Dynamic steady state of periodically driven quantum systems

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Using the density matrix formalism, we prove the existence of the periodic steady state for an arbitrary periodically driven system described by linear dynamic equations. This state has the same period as the modulated external influence, and it is realized as an asymptotic solution ($t \to +\infty$) due to relaxation processes. The presented derivation simultaneously contains a simple and effective computational algorithm (without using either the Floquet or Fourier formalisms), which automatically guarantees a full account of all frequency components. As a particular example, for three-level Λ system we calculate the line shape and field-induced shift of the dark resonance formed by the field with a periodically modulated phase. Also we have analytically solved a basic theoretical problem of the direct frequency comb spectroscopy, when the two-level system is driven by the periodic sequence of rectangular pulses. In this case, the radical dependence of the spectroscopy line shape on pulse area is found. Moreover, the existence of quasiforbidden spectroscopic zones, in which the Ramsey fringes are significantly reduced, is predicted. Our results have a wide area of applications in laser physics, spectroscopy, atomic clocks, and magnetometry. Also they can be useful for any area of quantum physics where periodically driven systems are considered.

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I. INTRODUCTION

For the last several decades rapid scientific and technological progress has been substantially connected with an expansion of the lasers and laser technologies at different areas of the science, engineering, and industry. In this process laser physics, nonlinear optics, and laser spectroscopy take a special place. Many impressive successes in these directions are due to the theoretical support, motivation, and interpretation of experimental researches. In this context, of paramount importance is the formulation of mathematical models (equations) and finding of their solutions, which adequately describe the physical picture of investigated problems. As an example, for atomic mediums the density matrix formalism is the most widespread approach describing the atom-field interaction and different relaxation processes (spontaneous, collisional, etc.). Especial significance has the so-called steady state, which arises under the interaction of a quantum system with stationary external fields.

Recall that the steady-state concept has the following theoretical justification. For an arbitrary quantum system let us consider the density matrix $\hat{\rho}(t)$ and its normalization condition (integral of motion describing the conservation of probability), which in a basis of states $\{|j\rangle\}$ can be presented as

$$\hat{\rho}(t) = \sum_{a,b} |a\rangle \rho_{ab}(t) \langle b|; \quad \mathrm{Tr}\{\hat{\rho}(t)\} = \sum_{j} \rho_{jj}(t) = 1, \quad (1)$$

where $\rho_{ab}(t)$ are the matrix elements. The density matrix dynamic equation can be written as

$$\partial_t \hat{\rho}(t) = -(i/\hbar)[\widehat{H}, \hat{\rho}(t)] + \widehat{\Gamma}\{\hat{\rho}(t)\}; \quad \mathrm{Tr}\{\hat{\rho}(t)\} = 1. \quad (2)$$

Here we have separately selected the Hamiltonian term $[\widehat{H}, \hat{\rho}(t)]$ having the form of a commutator with Hamiltonian $\widehat{H}$, which describes the energy states of the quantum system and the interaction with external fields. The other operator functional $\widehat{\Gamma}\{\hat{\rho}(t)\}$ describes all possible relaxation processes (e.g., for atomic gases it can be spontaneous, collisional, time of flight, and other processes), and $\widehat{\Gamma}\{\hat{\rho}(t)\}$ cannot be presented in the commutator form.

If $\widehat{H} = \text{const.}$ and the initial state is $\hat{\rho}(0)$ (at $t = 0$, for definiteness), then during the dynamic process the asymptotic (i.e., under $t \to +\infty$) steady state $\hat{\rho}_{\text{st-st}} \equiv \hat{\rho}(+\infty)$ is formed. This state does not depend on the time ($\partial_t \hat{\rho}_{\text{st-st}} = 0$) and does not depend on the initial condition $\hat{\rho}(0)$, and it satisfies the equation

$$-(i/\hbar)[\widehat{H}, \hat{\rho}_{\text{st-st}}] + \widehat{\Gamma}\{\hat{\rho}_{\text{st-st}}\} = 0; \quad \mathrm{Tr}\{\hat{\rho}_{\text{st-st}}\} = 1. \quad (3)$$

Note that the existence of the single asymptotic state $\hat{\rho}(+\infty)$ is completely determined by the relaxation processes. Indeed, in the absence of relaxation (i.e., $\widehat{\Gamma}\{\ldots\} = 0$) the evolution equation (2) corresponds in essence to the Schrödinger equation, and its solution has the well-known form

$$\hat{\rho}(t) = \exp\{-i\widehat{H}t/\hbar\}\, \hat{\rho}(0)\, \exp\{i\widehat{H}t/\hbar\}, \quad (4)$$

which describes the undamped dynamics of the quantum system, starting from the initial state $\hat{\rho}(0)$. In this case, the mathematical expression (4) even for $t \to +\infty$ does not allow the existence of the asymptotically unique steady state $\hat{\rho}_{\text{st-st}}$, which is independent of the initial state $\hat{\rho}(0)$. Thus, the steady-state concept is directly connected with the non-Hamiltonian relaxation term $\widehat{\Gamma}\{\hat{\rho}(t)\}$ in the dynamic equation (2).

During long time, steady states play a key role in the theoretical description of the basic problems in laser physics and spectroscopy (for example, see [1–3]). However, in the last few years the devices in which different parameters of electromagnetic fields are periodically modulated have gained a greater importance. First of all, the so-called frequency comb generators use the periodic pulse modulation of a laser field. Such sources of pulse radiation are actively used now in

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modern atomic clocks for frequency measurements [4,5], and they have promising perspectives for direct frequency comb spectroscopy (e.g., see [6–9]). Also, the phase (frequency) and/or amplitude periodic modulation of the laser field is now widely used for different tasks and applications (including atomic clocks and magnetometers) [10–16]. Other promising and interesting directions in atomic clocks and magnetometers are connected with periodically modulated polarization of the laser field [17–22]. Furthermore, a periodically driven quantum system displays many curious features, e.g., dynamic suppression of tunneling in a double well [23], Bloch-Siegert shift [24], and effects of counter-rotating terms. Of late, the understanding of periodically driven systems is also one of the most active areas of research in many-body physics (e.g., see [25–38]). In particular, there is the problem of a stationary state existence. In all these examples the *standard* concept of steady state based on the time-independent equation (3) is inapplicable, generally speaking, because of time dependence $\widehat{H}(t)$.

In this paper, we generalize the steady-state concept for an arbitrary quantum system under arbitrary periodic external influence. In this way we prove the following existence theorem: if the coefficients of density matrix dynamic equation (2) have the period T, then the periodic solution with the same period T exists always. A completely unexpected result is that so universal and fundamental a statement is based only on the normalization condition for the density matrix. Due to the relaxation processes this solution is realized as an asymptotics ($t \to +\infty$) and, therefore, can be characterized as a periodic steady state. The developed simple algorithm allows us to directly construct this solution independently of initial conditions and without the use of either Floquet or Fourier formalisms. Our approach considerably simplifies the analysis regardless of the periodic modulation character: from smoothly harmonic type to ultrashort pulses. As a striking example, for two-level atoms we have analytically calculated the signal and have found unknown features of the direct frequency comb spectroscopy formed by the periodic sequence of rectangular pulses.

II. GENERAL THEORY

The general mathematical formulation of the steady-state problem for periodically driven systems is the following. First of all, let us rewrite the differential equation for the density matrix (2) in the vector form:

$$\partial_t \vec{\rho}(t) = \widehat{L}(t)\,\vec{\rho}(t); \quad \mathrm{Tr}\{\hat{\rho}(t)\} = (\vec{n},\vec{\rho}(t)) = 1, \tag{5}$$

where the column vector $\vec{\rho}(t)$ is formed by the matrix elements $\rho_{ab}(t)$ using some definite rule, the linear operator $\widehat{L}(t)$ corresponds to the right-hand member of Eq. (2). As an example, for a two-level system with basis states $|1\rangle$ and $|2\rangle$ there are four matrix elements $\rho_{ab}(t)$ ($a,b = 1, 2$), which can be ordered as the following vector:

$$\vec{\rho}(t) = \begin{pmatrix} \rho_{22}(t) \\ \rho_{21}(t) \\ \rho_{12}(t) \\ \rho_{11}(t) \end{pmatrix}. \tag{6}$$

Besides we define the supplementary column vector $\vec{n}$ permitting us to express the value $\mathrm{Tr}\{\hat{\rho}(t)\}$ as the dot product in Eq. (5), where we use the standard definition of the dot product for arbitrary complex-valued vectors: $(\vec{x},\vec{y}) = \sum_m x_m^* y_m$. The vector $\vec{n}$ is formed in the following way: for the positions corresponding to the diagonal elements $\rho_{jj}(t)$ [as components of the vector $\vec{\rho}(t)$] the vector $\vec{n}$ has the value 1, and it equals 0 for all other positions. In particular, for two-level systems, according to the definition (6), we obtain

$$\vec{n} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 1 \end{pmatrix}. \tag{7}$$

The normalization condition in Eq. (5) implies the degeneracy of equations [right-hand members, $\widehat{L}(t)\vec{\rho}(t)$] which corresponds to the following expression:

$$(\vec{n},\widehat{L}(t)\vec{\rho}) = 0 \tag{8}$$

for an arbitrary vector argument $\vec{\rho}$. Note that the existence of the time-independent vector $\vec{n}$ is a base point for the next reasonings.

Let us suppose an existence of the time period T in the operator $\widehat{L}(t)$:

$$\widehat{L}(t+T) = \widehat{L}(t). \tag{9}$$

In this case, as it will be shown below, Eq. (5) always has a periodic solution with the same period T:

$$\vec{\rho}(t+T) = \vec{\rho}(t) \tag{10}$$

for each t.

To begin the proof of this theorem, we assume that at some instant of time t_1 we have an arbitrary vector $\vec{\rho}(t_1)$. Then, in accordance with Eq. (5), for other instant of time t_2 we can write

$$\vec{\rho}(t_2) = \widehat{A}(t_2,t_1)\,\vec{\rho}(t_1), \tag{11}$$

where the two-time evolution operator $\widehat{A}(t_2,t_1)$ is determined by the matrix $\widehat{L}(t)$. Note that in the case of periodicity condition (9) the following relationship takes place:

$$\widehat{A}(t_2+T,t_1+T) = \widehat{A}(t_2,t_1) \tag{12}$$

for arbitrary t_1,t_2.

However, before the consideration of the periodic case, let us prove several general statements. For this purpose, we multiply Eq. (11) by the vector $\vec{n}$ as the dot product:

$$(\vec{n},\vec{\rho}(t_2)) = (\vec{n},\widehat{A}(t_2,t_1)\,\vec{\rho}(t_1)) = (\widehat{A}^\dagger(t_2,t_1)\,\vec{n},\vec{\rho}(t_1)), \tag{13}$$

where $\widehat{A}^\dagger(t_2,t_1)$ is the Hermitian conjugate operator to $\widehat{A}(t_2,t_1)$. Because $(\vec{n},\vec{\rho}(t_2)) = (\vec{n},\vec{\rho}(t_1))$, from Eq. (13) we obtain

$$(\vec{n},\vec{\rho}(t_1)) = (\widehat{A}^\dagger(t_2,t_1)\,\vec{n},\vec{\rho}(t_1)) \tag{14}$$

for arbitrary $\vec{\rho}(t_1)$. This formula directly implies

$$\widehat{A}^\dagger(t_2,t_1)\,\vec{n} = \vec{n}, \tag{15}$$

i.e., the operator $\widehat{A}^\dagger(t_2,t_1)$ always has the eigenvector with the real eigenvalue 1. Therefore, the same eigenvalue 1 also exists for the direct operator $\widehat{A}(t_2,t_1)$, i.e., there is always the eigenvector $\vec{r}(t_2,t_1)$, which satisfies the equation

$$\widehat{A}(t_2,t_1)\,\vec{r}(t_2,t_1) = \vec{r}(t_2,t_1). \tag{16}$$

In contrast to Eq. (15), the eigenvector $\vec{r}(t_2,t_1)$ depends on t_2,t_1 in the general case.

Let us return to the case of periodicity [Eq. (9)]. Consider $\vec{\rho}(t)$ at arbitrary instant of time t. In conformity with Eq. (11), the vector $\vec{\rho}(t+T)$ is determined as

$$\vec{\rho}(t+T) = \widehat{A}(t+T,t)\,\vec{\rho}(t). \tag{17}$$

Supposing the existence of the periodic solution $\vec{\rho}(t+T) = \vec{\rho}(t)$, it follows from Eq. (17) that this solution satisfies the equation

$$\vec{\rho}(t) = \widehat{A}(t+T,t)\,\vec{\rho}(t); \quad (\vec{n},\vec{\rho}(t)) = 1, \tag{18}$$

which always has a nonzero solution due to the above proven statement of existence of eigenvector with eigenvalue 1 for the operator $\widehat{A}(t_2,t_1)$ for arbitrary t_1,t_2 [see Eq. (16)]. Using Eqs. (12), (17), and (18), it can be easily shown that $\vec{\rho}(t+lT) = \vec{\rho}(t)$ ($l = \pm 1, \pm 2, \ldots$). Taking into account an arbitrariness of the time t in Eqs. (17) and (18), we can assert that the existence theorem of the periodic solution (10) is proven.

Due to the relaxation processes there is a unique (in the majority of cases) periodic solution which is realized during the time evolution as an asymptotic state ($t \to +\infty$) independently of initial conditions [similar to the well-known stationary case in Eq. (20) below]. The solution (10) can be called a *dynamic* steady state because of its dependence on t.

However, taking into account a mathematical generality, for some theoretical models we can hypothetically assume the possibility of several solutions $\{\vec{\rho}_1(t), \vec{\rho}_2(t), \ldots, \vec{\rho}_Q(t)\}$ for Eq. (18), when the eigenvalue 1 for the matrix $\widehat{A}(t+T,t)$ is degenerated in spite of relaxation processes. In this case, the general periodic solution has a form of superposition:

$$\vec{\rho}(t) = \sum_{q=1}^{Q} \alpha_q \vec{\rho}_q(t); \quad (\vec{n},\vec{\rho}_q(t)) = 1; \quad \sum_{q=1}^{Q} \alpha_q = 1, \tag{19}$$

where the number set $\{\alpha_1, \ldots, \alpha_Q\}$ for the asymptotic solution ($t \to +\infty$) will depend on initial conditions (under the dynamic consideration).

Let us show that the found solution (18) is a generalization of the well-known steady state [see the matrix $\hat{\rho}_{\text{st-st}}$ in Eq. (3)] for the constant operator $\widehat{L}$ in Eq. (5):

$$\widehat{L}\vec{\rho}_{\text{st-st}} = 0; \quad \partial_t \vec{\rho}_{\text{st-st}} = 0; \quad (\vec{n},\vec{\rho}_{\text{st-st}}) = 1. \tag{20}$$

It is obvious that the stationary case can be considered as a periodic case with an arbitrary value of the period T. Therefore, according to the above analysis, the state $\vec{\rho}_{\text{st-st}}$ also must satisfy Eq. (18) for arbitrary t,T. Indeed, in the case of $\widehat{L} = \text{const.}$, we have for arbitrary t,T

$$\widehat{A}(t+T,t) = e^{\widehat{L}T} = 1 + \sum_{k=1}^{+\infty} \frac{1}{k!} T^k \widehat{L}^k. \tag{21}$$

Taking into account Eqs. (20) and (21), we obtain perfect compliance with Eq. (18): $\widehat{A}(t+T,t)\vec{\rho}_{\text{st-st}} = \vec{\rho}_{\text{st-st}}$.

Let us consider our results in relation to the general Floquet theory (e.g., see [39]). According to this theory, the solutions of the differential equation system (5) with periodic coefficients can be presented in the following form:

$$\vec{\rho}(t) = \widehat{D}(t) e^{\widehat{K}t} \vec{a}, \tag{22}$$

where the matrix $\widehat{D}(t) = \widehat{D}(t+T)$ is periodic, $\widehat{K}$ is a certain constant matrix connected with the matrix $\widehat{L}(t)$, and $\vec{a}$ is an arbitrary constant vector. Using the set of eigenvectors $\widehat{K}\vec{a}_j = \lambda_j \vec{a}_j$, expression (22) can be rewritten as the superposition of independent solutions:

$$\vec{\rho}(t) = \widehat{D}(t) \sum_j C_j e^{\lambda_j t} \vec{a}_j, \quad C_j = \text{const.} \tag{23}$$

From this formula it follows that the existence of the periodic solution $\vec{\rho}(t+T) = \vec{\rho}(t)$ seems to be an exception to the general rule, because such solution can be realized only if at least one eigenvalue $\lambda_s = i2\pi m/T$ exists [where m is an arbitrary integer number (including 0)]. However, in the framework of the general Floquet theory the universal approach to this problem is absent, i.e., the special consideration is required for each concrete task. In this context, our result is that the equation class, describing the density matrix dynamics for different quantum systems with arbitrary relaxation processes, always has a periodic solution. Our theorem proof is based only on the normalization condition and it is done without the Floquet expansion (23) [instead of this, we use the two-time evolution operator (11)]. Apart from the density matrix equations (2), the developed approach can be applied to prove a similar existence theorem of periodic solutions for other differential equation systems, for which some *linear* integrals of motion exist.

In regard to the other states in the superposition (23) with $\lambda_j \neq i2\pi m/T$, for the majority of the adequate mathematic models of quantum systems with relaxation these states should be damped under $t \to +\infty$, i.e., $\text{Re}\{\lambda_j\} < 0$ (see also comments [40,41]). In contrast, without relaxation (i.e., $\widehat{\Gamma}\{\ldots\} = 0$) the periodically driven systems can be described by the Schrödinger equation (e.g., see the classical paper [42]) and all possible solutions in Eq. (23) will be undamped: $\text{Re}\{\lambda_j\} = 0$. In this case, the unique asymptotic steady state $\vec{\rho}(+\infty)$ does not exist. It is precisely this fact that explains the base difference between damped and undamped quantum systems with respect to the long-time dynamics.

Note that some authors supposed (without proof) the existence of the periodic steady-state solution [Eq. (10)] for some certain problems. In this case they usually used the Fourier analysis for numerical calculations (e.g., see [12,43]). However, an intuitive assumption about the periodic steady state is now rigorously substantiated. At the same time, of special interest is the direct and simple method, which allows us to construct the periodic solution on the base of Eq. (18) without Fourier expansion. Thus, our approach automatically takes a full account of all frequency components and can radically simplify the calculations (numerical and analytical).

Let us describe one possible numerical algorithm based on Eq. (18). We consider an arbitrary periodic dependence of the operator $\widehat{L}(t)$ (see Fig. 1). For instance, under an atom-field interaction such a dependence can be produced by the modulation of the field parameters (amplitude, phase, polarization, etc.). The selected time interval $[t_0,t_0+T]$ is divided into N small subintervals, where $t_N = t_0 + T$. The character of partition (uniform or nonuniform discrete mesh) and number of subintervals are determined in conformity with the studied problem. The dependence $\widehat{L}(t)$ we will approximate by step function (see Fig. 1), where the matrix

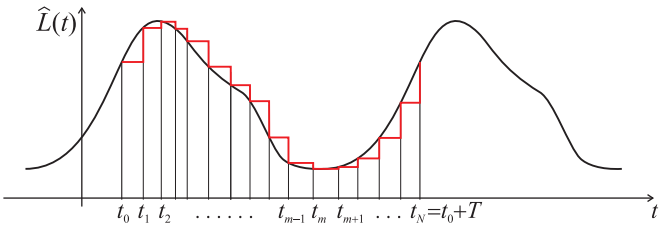

FIG. 1. Partition of the time interval $[t_0,t_0+T]$ at N subintervals and symbolic approximation of the dependence $\widehat{L}(t)$ by the step function (red step line).

$\widehat{L}(t)$ has the constant value $\widehat{L}(t_{m-1})$ inside of subinterval $(t_{m-1},t_m]$. In this case the vector $\vec{\rho}(t_0)$ in initial point t_0 is determined by Eq. (18), where the evolution operator $\widehat{A}(t_0+T,t_0)$ has the form of a chronologically ordered product of the matrix exponents (in the manner of [44]):

$$\widehat{A}(t_0+T,t_0) \approx \prod_{m=1}^{m=N} e^{(t_m-t_{m-1})\widehat{L}(t_{m-1})} = e^{(t_N-t_{N-1})\widehat{L}(t_{N-1})} \times \cdots \times e^{(t_1-t_0)\widehat{L}(t_0)}. \quad (24)$$

The vectors $\vec{\rho}(t_m)$ in other points of the interval $[t_0,t_0+T]$ are determined by the recurrence relation

$$\vec{\rho}(t_m) = e^{(t_m-t_{m-1})\widehat{L}(t_{m-1})}\vec{\rho}(t_{m-1}). \quad (25)$$

Below we consider several important examples (related to the atomic clock and spectroscopy of atomic transitions), which demonstrate the simplicity and high efficiency of our method.

III. DARK RESONANCES IN THREE-LEVEL Λ SYSTEM DRIVEN BY THE PHASE-MODULATED FIELD

The first example is the so-called dark resonance, which is formed in a three-level Λ system by a bichromatic field (see Fig. 2). This resonance occurs when a difference between the optical frequencies $(\omega_1-\omega_2)$ is varied near the transition frequency between the lower energy levels $|1\rangle$ and $|2\rangle$: $\omega_1-\omega_2 \approx \Delta$. Currently, such resonances at the hyperfine structure of the alkali-metal atoms (first of all, for ^{87}Rb and ^{133}Cs) underlie the chip-scale atomic clocks,

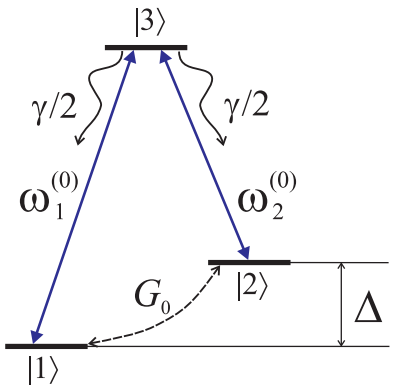

FIG. 2. Three-level system where the dark resonance can be formed.

which have a great practical importance. At the same time the harmonic phase modulation (frequency deviation) of miniature semiconductor lasers (so-called VCSEL) is ordinarily used in practice. In this case the field can be written as

$$E(t) = E_0 e^{-i[\omega t+\varphi(t)]} + \text{c.c.}; \quad \varphi(t) = A\sin(\nu t), \quad (26)$$

where E_0 is the field amplitude, ω is the central frequency of laser, and A and ν are the amplitude and frequency of phase modulation, respectively. It is obvious that this field is polychromatic, in which the frequency difference between the adjacent components equals ν. The phase modulation at full or half of the hyperfine splitting is usually used: $\nu \approx \Delta$ or $\Delta/2$. However, the dark resonance can be observed in the more general case of $\nu \approx \Delta/l$ $(l = 1,2,3,\ldots)$.

For theoretical description of this problem the Fourier expansion of the field (26) is traditionally used. In this case, to solve the density matrix equation one can extract two resonant components (e.g., see [12]), which are involved in the absorption and directly form the dark resonance. While the remaining frequency components are taken into account only from the viewpoint of field-induced shifts of the clock transition. It is evident that such an approach is quite cumbersome and incomplete. In contrast, our method does not use the Fourier expansion and we directly calculate the time-dependent periodic $\hat{\rho}(t)$. In the resonance approximation, the equations for the density matrix components $\rho_{jk}(t)$ have the following form:

$$\begin{aligned}
[\partial_t+\Gamma-i\delta_1-i\varphi'(t)]\rho_{31} &= i\Omega_1(\rho_{11}-\rho_{33})+i\Omega_2\rho_{21},\\
[\partial_t+\Gamma-i\delta_2-i\varphi'(t)]\rho_{32} &= i\Omega_2(\rho_{22}-\rho_{33})+i\Omega_1\rho_{12},\\
[\partial_t+G_0-i\Delta]\rho_{12} &= i(\Omega_1^*\rho_{32}-\rho_{13}\Omega_2),\\
[\partial_t+G_0]\rho_{11} &= \gamma\rho_{33}/2+G_0\mathrm{Tr}\{\hat{\rho}\}/2+i(\Omega_1^*\rho_{31}-\rho_{13}\Omega_1),\\
[\partial_t+G_0]\rho_{22} &= \gamma\rho_{33}/2+G_0\mathrm{Tr}\{\hat{\rho}\}/2+i(\Omega_2^*\rho_{32}-\rho_{23}\Omega_2),\\
[\partial_t+G_0+\gamma]\rho_{33} &= i(\Omega_1\rho_{13}-\rho_{31}\Omega_1^*)+i(\Omega_2\rho_{23}-\rho_{32}\Omega_2^*),\\
\rho_{jk} &= \rho_{kj}^* \ (j,k=1,2,3);\ \ \mathrm{Tr}\{\hat{\rho}\}=\rho_{11}+\rho_{22}+\rho_{33}=1.
\end{aligned} \quad (27)$$

Here $\delta_{1,2} = (\omega-\omega_{1,2}^{(0)})$ are one-photon detunings, $\Omega_1 = d_{31}E_0/\hbar$ and $\Omega_2 = d_{32}E_0/\hbar$ are the Rabi frequencies (d_{31} and d_{32} are the reduced matrix elements of the dipole moment for the respective transitions), γ is the decay rate of excited level $|3\rangle$, Γ is the total decoherence rate (spontaneous, collision, time of flight) of optical transitions $|1\rangle \to |3\rangle$ and $|2\rangle \to |3\rangle$, G_0 is the relaxation rate of lower energy levels to the equilibrium isotropic state: $(|1\rangle\langle 1|+|2\rangle\langle 2|)/2$. Note, if we delete the

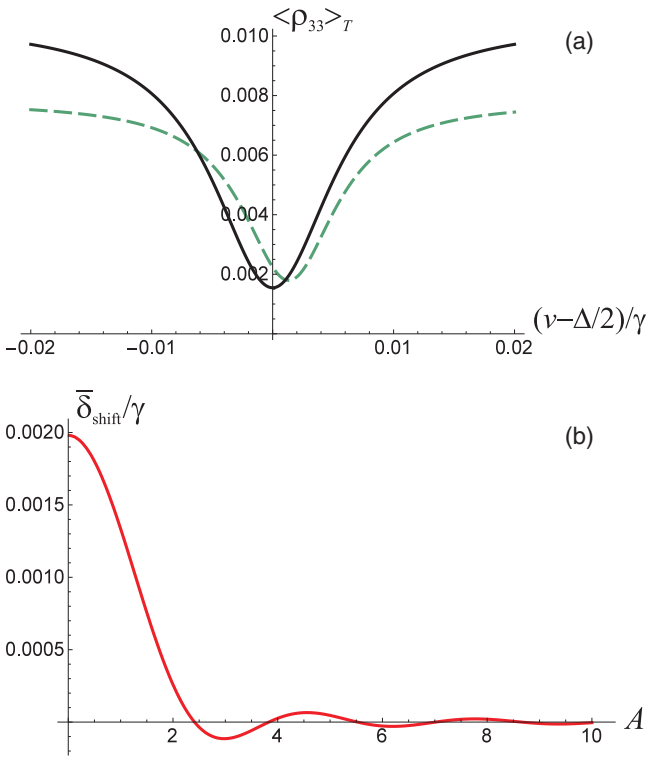

FIG. 3. (a) The dark resonance line shape in the case when ν is varied near $\Delta/2$ for different values of the phase modulation amplitude A: $A = 1.0$ (green dashed line) and $A = 2.4$ (solid black line). (b) The dependence of the dark resonance position (relative to $\Delta/2$) on parameter A. Calculations are done for $\Omega_1/\gamma = \Omega_2/\gamma = 1$; $G_0/\gamma = 1.5 \times 10^{-3}$; $\Gamma/\gamma = 50$; $\Delta/\gamma = 1000$, $\delta_2 = -\delta_1 = \Delta/2$.

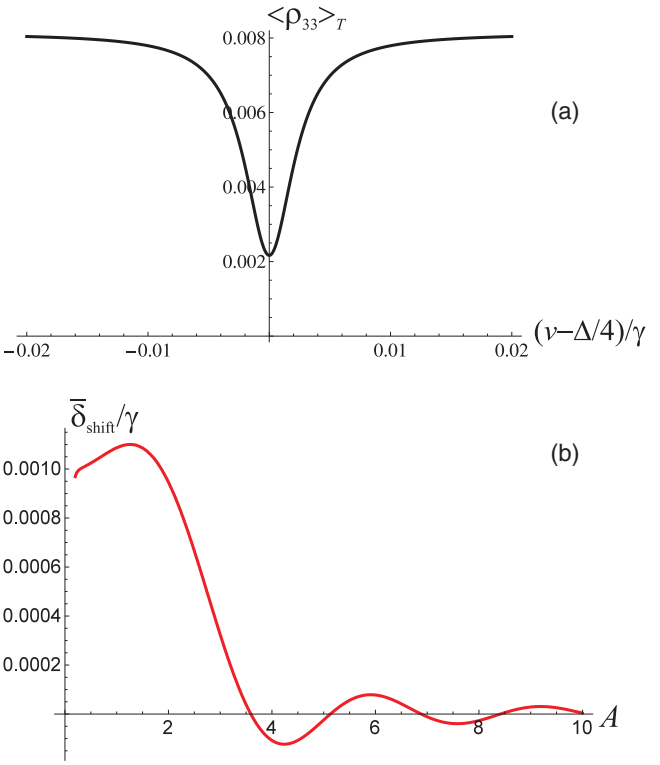

FIG. 4. (a) The dark resonance line shape in the case when ν is varied near $\Delta/4$ for the phase modulation amplitude $A = 3.59$. (b) The dependence of the dark resonance position (relative to $\Delta/4$) on parameter A. Calculations are done for $\Omega_1/\gamma = \Omega_2/\gamma = 1$; $G_0/\gamma = 1.5 \times 10^{-3}$; $\Gamma/\gamma = 50$; $\Delta/\gamma = 1000$, $\delta_2 = -\delta_1 = \Delta/2$.

derivative by phase modulation $\varphi'(t)$, then Eqs. (27) formally coincide with the description of interaction between the monochromatic field (at the frequency ω) and the three-level Λ system.

In particular, we have calculated by our method the resonance line shapes as dependencies on the frequency of phase modulation $\nu \approx \Delta/2$ for different values of amplitude A [see Fig. 3(a)]. The taken parameters are typical for atomic cells with vapor of ^{87}Rb in the buffer gas. Figure 3(b) displays the dependence of the field shift of the dark resonance $\bar{\delta}_{\text{shift}}$ (with respect to $\Delta/2$) on the modulation amplitude A. There is the value set of A, for which the field shift vanishes (see also [45]). In Fig. 4 similar graphics are shown for the case of phase modulation frequency $\nu \approx \Delta/4$. From Fig. 4(a) it is seen that the amplitude of dark resonance can be comparable with the case of $\nu \approx \Delta/2$ [compare with Fig. 3(a)].

IV. TWO-LEVEL SYSTEM DRIVEN BY THE PULSE-MODULATED FIELD

For another example, let us consider a two-level system (with unperturbed frequency ω_0) interacting with the field at the frequency ω:

$$E(t) = \text{Re}\{\mathcal{E}(t)\, e^{-i\omega t}\}, \tag{28}$$

where the amplitude modulation $\mathcal{E}(t)$ has the form of rectangular-pulse periodic sequence [see Fig. 5(a)], which is a basic model of direct frequency comb spectroscopy and can be considered as a limiting case of Ramsey spectroscopy. In this case the field has an equidistant spectrum $\omega_m = \omega + mf_r$ ($m = 0, \pm 1, \pm 2, \ldots$) (so-called frequency comb), where $f_r = 2\pi/T$ is the repetition frequency, and a spectral width (with respect to the central frequency ω) is determined by the pulse duration as $1/\tau$. The dynamic equations for density

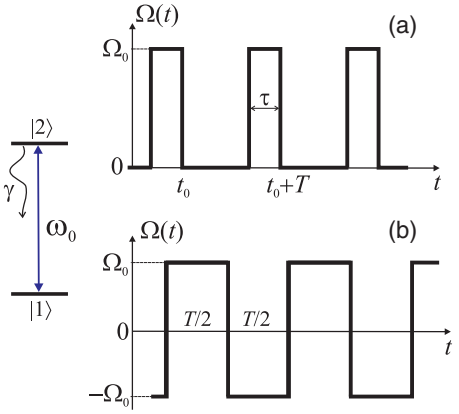

FIG. 5. Two-level system interacting with (a) periodic sequence of rectangular pulses and (b) periodic sequence of ($\pm$)-phase jumps.

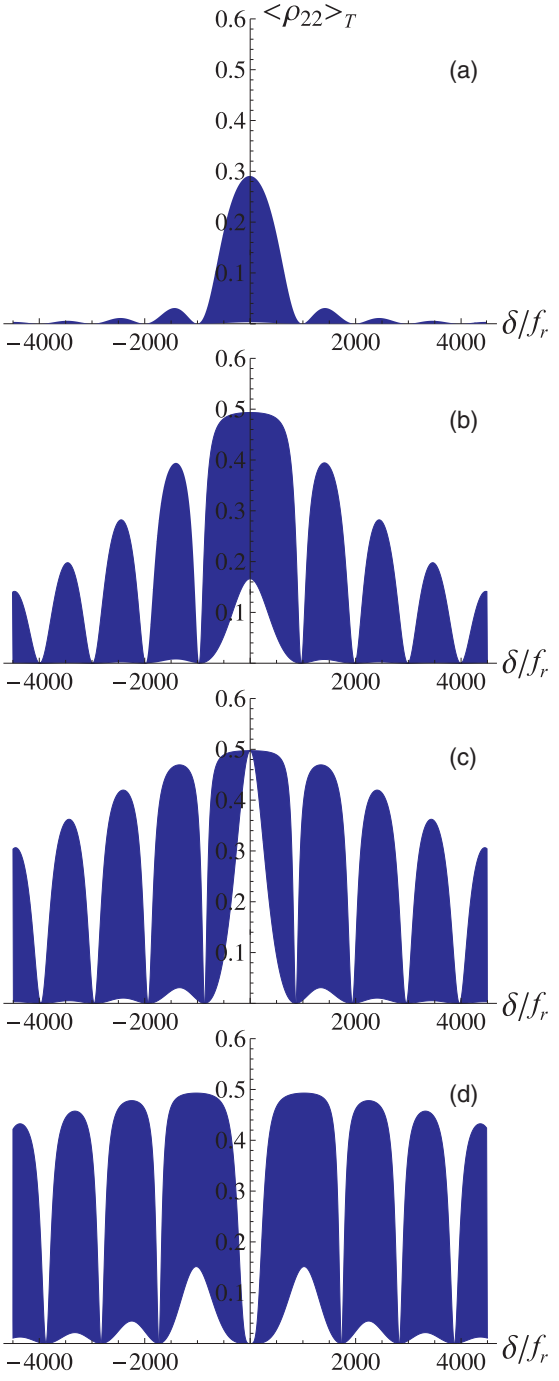

FIG. 6. The line shapes for direct frequency comb spectroscopy [see Fig. 5(a)] over large interval of detuning δ, where the narrow resonances are visually inseparable (dark areas). Calculations are done for $\Gamma = \gamma/2 = 0.02 f_r$, $\tau/T = 0.001$ and for different pulse areas: (a) $\Omega_0\tau = \pi/15$; (b) $\Omega_0\tau = \pi/2$; (c) $\Omega_0\tau = \pi$; and (d) $\Omega_0\tau = 2\pi$.

matrix components are given by

$$
\begin{aligned}
&[\partial_t + \Gamma - i\delta]\rho_{21} = i\Omega(t)(\rho_{11} - \rho_{22})/2,\\
&[\partial_t + \Gamma + i\delta]\rho_{12} = -i\Omega^*(t)(\rho_{11} - \rho_{22})/2,\\
&[\partial_t + \gamma]\rho_{22} = i[\Omega(t)\rho_{12} - \rho_{21}\Omega^*(t)]/2,\\
&\partial_t\rho_{11} = \gamma\rho_{22} + i[\Omega^*(t)\rho_{21} - \rho_{12}\Omega(t)]/2,\\
&\mathrm{Tr}\{\hat{\rho}\} = \rho_{11} + \rho_{22} = 1,
\end{aligned}
\tag{29}
$$

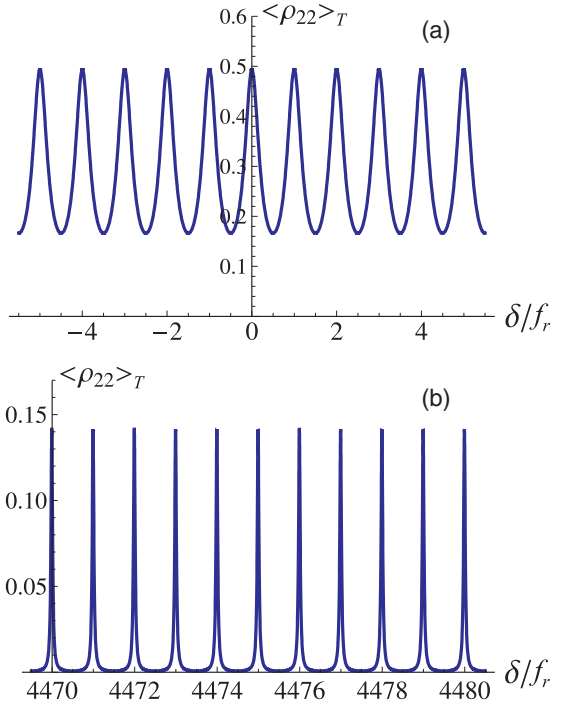

FIG. 7. The line shapes for direct frequency comb spectroscopy [see Fig. 5(a)] over two small intervals of detuning δ, where the narrow resonances are visually separated. Calculations are done for $\Gamma = \gamma/2 = 0.02 f_r$, $\tau/T = 0.001$, $\Omega_0\tau = \pi/2$.

where $\Omega(t) = d\mathcal{E}(t)/\hbar$ is the time-dependent Rabi frequency (d is the dipole matrix element), and $\delta = (\omega - \omega_0)$ is the detuning of the central frequency ω. The constants γ and Γ describe the relaxation rates of the exited state population (ρ_{22}) and optical coherence (ρ_{12}, ρ_{12}), respectively. Note that for pure spontaneous relaxation the condition $\Gamma = \gamma/2$ takes place.

Of most interest is a regime in which the repetition frequency significantly exceeds the relaxation rate of atomic system: Γ, $\gamma \ll f_r$. Figure 6 demonstrates the dependences of period-averaged value $\langle\rho_{22}\rangle_T$ as a function of detuning δ for different pulse areas. The structure of these line shapes consist of large number of resonances at the frequencies $\omega = \omega_0 + mf_r$ ($m = 0, \pm 1, \pm 2, \ldots$). Their amplitudes lie between two envelope curves. The dark areas in Fig. 6 correspond to the resonances, which are visually inseparable for the given scale. A more detailed picture (using small frequency intervals) is presented in Fig. 7, where the individual resonances are visible.

As seen in Figs. 6(b)–6(d), under condition $\Omega_0\tau \geqslant 1$ the dependences become quite unusual. We have found that the lower envelope has a typical width of the order of $2\Omega_0$ [see in Figs. 6(b)–6(d)] and it looks similar to the line shape on δ in the case of single-pulse Rabi spectroscopy. The upper envelope is much more broadened (with width of the order of $\Omega_0 f_r/\gamma \gg \Omega_0$ in the case of $\Omega_0\tau \geqslant 1$). Moreover, this envelope curve has a series of relatively narrow resonance-like dips (practically to zero), which can be characterized as quasiforbidden spectroscopic zones. The positions of these zones are determined from the condition $\tau\sqrt{\Omega_0^2 + \delta^2} = 2k\pi$ ($k = 1,2,\ldots$), and their widths are proportional to γ, i.e., they tend to zero if $\gamma \to 0$.

Note the high efficiency and simplicity of our method in the case of periodic pulse modulation. Indeed, for the above considered task with rectangular pulses [see Fig. 5(a)] the operator $\widehat{A}(t_0+T,t_0)$ includes the product of only two matrix exponents:

$$\widehat{A}(t_0+T,t_0)=e^{\tau\widehat{L}(\Omega=\Omega_0)}\,e^{\mathcal{T}\widehat{L}(\Omega=0)}, \tag{30}$$

where $\mathcal{T}=(T-\tau)$ is a free evolution time between pulses. As a result, the calculation of the dynamic steady state in concordance with Eq. (18) becomes quite simple. Moreover, for a two-level atom we have obtained the analytical expressions (exact and approximative) for the operator $\widehat{A}(t_0+T,t_0)$ and vector $\vec{\rho}(t_0)$. In particular, for the short pulses ($\tau \ll \min\{1/\gamma; 1/\Gamma; T\}$) we can neglect the relaxation processes during the pulse duration τ. In this case, the atomic evolution is described by the Schrödinger equation, which leads to the following approximation:

$$e^{\tau\widehat{L}(\Omega=\Omega_0)} \approx \begin{pmatrix} |w|^2 & \frac{\Omega_0 w^*\mathrm{Re}\{w\}}{\delta} & \frac{\Omega_0 w\mathrm{Re}\{w\}}{\delta} & \frac{\Omega_0^2\mathrm{Re}^2\{w\}}{\delta^2} \\ \frac{\Omega_0 w^*\mathrm{Re}\{w\}}{\delta} & -(w^*)^2 & \frac{\Omega_0^2\mathrm{Re}^2\{w\}}{\delta^2} & -\frac{\Omega_0 w^*\mathrm{Re}\{w\}}{\delta} \\ \frac{\Omega_0 w\mathrm{Re}\{w\}}{\delta} & \frac{\Omega_0^2\mathrm{Re}^2\{w\}}{\delta^2} & -w^2 & -\frac{\Omega_0 w\mathrm{Re}\{w\}}{\delta} \\ \frac{\Omega_0^2\mathrm{Re}^2\{w\}}{\delta^2} & -\frac{\Omega_0 w^*\mathrm{Re}\{w\}}{\delta} & -\frac{\Omega_0 w\mathrm{Re}\{w\}}{\delta} & |w|^2 \end{pmatrix}, \tag{31}$$

where the value w is determined as

$$w=\frac{\delta}{V}\sin(V\tau/2)+i\cos(V\tau/2);\quad V=\sqrt{\Omega_0^2+\delta^2}. \tag{32}$$

The atomic evolution between pulses is determined only by the relaxation processes:

$$e^{\mathcal{T}\widehat{L}(\Omega=0)}=\begin{pmatrix} e^{-\gamma\mathcal{T}} & 0 & 0 & 0 \\ 0 & e^{-(\Gamma-i\delta)\mathcal{T}} & 0 & 0 \\ 0 & 0 & e^{-(\Gamma+i\delta)\mathcal{T}} & 0 \\ 1-e^{-\gamma\mathcal{T}} & 0 & 0 & 1 \end{pmatrix}. \tag{33}$$

Note that Eqs. (31) and (33) are written according to definition (6) for the vector $\vec{\rho}(t)$.

Using Eqs. (31)–(33) for the operator (30), we find the solution $\vec{\rho}(t_0)$ of Eq. (18) in analytical form (see also comments [46]). For example, the exited state population has the following approximation:

$$\begin{aligned}\rho_{22}(t_0)\approx{}&\Omega_0^2 e^{\gamma\mathcal{T}}(e^{2\Gamma\mathcal{T}}-1)\sin^2(V\tau/2)\big\{(e^{(2\Gamma+\gamma)\mathcal{T}}-1)V^2\\ &+2e^{\Gamma\mathcal{T}}(e^{\gamma\mathcal{T}}-1)V\delta\sin(\delta\mathcal{T})\sin(V\tau)\\ &-e^{\Gamma\mathcal{T}}(e^{\gamma\mathcal{T}}-1)\cos(\delta\mathcal{T})\big[\Omega_0^2+\big(2\delta^2+\Omega_0^2\big)\cos(V\tau)\big]\\ &-(e^{2\Gamma\mathcal{T}}-e^{\gamma\mathcal{T}})\big[\Omega_0^2\cos(V\tau)+\delta^2\big]\big\}^{-1}.\end{aligned} \tag{34}$$

For the pure spontaneous relaxation ($\Gamma=\gamma/2$) expression (34) can be transformed to the form

$$\begin{aligned}\rho_{22}(t_0)\approx{}&\Omega_0^2\exp(\gamma\mathcal{T}/2)\sin^2(V\tau/2)\\ &\times\big\{2V^2\cosh(\gamma\mathcal{T}/2)+2V\delta\sin(\delta\mathcal{T})\sin(V\tau)\\ &-\cos(\delta\mathcal{T})\big[\Omega_0^2+\big(2\delta^2+\Omega_0^2\big)\cos(V\tau)\big]\big\}^{-1}.\end{aligned} \tag{35}$$

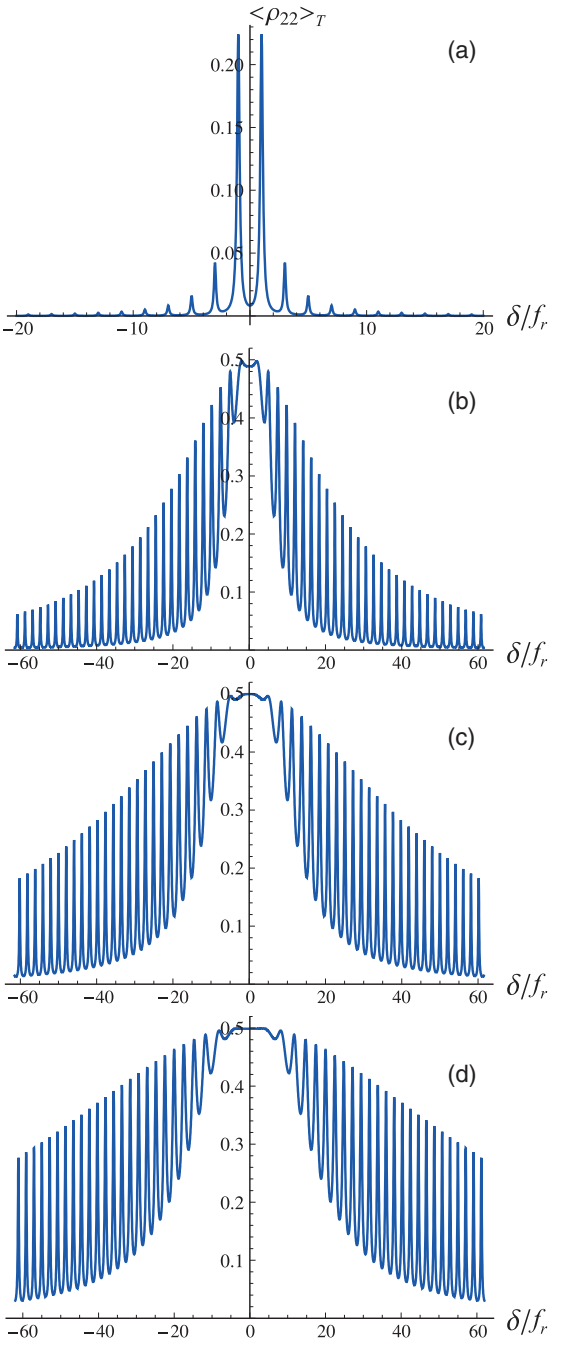

FIG. 8. The spectroscopic line shapes for the field modulation which is formed by the (±)-phase jumps [see Fig. 5(b)]. Calculations are done for $\Gamma=\gamma/2=0.1\,f_r$ and for different Rabi frequencies: (a) $\Omega_0=0.2\,f_r$; (b) $\Omega_0=5\,f_r$; (c) $\Omega_0=10\,f_r$; and (d) $\Omega_0=15\,f_r$.

Note that Eq. (34) (i.e., if $\Gamma\neq\gamma/2$) allows us to model the nonzero spectral width of the laser field at the carrier frequency ω. In this case, we can use $\Gamma=(\gamma/2+\sigma)$, where the parameter σ is proportional to the given spectral width. If the value σ is comparable with γ (or exceeds it), then Eq. (34) will be more adequate than Eq. (35).

In addition, under condition of $\tau \ll T$ we have a close approximation:

$$\langle\rho_{22}\rangle_T\approx\rho_{22}(t_0)[1-e^{-\gamma T}]/\gamma T, \tag{36}$$

with an absolute inaccuracy less than small value $(\tau/T) \ll 1$. Thus, to reproduce with good accuracy the line shape $\langle\rho_{22}\rangle_T$ we can calculate only the dependence $\rho_{22}(t_0)$ on δ. Note that Eq. (36) can be used for an arbitrary pulse form (not only rectangular).

In contrast, using the Fourier analysis for the frequency comb spectroscopy to numerically solve Eq. (5) we should use the following decomposition:

$$\vec{\rho}(t) = \sum_m \vec{\rho}_m e^{mf_r t} \quad (m = 0, \pm 1, \pm 2, \dots), \tag{37}$$

where the components $\vec{\rho}_m$ satisfy certain recurrent relations. For example, the direct Fourier calculation of the dependences in Fig. 6 requires a huge computational burden.

The last numerical example is shown in Fig. 8. There are the spectroscopy line shapes for the field modulation, which is formed by $(\pm)$-phase jumps [see Fig. 5(b)]. In the case of $\Omega_0 \gg \gamma$, these interesting dependences have the lower resonance-like envelope, which has a width of the order of $2\Omega_0$ [see Figs. 8(b)–8(d)].

V. CONCLUSION

Note that for a free atom gas it is necessary to take into account the motion of atoms, which results in dependence of the density matrix on velocity $\mathbf{v}$. In this case the periodic steady state $\hat{\rho}(t+T,\mathbf{v}) = \hat{\rho}(t,\mathbf{v})$ should be found for each velocity group, and then the spectroscopic signal is computed with the use of velocity averaging. Also our consideration of temporal periodic modulation can be adapted for the case of spatially periodic modulation of field parameters (amplitude, phase, polarization, etc.). It implies the proof of existence and the calculation of the spatially periodic steady state of the atoms (including the problems of laser cooling and trapping of atoms in optical lattices). In addition, apart from the density matrix equations (2), the developed approach can be applied to prove the existence theorem of periodic solution for other differential equation systems for which some linear integrals of motion exist.

In summary, in the framework of density matrix formalism we have rigorously proven the existence theorem of the periodic steady state for an arbitrary periodically driven system. Due to the relaxation processes this state is realized as an asymptotics $(t \to +\infty)$ independently of initial conditions, i.e., periodicity is the main attribute of steady state. The presented proof simultaneously contains a computational algorithm, which uses neither Floquet nor Fourier theories. Our method radically simplifies the calculations for arbitrary types of periodic modulation (including the ultrashort pulses) and opens up great possibilities for analysis and development of new methods in laser physics, nonlinear optics, and spectroscopy. As an important example, we have analytically solved a basic theoretical model of the direct frequency comb spectroscopy, when two-level atoms are driven by the periodic sequence of rectangular field pulses. Also our results are applicable to any area of quantum physics where periodically driven systems are considered. The significance of the obtained results becomes especially obvious due to the infinite variety of possible periodic actions for different quantum systems.

ACKNOWLEDGMENTS

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