel their counterparts corresponding to $(-\vec{k}_\nu)$. Hence, we have simply

$$-\frac{1}{c}\frac{\partial}{\partial t}\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) = \vec{E}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}); \tag{35}$$

in consonance with the correspondence principle between the classical and quantum electrodynamics. In passing we note that also the associated magnetic field results from the standard formula

$$\vec{\nabla}_{\vec{r}} \times \vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) = \vec{B}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}); \tag{36}$$

as follows immediately by direct calculation.

From now on, our attention will be focused on finding an overall equation of motion for the vector field $\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\})$. An adjective overall emphasizes here our decision to concentrate on the space-time evolution of the whole field, without any explicit reference to its amplitudes $\{\alpha_{\nu\wp}(t)\}$. The mentioned way of investigation is, again, motivated by an anticipated correspondence with the formalism of classical electrodynamics.

An action of the Laplacian on the vector potential field is evaluated as

$$\begin{aligned}
&\Delta_{\vec{r}}\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) \\
&\quad = -\frac{1}{c^2}\sum_{\nu\wp}\omega_\nu^2\sqrt{\frac{2\pi\hbar c^2}{\omega_\nu V}}[e^{-i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\alpha_{\nu\wp}(t) \\
&\qquad + e^{+i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\alpha^*_{\nu\wp}(t)]\vec{\varepsilon}_{\nu\wp}.
\end{aligned} \tag{37}$$

In addition, we differentiate Eq. (35) with respect to time, to obtain the formula

$$\begin{aligned}
&-\frac{1}{c}\frac{\partial^2}{\partial t^2}\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) \\
&\quad = \frac{\partial}{\partial t}\vec{E}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) \\
&\quad = \frac{1}{c}\sum_{\nu\wp}\omega_\nu^2\sqrt{\frac{2\pi\hbar c^2}{\omega_\nu V}}[e^{-i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\alpha_{\nu\wp}(t) \\
&\qquad + e^{+i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\alpha^*_{\nu\wp}(t)]\vec{\varepsilon}_{\nu\wp} + \frac{i}{c}\sum_{\nu\wp}\omega_\nu\sqrt{\frac{2\pi\hbar c^2}{\omega_\nu V}} \\
&\qquad \times [e^{-i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\dot{\alpha}_{\nu\wp}(t) - e^{+i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\dot{\alpha}^*_{\nu\wp}(t)]\vec{\varepsilon}_{\nu\wp}.
\end{aligned} \tag{38}$$

The combination of the expressions (37) and (38) yields consequently the well-known classical wave equation

$$\begin{aligned}
&\Delta_{\vec{r}}\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) \\
&\quad = -\frac{4\pi}{c}\vec{J}^{\perp}(\vec{r},t);
\end{aligned} \tag{39}$$

with an inhomogeneous term

$$\begin{aligned}
\vec{J}^{\perp}(\vec{r},t) = -i\sum_{\nu\wp}\sqrt{\frac{\hbar\omega_\nu}{8\pi V}}[e^{-i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\dot{\alpha}_{\nu\wp}(t) \\
- e^{+i(\omega_\nu t-\vec{k}_\nu\cdot\vec{r})}\dot{\alpha}^*_{\nu\wp}(t)]\vec{\varepsilon}_{\nu\wp}.
\end{aligned} \tag{40}$$

What remains to be done is to ascribe some physical meaning to the right-hand side of Eq. (39). For this purpose, we transform the contribution (40) by the insertion of the explicit functional form (28) of the derivative $\dot{\alpha}_{\nu\wp}(t)$. It shows that

$$\begin{aligned}
\vec{J}^{\perp}(\vec{r},t) = \frac{1}{V}\sum_{j=1}^{N}\sum_{\nu\wp}{}_A\langle\varphi_{\overline{n}}(t)|\cos[\vec{k}_\nu\cdot(\vec{r}-\vec{\mathbf{r}}_j)] \\
\times\left(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\cdot\vec{\varepsilon}_{\nu\wp}\right)\vec{\varepsilon}_{\nu\wp}|\varphi_{\overline{n}}(t)\rangle_a;
\end{aligned} \tag{41}$$

or more conveniently,

$$\begin{aligned}
&\vec{J}^{\perp}(\vec{r},t) \\
&\quad = \frac{1}{2V}\sum_{j=1}^{N}\sum_{\nu\wp}{}_A\langle\varphi_{\overline{n}}(t)|\cos[\vec{k}_\nu\cdot(\vec{r}-\vec{\mathbf{r}}_j)]\left(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\cdot\vec{\varepsilon}_{\nu\wp}\right) \\
&\qquad \times\vec{\varepsilon}_{\nu\wp}|\varphi_{\overline{n}}(t)\rangle_A + \frac{1}{2V}\sum_{j=1}^{N}\sum_{\nu\wp}{}_A\langle\varphi_{\overline{n}}(t)| \\
&\qquad \times\left(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\cdot\vec{\varepsilon}_{\nu\wp}\right)\cos[\vec{k}_\nu\cdot(\vec{r}-\vec{\mathbf{r}}_j)]\vec{\varepsilon}_{\nu\wp}|\varphi_{\overline{n}}(t).
\end{aligned} \tag{42}$$

The passage to the symmetrized version (42) of equation (41) is based upon the transversal property $\vec{k}_\nu\cdot\vec{\varepsilon}_{\nu\wp}=0$ and relation (19). Another possibility is to formally symmetrize the real quantity $\vec{J}^{\perp}(\vec{r},t)$ as $[\vec{J}^{\perp}(\vec{r},t)+\vec{J}^{\perp *}(\vec{r},t)]/2$.

Relations $\vec{k}_\nu\cdot\vec{\varepsilon}_{\nu\wp}=0$ imply that the quantity (42) is purely transversal,

$$\vec{\nabla}_{\vec{r}}\cdot\vec{J}^{\perp}(\vec{r},t)=0. \tag{43}$$

Hence, each particular solution $\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\})$ of the wave equation (39) is also purely transversal, in full agreement with the definition formula (7). However, we are free to add an arbitrary strictly longitudinal component $\vec{J}^{\parallel}(\vec{r},t)$ to $\vec{J}^{\perp}(\vec{r},t)$, keeping implicitly in mind that we shall project it out when dealing with Eq. (39). Such a kind of manipulation is justified via the Helmholtz theorem [19,22]. Following this argument, we introduce a purely longitudinal term

$$\begin{aligned}
\vec{J}^{\parallel}(\vec{r},t) = \frac{1}{2V}\sum_{j=1}^{N}\sum_{\nu\wp}{}_A\langle\varphi_{\overline{n}}(t)|\cos[\vec{k}_\nu\cdot(\vec{r}-\vec{\mathbf{r}}_j)] \\
\times\left(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\cdot\vec{\kappa}_\nu\right)\vec{\kappa}_\nu|\varphi_{\overline{n}}(t)\rangle_A + \frac{1}{2V}\sum_{j=1}^{N}\sum_{\nu\wp}{}_A\langle\varphi_{\overline{n}}(t)| \\
\times\left(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\cdot\vec{\kappa}_\nu\right)\cos[\vec{k}_\nu\cdot(\vec{r}-\vec{\mathbf{r}}_j)]\vec{\kappa}_\nu|\varphi_{\overline{n}}(t)\rangle_A;
\end{aligned} \tag{44}$$

where $\vec{\kappa}_\nu=\vec{k}_\nu/k_\nu$. The longitudinal property

$$\vec{\nabla}_{\vec{r}}\times\vec{J}^{\parallel}(\vec{r},t)=\vec{0}, \tag{45}$$

of the vector function (44) results trivially. To complete the task, we compose a sum

$$\vec{J}(\vec{r},t)=\vec{J}^{\perp}(\vec{r},t)+\vec{J}^{\parallel}(\vec{r},t); \tag{46}$$

and concentrate on the wave equation

$$\Delta_{\vec{r}}\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{A}_L^{\perp}(\vec{r},t,\{\alpha_{\nu\wp}(t)\}) = -\frac{4\pi}{c}\vec{J}(\vec{r},t); \tag{47}$$

rather than on the original problem (39). In spite of the fact that the quantity $\vec{J}(\vec{r},t)$ possesses a nonzero longitudinal part, we still attach the transversal superscript $\perp$ to the vector potential, since the longitudinal part of the solution is ignored as being unphysical.

We continue by the simplification of the expression for $\vec{J}(\vec{r},t)$. Putting together the two components (41) and (44) and employing tensor notation, we arrive at the formula

$$\begin{aligned}\vec{J}(\vec{r},t) &= \frac{1}{2V}\sum_{j=1}^{N}\sum_{\nu} {}_A\langle\varphi_{\bar{n}}(t)|\cos[\vec{k}_\nu\cdot(\vec{r}-\vec{\mathbf{r}}_j)]\left[\left(\vec{\varepsilon}_{\nu 1}\vec{\varepsilon}_{\nu 1}^{T}+\vec{\varepsilon}_{\nu 2}\vec{\varepsilon}_{\nu 2}^{T}+\vec{\kappa}_\nu\vec{\kappa}_\nu^{T}\right)\cdot\left(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\right)\right]|\varphi_{\bar{n}}(t)\rangle_A \\ &\quad+\frac{1}{2V}\sum_{j=1}^{N}\sum_{\nu} {}_A\langle\varphi_{\bar{n}}(t)|\left[\left(\vec{\varepsilon}_{\nu 1}\vec{\varepsilon}_{\nu 1}^{T}+\vec{\varepsilon}_{\nu 2}\vec{\varepsilon}_{\nu 2}^{T}+\vec{\kappa}_\nu\vec{\kappa}_\nu^{T}\right)\cdot\left(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\right)\right]\cos[\vec{k}_\nu\cdot(\vec{r}-\vec{\mathbf{r}}_j)]|\varphi_{\bar{n}}(t)\rangle_A \\ &= \frac{1}{2}\sum_{j=1}^{N} {}_A\langle\varphi_{\bar{n}}(t)|\left\{\frac{1}{V}\sum_{\nu}\cos[\vec{k}_\nu\cdot(\vec{r}-\vec{\mathbf{r}}_j)]\right\}\left(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\right)|\varphi_{\bar{n}}(t)\rangle_A \\ &\quad+\frac{1}{2}\sum_{j=1}^{N} {}_A\langle\varphi_{\bar{n}}(t)|\left(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\right)\left\{\frac{1}{V}\sum_{\nu}\cos[\vec{k}_\nu\cdot(\vec{r}-\vec{\mathbf{r}}_j)]\right\}|\varphi_{\bar{n}}(t)\rangle_A. \end{aligned} \tag{48}$$

Our last step consists in the replacement of the summation over the discrete modal index ν by an integration over a continuous vector variable $\vec{k}$. Equation (48) is transformed into

$$\begin{aligned}\vec{J}(\vec{r},t) &= \frac{1}{2}\sum_{j=1}^{N} {}_A\langle\varphi_{\bar{n}}(t)|\left\{\frac{1}{(2\pi)^3}\int_{\mathcal{R}^3}\cos[\vec{k}\cdot(\vec{r}-\vec{\mathbf{r}}_j)]d^3k\right\} \\ &\quad\times\left(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\right)|\varphi_{\bar{n}}(t)\rangle_A+\frac{1}{2}\sum_{j=1}^{N} {}_A\langle\varphi_{\bar{n}}(t)|\left(q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\right) \\ &\quad\times\left\{\frac{1}{(2\pi)^3}\int_{\mathcal{R}^3}\cos[\vec{k}\cdot(\vec{r}-\vec{\mathbf{r}}_j)]d^3k\right\}|\varphi_{\bar{n}}(t)\rangle_A; \end{aligned} \tag{49}$$

so that finally

$$\begin{aligned}\vec{J}(\vec{r},t) &= \frac{1}{2}\sum_{j=1}^{N} {}_A\langle\varphi_{\bar{n}}(t)|\left[\delta^3(\vec{r}-\vec{\mathbf{r}}_j)q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\right. \\ &\quad\left.+q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\delta^3(\vec{r}-\vec{\mathbf{r}}_j)\right]|\varphi_{\bar{n}}(t)\rangle_A. \end{aligned} \tag{50}$$

Symbol $\delta^3(\vec{r}-\vec{r}')$ stands here for the Dirac δ function.

The physical interpretation of quantity $\vec{J}(\vec{r},t)$ [i.e., the meaning of its transversal component $\vec{J}^{\perp}(\vec{r},t)$] can be now easily understood. Expression (50) is recognized as an expectation value of the semiclassical electric current operator

$$\vec{\mathbf{J}}^{\mathrm{sc}}(\vec{r},t) = \frac{1}{2}\sum_{j=1}^{N}\left[\delta^3(\vec{r}-\vec{\mathbf{r}}_j)q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)+q_j\vec{\mathbf{v}}_j^{\mathrm{sc}}(t)\delta^3(\vec{r}-\vec{\mathbf{r}}_j)\right], \tag{51}$$

over the given atomic state $|\varphi_{\bar{n}}(t)\rangle_A$.

Summarizing the contents of the present section, we may conclude that our previous considerations established a rigorous theoretical ground for the semiclassical treatment of "atom-laser" interaction, based upon solving the coupled system of Schrödinger plus Maxwell equations (15) and (47). We demonstrated that the mentioned semiclassical approach is equivalent to neglecting the terms (21) and (22) of the exact Hamiltonian (18).

It is important to emphasize in this context that the Schrödinger equation (15) for the dynamical state $|\varphi_{\bar{n}}(t)\rangle_A$ must be solved simultaneously with the just discussed wave equation (47) for the field (7), since the two problems are coupled. We recall for clarity that the c-number field (7) enters into the semiclassical Hamiltonian (16) of the Schrödinger equation (15), while the Maxwell equation (47) is parametrized by an expectation value (50) over the propagated atomic state vector $|\varphi_{\bar{n}}(t)\rangle_A$.

VII. APPLICABILITY AND LIMITATIONS OF THE SEMICLASSICAL APPROXIMATION

The physical correctness of the semiclassical approach stands and falls on the (in)adequacy of the previously made semiclassical approximations (a) and (b) represented by Eqs. (21) and (22). It is more than evident that a critical analysis of these approximative steps must be performed. Such an analysis will be developed in full detail elsewhere [23]. In the present section, we shall restrict ourselves to a brief inspection of a simplified but instructive model problem. (An additional and very explicit discussion of a much more concrete model system is elaborated in the Appendix.) From now on we shall suppose that:

(i) The potential function $V(\vec{r}^{\,N})$ of the Hamiltonian (16) does not allow the "atom" to ionize. That is, $V(\vec{r}^{\,N})$ corresponds to the infinite potential wells of a trap where the charged

particles are confined to move (like, e.g., electrons in a simple model of a quantum dot).

(ii) The incoming laser light (9) behaves as a monochromatic adiabatically switched continuous wave (CW) pulse with some prescribed wave vector $\vec{k}_L$ and the corresponding circular frequency $\omega_L = (2\pi/T) = c|k_L|$. Formulated mathematically, we shall take the c-number light term (9) of the form

$$\vec{A}_L^{\perp}(\vec{r}, t, \{\alpha_{\nu\wp}^{\mathrm{inc}}\}) = \eta(t)\vec{A}_L^0 \cos(\omega_L t - \vec{k}_L \cdot \vec{r}). \tag{52}$$

Here, $\eta(t)$ is a very slowly varying envelope of the pulse vanishing at infinite past and future, whereas $\vec{A}_L^0$ stands for a constant vector satisfying the transversality condition $\vec{A}_L^0 \cdot \vec{k}_L = 0$. Under the above circumstances (i) and (ii), the overall (incoming plus emitted) c-number field (7) resulting from the solution of the coupled problems (15) and (47) will behave as a superposition of adiabatically switched CW pulses whose frequencies are integer multiples of ω_L. That is,

$$\vec{A}_L^{\perp}(\vec{r}, t, \{\alpha_{\nu\wp}(t)\}) = \vec{A}_L^{\perp}(\vec{r}, t, \{\alpha_{\nu\wp}^{\mathrm{inc}}\}) + \sum_{a=1}^{\infty} \vec{A}_L^{(a)}(\vec{r}, t, \eta(t)); \tag{53}$$

where the ath order emission term is almost time periodic with the circular frequency $a\omega_L$, the time periodicity is slightly violated here due to the slowly varying envelope function $\eta(t)$. In other words,

$$\vec{A}_L^{(a)}(\vec{r}, t, \eta(t)) = \vec{A}_L^{(a)}(\vec{r}, t + T/a, \eta(t)). \tag{54}$$

Relations (53) and (54) are direct consequences of symmetry of the coupled Eqs. (15) and (47) with respect to time translations, combined with the near symmetry of an initial condition (52) with respect to specific time translations of the form $t \to t + T$. This near symmetry can be made arbitrarily exact by varying the envelope function $\eta(t)$ slowly enough.

An almost exact time periodicity of the c-number field (53) enables us to exploit the adiabatic Floquet theory [24] and to extract in this way a substantial amount of additional information concerning the solutions $\{|\varphi_n(t)\rangle_A\}$ of the Schrödinger equation (15). More explicitly, one finds that the dynamical atomic states $|\varphi_n(t)\rangle_A$ are expressible by the formula

$$|\varphi_n(t)\rangle_A = e^{-(i/\hbar)\int_{t_0}^{t} \mathcal{E}_n^{\mathrm{QE}}(\eta(\tau))d\tau} |\psi_n(t, \eta(t))\rangle_A. \tag{55}$$

Here, the so-called Floquet state $|\psi_n(t, \eta)\rangle_A$ and the so-called quasienergy $\mathcal{E}_n^{\mathrm{QE}}(\eta)$ depend parametrically upon the instantaneous value of the envelope function $\eta(t)$ and are obtained by solving the generalized eigenvalue problem

$$\left[\sum_{j=1}^{N} \frac{1}{2\mu_j}\left(\vec{\mathbf{p}}_j - \frac{q_j}{c}\vec{A}_L^{\perp}(\vec{\mathbf{r}}_j, t, \{\alpha_{\nu\wp}(t)\})\right)^2 + V(\vec{\mathbf{r}}^N) - i\hbar\frac{\partial}{\partial t}\right] \times |\psi_n(t, \eta)\rangle_A = \mathcal{E}_n^{\mathrm{QE}}(\eta)|\psi_n(t, \eta)\rangle_A; \tag{56}$$

with variable t playing the role of an additional coordinate running through an interval of one optical cycle. Consistently with such a viewpoint one imposes a periodic boundary condition

$$|\psi_n(t + T, \eta)\rangle_A = |\psi_n(t, \eta)\rangle_A. \tag{57}$$

In Eq. (56), the c-number field term corresponds to expression (53) evaluated at a fixed value of the envelope parameter η. We recall in this context that the envelope function remains almost constant on the time scale of one optical cycle. Different eigensolutions of the Floquet problem (56) are labeled by subscript n in such a way that $\mathcal{E}_n^{\mathrm{QE}}(\eta = 0)$ and $|\psi_n(t, \eta = 0)\rangle_A$ become indistinguishable from their zero-field counterparts $\mathcal{E}_n^0$ and $|\varphi_n^0(t)\rangle_A$ introduced before in Sec. IV. For $\eta \neq 0$, each Floquet state $|\psi_n(t, \eta)\rangle_A$ is expressible by the basis set expansion

$$|\psi_n(t, \eta)\rangle_A = \sum_{m'=-\infty}^{m'=+\infty} \sum_{n'} C_{m'n'}^n(\eta)\left|\varphi_{n'}^0\right\rangle_A e^{+im'\omega_L t}; \tag{58}$$

in consonance with the time periodicity requirement (57).

The expression (55) for the dressed atomic states $\{|\varphi_n(t)\rangle_A\}$ enables us to discuss an applicability of the semiclassical approximation by examining the relative importance of the as yet neglected off-diagonal contributions (21) to the exact Hamiltonian (18). The discussion of an extra diagonal term (22) is postponed to Ref. [23]. It is very instructive to inspect first the situation before the arrival of the laser pulse, that is, the infinite past limit $\lim_{t\to-\infty} \tilde{\mathbf{H}}(t) = \mathbf{H}$ where $\mathbf{H}$ corresponds to Eq. (1) and the quantum state of the system is identified with the ground state of $\mathbf{H}$. An eigenproblem of $\mathbf{H}$ is encountered in numerous basic applications dealing with a small but measurable effect of the quantum vacuum on atomic or molecular species. Mentioned applications include e.g. the calculation of the the nonrelativistic Lamb shift, the evaluation of the spontaneous emission rates, or the studies of the Casimir forces (van der Waals interactions) between atoms, see Ref. [19] for more details. In all of the previous examples, as well as in the present case of our interest, the atomic wave function remains relatively well separable from the field degrees of freedom even after the effect of vacuum fluctuations is taken into account. This shows that the semiclassical approximation (29) is well justified for $t \to -\infty$. Nevertheless, the mere validity of the semiclassical approach does not imply that the off-diagonal matrix elements (21) are here smaller in magnitude than their diagonal counterparts. The true justification for neglecting these off-diagonal terms consists in the fact that each element (21) carries an oscillating phase factor $e^{+(i/\hbar)(\mathcal{E}_n^0 - \mathcal{E}_{n'}^0)(t-t_0)}$. Provided that the energy separations $|\mathcal{E}_n^0 - \mathcal{E}_{n'}^0|$ between the involved atomic energy levels are large enough, the phase oscillations of $e^{+(i/\hbar)(\mathcal{E}_n^0 - \mathcal{E}_{n'}^0)(t-t_0)}$ are so rapid that the associated Hamiltonian terms (21) become dynamically irrelevant and thus negligible to a good approximation. Making an intermediate summary, one may state that an ansatz $|\varphi_n^0\rangle_A|\{0\}\rangle_R$ represents a good approximation to a stationary state of the full Hamiltonian $\mathbf{H}$ whenever all the relevant energy gaps $|\mathcal{E}_n^0 - \mathcal{E}_{\bar{n}}^0|$ are large enough compared to the energy contents of the quantum vacuum fluctuations.

Importantly, an analysis of the exact Hamiltonian (18) at finite times (when the laser has arrived and interacts with the atom) turns out to be entirely analogical to the considerations of the previous paragraph. Again, the off-diagonal Hamiltonian terms (21) are not smaller in magnitude than their diagonal counterparts, but they carry an oscillating phase

factor

$$e^{+(i/\hbar)\int_{t_0}^{t}\{\mathcal{E}_n^{\mathrm{QE}}(\eta(\tau))-\mathcal{E}_{n'}^{\mathrm{QE}}(\eta(\tau))\}d\tau}; \tag{59}$$

as follows directly from the adiabatic Floquet formula (55) for the dressed atomic states $\{|\varphi_n(t)\rangle_A\}$. Provided that the quasienergy separations $|\mathcal{E}_n^{\mathrm{QE}}(\eta(t)) - \mathcal{E}_{\bar{n}}^{\mathrm{QE}}(\eta(t))|$ are large enough throughout the entire time interval of the interaction between the c-number field (7) and the atom, the Floquet phase oscillations (59) are so rapid that the associated Hamiltonian terms (21) become dynamically irrelevant and thus negligible to a good approximation. Under such circumstances, the semiclassical ansatz (29) appears to be well theoretically justified. One may thus state that the semiclassical treatment represents a good approximation to the exact quantum dynamics governed by the full Hamiltonian (18) as long as all the relevant quasienergy gaps $|\mathcal{E}_n^{\mathrm{QE}}(\eta(t)) - \mathcal{E}_{\bar{n}}^{\mathrm{QE}}(\eta(t))|$ are large enough compared to the energy contents of the quantum vacuum fluctuations.

On the other hand, it is well known from numerical calculations (even for open systems allowing field induced ionization, see Refs. [25–27]) that the frequency and intensity of the incoming laser field can be tuned in such a way as to make certain quasienergies accidentally near degenerate. When some quasienergy gap $|\mathcal{E}_n^{\mathrm{QE}}(\eta(t)) - \mathcal{E}_{\bar{n}}^{\mathrm{QE}}(\eta(t))|$ is not sufficiently large or even zero, the phase factor (59) does not oscillate rapidly enough and there turns out to be no reason to neglect the associated off-diagonal Hamiltonian terms (21). Hence, a single channel direct product ansatz of the form (29) no longer constitutes an adequate approximation for the exact dynamical quantum state of a strongly entangled system "atom plus quantized radiation field." *Under these conditions one should expect a complete breakdown of the semiclassical theory.*

In the case when the semiclassical approach is justified, one possible strategy for correcting systematically the solution (29) can be based upon the perturbation method, to be implemented in a similar way as in conventional studies of quantum vacuum effects on atoms in the absence of laser light. An application of the perturbation technique enables us also to derive a rigorous quantitative criterion (to be published elsewhere in Ref. [23]), which shows how to recognize whether or not the quasienergy separations $|\mathcal{E}_n^{\mathrm{QE}}(\eta(t)) - \mathcal{E}_{\bar{n}}^{\mathrm{QE}}(\eta(t))|$ are sufficiently large enough to justify the semiclassical treatment. In the case when the semiclassical approximation breaks down completely as being anticipated within the previous paragraph, some variant of nonperturbative quantum electrodynamics treatment must be adopted to properly describe the problem. The development of such a nonperturbative mode of description is currently in progress.

Importantly, the qualitative considerations of the present section can be supported by an explicit QED treatment of a simple model of a two-level atom driven by a laser. The solution of this problem is elaborated in detail within the Appendix. The derivation presented there is based upon mathematically well-defined and physically well-justifiable approximations and leads finally to a simple formula (A44), which serves as a quantitative indicator of (in)adequacy of the semiclassical approach. This formula demonstrates explicitly that the quasienergy separations $|\mathcal{E}_n^{\mathrm{QE}}(\eta(t)) - \mathcal{E}_{\bar{n}}^{\mathrm{QE}}(\eta(t))|$ referred to previously represent indeed the crucial criterion according to which the validity of the semiclassical theory should be judged.

Summarizing the most important finding of the present section, we may conclude that we predicted a complete breakdown of the conventional "quantized atom-classical light" treatment of the matter-light interaction problem under special but well-defined and achievable conditions. The breakdown of the semiclassical theory is attributed to accidental (near) degeneracies in the energy spectrum of the atomic system dressed by the laser. Failure of the semiclassical treatment arises here due to an unhindered transfer of quantum population between the (near) degenerate atomic channels and suggests a possible encounter with new, as yet unobserved, physical phenomena observable in harmonic generation spectra. It is clear at least that the harmonic generation spectrum calculated using the semiclassical approach will not agree in this case with the QED result obtained using the exact dynamical solution of the full Schrödinger equation (20).

In passing we note that an experimental measurement and control of the relevant quasienergy gaps might be achieved using the technique of Floquet spectroscopy (see Refs. [28, 29]). More explicitly, by carrying out Floquet spectroscopy measurements on atoms strongly driven by intense light one may adjust the frequency and intensity of the driving intense laser in such a way as to produce the desired quasienergy (near) degeneracy. Consequently, the corresponding harmonic generation spectrum is anticipated to differ strongly from the prediction of semiclassical (SC) theory (see also the last paragraph of Sec. A.5 for further comments).

VIII. CONCLUDING REMARKS

The main purpose of the present work is to examine the validity of the semiclassical treatment of "atom-laser" interaction, that is, to derive the corresponding semiclassical system of coupled Maxwell-Schrödinger equations from the first principles of quantum electrodynamics along with the critical examination of the validity of such an approach. We believe that this program was successfully accomplished. More explicitly, we arrived at two basic results:

(1) We show that the semiclassical set of coupled Maxwell-Schrödinger equations results from the developed formalism as a well-defined approximation, obtained by neglecting certain terms of the exact Hamiltonian.

(2) Assuming a special arrangement when the model atomic system is spatially confined and the incoming laser light corresponds to an adiabatically switched monochromatic CW pulse, we find a simple criterion of the validity of the semiclassical approach. We also demonstrate, on the basis of the derived criterion, that the semiclassical treatment does break down completely in some special but achievable situations. This becomes apparent most explicitly from the formula (A44) derived in the Appendix for a simple two-level model. The possibility of observing new physical phenomena emerges whenever the breakdown of the semiclassical theory takes place.

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APPENDIX: GOING BEYOND THE SC APPROACH FOR A SIMPLE TWO LEVEL ATOMIC SYSTEM

The purpose of this Appendix is to analyze more explicitly the limitations of the SC approximation, using a simple model problem of a two-level atom. While a two-level atomic system represents *per se* a legitimate and frequently considered model problem from a theoretical standpoint, one can also find realistic physical systems for which the two-level approximation is adequate. For example, the lowest two eigenstates of a double well potential (which has a high enough barrier between its two minima) are energetically very close to each other but stay separated from all the other states by a much larger energy gap. Two-level atoms strongly driven by an intense laser field were used by several authors to study harmonic generation and other phenomena, see, e.g., Refs. [30,31].

1. Basic definitions

We shall consider a two-level atomic system coupled to a quantum electromagnetic field. The total Hamiltonian takes the form

$$\mathbf{H} = \mathbf{H}_A + \mathbf{H}_R + \mathbf{H}_{SR}(t). \tag{A1}$$

Here,

$$\mathbf{H}_A = |1\rangle_A \mathcal{E}^1_{A\,A}\langle 1| + |2\rangle_A \mathcal{E}^2_{A\,A}\langle 2|, \tag{A2}$$

represents the Hamiltonian of a free two-level atom, whereas $\mathbf{H}_R = \sum_\nu \hbar\omega_\nu \mathbf{a}^\dagger_\nu \mathbf{a}_\nu$ stands for the Hamiltonian of a free electromagnetic field. For the sake of conceptual simplicity we shall assume here that the field modes can be labeled just by a one-dimensional wave vector k_ν, although such an assumption is not essential for our forthcoming derivations. The corresponding electric and magnetic field operators are defined on one-dimensional position space by the formulas

$$\mathbf{E}(x) = i\sum_\nu \sqrt{\frac{\hbar\omega_\nu}{4L}} e^{+ik_\nu x}\mathbf{a}_\nu + \text{c.c.}, \tag{A3}$$

$$\mathbf{B}(x) = i\sum_\nu ck_\nu \sqrt{\frac{\hbar}{4\omega_\nu L}} e^{+ik_\nu x}\mathbf{a}_\nu + \text{c.c.}, \tag{A4}$$

and determine the field Hamiltonian as $\mathbf{H}_R = \int_{-L/2}^{+L/2}[\mathbf{E}^2(x) + \mathbf{B}^2(x)]dx$, with an implicit omission of the zero point energy term. Symbol L is, of course, understood as the box quantization length giving the modal periodic boundary conditions. Coupling between the two-level atom and the quantum field is within the present model defined via the length gauge prescription

$$\mathbf{H}_{SR}(t) = \int_{-L/2}^{+L/2} \mathbf{d}(x,t)\mathbf{E}(x)\,dx. \tag{A5}$$

The atomic dipole function $\mathbf{d}(x,t)$ used here is for later convenience assumed to vary adiabatically in time, such that the atom and quantum field become uncoupled in the infinite past limit. More explicitly, we shall take

$$\mathbf{d}(x,t) = f(t)\sqrt{\zeta/\pi}e^{-\zeta x^2}\mathbf{w},\ \mathbf{w} = |1\rangle_{A\,A}\langle 2| + |2\rangle_{A\,A}\langle 1|; \tag{A6}$$

with $f(t)$ being an auxiliary adiabatic switching function which vanishes as $t \to -\infty$ and reaches a constant nonzero value f_a before the arrival of the laser pulse. As a concrete example one may take $f(t) = f_a e^{-\varepsilon|t|}$ where $\varepsilon \to +0$. The meaning of the parameter ζ is as follows. In the limit of large ζ one will recover from Eq. (A5) the usual dipole approximation formula. Nevertheless, we prefer to choose ζ in Eq. (A6) in such a way that the spatial extension of $\mathbf{d}(x,t)$ is finite. This introduces an effective cutoff of very high modal frequencies ω_ν and allows us therefore to avoid delving into the matters of renormalization in our subsequent elaborations. Closely related is an observation that a finite value of ζ implies a smooth regular source term in the Maxwell wave equation, to be seen later.

2. Canonical transformations and the SC approach

Let us apply now the general theoretical formalism developed in the main part of the article. First, the displacement operator $\mathbf{D}(\{\alpha_\nu(t)e^{-i\omega_\nu t}\})$ and the free field time evolution operator transform the total Hamiltonian (A1) into

$$\mathbf{H}(t) = \mathbf{H}_A + \int_{-L/2}^{+L/2} \mathbf{d}(x,t)E_L(x,t)dx + \int_{-L/2}^{+L/2} \mathbf{d}(x,t) \times \mathbf{E}(x,t)\,dx - i\hbar\sum_\nu (\dot\alpha_\nu(t)\mathbf{a}^\dagger_\nu - \dot\alpha^*_\nu(t)\mathbf{a}_\nu). \tag{A7}$$

Here

$$E_L(x,t) = i\sum_\nu \sqrt{\frac{\hbar\omega_\nu}{4L}} e^{-i(\omega_\nu t - k_\nu x)}\alpha_\nu(t) + \text{c.c.}, \tag{A8}$$

is a c-number field characterized by its as yet unspecified amplitudes $\{\alpha_\nu(t)e^{-i\omega_\nu t}\}$ and $\mathbf{E}(x,t)$ stands, of course, for an interaction picture counterpart of Eq. (A3). Second, we define the Hamiltonian of the SC approach,

$$\mathbf{H}^{\text{sc}}_A(t) = \mathbf{H}_A + \int_{-L/2}^{+L/2} \mathbf{d}(x,t)E_L(x,t)dx; \tag{A9}$$

and solve the associated problem of a laser-driven two-level atom

$$i\hbar\frac{d}{dt}|\varphi_n(t)\rangle_A = \mathbf{H}^{\text{sc}}_A(t)|\varphi_n(t)\rangle_A,\ |\varphi_n(t_{\text{init}})\rangle_A = |n\rangle_A. \tag{A10}$$

Consequently, we eliminate the $\mathbf{H}^{\text{sc}}_A(t)$ contribution from Eq. (A7) by a canonical transformation (see Sec. IV of the

main text), to get

$$\widetilde{\mathbf{H}}(t) = \int_{-L/2}^{+L/2} \mathbf{d}_{\mathrm{I}}(x,t)\mathbf{E}(x,t)\,dx - i\hbar \sum_{\nu} \left(\dot{\alpha}_{\nu}(t)\mathbf{a}_{\nu}^{\dagger} - \dot{\alpha}_{\nu}^{*}(t)\mathbf{a}_{\nu}\right). \tag{A11}$$

The relevant dipole term is now defined through its matrix elements

$$\begin{aligned} {}_A\langle n|\mathbf{d}_{\mathrm{I}}(x,t)|n'\rangle_A &= f(t)\sqrt{\zeta/\pi}e^{-\zeta x^2} w_{nn'}(t), \\ w_{nn'}(t) &= {}_A\langle \varphi_n(t)|\mathbf{w}|\varphi_{n'}(t)\rangle_A. \end{aligned} \tag{A12}$$

Note that besides an obvious property $w_{12}(t) = w_{21}^{*}(t)$ we have also $w_{11}(t) = -w_{22}(t)$ for the case of a two-level atom. Finally, the as yet unspecified complex amplitudes $\alpha_{\nu}(t)$ are chosen to evolve in time according to

$$\begin{aligned} i\hbar\dot{\alpha}_{\nu}(t) &= -L^{-1/2} i\sqrt{\hbar\omega_{\nu}/4} D_{\nu}^{-} e^{+i\omega_{\nu}t} f(t) w_{11}(t), \\ \lim_{t\to-\infty} \alpha_{\nu}(t) &= \alpha_{\nu}^{\mathrm{inc}}; \end{aligned} \tag{A13}$$

where

$$D_{\nu}^{\pm} = \int_{-L/2}^{+L/2} \sqrt{\zeta/\pi} e^{-\zeta x^2} e^{\pm i k_{\nu} x} dx = e^{-k^2/(4\zeta)}. \tag{A14}$$

If so, manipulations completely analogical to those of Sec. VI of the main text show that the c-number field (A8) satisfies the Maxwell wave equation of the SC approach, namely,

$$\frac{\partial^2}{\partial x^2} E_L(x,t) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2} E_L(x,t) = -\frac{f(t) w_{11}(t)}{2} \frac{\partial^2}{\partial x^2}\left(\sqrt{\zeta/\pi} e^{-\zeta x^2}\right). \tag{A15}$$

Moreover, the transformed Hamiltonian (A11) can be now cast into a simple matrix form

$$\widetilde{\mathbf{H}}(t) = \begin{pmatrix} \mathbf{0} & \mathbf{h}_{12}(t) \\ \mathbf{h}_{21}(t) & \mathbf{h}_{22}(t) \end{pmatrix}; \tag{A16}$$

with the matrix elements

$$\mathbf{h}_{n_1 n_2}(t) = L^{-1/2} \sum_{\nu} \left(h_{n_1 n_2}^{(+)\nu}(t)\mathbf{a}_{\nu}^{\dagger} + h_{n_1 n_2}^{(-)\nu}(t)\mathbf{a}_{\nu}\right), \tag{A17}$$

being explicitly determined through the coefficients

$$h_{12}^{(\pm)\nu}(t) = \mp 1 i\sqrt{\hbar\omega_{\nu}/4} D_{\nu}^{\mp} e^{\pm i\omega_{\nu}t} f(t) w_{12}(t) = h_{21}^{(\mp)\nu *}(t), \tag{A18}$$

$$h_{22}^{(\pm)\nu}(t) = \pm 2 i\sqrt{\hbar\omega_{\nu}/4} D_{\nu}^{\mp} e^{\pm i\omega_{\nu}t} f(t) w_{11}(t) = h_{22}^{(\mp)\nu *}(t). \tag{A19}$$

The previously used row and column labels $n_{1,2}$ refer, of course, to the previously constructed atomic basis functions $|\varphi_{n_{1,2}}(t)\rangle_A$.

3. Going beyond the SC approach

Within the theoretical framework introduced in A1 and A2, exact quantum dynamics of the studied model system (two-level atom coupled to quantum electromagnetic field) is described by a coupled channel problem

$$i\hbar\frac{d}{dt}\begin{pmatrix} |\tilde{\Psi}_1(t)\rangle_R \\ |\tilde{\Psi}_2(t)\rangle_R \end{pmatrix} = \begin{pmatrix} \mathbf{0} & \mathbf{h}_{12}(t) \\ \mathbf{h}_{21}(t) & \mathbf{h}_{22}(t) \end{pmatrix} \begin{pmatrix} |\tilde{\Psi}_1(t)\rangle_R \\ |\tilde{\Psi}_2(t)\rangle_R \end{pmatrix}. \tag{A20}$$

Since the adiabatic switching function $f(t)$ is supposed to vanish in the infinite past, the evolution (A20) becomes trivial for $t \to -\infty$. An initial condition of our interest corresponds to the atom prepared in its ground state in the infinite past. That is,

$$\begin{pmatrix} |\tilde{\Psi}_1(t_{\mathrm{init}})\rangle_R \\ |\tilde{\Psi}_2(t_{\mathrm{init}})\rangle_R \end{pmatrix} = \begin{pmatrix} |\{0\}\rangle_R \\ |\emptyset\rangle_R \end{pmatrix}, \quad t_{\mathrm{init}} \to -\infty; \tag{A21}$$

where $|\emptyset\rangle_R$ denotes a zero vector. Even for finite values of t, the SC approximation of the sought dynamical quantum state vector takes a simple form

$$\begin{pmatrix} |\tilde{\Psi}_1(t)\rangle_R \\ |\tilde{\Psi}_2(t)\rangle_R \end{pmatrix} = \begin{pmatrix} |\{0\}\rangle_R \\ |\emptyset\rangle_R \end{pmatrix}. \tag{A22}$$

The purpose of this Appendix is to critically examine an (in) accuracy of the SC approximation. Clearly, the most satisfactory way to facilitate such an investigation will be to solve directly the initial value problems (A20) and (A21), and to compare the outcome with the approximative solution (A22). However, the only known route to obtain an explicit solution of Eqs. (A20) and (A21) is via carrying out a large-scale numerical calculation. Fortunately, in spite of this fact, it turns out that for the present simple model an important amount of physical insight can be gained using analytic methods and well-justified approximations, as elaborated in detail in the following.

Without any limitations of generality let us express the first component $|\tilde{\Psi}_1(t)\rangle_R$ as

$$|\tilde{\Psi}_1(t)\rangle_R = C(t)|\psi_1(t)\rangle_R. \tag{A23}$$

Here, the vector $|\psi_1(t)\rangle_R$ is required to satisfy the conditions

$${}_R\langle\psi_1(t_{\mathrm{init}})|\psi_1(t_{\mathrm{init}})\rangle_R = 1, \quad {}_R\langle\psi_1(t)|(d/dt)|\psi_1(t)\rangle_R = 0; \tag{A24}$$

meaning that its unit norm and its overall phase are held fixed in time. If so, then $|C(t)|^2$ is interpreted as the population of the first channel. The second component $|\tilde{\Psi}_2(t)\rangle_R$ is left as it stands. It is a simple matter to write down the equations of motion for $C(t)$, $|\psi_1(t)\rangle_R$, and $|\tilde{\Psi}_2(t)\rangle_R$. One has

$$i\hbar\frac{d}{dt}C(t) = {}_R\langle\psi_1(t)|\mathbf{h}_{12}(t)|\tilde{\Psi}_2(t)\rangle_R, \tag{A25}$$

$$i\hbar C(t)\frac{d}{dt}|\psi_1(t)\rangle_R = \mathbf{h}_{12}(t)|\tilde{\Psi}_2(t)\rangle_R - {}_R\langle\psi_1(t)|\mathbf{h}_{12}(t)|\tilde{\Psi}_2(t)\rangle_R|\psi_1(t)\rangle_R, \tag{A26}$$

$$i\hbar\frac{d}{dt}|\tilde{\Psi}_2(t)\rangle_R = \mathbf{h}_{22}(t)|\tilde{\Psi}_2(t)\rangle_R + C(t)\mathbf{h}_{21}(t)|\psi_1(t)\rangle_R. \tag{A27}$$

Note that Eq. (A26) conserves automatically the properties in Eq. (A24).

Assume now that we do not solve Eq. (A26) coupled to Eqs. (A25) and (A27), but instead we prescribe $|\psi_1(t)\rangle_R$ externally in some way, while keeping in mind that the requirements (A24) must be maintained. Suppose then that one propagates $C(t)$ and $|\tilde{\Psi}_2(t)\rangle_R$ according to the coupled differential Eqs. (A25) and (A27), which contain the just mentioned external choice of $|\psi_1(t)\rangle_R$. Importantly, it turns out that even under such circumstances the norm of the corresponding dynamical state vector remains unity at all times, meaning that

the population of the first channel is still $|C(t)|^2$. Indeed, let us take the self-overlap $S(t) = C^*(t)C(t)_R\langle\psi_1(t)|\psi_1(t)\rangle_R + R\langle\tilde{\Psi}_2(t)|\tilde{\Psi}_2(t)\rangle_R$ and calculate its time derivative. It holds

$$\begin{aligned}\dot{S}(t) &= \dot{C}^*(t)C(t)_R\langle\psi_1(t)|\psi_1(t)\rangle_R + C^*(t)\dot{C}(t)_R\langle\psi_1(t)|\psi_1(t)\rangle_R \\ &\quad + C^*(t)C(t)_R\langle\dot{\psi}_1(t)|\psi_1(t)\rangle_R + C^*(t)C(t)_R\langle\psi_1(t)|\dot{\psi}_1(t)\rangle_R \\ &\quad + {}_R\langle\dot{\tilde{\Psi}}_2(t)|\tilde{\Psi}_2(t)\rangle_R + {}_R\langle\tilde{\Psi}_2(t)|\dot{\tilde{\Psi}}_2(t)\rangle_R. \end{aligned} \tag{A28}$$

The second line of Eq. (28) vanishes due to property (A24), whereas the remaining terms cancel each other after a substitution is made from Eqs. (A25) and (A27) for the involved time derivatives. Note in passing that the issue of initial conditions is completely immaterial here.

From now on we shall adopt the following strategy. We shall identify $|\psi_1(t)\rangle_R$ with the vacuum state $|\{0\}\rangle_R$ [this choice satisfies automatically the conditions (A24)] and search for $C(t)$ and $|\tilde{\Psi}_2(t)\rangle_R$ under such a restriction. In other words, we shall approximate the exact solution of the initial value problems (A20) and (A21) by an ansatz

$$\begin{pmatrix} |\tilde{\Psi}_1(t)\rangle_R \\ |\tilde{\Psi}_2(t)\rangle_R \end{pmatrix} = \begin{pmatrix} C(t)|\{0\}\rangle_R \\ |\tilde{\Psi}_2(t)\rangle_R \end{pmatrix}. \tag{A29}$$

Here, the time evolution of $C(t)$ and $|\tilde{\Psi}_2(t)\rangle_R$ is governed by coupled Eqs. (A25) and (A27), with the initial conditions

$$C(t_{\text{init}}) = 1, \quad |\tilde{\Psi}_2(t_{\text{init}})\rangle_R = |\emptyset\rangle_R. \tag{A30}$$

As already discussed, the approximative solution (A29) remains unit normalized in the course of time. It is not hard to trace out our motivation behind an ansatz (A29). As long as the much more crude SC approximation (A22) is valid, our solution of the problems (A25), (A27), and (A30) must provide $C(t) \approx 1$ and $|\tilde{\Psi}_2(t)\rangle_R \approx |\emptyset\rangle_R$. The breakdown of the SC approximation takes place whenever this ceases to be true. Hence, *an ansatz* (A29) *enables us to strictly falsify the SC approach in a nonperturbative manner.* Note that we do not claim that our aforementioned ansatz (A29) always represents a good approximation to the exact solution in the case when the SC approach proves to be inadequate. We do claim, however, that an observation of $C(t) \not\approx 1$ and $|\tilde{\Psi}_2(t)\rangle_R \not\approx |\emptyset\rangle_R$ implies the breakdown of the SC approximation. In passing we note that, as long as the SC approach is approximately valid, an identification $|\psi_1(t)\rangle_R = |\{0\}\rangle_R$ represents the leading order of the exact solution $|\psi_1(t)\rangle_R$. Using this leading-order approximation in an ansatz (A29) one should be able to approximate well the leading order of the exact solution $|\tilde{\Psi}_2(t)\rangle_R$. The c-number factor $C(t)$ must be, in any approximative treatment, necessarily time propagated as well to account for the overall norm conservation and for the dynamical phase. These considerations show that our ansatz (A29) is rather well physically motivated.

After an ansatz (A29) is combined with the time evolution Eqs. (A25) and (A27) one gets

$$i\hbar\frac{d}{dt}C(t) = {}_R\langle\{0\}|\mathbf{h}_{12}^{(-)}(t)|\tilde{\Psi}_2(t)\rangle_R, \tag{A31}$$

$$i\hbar\frac{d}{dt}|\tilde{\Psi}_2(t)\rangle_R = \mathbf{h}_{22}(t)|\tilde{\Psi}_2(t)\rangle_R + C(t)\mathbf{h}_{21}^{(+)}(t)|\{0\}\rangle_R, \tag{A32}$$

where $\mathbf{h}_{12}^{(-)}(t) = L^{-1/2}\sum_\nu h_{12}^{(-)\nu}(t)\mathbf{a}_\nu = \mathbf{h}_{21}^{(+)\dagger}(t)$. Let us consider now separately Eq. (A32). It turns out that an explicit closed-form solution to this equation can be worked out. Namely, the $\mathbf{h}_{22}(t)$ term can be eliminated using the displacement operator method (in a similar fashion as being done in Sec. III and in A.2) and the remaining differential equation can be easily integrated. After straightforward manipulations, one arrives at the result

$$\begin{aligned}|\tilde{\Psi}_2(t)\rangle_R = -\frac{i}{\hbar}\int_{t_{\text{init}}}^{t} d\tau\, e^{+(i/\hbar)(F(\tau)-F(t))}C(\tau) \\ \times e^{-i\sum_{\nu''}\text{Im}(\beta_{\nu''}(t)\beta^*_{\nu''}(\tau))}L^{-1/2}\sum_\nu h_{21}^{(+)\nu}(\tau) \\ \times(\mathbf{a}_\nu^\dagger - \beta_\nu^*(t) + \beta_\nu^*(\tau))|\{\beta_{\nu'}(t) - \beta_{\nu'}(\tau)\}\rangle_R,\end{aligned} \tag{A33}$$

where the coherent-state amplitudes $\beta_\nu(t) = L^{-1/2}b_\nu(t) = -2(\alpha_\nu(t) - \alpha_\nu^{\text{inc}})$, the function $F(t) = \frac{1}{2}\int_{t_{\text{init}}}^{t}\tilde{h}_{22}(\tau)d\tau$, and $\tilde{h}_{22}(t) = L^{-1/2}\sum_\nu(h_{22}^{(+)\nu}(t)\beta_\nu^*(t) + h_{22}^{(-)\nu}(t)\beta_\nu(t))$. Formula (A33) gives $|\tilde{\Psi}_2(t)\rangle_R$ in terms of the still unknown c-number function $\{C(\tau), t_{\text{init}} \leqslant \tau < t\}$. By substituting Eq. (A33) into Eq. (31) one obtains, after some simple rearrangements, a self-contained time evolution equation for $C(t)$. It holds

$$i\hbar\frac{d}{dt}C(t) = \int_{t_{\text{init}}}^{t} K(t,\tau)C(\tau)d\tau, \tag{A34}$$

where the corresponding integration kernel can be written as follows

$$K(t,\tau) = \ell(t,\tau)\mathcal{H}(t,\tau). \tag{A35}$$

The two constituents of $K(t,\tau)$ are given by explicit formulas

$$\begin{aligned}\ell(t,\tau) &= \frac{i}{\hbar}e^{+(i/\hbar)(F(\tau)-F(t))} \\ &\quad\times\exp\left\{-\frac{1}{2}L^{-1}\sum_\nu(|b_\nu(t)|^2 + |b_\nu(\tau)|^2 - 2b_\nu^*(t)b_\nu(\tau))\right\} \\ &= \ell^*(\tau,t),\end{aligned} \tag{A36}$$

$$\begin{aligned}\mathcal{H}(t,\tau) = \left(L^{-1}\sum_{\nu_1}h_{12}^{(-)\nu_1}(t)(b_{\nu_1}(t) - b_{\nu_1}(\tau))\right) \\ \times\left(L^{-1}\sum_{\nu_2}h_{21}^{(+)\nu_2}(\tau)(b_{\nu_2}^*(t) - b_{\nu_2}^*(\tau))\right) \\ - L^{-1}\sum_\nu h_{12}^{(-)\nu}(t)h_{21}^{(+)\nu}(\tau) = \mathcal{H}^*(\tau,t),\end{aligned} \tag{A37}$$

and possess a well-defined continuum mode limit.

Importantly, an integro-differential Eq. (A34) for a single c-number function $C(t)$ lends itself much better to numerical investigations than the original coupled-channel problem (A20), which deals with an infinite number of degrees of freedom of the quantum field. In other words, our ansatz (A29) enables one to go beyond the SC approach in an approximative yet physically well-motivated fashion, while offering a tremendous computational advantage.

4. An explicit criterion of validity of the SC approach

In the present text we shall not pursue the numerical solution of Eq. (A34). Instead, we prefer to simplify the problem even further to be able to obtain physical insights

in an analytic yet well-defined approximative manner. Our first simplification is based upon a very realistic assumption that the coupling between an atom and the radiation field (as determined by the strength parameter f_a, see A.1) is weak in magnitude. This of course does not prevent the strong driving of an atom by an intense enough laser field. Provided that the value of f_a is small, then also the c-number radiation emitted from the atom is very weak in magnitude, as follows from the Maxwell wave equation (A15). Now, observe that the exponential factors in $\ell(t,\tau)$ are proportional to the amount of emitted radiation since $b_\nu(t)$ are proportional to f_a. Hence, for the hereafter considered case of weak emission, the exponentials involved in $\ell(t,\tau)$ can be ignored, to give an approximation $\ell(t,\tau)=(i/\hbar)$. Moreover, the first line of Eq. (A37) is proportional to $b_\nu^2(t)f_a^2$, contrary to the second line, which is proportional only to f_a^2. This means that the first line of Eq. (A37) can be again neglected as a well-defined approximation. Having implemented these simplifications, the kernel (A35) boils down into

$$K(t,\tau)=-\frac{i}{\hbar}L^{-1}\sum_\nu h_{12}^{(-)\nu}(t)h_{21}^{(+)\nu}(\tau)$$
$$=\frac{(-i)}{8\pi}f(t)f(\tau)w_{12}(t)w_{12}^*(\tau)I(\tau-t), \qquad \text{(A38)}$$

where

$$I(t)=\int_{-\infty}^{+\infty}\omega_k|D_k^+|^2e^{+i\omega_k t}dk. \qquad \text{(A39)}$$

Note that the above-made simplification has completely erased the effect of $\mathbf{h}_{22}(t)$ on $C(t)$. This is not so surprising since the major players changing the population of the first channel are expected to be the off-diagonal matrix elements $\mathbf{h}_{12}(t)$ and $\mathbf{h}_{21}(t)$.

So far we did not impose any restrictions regarding the shape of the incoming c-number light pulse. Hereafter we shall consider the situation when the atom interacts with a CW-like laser pulse whose central frequency is ω_L. If so, the factor $w_{12}(t)$ can be conveniently expressed using the formalism of the adiabatic Floquet theory [24], which was discussed in Sec. VII of the main text. Namely,

$$w_{12}(t)=e^{+(i/\hbar)\int_{t_{\rm init}}^t\left(\mathcal{E}_1^{\rm QE}(\eta(t'))-\mathcal{E}_2^{\rm QE}(\eta(t'))\right)dt'}$$
$$\times\sum_{m=-\infty}^{m=+\infty}w_{12}^m(\eta(t))e^{+im\omega_L t}. \qquad \text{(A40)}$$

Here, as we recall, $\eta(t)$ characterizes the adiabatically varying laser envelope and $\mathcal{E}_{1,2}^{\rm QE}(\eta(t))$ are the Floquet quasienergies pertaining, respectively, to the two atomic dressed states. To avoid confusion during our forthcoming argumentations, one additional insight is worth noting in this context: Under well-defined conditions, the adiabatic Floquet formulas (55), (58), and (A40) are applicable without change even for the case when the quasienergies $\mathcal{E}_{1,2}^{\rm QE}(\eta(t))$ come close to each other, or even pass through a degeneracy point, $\mathcal{E}_1^{\rm QE}(\eta_{\rm deg})=\mathcal{E}_2^{\rm QE}(\eta_{\rm deg})$, at some time instant during the action of the light pulse. Let us explain this claim more explicitly. The adiabatic theorem for Floquet states [24] ensures that one can get to an arbitrarily close proximity $\eta_{\rm deg}-\Delta\eta$ of $\eta_{\rm deg}$ by varying the laser envelope $\eta(t)$ slowly enough. On the other hand, the corresponding Floquet wave functions $|\psi_{1,2}(t,\eta)\rangle_A$ depend continuously upon η even in the vicinity of $\eta_{\rm deg}$ and remain always mutually orthonormal. As long as both wave functions $|\psi_{1,2}(t,\eta)\rangle_A$ remain almost constant throughout the small interval $\eta\in(\eta_{\rm deg}-\Delta\eta,\eta_{\rm deg}+\Delta\eta)$, a sudden yet small change from $\eta_{\rm deg}-\Delta\eta$ to $\eta_{\rm deg}$ (or even to $\eta_{\rm deg}+\Delta\eta$) will not induce any nonadiabatic transitions. Hence, an applicability of the formulas (55), (58), and (A40) is not compromised for the just-described arrangement of the light pulse.

Expressions (A38) through (A40) represent certainly an additional substantial simplification of the studied problem. Nevertheless, the associated time evolution Eq. (A34) still does not seem to admit a simple analytic solution. We resort therefore to another (and last) approximative step, based upon the following observation. Even when atoms are exposed to very intense laser fields capable of manipulating significantly with the quasienergies, it often happens that the wave functions of the associated Floquet states are not much different from their field-free counterparts. In such a case (which occurs also for our numerical example, to be seen later in A.6), the zeroth Fourier component $w_{12}^0(\eta(t))$ in Eq. (A40) will be much larger (and close to unity) than all the other $m\neq 0$ components $w_{12}^m(\eta(t))$. Under these circumstances one may simplify the kernel (A38) into

$$K(t,\tau)=\frac{(-i)}{8\pi}f(t)f(\tau)e^{+(i/\hbar)\int_\tau^t\left(\mathcal{E}_1^{\rm QE}(\eta(t'))-\mathcal{E}_2^{\rm QE}(\eta(t'))\right)dt'}I(\tau-t). \qquad \text{(A41)}$$

Now we are able to solve the problem (A34) analytically. It turns out that the sought solution takes an explicit form

$$C(t)=\mathcal{C}(t)e^{+i\int_{t_{\rm init}}^t\tilde{\omega}(t')dt'}, \qquad \text{(A42)}$$

where the as yet unspecified quantities $\mathcal{C}(t)$ and $\tilde{\omega}(t)$ are allowed to depend only adiabatically upon time.

The substitution of Eqs. (41) and (A42) into Eq. (A34) reveals the internal consistency of the approach and leads toward the condition

$$\tilde{\omega}(t)=\frac{f^2(t)}{8\pi}\int_{-\infty}^{+\infty}\frac{\omega_k|D_k^+|^2}{\hbar\left(\omega_{21}^{\rm QE}(\eta(t))+\omega_k+\tilde{\omega}(t)\right)}dk, \qquad \text{(A43)}$$

with $\hbar\omega_{21}^{\rm QE}(\eta(t))=\mathcal{E}_2^{\rm QE}(\eta(t))-\mathcal{E}_1^{\rm QE}(\eta(t))$. Quantity $\hbar\tilde{\omega}(t)$ is interpreted physically as an experimentally unobservable (yet theoretically calculable) shift of the SC quasienergy $\mathcal{E}_1^{\rm QE}(\eta(t))$, arising due to the quantized nature of the radiation field. A moment of reflection reveals that the shift $\hbar\tilde{\omega}(t)$ is small compared to the atomic energy scale when f_a is small enough, and simultaneously, when the modal frequency cutoff (controlled by parameter ζ) is held finite. Before proceeding further, let us point out that, for any finite values of f_a and $\hbar\omega_{21}^{\rm QE}(\eta(t))$, an infinitely large energy shift $\hbar\tilde{\omega}(t)$ will be encountered in the limit $\zeta\to+\infty$ of an infinite modal cutoff. In such a case one will need to remove infinities by applying an appropriate renormalization technique, but we shall not delve into these matters here.

Having determined $\tilde{\omega}(t)$, the fastest route toward the population $|C(t)|^2 = |\mathcal{C}(t)|^2$ of the first channel is via the normalization condition $|\mathcal{C}(t)|^2 + {}_R\langle\tilde{\Psi}_2(t)|\tilde{\Psi}_2(t)\rangle_R = 1$. The self-overlap ${}_R\langle\tilde{\Psi}_2(t)|\tilde{\Psi}_2(t)\rangle_R$ can be evaluated using Eq. (A33) while incorporating the same simplifications as those employed in the present section A.4. One finds that

$$|\mathcal{C}(t)|^2 = \left\{1 + \frac{f^2(t)}{8\pi\hbar}\int_{-\infty}^{+\infty}\frac{\omega_k|D_k^+|^2}{\left(\omega_{21}^{\mathrm{QE}}(\eta(t)) + \tilde{\omega}(t) + \omega_k\right)^2}dk\right\}^{-1}. \tag{A44}$$

This is the most important outcome of the Appendix, demonstrating how the population $|\mathcal{C}(t)|^2$ of the first channel is affected by QED effects and how it depends upon the quasienergy gap $\hbar\omega_{21}^{\mathrm{QE}}(\eta(t))$. We recall in this context that $|\mathcal{C}(t)|^2 = 1$ within the SC approximation.

For the sake of clarity we note explicitly at this point that our derivation leading to Eq. (A44) is, of course, applicable also in the case when no laser light appears in the system, that is, in the case of a two-level atom embedded in the quantum vacuum. For such a situation, the additional simplifying assumptions made previously in the present section A.4 (weak emission, $|w_{12}(t)| = 1$) are not approximations but merely trivial statements. One arrives to the same result as Eq. (A44), with $\hbar\omega_{21}^{\mathrm{QE}}(\eta(t))$ being replaced by the bare energy difference $(\mathcal{E}_A^2 - \mathcal{E}_A^1)$.

Let us discuss now the physical contents of the just-derived formula (A44). It is evident that the population $|\mathcal{C}(t)|^2$ remains close to unity consistently with the SC prediction as long as the QED corrected quasienergy gap $\hbar\omega_{21}^{\mathrm{QE}}(\eta(t)) + \hbar\tilde{\omega}(t)$ is large compared to f_a^2 (the modal frequency cutoff is implicitly kept finite here, see the discussion in the previous paragraphs). *A radically different picture emerges, however, when the gap $\hbar\omega_{21}^{\mathrm{QE}}(\eta(t)) + \hbar\tilde{\omega}(t)$ is made small enough (but not necessarily zero) compared to f_a^2*, either by choosing correspondingly the parameters $(\mathcal{E}_A^2 - \mathcal{E}_A^1)$ and f_a of the two-level atomic system in the absence of laser light, or more importantly, by a suitable tailoring of the frequency and intensity of the incoming CW laser light. The breakdown of the SC approximation is implied here by the fact that the term

$$\frac{f^2(t)}{8\pi\hbar}\int_{-\infty}^{+\infty}\frac{\omega_k|D_k^+|^2}{\left(\omega_{21}^{\mathrm{QE}}(\eta(t)) + \tilde{\omega}(t) + \omega_k\right)^2}dk, \tag{A45}$$

appearing in Eq. (A44) becomes divergent in the limit when the gap $\hbar\omega_{21}^{\mathrm{QE}}(\eta(t)) + \hbar\tilde{\omega}(t)$ is made to vanish (while keeping a small nonzero value of f_a and a constant finite modal frequency cutoff). *This behavior then forces the population* (A44) *to approach zero, in a sharp deviation from the SC prediction.* Whenever the population $|\mathcal{C}(t)|^2$ differs significantly from unity, the dynamical quantum state vector of the whole system "atom and quantum field" cannot be accurately expressed as a direct product of a well-defined atomic state and a well-defined field state, contrary to the conceptual basis behind the SC description. Instead, the atomic and the field degrees of freedom become very strongly entangled due to QED effects.

The physical intuition suggests that the breakdown of the SC approach should be expected as soon as the gap $\hbar\omega_{21}^{\mathrm{QE}}(\eta(t)) + \hbar\tilde{\omega}(t)$ is made smaller than the spontaneous emission linewidth of the upper atomic level. We shall, however, not further examine this plausible hypothesis within the present article.

For the sake of completeness and clarity, we note in passing that the previously referred to singular situation

$$\hbar\omega_{21}^{\mathrm{QE}}(\eta(t)) + \hbar\tilde{\omega}(t) = 0, \tag{A46}$$

is perfectly consistent with Eq. (A43). Indeed, to satisfy Eq. (A46) one needs only to choose $\omega_{21}^{\mathrm{QE}}(\eta(t)) = (-f^2(t)/(8\pi\hbar))\int_{-\infty}^{+\infty}|D_k^+|^2dk = ((-f_a^2)/(8\pi\hbar))\sqrt{2\pi\zeta}$. The corresponding frequency shift comes out to be then $\tilde{\omega}(t) = -\omega_{21}^{\mathrm{QE}}(\eta(t))$ exactly as was stated by Eq. (A46). Note, however, that the required value of $\omega_{21}^{\mathrm{QE}}(\eta(t))$ is negative and refers thus to the light intensity for which the quasienergies have already passed through an intersection point. The discussion made after Eq. (A40) shows that the just-mentioned fact does not represent a serious obstacle for our argumentation.

5. An impact of QED effects on the emission of radiation

To complete our theoretical analysis, let us explore how the previously discussed breakdown of the SC approximation affects the emission of radiation from the atom. Formulated mathematically, we are interested in calculating the quantum mechanical expectation value of the electric field operator $\mathbf{E}(x,t)$ over the dynamical state vector (A29). That is,

$$E_{\mathrm{QED}}(x,t) = {}_R\langle\tilde{\Psi}_2(t)|\mathbf{E}(x,t)|\tilde{\Psi}_2(t)\rangle_R. \tag{A47}$$

This quantity represents the sought QED correction of the SC result $E_L(x,t)$. In principle one may evaluate $E_{\mathrm{QED}}(x,t)$ directly by substituting the formula (A33). We shall, however, not undertake such a direct yet tediously looking route. Instead, we proceed in a much more elegant and insightful fashion.

Namely, we shall exploit the fact that the total electric field $E_{\mathrm{TOT}}(x,t) = E_L(x,t) + E_{\mathrm{QED}}(x,t)$ corresponds to the quantum mechanical expectation value of the Heisenberg picture electric field operator $\mathbf{E}_R^H(x,t)$. Straightforward manipulations starting from the formulas (A1) through (A6) reveal that the dynamical time evolution of $\mathbf{E}_R^H(x,t)$ is governed by an operator equation of motion

$$\frac{\partial^2}{\partial x^2}\mathbf{E}_R^H(x,t) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{E}_R^H(x,t) = -\frac{f(t)\mathbf{w}^H(t)}{2}\frac{\partial^2}{\partial x^2}\left(\sqrt{\zeta/\pi}e^{-\zeta x^2}\right). \tag{A48}$$

Symbol $\mathbf{w}^H(t)$ stands here, of course, for the Heisenberg picture counterpart of an operator $\mathbf{w}$, which was introduced previously in Eq. (A6). Within the framework of the model problem considered in the present Appendix, the result in Eq. (A48) constitutes an exact nonapproximative QED counterpart of the SC Maxwell wave Eq. (A15). By taking the quantum expectation value over both sides of Eq. (A48) one finds that the sought total electric field $E_{\mathrm{TOT}}(x,t) = \langle\mathbf{E}_R^H(x,t)\rangle$ obeys the Maxwell wave equation

$$\frac{\partial^2}{\partial x^2}E_{\mathrm{TOT}}(x,t) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}E_{\mathrm{TOT}}(x,t) = -\frac{f(t)\langle\mathbf{w}^H(t)\rangle}{2}\frac{\partial^2}{\partial x^2}\left(\sqrt{\zeta/\pi}e^{-\zeta x^2}\right). \tag{A49}$$

This equation involves only c-numbers and should be confronted again versus its SC approximation (A15).

Importantly, the only difference between the wave equations (A15) and (A49) consists in the fact that $\langle \mathbf{w}^H(t)\rangle \neq w_{11}(t)$. It is an easy matter to explicitly translate $\langle \mathbf{w}^H(t)\rangle$ into the Schrödinger picture formalism developed above in A.1 through A.4. One has

$$\begin{aligned}\langle \mathbf{w}^H(t)\rangle = {} & |\mathcal{C}(t)|^2 w_{11}(t) + w_{22}(t)_R\langle\tilde{\Psi}_2(t)|\tilde{\Psi}_2(t)\rangle_R \\ & + C^*(t)w_{12}(t)_R\langle\{0\}|\tilde{\Psi}_2(t)\rangle_R \\ & + C(t)w_{21}(t)_R\langle\tilde{\Psi}_2(t)|\{0\}\rangle_R. \end{aligned} \tag{A50}$$

Using formula (A33) one can verify immediately that the overlap matrix element ${}_R\langle\{0\}|\tilde{\Psi}_2(t)\rangle_R$ is proportional to $b_v(t)f_a$. Correspondingly, at least in the weak emission regime, which we chose to follow at the beginning of A.4, the second line contribution to Eq. (A50) turns out to be negligible compared to the first line. Thus, to a good approximation, we may rewrite Eq. (A50) just into

$$\langle \mathbf{w}_H(t)\rangle = |\mathcal{C}(t)|^2 w_{11}(t) + \mathbf{(}1 - |\mathcal{C}(t)|^2\mathbf{)}w_{22}(t), \tag{A51}$$

with the normalization condition ${}_R\langle\tilde{\Psi}_2(t)|\tilde{\Psi}_2(t)\rangle_R = 1 - |\mathcal{C}(t)|^2$ being incorporated along the way. In addition, for our two-level atomic system we have $w_{22}(t) = -w_{11}(t)$, as already pointed out in A.2 after Eq. (A12). One arrives therefore toward a very simple final outcome

$$\langle \mathbf{w}_H(t)\rangle = \mathbf{(}2|\mathcal{C}(t)|^2 - 1\mathbf{)}w_{11}(t). \tag{A52}$$

Returning back to a confrontation between the two wave equations (A15) and (A49), *we may conclude now that the radiation emitted from the atomic source is scaled by factor*

$$Q(t) = \mathbf{(}2|\mathcal{C}(t)|^2 - 1\mathbf{)}, \tag{A53}$$

relatively to the prediction of the SC theory. This is the second most important result of the present Appendix.

The just-derived QED scaling factor (A53) reduces, of course, to 1 within the SC approximation of $|\mathcal{C}(t)|^2 = 1$. However, in the preceding section A.4 we demonstrated that $|\mathcal{C}(t)|^2$ may actually differ very significantly from 1 for certain well-defined and attainable conditions. *Under these conditions, the scaling factor $Q(t)$ differs very significantly from its SC approximation, showing a pronounced impact of QED effects on the experimentally measurable emitted field. Note, for example, that the emission is totally suppressed for the case of $|\mathcal{C}(t)|^2 = 1/2$.*

One may anticipate that the situation will be even more interesting for the case of a multilevel atom, where the time dependencies of the dipole functions are not constrained via the property $w_{22}(t) = -w_{11}(t)$. It seems likely that an analog of Eq. (A51) applies also in this more general system, with the corresponding dipole (or the electric current) being expressed as a weighted superposition over differently looking SC contributions. If so, the emitted radiation $E_{\mathrm{TOT}}(x,t)$ is also given as such a weighted superposition. Accordingly, QED effects are expected to have a pronounced impact on $E_{\mathrm{TOT}}(x,t)$ whenever a near degeneracy in quasienergies occurs.

6. An illustrative numerical example

Finally, let us illustrate the theoretical results of this Appendix by a simple numerical calculation. As already pointed out before, a two-level system can be realized in practice by the two lowest-energy eigenstates of a quantum particle moving in a double well potential. The height of an intermediate potential well controls here the energy splitting $\hbar\omega_{21}^A = (\mathcal{E}_A^2 - \mathcal{E}_A^1)$, whereas the shape of a spatial extension of the two potential minima determine the magnitude f_a of the dipole coupling between the two mentioned states.

Our particular choice of all the system parameters must be made consistently with the theoretical assumptions made previously. That is, we need to use a CW-like light pulse leading to an intersection of Floquet quasienergies for some laser field intensity. Moreover, we have to respect the approximations introduced within section A.4 (weak emission, $|w_{12}(t)| \approx 1$). As a matter of fact, the crucial condition for these requirements to be satisfied turns out to be $\omega_L \gg \omega_{21}^A$. Hereafter we shall adopt the following numerical parameters

$$\omega_{21}^A = (2\pi c/\lambda_A), \quad \lambda_A = 20\,000\,\mathrm{nm},$$

$$\text{i.e.,} \quad \omega_{21}^A = 0.002278\,\mathrm{a.u.}, \tag{A54}$$

$$f_a = 1.0\,\mathrm{a.u.}, \quad \zeta = 10^{-5}\,\mathrm{a.u.}, \tag{A55}$$

$$\omega_L = (2\pi c/\lambda_L), \quad \lambda_L = 800\,\mathrm{nm},$$

$$I \leqslant 2.5 \cdot 10^{14}\,\mathrm{W/cm^2}. \tag{A56}$$

Symbol I stands here, of course, for the intensity of the CW laser field, which is related to the electric field amplitude E_0 by the well-known formula $I[\mathrm{W/cm^2}] = 3.509 \cdot 10^{16}\,(E_0[\mathrm{a.u.}])^2$.

Figure 1 shows how the calculated quasienergies $\mathcal{E}_{1,2}^{\mathrm{QE}}(\eta)$ vary when the laser envelope is adiabatically switched on, the quasienergy intersection is detected for $I_{\mathrm{deg}} \approx 1.645 \cdot 10^{14}\,\mathrm{W/cm^2}$.

In Fig. 2 we display the Fourier components of the induced dipole $w_{11}(t)$, calculated for the maximum field intensity $I = 2.5 \cdot 10^{14}\,\mathrm{W/cm^2}$. We recall in this context that $w_{11}(t)$ enters as a source into the Maxwell wave equation (A15) and its frequency spectrum determines thereby the harmonics emitted from the atom according to the SC approach. One

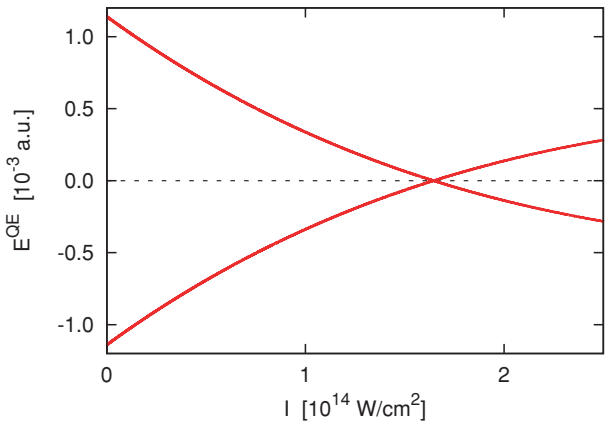

FIG. 1. (Color online) Dependence of the Floquet quasienergies $\mathcal{E}_{1,2}^{\mathrm{QE}}(\eta)$ of a two-level atom upon the intensity I of the CW laser wave. Laser light forces the dressed atomic energies to approach each other and to intersect. Numerical parameters characterizing the studied problem are given by Eqs. (A54) through (A56).

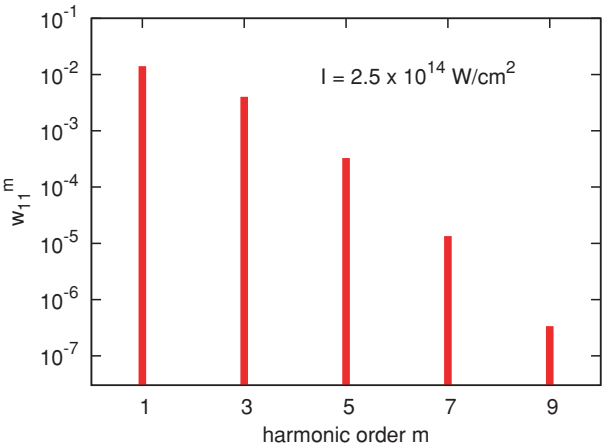

FIG. 2. (Color online) Fourier components w_{11}^m of the induced dipole function $w_{11}(t)$ entering into the Maxwell wave equation (A15). Note that $w_{11}^{-m} = w_{11}^{+m}$.

can see that the magnitude of $w_{11}(t)$ is of the order $O(10^{-2})$, consistently with our previously introduced weak emission assumption.

Next, in Fig. 3 we plot the Fourier components w_{12}^m defined by the formula (A40). The used field intensity corresponds again to the maximum value $I = 2.5 \cdot 10^{14}$ W/cm^2. It is evident that our approximation of $|w_{12}(t)| \approx 1$, made in A.4, is very well justified.

Having discussed the SC results, let us present now an outcome of our QED numerical calculation. Figure 4 is obtained by direct application of the QED formula (A44) and demonstrates explicitly how the population $|\mathcal{C}|^2$ of the first atomic channel depends upon the dressed quasienergy difference $\Delta E^{\mathrm{QE}} = \hbar(\omega_{21}^{\mathrm{QE}}(\eta) + \tilde{\omega})$. Degeneracy condition (A46) for the QED corrected quasienergy gap ΔE^{QE} is fulfilled when one takes $\omega_{21}^{\mathrm{QE}}(\eta) = -0.000315$ a.u., that is, for the laser field intensity $I_{\mathrm{crit}} \approx 1.72 \cdot 10^{14}$ W/cm^2. An inspection of Fig. 4 leads to the following important conclusions. For those laser field intensities $I < I_{\mathrm{crit}}$, which give significantly nonzero values of ΔE^{QE}, one finds that $|\mathcal{C}|^2 > 0.99$, validating thus

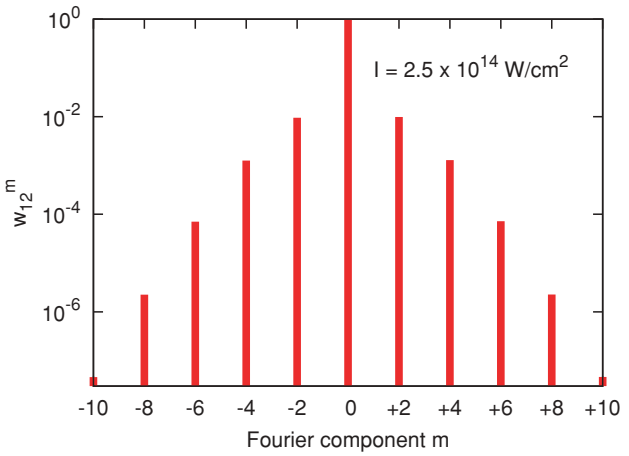

FIG. 3. (Color online) Fourier components w_{12}^m of the quantity $w_{12}(t)$ as defined by formula (A40). One can see that $|w_{12}(t)|$ equals to unity to a good approximation.

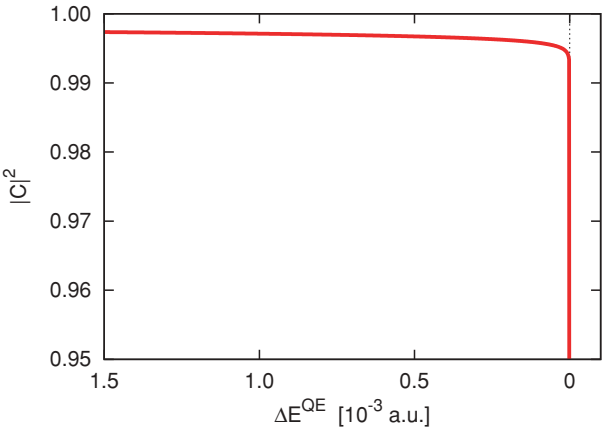

FIG. 4. (Color online) Population $|\mathcal{C}|^2$ of the first atomic channel plotted as a function of the quasienergy difference ΔE^{QE}. An abrupt decrease of $|\mathcal{C}|^2$ is evident as the quasienergy gap ΔE^{QE} approaches zero, indicating the breakdown of SC theory. An impact of this breakdown on the radiation emitted from the atom is documented by Eqs. (A49) and (A52).

an application of the SC theory. On the other hand, however, *a very abrupt decrease of* $|\mathcal{C}|^2$ *is observed in Fig. 4 as* ΔE^{QE} *approaches zero. In such a situation, the SC approximation of* $|\mathcal{C}|^2 = 1$ *becomes grossly inadequate,* consistently with our theoretical claims made in A.4. Moreover, the QED scaling factor (A53) derived in A.5 becomes substantially different from unity and hence *the emission of radiation from the atom is strongly influenced by QED effects*, as predicted by the relevant Eqs. (A49) and (A52). It is worth noting in this context that an explicit appearance of the QED source term (A52) can be recovered immediately through a combination of Figs. 2 and 4.

7. Summary

Summarizing the contents of this Appendix, we examined an (in)adequacy of the SC approach using a simple model of a two-level atom coupled to quantum radiation field. Our derivation is based upon mathematically well-defined and physically well-justifiable approximations and leads finally to simple formulas (A44) and (A52), which serve as indicators of nonseparability between the atomic and the field degrees of freedom. The SC approach, based implicitly upon the direct product separation between the atomic and the field variables, is falsified by formula (A44) whenever an accidental (near)degeneracy occurs between the two atomic energy levels dressed by the c-number laser light and by the QED effects. The breakdown of the SC theory impacts also significantly on the radiation emitted from the atom, see Eqs. (A49) and (A52) and the accompanying discussion. The analytical and numerical findings of this Appendix agree completely with the qualitative claims made in the main text.

In passing we note that the usual argumentation regarding the "large number of photons in the laser beam" is never encountered during the theoretical analysis made previously. Indeed, (in)validity of the SC approach seems to be completely unrelated to the number of photons, when the problem is examined from the theoretical perspective adopted in the present article.

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