FEATURES OF DYNAMICS OF STIMULATED ATOMIC - MOLECULAR RAMAN CONVERSION IN A BOSE - EINSTEIN CONDENSATE

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A set of nonlinear evolution equations describing the dynamics of atoms, molecules, and photons in the course of stimulated atomic-molecular conversion in a Bose-Einstein condensate is derived and studied in the mean-field approximation. It is shown that conversion can be periodic or aperiodic in time, the rate of the process being determined, to a considerable extent, by the initial density of particles and by the initial phase difference. Depending on initial conditions, various conversion modes can be realized. The possibility of stabilization of a special state (of rest) of the system for nonzero initial number densities of particles is predicted. It is pointed out that coherence of a Bose condensate of atoms, molecules, and photons predetermines the possibility of phase control of the conversion process.

1. Introduction

The first observation of Bose-Einstein condensation in low-density atomic vapors [1-5] stimulated substantial progress in experimental and theoretical studies of physical properties of Bose condensate in magnetic traps [6-9] and its dynamics in two-well traps [10-15]. In recent years, investigations into the dynamics of bound atomic-molecular condensates under the conditions when Feschbach resonances are manifested or under the conditions of the atomic-molecular Raman conversion involving two electromagnetic radiation pulses have been attracting special attention.

If the total energy of two colliding atoms is equal to the energy of a quasi-bound diatomic state (a molecule in a strongly excited state), the emergence of resonant bonding of atoms is referred to as Feschbach resonance. The dependence of the Feschbach resonance position on the applied magnetic field determines the unique possibility of continuous variation of inter-atomic scattering length and even sign reversal of the interatomic interaction upon a variation of the field [16]. The magnetic field tunes the levels of the diatomic molecular system so that pairs of atoms are combined into a molecule in a strongly excited state. It was noted in [17] that the concepts atomic pair and molecule in a strongly interacting many-particle system in the region of the Feschbach resonance become conditional to a certain extent. Such resonances were observed, for example, in the Bose condensate of sodium atoms in magnetic fields of 853 G [18] and in cold the gas of ^{85}Rb in a field of 164 G [19]. In sodium, the scattering length a as a function of magnetic field B varied in accordance with theoretical predictions [16]. It was shown in [20-22] that the lifetime of the molecules formed in a strongly excited state is much longer than the time of their relaxation to the lower energy levels. This property was explained in [23] by the action of suppression of inelastic processes of interaction leading to transitions of molecules to deeper levels. However, it was shown in [24, 25] that mechanisms of inelastic collisions (e.g., collision of a molecule with a third atom [25]) exist for which the collision rate

increases. Such collisions determine actual losses in the system of condensed atoms and molecules. In [26], the Feschbach resonance in a time-dependent magnetic field was investigated. In this case, damping of condensate also takes place due to transition of a part of atoms into non-condensate states.

At present, the production of ultracold Bose-condensed molecules of K_2 [27-29], Li_2 [3, 0-22, 30-32], Cs_2 [33-35], Cs_4 [36], Na_2 [2, 18, 37], and Rb_2 [1, 19, 38-41] has been observed.

In [42-45], the idea of optical manipulation of the magnitude and sign of the scattering length in low-temperature atomic gases (i.e., the ideal of the optical Feschbach resonance) was developed. Two-photon atomic photoassociation was observed in [41, 46] when two Raman pulses, which led to binding of pairs of interacting atoms into molecules, acted on the system. The scattering length in this case turned out to be noticeably dependent on the intensity of laser beams and detuning of their frequencies from resonance.

The publications [47-51] by different authors have similar formulations of the problem, the method of their solution, and results. The general approach used by the authors of these publications is the use of the system of coupled Gross-Pitaevskii equations in the mean-field approximation for the amplitudes of atomic and molecular condensate and numerical solution of these equations. The results and conclusions drawn in [47-51] are identical in many respects.

In [47], the coherent formation of a molecular Bose condensate from the atomic one was studied using the parametric interaction, in which the processes of formation and dissociation of molecules are analogous to the processes of second harmonic generation and parametric down-conversion, respectively. The possibility of oscillations of the number densities of atoms and molecules in the system was noted and the existence of a 3D bound atomic-molecular condensate in the form of a localized solitonlike structure even in the absence of a trap was predicted.

It should be noted that quantum dynamics of a two mode atomic-molecular condensate was studied in [52] on the basis of a set of coupled equations for the amplitudes of condensed atoms and molecules. As in [47] only the parametric interaction between the condensates was taken into account.

In [48], the properties of the atomic Bose condensate, which was parametrically coupled with a molecular Bose condensate through stimulated coherent Raman photoassociation, were studied. It is shown that a hybrid atomic-molecular condensate, which is captured in a trap and is dynamically stable even in the absence of the trap, is formed owing to the interaction between particles. If, however, the condensate in the trap turns out to be unstable, self-localized stable drops of an atomic-molecular condensate can be formed after the condensate leaves the trap. Such drops of coherent material waves resemble self-localized optical solitons sustained by the parametric interaction.

In [50], the possibility of forming a hybrid atomic-molecular condensate with a self-consistent density, which exhibits liquidlike properties, was also proved. The energy of the ground state of the condensate per particle was calculated as a function of the density for various resonance detunings. The existence of the energy minimum at a certain value of density was proved.

The Raman photoassociation process was also studied in [53]. It was found that the rate of binding of atoms into a molecule and the rate of its dissociation are on the order of $10^{-10}\, m^{-3}/s$, which is much higher than the rate of spontaneous transitions from excited states of the molecule. This indicates the possibility of rapid coherent atomic-molecular conversion in the condensate.

The advances made in controlling the interaction of ultracold atoms raise the question of the possibility of similar control for ultracold molecules. The resonant interactions between molecules may lead to synthesis of more complex objects than atomic dimers. Moreover, scattering processes for molecules contain a large number of new reaction channels as com-

pared to atomic systems. Like atomic Feschbach resonance, magnetic tuning of interactions between molecules will lead to new processes, controlled chemical reactions, and the synthesis of complex ultracold molecules in regimes in which quantum coherence plays an extremely important role. The authors of [36] observed molecules, collisional resonances in the ultracold gas of Cs_2 which were produced from an atomic Bose-*E*instein condensate using CO_2 laser radiation and were tuned by a magnetic field. These resonances were interpreted in [36] as Feschbach resonances for ultracold Cs_2 molecules, leading to the formation of Cs_4 molecules.

It was shown in [51] that such processes may lead to basically new chemistry (so-called superchemistry), in which strong coherent stimulation of chemical reactions involving ultracold atoms and molecules take place. Atomic-molecular Raman conversion may be the predominant nonlinearity and might lead to new chemical reactions at ultralow temperatures. The theory of photoassociation of Bose-condensate atoms proposed in [41, 47-51, 53-58] ultimately boils down to the theory of atomic-molecular conversion induced by an incident electromagnetic wave. The role of the optical pulses ensuring a Raman transition is reduced to nonlinear coefficients in the interaction Hamiltonian, which depend on the field intensities (i.e., actually, to the use of the preset field approximation for optical pulses). However, photons of both pulses can also stimulate chemical reactions and, hence, their role may turn out to be much more important and diversified. It was shown in [51] that simple atomic-molecular conversion or atomic-molecular conversion induced by a Raman transition boils down to periodic processes of the binding of two atoms into a molecule and its dissociation, the rates of these processes being determined by the density of the initial atoms. In fact, the situation may prove to be more complicated if the time evolution of the photon density in Raman pulses is taken into account directly [59].

2. Formulation of the problem and basic equations

It is interesting to consider the dynamics of atomic-molecular Bose condensates. The Raman process of binding two atoms into a molecule is treated as a single process. We will consider below the results of analysis of this process. We will show that the number densities of initial atoms, molecules, and photons considerably affect the process rate, and we will observe that reactions may be either periodic or aperiodic in time. In addition, a significant role of the phase difference of the initial reaction components will be noted and the possibility of phase control of a chemical reaction will be predicted. Macroscopic coherence of Bose condensates of atoms, molecules, and photons of both pulses predetermines quantum interference of all reaction components and, accordingly, the importance of phase relations.

In principle, the process under investigation could also be referred to as optical Raman nutation under the conditions of atomic-molecular conversion, which involves periodic variation of the populations of the atomic and molecular states under the action of two coherent Raman pulses of laser radiation and periodic enhancement of one of the pulses and suppression of the other pulse.

Let us suppose that two identical free Bose-condensed atoms with zero kinetic energy (zero temperature) and with total energy $E_i=2\hbar\omega_0$ pass to a molecular state with energy $E_m=\hbar\Omega_0$ via an excited molecular state E_u by absorbing and emitting light quanta with energies $\hbar\omega_1$ and $\hbar\omega_2$ (Fig. 1). In this case, two phasecoherent pulses with frequencies ω_1 and ω_2 and with certain values of field amplitudes and phases are used. Binding of each pair of atoms into a molecule leads to a transfer of a photon from the first pulse to the second one. For this reason, enhancement of one pulse and suppression of the other during atomic-molecular conversion could serve as an indication of a coherent process. As regards the intermediate excited level with

energy E_u, it can be omitted from the analysis using the adiabatic passage principle [54]. In [60], stimulated Raman transport of an atomic Bose condensate from one trap to another via a common excited level of the system was studied. The atomic transport was treated as a two-stage process. Numerical solution of the set of equations for the populations of two lower levels and the upper excited level proved that the population of the latter level is negligibly low as compared to those of the lower levels.

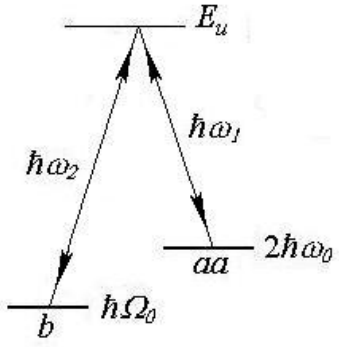

Fig. 1: Schematic diagram of stimulated Raman atomic-molecular conversion. Level $\alpha\alpha$ corresponds to the energy of two free atoms, while level b corresponds to the energy of a free molecule.

The process of stimulated Raman atomic-molecular conversion as a single (one-stage) process induced by short pulses of resonant laser radiation can be described by a model interaction Hamiltonian Hint

$$\hat{H}_{\text{int}} = \hbar g(\hat{\alpha}\hat{\alpha}\hat{b}^{+}\hat{c}_1\hat{c}_2^{+} + \hat{\alpha}^{+}\hat{\alpha}^{+}\hat{b}\hat{c}_1^{+}\hat{c}_2), \tag{1}$$

$\hat{\alpha}$ and $\hat{b}$ ($\hat{\alpha}^{+}$ and $\hat{b}^{+}$) are the boson operators of annihilation (production) of the atomic and molecular states, respectively, $\hat{c}_1$ and $\hat{c}_2$ ($\hat{c}_1^{+}$ and $\hat{c}_2^{+}$) are the annihilation (production) operators for photons with frequencies ω_1 and ω_2, and g is the interaction constant. Essentially, we are proposing a general type of nonlinearity, which may lead to the formation of a molecular Bose condensate via the emission of molecular bosons from the atomic Bose condensate, which is stimulated by two Raman pulses. A similar Hamiltonian describing the process of one-photon atomic-molecular photoassociation was discussed in [54]. Information on the amplitude and other parameters of the incident pulse was contained in the photoassociation coefficient.

As in [48-51, 54-58], we will use the mean-field approximation. The applicability conditions for the mean-field approximations were discussed in detail in [8]. The atomic-molecular Bose condensate is characterized by nonzero order parameters for atomic $\langle\hat{\alpha}\rangle = \alpha \neq 0$, molecular $\langle\hat{b}\rangle = b \neq 0$, and electromagnetic $\langle\hat{c}_{1,2}\rangle = c_{1,2} \neq 0$ fields, which will be henceforth referred to as the amplitudes of the material and electromagnetic fields. Averaging the Heisenberg equations of motion for operators $\hat{\alpha}$, $\hat{b}$, $\hat{c}_1$ and $\hat{c}_2$ ($i\cdot\hbar\cdot\dot{\hat{\alpha}} = [\hat{\alpha}, \hat{H}]$, etc.), we obtain in this approximation the equations of motion for the corresponding amplitudes α, b, c_1 and c_2. In this case, we take into account the fact that the mean value of the product of several operators (under the assumption of complete coherence of the system) is equal to the product of mean values of each operator (i.e., the product of the corresponding field amplitudes). The system of nonlinear differential equations obtained in this way using Hamiltonian (1) and describing the time evolution of the amplitudes of material and electromagnetic fields has the form

$$
\begin{aligned}
i\dot{\alpha} &= \omega_0\alpha + 2g\alpha^* bc_1^* c_2,\\
i\dot{b} &= \Omega_0 b + g\alpha\alpha c_1 c_2^*,\\
i\dot{c}_1 &= \omega_1 c_1 + g\alpha^*\alpha^* bc_2,\\
i\dot{c}_2 &= \omega_2 c_2 + g\alpha\alpha b^* c_1.
\end{aligned}
\tag{2}
$$

This system of equations should be supplemented with the initial conditions, which can be represented in the form

$$\alpha|_{t=0}=\alpha_0 e^{i\varphi_0},\ b|_{t=0}=b_0 e^{i\psi_0},\ c_1|_{t=0}=c_{10}e^{i\psi_{10}},\ c_2|_{t=0}=c_{20}e^{i\psi_{20}}, \tag{3}$$

where each variable is characterized by its own initial amplitude and phase.

It can be seen from Eqs. (2) that stimulated Raman atomic-molecular conversion in the condensate is significantly nonlinear in the amplitudes of both material and electromagnetic fields. Further, we introduce the particle number densities, $n=|\alpha|^2$, $N=|b|^2$, $f_1=|c_1|^2$, $f_2=|c_2|^2$, and two polarization components $Q=i\,(\alpha\alpha b^* c_1 c_2^* - \alpha^*\alpha^* b c_1^* c_2)$ and $R=\alpha\alpha b^* c_1 c_2^* + \alpha^*\alpha^* b c_1^* c_2$.

Using Eqs. (2), we arrive at the following set of nonlinear differential equations

$$\dot{n} = 2gQ,\quad \dot{N} = -gQ,\quad \dot{f}_1 = gQ,\quad \dot{f}_2 = -gQ, \tag{4}$$

$$\dot{Q} = \Delta R + 2gn\left[(4N-n)f_1 f_2 + Nn(f_2 - f_1)\right], \tag{5}$$

$$\dot{R} = -\Delta Q, \tag{6}$$

where $\Delta=2\omega_0-\Omega_0+\omega_1-\omega_2$ is the resonance detuning. Using conditions (2), we can write the initial condition for the particle number densities

$$n\big|_{t=0} \equiv n_0 = |\alpha_0|^2,\ N\big|_{t=0} \equiv N_0 = |b_0|^2,\ f_1\big|_{t=0} \equiv f_{10} = |c_{10}|^2,\ f_2\big|_{t=0} \equiv f_{20} = |c_{20}|^2, \tag{7}$$

and the expressions for the polarization components

$$Q\big|_{t=0} \equiv Q_0 = 2n_0\sqrt{N_0 f_{10} f_{20}}\,\sin\Theta_0,\quad R\big|_{t=0} \equiv R_0 = 2n_0\sqrt{N_0 f_{10} f_{20}}\,\cos\Theta_0, \tag{8}$$

where

$$\Theta_0=\psi_0-2\varphi_0+\psi_{20}-\psi_{10} \quad — \tag{9}$$

is the initial phase difference.

Solving the set of equations (4) - (6) under conditions (7) and (8), we obtain three independent integrals of motion for the number densities of particles

$$n + 2N = n_0 + 2N_0,\ f_1 + N = f_{10} + N_0,\ f_2 - N = f_{20} - N_0 \tag{10}$$

as well as the expressions for functions R and Q

$$R = \frac{\Delta}{g}(N - N_0) + 2n_0\sqrt{N_0 f_{10} f_{20}}\,\cos\Theta_0, \tag{11}$$

$$
\begin{aligned}
Q^2 = &-\left[\frac{\Delta}{g}(N - N_0) - 2n_0\sqrt{N_0 f_{10} f_{20}}\,\cos\Theta_0\right]^2 +\\
&+ 4N(n_0 + 2N_0 - 2N)^2(N_0 + f_{10} - N)(f_{20} - N_0 + N).
\end{aligned}
\tag{12}
$$

Using further the equation $\dot{N} = -gQ$ from set (4) and the expression for $Q(N)$ from (12), we can obtain a formal solution in quadratures for the density of molecules $N(t)$ in the form of the hyperelliptic integral

$$\int_{N_0}^{N(t)} dN\cdot\left[Q(N)\right]^{-1} = \pm gt. \tag{13}$$

Analyzing the process of atomic-molecular conversion as the chemical reaction $\alpha\alpha c_1 \leftrightarrow bc_2$, in which two atoms are combined into a molecule and which involves two Raman photons c_1 and c_2, we will be interested in the concentration $N(t)$ of the molecules formed in this

case as a function of time for various values of the densities of initial reaction components and other parameters of the system. In this case, the time variation of other system components (n, f_1, f_2) can be determined using integrals of motion (10). This reaction can proceed in both directions and can be self-stimulating, periodic, or aperiodic in time.

Let us analyze the time evolution of the system under exact resonance (Δ=0), when the difference $\hbar(2\omega_0-\Omega_0)$ between the energies of the diatomic and the molecular states is exactly equal to the difference $\hbar(\omega_2-\omega_1)$ of the two photons. Using expression (12), the time-evolution equation for molecules in this case can be written in the form

$$\left(\frac{dN}{dt}\right)^2 + W(N) = E_0, \tag{14}$$

where

$$W = -16N\left(N_0 + \tfrac{1}{2}n_0 - N\right)^2\left(N_0 + f_{10} - N\right)\left(f_{20} - N_0 + N\right), \tag{15}$$

$$E_0 = -4n_0^2 N_0 f_{10} f_{20}\, cos^2\,\Theta_0. \tag{16}$$

*E*xpression (14) can be treated as the equation describing the oscillations of a nonlinear oscillator, where $(dN/dt)^2$ plays the role of the kinetic energy of the oscillator, while $W(N)$ and E_0 are the potential and total energy, respectively. It can be seen from relation (16) that the total energy E_0 of the oscillator is zero if the density of one of the components of the system is equal to zero at the initial instant. The effect of the initial phase difference Θ_0 on the evolution of the system vanishes in this case. An important feature of (14) is that the evolution of the system is impossible if atoms are absent (n_0=0) at the initial instant; i.e., the system remains at rest even if the quantities of the remaining three components differ from zero for t=0. This means that the dissociation of molecules into atoms is impossible for n_0=0. If, however, we assume that one of three other quantities (N_0, f_{10} or f_{20}) is zero at t=0, the evolution of the system takes place (i.e., the densities of all particles vary with time). It can also be easily seen that if any of two densities are zero at the initial instant, the evolution of the system is also impossible. It follows from Hamiltonian (1) that it is due to complex inducting of the process.

3. Preliminary qualitative conclusions

The behavior of the $N(t)$ function can be explained qualitatively if we analyze the dependence of the potential energy of a nonlinear oscillator for various relations between the parameters. For zero total energy (E_0=0), the time variation of function $N(t)$ is possible in the domain of values of this function, where $W(N)\leq 0$. Figure 2 shows the potential energy curves $W(N)$ for various relations between the initial number densities of particles. Each potential energy curve corresponds to two types of the time evolution of function $N(t)$, which differ in the sign of the derivative $dN/dt\big|_{t=0} \equiv \dot{N}_0$ at the initial instant for the same value of density $\dot{N}_0$ of molecules at this instant. In turn, the sign of is determined by the sign of quantity $Q\big|_{t=0} = Q_0$ or by the sign of function $sin\Theta_0$ (this follows from the equation $\dot{N} = -gQ$ and the expression for Q_0 from (8). Thus, the initial phase difference is a substantial control parameter of the system. The time evolution of the system and, in particular, the direction of this evolution beginning from instant t=0 depend on the degree of readiness of the system at the initial instant (i.e., by the initial phase difference Θ_0). Consequently, the same value of the initial density N_0 of the molecules may correspond to two values of $\dot{N}_0$, which differ only in sign ($\pm\dot{N}_0$) for values of Θ_0 equal, for example, to $+\pi/2$ and $-\pi/2$.

The curves depicted in Fig. 2 show that the oscillatory mode of the evolution of function

$N(t)$ for $\Theta_0=\pm\pi/2$ is possible only provided that $f_{10}=0$, including the case $f_{10}\le n_0/2$. For any other relations between parameters n_0, N_0, f_{10} and f_{20} only aperiodic regimes of time evolution take place. It can be seen from Figs. 2a and 2b that aperiodic regimes of evolution are observed for $f_{20}>N_0$ and $f_{10}\ge n_0/2$; for $N_0>0$, the density of molecules increases with time and asymptotically tends to a value of $N_0+n_0/2$. If, however, $N_0<0$, the density of molecules first decreases to zero, after which it increases and again tends asymptotically to a value of $N_0+n_0/2$. For $f_{20}>N_0$ and $f_{10}<n_0/2$, the oscillatory mode of evolution of the molecular density takes place, the value of $N(t)$ varying from zero to N_0+f_{10} for $f_{20}>N_0$ (Fig. 2c) and from N_0-f_{20} to N_0+f_{10} for $f_{20}<N_0$ (Fig. 2i). In this case, two oscillatory modes for each of the $W(N)$ curves can emerge depending on the sign of N_0, the oscillation period being independent of the sign of N_0. For $f_{20}=N_0$ and $f_{10}\ge n_0/2$, evolution is aperiodic; in this case, function $N(t)$ increases monotonically with time from N_0 to $N_0+n_0/2$ for $N_0>0$ and decreases monotonically from N_0 to zero for $N_0<0$ (Figs. 2d, 2f). If, however, $f_{10}<n_0/2$, the density of molecules first increases, attains its maximum value N_0+f_{10}, and then monotonically decreases to zero for $N_0>0$ or monotonically decreases from N_0 to zero for $N_0<0$ (Fig. 2f). For $f_{20}<N_0$ and $f_{10}\ge n_0/2$, the density of molecules increases monotonically from N_0 to $N_0+n_0/2$ for $N_0>0$ or first decreases from N_0 to N_0-f_{20} and then increases, asymptotically approaching $N_0+n_0/2$ for $N_0<0$ (Figs. 2g, 2h). If, however, $f_{10}<n_0/2$, the oscillatory evolution mode takes place and function $N(t)$ oscillates between N_0-f_{20} and N_0+f_{10} (see Fig. 2i).

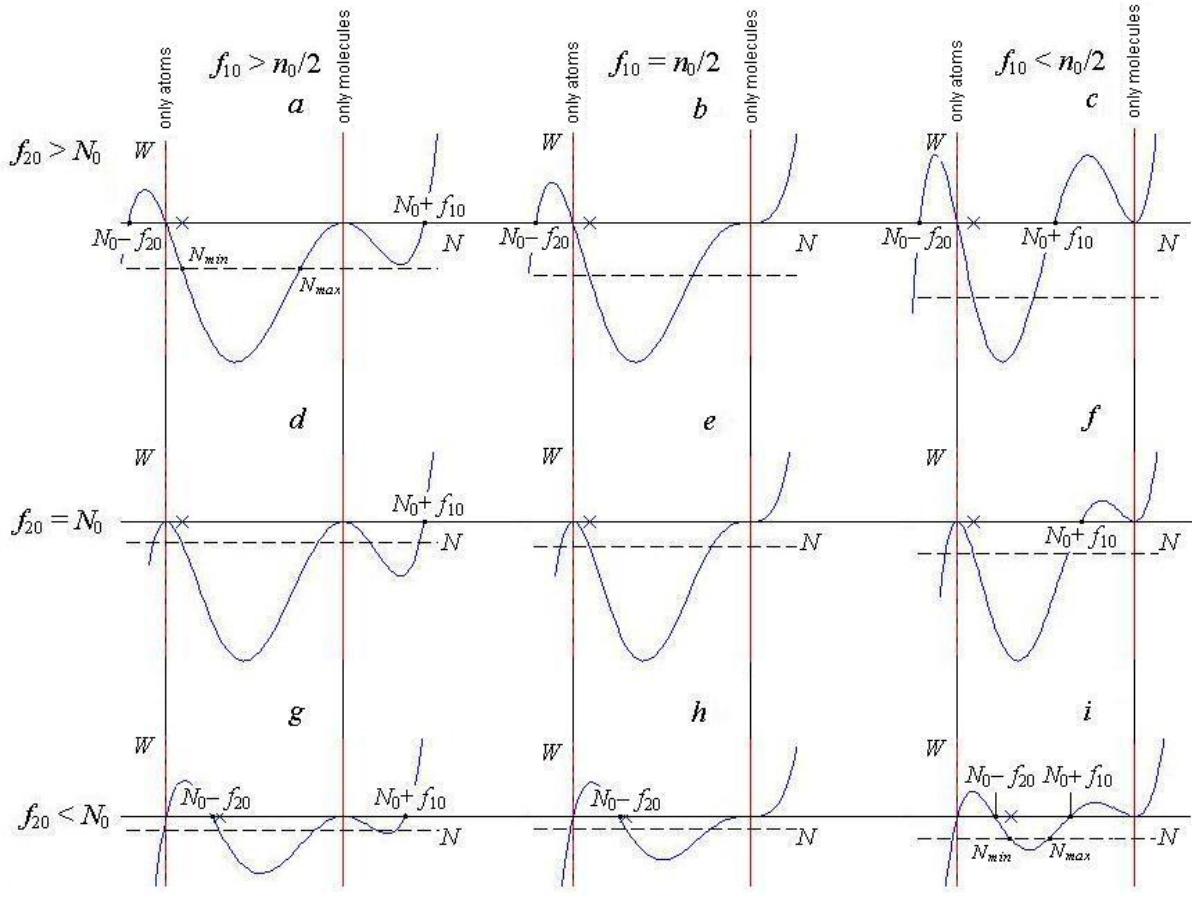

Fig. 2: Curves describing the potential energy $W(N)$ for $\Theta_0=\pi/2$ and for various relations between parameters f_{10}, f_{20}, N_0 n_0: a, b, c: $f_{20}>N_0$; d, e, f: $f_{20}=N_0$; g, h, i: $f_{20}<N_0$; a, d, g: $f_{10}>n_0/2$; b, e, h: $f_{10}=n_0/2$; c, f, i: $f_{10}<n_0/2$. The total energy E_0 of the oscillator is zero for $\Theta_0=\pi/2$ and is negative for $\Theta_0\ne\pi/2$. Dotted line shows maximal value of a total energy E_0, according to $\Theta_0=0$. The scales of various figures are different. The dagger designates an initial condition.

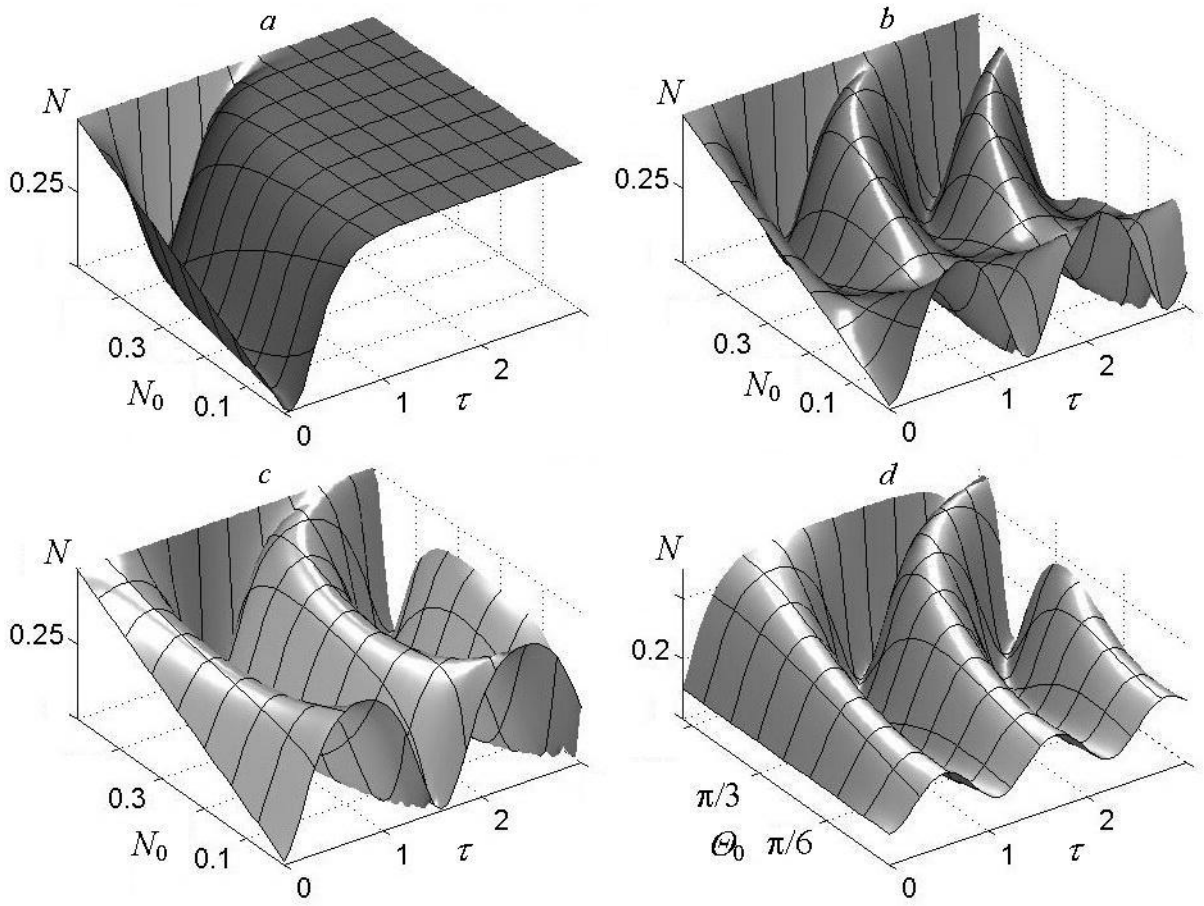

Fig. 3: Time evolution of normalized density of molecules $N(\tau)$ depending on: (a, b, c) the initial density of molecules N_0; (d) the initial phase difference Θ_0 at initial densities of photons of the first and second pulses $f_{10}=f_{20}=2$ in the approximation $f_{10}, f_{20} >> n_0, N_0$. The initial phase difference Θ_0 are: (a) $\pi/2$; (b) 0; (c) $\pi/6$.

It follows from expressions (14)-(16) that the total energy E_0 monotonically decreases with increasing difference from $\Theta_0=\pm\pi/2$ for the initial phase difference $-\pi/2<\Theta_0<\pi/2$ and attains its minimal value for $\Theta_0=0$ ($\Theta_0=\pm k\pi$, k=0, 1, 2, ...). The straight line $E_0<0$ passing below the zeroth level of the potential energy crosses all the curves corresponding to function $W(N)<0$. Two turning points corresponding to $N=N_{\min}$ and $N=N_{\max}$ are formed, between which function $N(t)$ varies. In this case ($\Theta_0=\pm\pi/2$), all aperiodic regimes represented in Fig. 2 are transformed into periodic regimes, the amplitude of oscillations being the smallest for $\Theta_0=0$ (or $\pm k\pi$) for all relations between parameters N_0, n_0, f_{10} and f_{20}. It should be noted that a special oscillatory mode is possible for $\Theta_0=2k\pi$; the amplitude of this mode decreases monotonically upon a change in the initial number densities of particles and is equal to zero for a certain ratio of these quantities. This means that, for a certain choice of the initial state, the system is at rest with nonzero number densities of particles.

4. Different approximations

4.1. Approximation of a preset density of photons of incident pulses

Let us consider solutions in the approximation of a preset density of photons in both pulses: $f_{10}, f_{20}>>n_0, N_0$. This approximation corresponds to the situation depicted in Fig. 2a for

$\Theta_0=+\pi/2$. For density $N(t)$ of molecules at $\Theta_0=\pm\pi/2$, the solution has the form

$$N=\left(N_0+\frac{n_0}{2}\right)\cdot\left[\frac{\sqrt{N_0}\pm\sqrt{N_0+n_0/2}\cdot th\left(2g\sqrt{f_{10}f_{20}\left(N_0+n_0/2\right)}\,t\right)}{\sqrt{N_0+n_0/2}\pm\sqrt{N_0}\cdot th\left(2g\sqrt{f_{10}f_{20}\left(N_0+n_0/2\right)}\,t\right)}\right]^2, \tag{17}$$

the plus and minus signs correspond to the initial conditions $N_0>0$ ($\Theta_0=-\pi/2$)and $N_0<0$ ($\Theta_0=+\pi/2$) (Fig. 3a). It follows from expression (17) that, in the long-time limit $\left(g\sqrt{f_{10}f_{20}\left(N_0+n_0/2\right)}t>>1\right)$, the density of molecules asymptotically tends to the value $N=N_0+n_0/2$. This means that, in the long run, all atoms are combined into molecules. The solution with the plus sign increases with time monotonically, the growth rate being the higher, the larger the number densities f_{10}, f_{20}, N_0 and n_0 (Fig. 3a). The solution with the minus sign first decreases, attains zero at the instant

$$t=t_0=arth\left(\sqrt{N_0/\left(N_0+n_0/2\right)}\right)\Big/\left(2g\sqrt{f_{10}f_{20}\left(N_0+n_0/2\right)}\right), \tag{18}$$

then begins to increase, and asymptotically approaches the value $N=N_0+n_0/2$ as in the case with the plus sign (Fig. 3a). Thus, over long periods of time, both solutions asymptotically approach the same value $N=N_0+n_0/2$ of the density of molecules. All atoms available in the system are bound into molecules, thus completing the process of evolution. The reverse reaction accompanied by the decomposition of molecules does not take place. Thus, we can state that, in this approximation, the reaction of binding of all atoms into molecules is aperiodic for $\Theta_0=\pm\pi/2$, although the solution with the minus sign describes the reaction of rapid dissociation of all molecules at the initial stage, followed by their recovery and generation of an additional number of molecules due to binding of the initial atoms.

The evolution of the system in this approximation f_{10}, $f_{20}>>n_0$, N_0, but for the initial phase difference $\Theta_0=0$, π ($\pm k\pi$, $k=0, 1, 2, ...$) is considerably different. It can be seen from expression (16) that the total energy in this case is $E_0=-4\mathrm{n}_0 N_0 f_{10} f_{20}$, while the potential energy is $W(N)=-16 f_{10} f_{20} N (N_0+n_0/2-N)^2$.

It should be noted that, for $\Theta_0=\pm k\pi$ ($k=0, 1, 2...$), the solutions with the plus and minus signs coincide. It can be seen from formulas (8) that $Q_0(k\pi)=0$ and, hence, $\dot{N}\big|_{t=0}=0$. In other words, the two solutions coincide due to the fact that one of the roots of the equation $W(N)=E_0$ coincides with the initial value $N=N_0$. Since this root lies on one of the slopes of the potential energy profile, further evolution of function $N(t)$ immediately after the initial instant occurs in only one direction either with an increase or a decrease in the value of $N(t)$ with time.

Depending on the relation between the initial number densities N_0 and n_0, different solutions exist. For $n_0<4 N_0$, the solution for the density $N(t)$ of molecules is an oscillating function of time (Fig. 3b),

$$N=N_-+\left(N_0-N_-\right)\cdot\frac{cn^2\left(2g\sqrt{f_{10}f_{20}\left(N_+-N_-\right)}\,t\right)}{dn^2\left(2g\sqrt{f_{10}f_{20}\left(N_+-N_-\right)}\,t\right)}, \tag{19}$$

where N_+ and N_- are defined by the formulas

$$N_\pm=\left(N_0+n_0\pm\sqrt{N_0\left(N_0+2n_0\right)}\right)\Big/2. \tag{20}$$

Here and below, *snx*, *cnx* and *dnx* are the Jacobi elliptic functions [61, 62] with modulus k. The modulus of the elliptic functions in expression (19) is defined as

$$k^2=\left(N_0-N_-\right)/\left(N_+-N_-\right). \tag{21}$$

The period of molecular density oscillations is defined by the formula

$$T=K(k)\Big/\left(g\sqrt{f_{10}f_{20}\left(N_+-N_-\right)}\right), \tag{22}$$

where $K(k)$ is a total first-order elliptic integral [61, 62] with modulus k.

For n_0>4 N_0, the solution has the form

$$N = N_0 + (N_- - N_0)\, sn^2\left(2g\sqrt{f_{10} f_{20}(N_+ - N_0)}\, t\right), \tag{23}$$

where modulus k of an elliptic function and the period of oscillations T are given by

$$k^2 = (N_- - N_0)/(N_+ - N_0),\ T = K(k)/\left(g\sqrt{f_{10} f_{20}(N_+ - N_0)}\right), \tag{24}$$

while quantities $N_\pm$ are defined by formula (20) as before. It can be seen from relations (24) and Fig. 3b that the period of oscillations decreases upon an increase in the number densities N_0 of molecules and in densities f_{10} and f_{20} of photons.

Finally, for n_0=4 N_0, the solution has the form

$$N=N_0. \tag{25}$$

It follows hence that, in the limit of preset number densities of photons in both pulses, the system is at rest provided that n_0=4 N_0 at the initial instant (Fig. 3b). This means that the processes of binding of atoms into molecules or dissociation of molecules do not take place.

Amplitude A of oscillations of function $N(t)$ for $\Theta_0=\pm k\pi$ is defined by the formula

$$A = \left|N_0 - n_0 + \sqrt{N_0(N_0 + 2n_0)}\right|/2, \tag{26}$$

which implies that the amplitude of oscillations for Θ_0=0 ($\pm k\pi$) first monotonically decreases upon an increase in parameter n_0/N_0, becomes zero for n_0/N_0=4, and then increases almost linearly.

Figure 3b illustrates the time evolution of the density of molecules in these cases. It can be seen that the density of molecules oscillates with time between $N_0 \le N \le N_-$ for n_0>4 N_0, as well as in the interval $N_- \le N \le N_0$ for n_0<4 N_0, and does not vary with time along straight line. The period T of oscillations decreases monotonically in this case upon an increase in the ratio N_0/n_0 and is equal to $T = \pi/\left(2g\sqrt{3N_0 f_{10} f_{20}}\right)$ for n_0=4 N_0. An increase in the photon number densities f_{10} and f_{20} in both pulses also leads to a monotonic decrease in the period of oscillations.

It should be noted once again that formulas (19), (23) and (25), as well as Fig. 3b, imply that the amplitude of oscillations of the density of molecules continuously decreases upon a gradual increase in the density n_0 of atoms at the initial instant beginning from the state when n_0<4 N_0 and vanishes for n_0=4 N_0. Analogously, beginning from the state when n_0>4 N_0, a gradual decrease in the initial density n_0 of atoms leads to a monotonic decrease in the amplitude of oscillations, which again becomes zero for n_0=4 N_0. In this case, the period of oscillations also varies monotonically. The effect of vanishing of the amplitude of oscillations of molecules upon a change in the initial density of atoms (or molecules) is interesting in that system does not oscillate (is at rest), although the number densities N_0, n_0, f_{10}, f_{20} of all particles differ from zero at the initial instant and the number densities of atoms and molecules satisfy the relation n_0=4 N_0. Apparently, we can state that the system is balanced in this case so that precisely the resultant conversion of particles is absent, although the conversions of each species of particles may take place (e.g., two atoms can combine into a molecule and a molecule can decompose into two atoms simultaneously). The general relation between the number densities of atoms, molecules, and photons in both pulses is preserved in time. It follows from analysis of the expressions for the potential and total energies of the oscillator that this effect takes place only for the initial phase difference $\Theta_0=\pm k\pi$ (k=0, 1, 2, ...). It turns out that only in this case the total energy E_0 of the oscillator is exactly equal to the potential energy minimum. Consequently, at the initial instant, the oscillator is in equilibrium at the bottom of the potential well and has zero initial velocity. Consequently, it cannot move from the equilibrium position and, hence, the oscillator (i.e., the density of molecules) does not oscillate.

For arbitrary values of the initial phase difference Θ_0, the dynamics of the atomic-molecular conversion (in particular, the amplitude and frequency of oscillations of the

density of molecules) is determined by quantity Θ_0 to a considerable extent. The critical values of the density of molecules can be determined from the solutions of the cubic equation

$$N^3\text{-}2N^2(N_0+n_0/2)+N(N_0+n_0/2)^2\text{-}(1/4)n_0^2 N_0 \cos^2\Theta_0=0, \quad (27)$$

which can be derived from the condition $W(N)=E_0$ in the approximation of a preset density of photons in both pulses ($f_{10}, f_{20} >> n_0, N_0$). In exact resonance ($\Delta=0$), the total energy E_0 of a nonlinear oscillator is a function of Θ_0 $E_0 = -4n_0^2 N_0 f_{10} f_{20} \cos^2\Theta_0$, while the potential energy $W(N)$ is independent of Θ_0. Analysis of cubic equation (27) shows that in the entire range of parameters N_0, n_0 and Θ_0 it has three real positive roots, two of which ($N_{\min}$ and $N_{\max}$) determine the amplitude $N_{\max}-N_{\min}$ of oscillations of function $N(t)$. The third root N_1 is always positive and $N_1>N_{\max}>N_{\min}$. The initial value N_0 of the density of molecules lies between $N_{\min}$ and $N_{\max}$. As the value of Θ_0 increases from zero to $\pi/2$, the level of total energy E_0 is gradually displaced upwards, approaching zero when $\Theta_0=\pi/2$. This leads to a change in the coordinates of points of intersection of straight line E_0 with the cubic parabola $W(N)$. The value of the root $N_{\max}$ increases with Θ_0, while the roots N_1 and $N_{\min}$ decrease in value so that $N_{\min}=0$ and $N_1=N_{\max}=N_0+n_0/2$ for $\Theta_0=\pi/2$. If $n_0=4N_0$, the roots $N_{\min}$ and $N_{\max}$ coincide and are equal to N_0 for $\Theta_0=0$, while $N_1=4N_0$; for $\Theta_0=\pi/2$, the roots N_1 and $N_{\max}$ coincide and are equal to $3N_0$, while $N_{\min}=0$. It should be noted that the roots $N_{\max}$ and $N_{\min}$ for $\Theta_0=\pm k\pi$ ($k=0, 1, 2, ...$) are equal to N_+ and N_-, respectively (see formula (20)). In the general case, the above three roots are defined as

$$N_1/N_0 = \frac{1}{3}(2+x)\cdot\left(1+\cos\frac{\alpha}{3}\right), \quad N_{\max}/N_0 = \frac{1}{3}(2+x)\cdot\left(1+\cos\frac{\alpha-2\pi}{3}\right),$$
$$N_{\min}/N_0 = \frac{1}{3}(2+x)\cdot\left(1+\cos\frac{\alpha+2\pi}{3}\right), \quad \cos\alpha = -1+\frac{27x^2}{(2+x)^3}\cos^2\Theta_0, \quad x=\frac{n_0}{N_0}. \quad (28)$$

Function $N(t)$ oscillated in the limits $N_{\min}\le N\le N_{\max}$ so that amplitude of oscillations (Fig. 4a) is defined by formula

$$A = \left(N_0/\sqrt{3}\right)\cdot(2+x)\cdot sin(\alpha/3). \quad (29)$$

The solution to the equation for $N(t)$ in this case has the form

$$N = N_{min} + (N_0 - N_{min})\cdot\left[\frac{cnu\cdot dnu \pm \sqrt{\frac{N_{max}-N_0}{N_0-N_{min}}\cdot\frac{N_1-N_0}{N_1-N_{min}}}snu}{1-\frac{N_0-N_{min}}{N_1-N_{min}}sn^2u}\right]^2, \quad (30)$$

where the variable u, the modulus k of the elliptic functions, and the oscillation period T are defined by the formulas

$$u = 2g\sqrt{f_{10}f_{20}(N_1-N_{\min})}\,t, \quad k^2 = (N_{\max}-N_{\min})/(N_1-N_{\min}),$$
$$T = K(k)\Big/\left(g\sqrt{f_{10}f_{20}(N_1-N_{\min})}\right). \quad (31)$$

Solutions with the plus and minus signs correspond to the initial conditions $N_0>0$ and $N_0<0$. It can be seen from solution (30) that atomic-molecular conversion is a periodic process (Figs. 3c, 3d). The period T of oscillations (Fig. 4b), as well as the maximal ($N_{\max}$) and the minimal ($N_{\min}$) number densities of molecules, depends, to a considerable extent, on the initial values of number densities f_{10}, f_{20}, N_0 and n_0, as well as on the phase difference Θ_0. Assuming that the initial number densities of particles are unchanged and varying only the initial phase difference Θ_0, we can control both the oscillation period and the values of $N_{\min}$ and $N_{\max}$ (i.e., the amplitude of oscillations of the density of molecules).

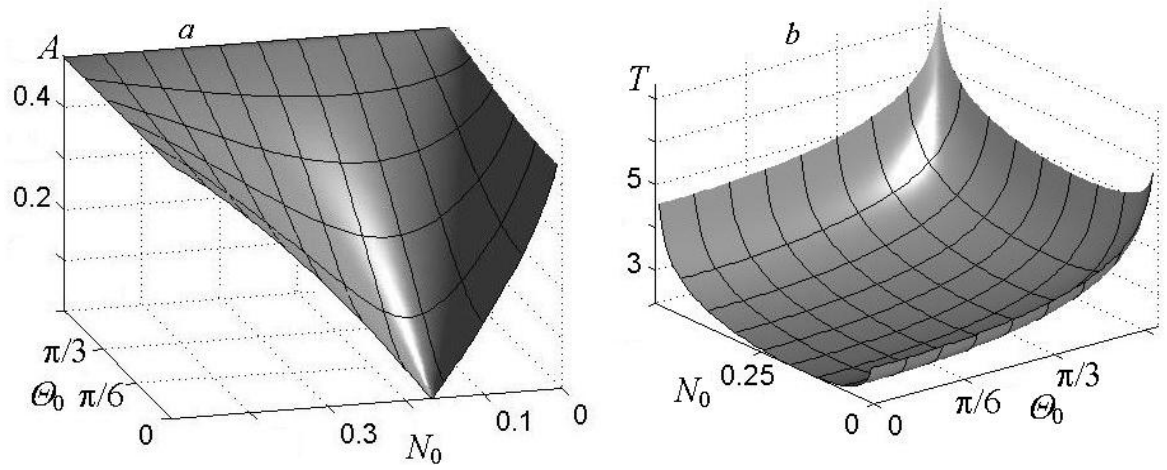

Fig. 4: Dependence of the normalized amplitude of oscillations of the molecule density A (a) and their period T (b) on the normalized density of molecules N_0 and the initial phase difference Θ_0 in the approximation $f_{10}, f_{20} >> n_0, N_0$.

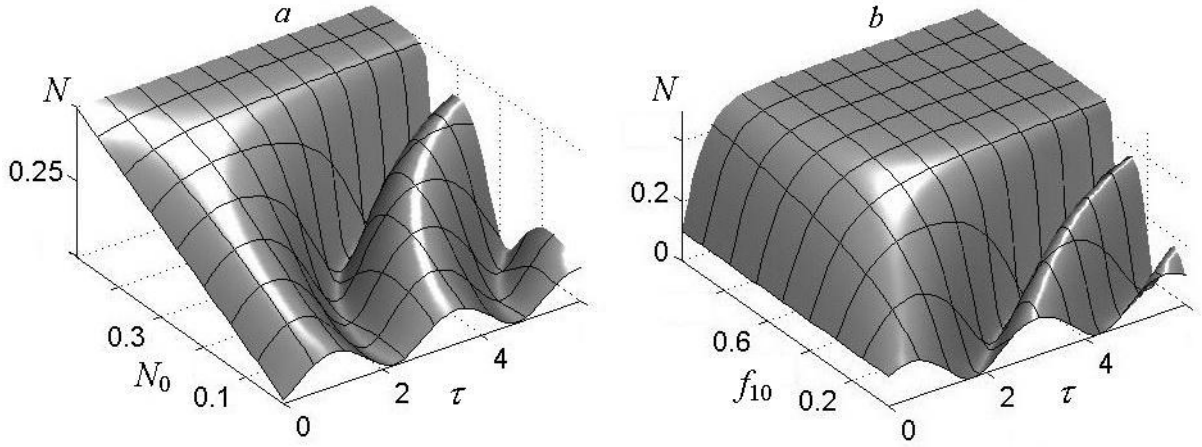

Fig. 5: Time evolution of normalized density of molecules $N(\tau)$ depending on: (a) the initial density of molecules N_0 at f_{10}=0.1; (b) the initial density of photons of the first pulse f_{10} at N_0=0.1 in approximation $f_{20}>>n_0, N_0, f_{10}$. The initial phase difference is $\Theta_0=\pi/2$; the normalized density of photons of the second pulse is f_{20}=2.

4.2. Approximation of a preset density of second-pulse photons

The solutions can also be obtained in the limit of a preset density of photons in each pulse separately. Let us first consider the evolution of the system in the approximation of a preset density of second-pulse photons ($f_{20} >> n_0, N_0, f_{10}$) for the initial phase difference $\Theta_0=\pm(2k+1)\pi/2$ (k=0, 1, 2,...). It follows from formula (12) that three types of solutions exist in this approximation depending on the relation between the parameters f_{10} and n_0.

For $f_{10}<n_0/2$, the solution to the equation for the density $N(t)$ of the molecules has the form

$$N = \left(N_0 + f_{10}\right)\cdot \frac{sin^2\left(2g\sqrt{f_{20}\left(N_0 + n_0/2\right)\left(n_0/2 - f_{10}\right)}\,t \pm C\right)}{1 - \dfrac{N_0 + f_{10}}{N_0 + n_0/2}\cdot cos^2\left(2g\sqrt{f_{20}\left(N_0 + n_0/2\right)\left(n_0/2 - f_{10}\right)}\,t \pm C\right)}, \qquad (32)$$

where

$$C = arctg\sqrt{(N_0/f_{10})(n_0/2-f_{10})/(n_0/2+N_0)}. \quad (33)$$

Figure 5 (a, b) describes the oscillatory mode of the time evolution of the density of molecules in this case. The solutions with the plus and minus signs in the arguments of the trigonometric functions correspond to the conditions $\Theta_0=\pm(2k+1)\pi/2$. It can be seen that the density of molecules oscillates with a phase shift, which is determined by the initial number densities of particles. The period of these oscillations is defined by the formula

$$T = \pi/\left(2g\sqrt{f_{20}(N_0+n_0/2)(n_0/2-f_{10})}\right). \quad (34)$$

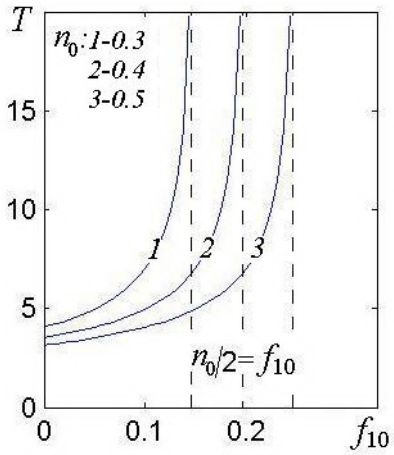

Fig. 6: The period of oscillations of molecule density depending on initial density of photons of the first pulse f_{10} at various values of the initial density of atoms n_0.

As the value of f_{10} increases from zero to $n_0/2$, the oscillation period monotonically increases, turning to infinity for $f_{10}=n_0/2$ (Figs. 5, 6). However, the oscillation period decreases monotonically upon an increase in parameters f_{20}, N_0 and n_0 (Fig. 5a).

For $f_{10}=n_0/2$, we obtain

$$N = (N_0+f_{10})\cdot\frac{\left(\sqrt{N_0}\pm 2g\sqrt{f_{10}f_{20}}(N_0+f_{10})t\right)^2}{f_{10}+\left(\sqrt{N_0}\pm 2g\sqrt{f_{10}f_{20}}(N_0+f_{10})t\right)^2}. \quad (35)$$

The solution with the plus sign monotonically increases with time from the value $N=N_0$ at $t=0$, asymptotically approaching the density N_0+f_{10} for large values of t (Fig. 5b). The solution with the minus sign first decreases from N_0 to the minimal value $N=0$ at $t=t_0$, where

$$t_0 = \sqrt{N_0}/\left(2g\sqrt{f_{10}f_{20}}(N_0+f_{10})\right), \quad (36)$$

after which it begins to increase and asymptotically tends to the value N_0+f_{10} like the solution with the plus sign (Fig. 3b). Thus, over long time periods, all atoms are bound into molecules, thus completing the evolution of the system.

For $f_{10}>n_0/2$, the evolution of the system is also aperiodic. The density of molecules is expressed by the formula

$$N = (N_0+n_0/2)\cdot\frac{sh^2\left(2g\sqrt{f_{20}(N_0+n_0/2)(f_{10}-n_0/2)}\,t\pm D\right)}{ch^2\left(2g\sqrt{f_{20}(N_0+n_0/2)(f_{10}-n_0/2)}\,t\pm D\right)-\dfrac{N_0+n_0/2}{N_0+f_{10}}}, \quad (37)$$

where

$$D = arth\sqrt{(N_0/f_{10})(f_{10}-n_0/2)/(N_0+n_0/2)}. \quad (38)$$

The solution with the plus sign monotonically increases with time from N_0 to $N_0+n_0/2$ (Fig. 5a). The solution with the minus sign first decreases, attains its minimal value $N=0$ for $t=t_0$, which is defined by the expression

$$2g\sqrt{f_{20}(N_0+n_0/2)(f_{10}-n_0/2)}\,t_0 = D\,, \tag{39}$$

and then increases and asymptotically tends to the value $N_0+n_0/2$ for large values of t. Thus, it can be seen that the evolution of the system is aperiodic in time and is completed when all atoms are converted into molecules. The reverse process (i.e., dissociation of molecules) does not occur.

If we assume that the system did not contain molecules ($N_0=0$) at the initial instant, formulas (32) and (34) are substantially simplified and assume the form

$$N = f_{10}\cdot\frac{sin^2\left(g\sqrt{n_0 f_{20}(n_0-2f_{10})}\,t\right)}{1-(2f_{10}/n_0)sin^2\left(g\sqrt{n_0 f_{20}(n_0-2f_{10})}\,t\right)}, \tag{40}$$

$$T = \pi\Big/\left(g\sqrt{n_0 f_{20}(n_0-2f_{10})}\right). \tag{41}$$

It can be seen that the density of molecules for $f_{10}<n_0/2$ varies periodically with period T and amplitude f_{10}. Since $f_{10}<n_0/2$, all photons with frequency ω_1 completely disappear over a period. If $f_{10}<n_0/2$, we have

$$N = f_{10}\,sin^2\left(gn_0\sqrt{f_{20}}\,t\right). \tag{42}$$

In this simplest case, period T_0 of oscillations is defined as

$$T_0 = \pi\Big/\left(gn_0\sqrt{f_{20}}\right). \tag{43}$$

It should be noted that formula (37) leads to formula (17) in the approximation ($f_{10}, f_{20} >> n_0, N_0$).

4.3. Approximation of a preset density of first-pulse photons

Let us now consider the evolution of the system in the approximation of a preset density of first-pulse photons $f_{10} >> n_0, N_0, f_{20}$ for the initial phase difference $\Theta_0=\pm(2k+1)\pi/2$ (k=0, 1, 2, ...). In this approximation, three types of solutions also exist depending on the relation between parameters f_{20} and $N0$. It should be noted that, in accordance with expression (12) and the qualitative results depicted in Fig. 2, the evolution of the system in this approximation is aperiodic for any relations between parameters f_{20}, N_0 and n_0. To be more precise, for all three types of solutions, the evolution of the system boils down to aperiodic binding of all atoms into molecules. True, for $N_0=f_{20}$, the evolution involving the dissociation of all molecules into atoms is possible for the initial condition $N_0<0$ (Figs. 7a, 7b).

Let us briefly consider the above solutions. For $f_{20}>N_0$, we have

$$N = (f_{20}-N_0)\cdot\frac{(2N_0+n_0)\,th^2\left(g\sqrt{f_{10}(2N_0+n_0)(2f_{20}+n_0)}\,t\pm P\right)}{(2f_{20}+n_0)-(2N_0+n_0)\,th^2\left(g\sqrt{f_{10}(2N_0+n_0)(2f_{20}+n_0)}\,t\pm P\right)}, \tag{44}$$

where

$$P = arth\sqrt{(N_0/f_{20})(2f_{20}+n_0)/(2N_0+n_0)}. \tag{45}$$

It can easily be seen that expression (44) can be transformed to (17) in the limit $f_{20}>>N_0$.

It follows from (44) that the solution with the plus sign monotonically increases with time from $N=N_0$ at $t=0$ and asymptotically approaches a limiting density $N_0+n_0/2$. The solution with the minus sign first decreases, attains its minimal value $N=0$ at instant $t=t_0$, and then increases, asymptotically approaching the value $N=N_0+n_0/2$ (Fig. 7b). In this case, the value of t_0 is defined by the formula

$$g\sqrt{f_{10}(2N_0+n_0)(2f_{20}+n_0)}\,t_0 = P. \tag{46}$$

Thus, both solutions (with the plus and minus signs) for large values of time assume the same limiting value of the density of molecules.

For $f_{20}<N_0$, we obtain

$$N = \frac{N_0 - f_{20}}{1 - \dfrac{2f_{20}+n_0}{2N_0+n_0} th^2\left(g\sqrt{f_{10}(2N_0+n_0)(2f_{20}+n_0)}\,t \pm S\right)}, \tag{47}$$

where

$$S = arth\sqrt{(f_{20}/N_0)(2N_0+n_0)/(2f_{20}+n_0)}. \tag{48}$$

Solution (47) exhibits a behavior qualitatively similar to that of solution (44) (Figs. 7a, 7b); however, the solution with the minus sign attains a nonzero minimal value of the molecular density, which is equal to N_0-f_{20} at instant $t=t_0$ (Figs. 7a, 7b), where

$$g\sqrt{f_{10}(2N_0+n_0)(2f_{20}+n_0)}\,t_0 = S. \tag{49}$$

In this case, the evolution is also completed when all atoms are bound into molecules.

Finally, for $f_{20}=N_0$, we obtain

$$N = (N_0 + n_0/2)\Big/\left(1 + \frac{n_0}{2N_0} exp\left(\pm 2g(2N_0+n_0)\sqrt{f_{10}}t\right)\right). \tag{50}$$

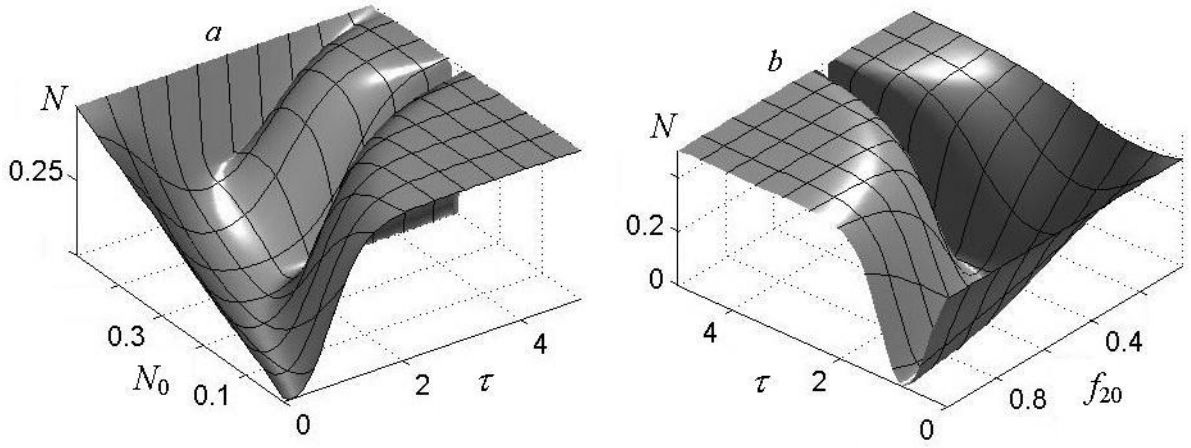

Fig. 7: Time evolution of normalized density of molecules $N(\tau)$ depending on: (a) the initial density of molecules N_0 at $f_{10}=2$, $f_{20}=0.3$; (b) the initial density of photons of the second pulse f_{20} at $f_{10}=2$, $N_0=0.4$ in approximation $f_{10}>>n_0, N_0, f_{20}$. The initial phase difference is $\Theta_0=\pi/2$.

This formula is the limiting case of expressions (44) and (47) for $f_{20}=N_0$. Hence, it follows that the solution with the plus (minus) sign monotonically decreases (increases), asymptotically approaching zero (the value $N_0+n_0/2$) (Figs. 7a, 7b).

Thus, the solutions obtained in the approximation of a preset density of first-pulse photons for $\Theta_0=\pm(2k+1)\pi/2$ indicate that the evolution of the system is an aperiodic process of conversion of all atoms into molecules (or dissociation of all molecules for $f_{20}=N_0$).

4.4. Approximation of a preset density of atoms

Let us now consider the approximation of a preset density of atoms $n_0 >> f_{10}, f_{20}, N_0$ for the initial phase difference $\Theta_0=\pm(2k+1)\pi/2$ ($k=0, 1, 2,...$). This approximation corresponds to the qualitative results represented in Fig. 2 (c, f, i). Detailed behavior of function $N(t)$ is determined

by the relation between number densities N_0 and f_{20}. For $f_{20}<N_0$, the solution to the equation for $N(t)$ has the form

$$N = N_0 \cdot \left(\frac{1 - \frac{f_{20}}{N_0} \cdot sn^2\left(gn_0\sqrt{N_0 + f_{10}}\,t\right)}{dn\left(gn_0\sqrt{N_0 + f_{10}}\,t\right) \pm \sqrt{\frac{f_{10}f_{20}}{N_0\left(N_0 + f_{10}\right)}} \cdot sn\left(gn_0\sqrt{N_0 + f_{10}}\,t\right) \cdot cn\left(gn_0\sqrt{N_0 + f_{10}}\,t\right)} \right)^2, \quad (51)$$

where the period T of oscillations and modulus k of elliptic functions are defined by the formulas

$$T = 2K(k)\Big/\left(gn_0\sqrt{N_0 + f_{10}}\right), \quad k^2 = \left(f_{10} + f_{20}\right)\Big/\left(N_0 + f_{10}\right). \quad (52)$$

For $f_{20}>N_0$, the solution has the form

$$N = N_0 \cdot \left(\frac{cn\left(gn_0\sqrt{f_{10} + f_{20}}\,t\right) \pm \sqrt{\frac{f_{10}f_{20}}{N_0\left(f_{10} + f_{20}\right)}} \cdot sn\left(gn_0\sqrt{f_{10} + f_{20}}\,t\right) \cdot dn\left(gn_0\sqrt{f_{10} + f_{20}}\,t\right)}{1 - \frac{f_{10}}{f_{10} + f_{20}} \cdot sn^2\left(gn_0\sqrt{f_{10} + f_{20}}\,t\right)} \right)^2, \quad (53)$$

where

$$T = 2K(k)\Big/\left(gn_0\sqrt{f_{10} + f_{20}}\right), \quad k^2 = \left(N_0 + f_{10}\right)\Big/\left(f_{10} + f_{20}\right). \quad (54)$$

If, however, $N_0=f_{20}$, the solution has the simple form

$$N = \frac{N_0}{\left(ch\left(gn_0\sqrt{N_0 + f_{10}}\,t\right) \pm \sqrt{\frac{f_{10}}{N_0 + f_{10}}} \cdot sh\left(gn_0\sqrt{N_0 + f_{10}}\,t\right) \right)^2}. \quad (55)$$

It can be seen from the above solutions that the density of molecules for $N_0\neq f_{20}$ oscillates with time (Figs. 8b, 8c). For $f_{20}<N_0$, the density $N(t)$ of molecules varies from N_0-f_{20} to N_0+f_{10}. With increasing f_{20}, the minimal (background) density of molecules decreases, while the period of oscillations increases (Fig. 8c). For $f_{20}=N_0$, the background density of molecules becomes zero (Figs. 8b, 8c). The density of molecules in this case monotonically tends to zero under the initial conditions $N_0>0$; for $N_0<0$, it first increases, attains its maximal value equal to N_0+f_{10} at instant $t = t_0 = arth\left(\sqrt{f_{10}/\left(N_0 + f_{10}\right)}\right)\Big/\left(gn_0\sqrt{\left(N_0 + f_{10}\right)}\right)$ and then monotonically decreases, so that both solutions (with the plus and minus signs) vanish over long time periods. For $f_{20}>N_0$, the density of molecules oscillates from zero to N_0+f_{10} (Figs. 8b, 8c). As regards the period of oscillations, it increases with the ratio f_{20}/n_0, turns to infinity for $f_{20}=N_0$, and then monotonically decreases (Fig. 9).

Let us now consider the evolution of the system for the initial phase difference $\Theta_0=\pm k\pi$ (k=0, 1, 2, ...). In this case, the potential energy $W(N)$ and total energy E_0 of a nonlinear oscillator are defined as

$$W(N) = -4n_0^2 N\left(N_0 + f_{10} - N\right)\cdot\left(f_{20} - N_0 + N\right), \; E_0 = -4n_0^2 N_0 f_{10} f_{20}.$$

The turning points of a classical trajectory can be determined solving the cubic equation $W(N)=E_0$. One of the roots of this equation is the initial value $N=N_0$ of the molecular density. Two other roots of the cubic equations are expressed by the formulas

$$N_{\pm} = \frac{1}{2}\left(N_0 + f_{10} - f_{20} \pm \sqrt{\left(N_0 + f_{10} - f_{20}\right)^2 + 4 f_{10} f_{20}} \right). \quad (56)$$

Root N_- is negative, while the value of N_+ determines the upper or lower limit of oscillations of the molecular density. If parameters N_0, n_0, f_{10} and f_{20} are such that $f_{20}<f_{10}+f_{10}f_{20}/N_0$,

then $N_+ > N_0$. In this case, function $N(t)$ oscillates (Figs. 8a, 8d) in the interval $N_0 < N < N_+$. The solution of the equation for $N(t)$ in this case has the form

$$N = N_- + \left(N_0 - N_-\right) \Big/ dn^2\left(gn_0\sqrt{N_+ - N_-}\,t\right). \tag{57}$$

Modulus k of the elliptic function and period T of oscillations are defined by the formulas

$$k^2 = \left(N_+ - N_0\right)/\left(N_+ - N_-\right),\ T = 2K(k)\Big/\left(gn_0\sqrt{N_+ - N_-}\right). \tag{58}$$

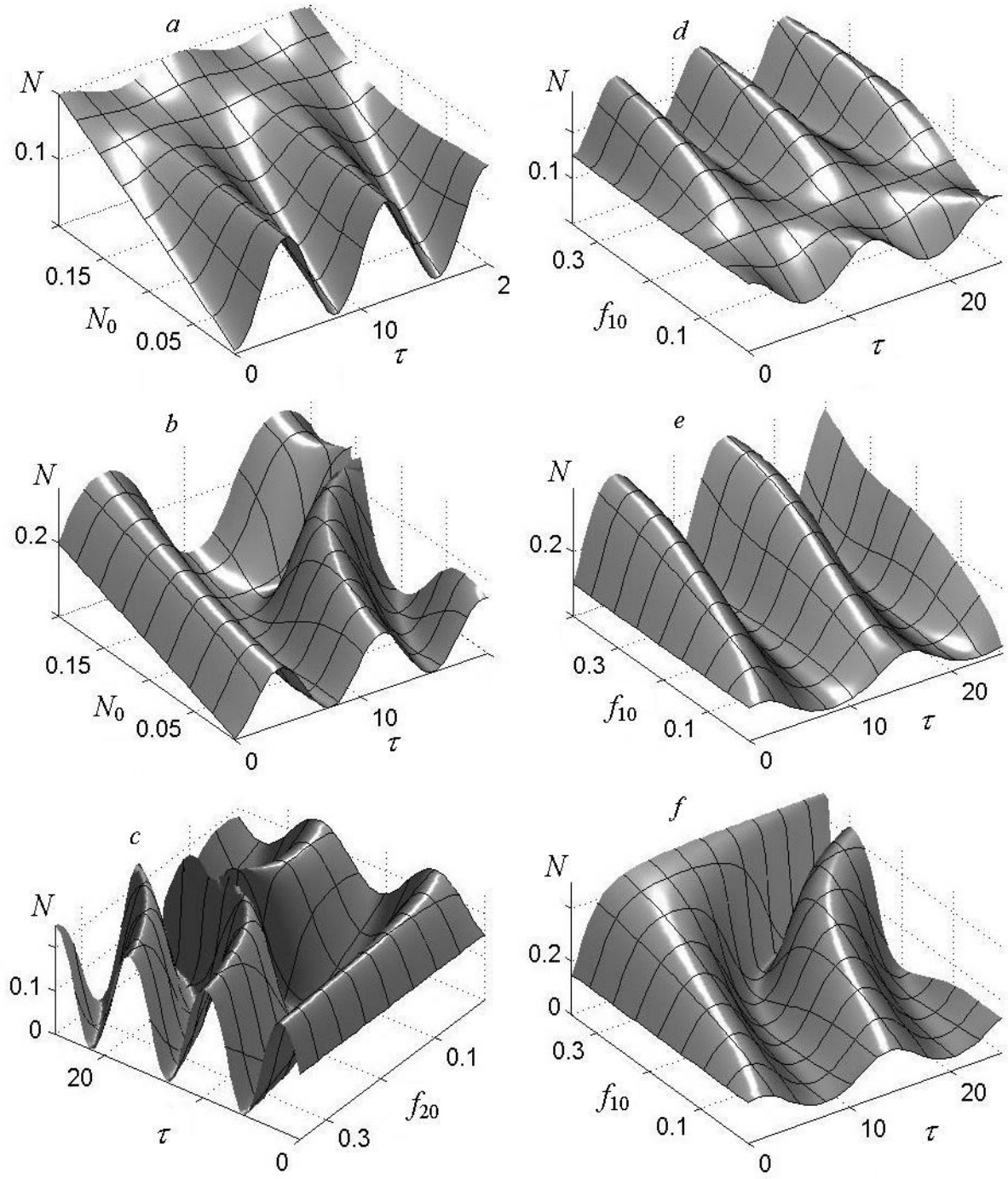

Fig. 8: Time evolution of normalized density of molecules $N(\tau)$ depending on: (a, b) the initial density of molecules N_0 at $f_{10}=f_{20}=0.15$; (c) the initial density of photons of the second pulse f_{20} at $f_{10}=0.1$; (d, e, f) the initial density of photons of the first pulse f_{10} at $f_{20}=0.1$ in the approximation $n_0 >> f_{10}, f_{20}, N_0$. The initial phase difference Θ_0 are: (a, d) 0; (b, c, f) $\pi/2$; (e) $\pi/6$.

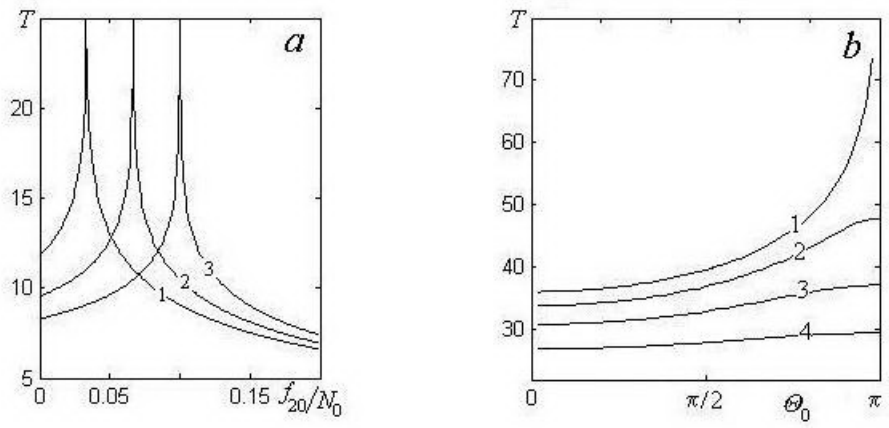

Fig. 9: Dependence of normalized period of oscillations T on: (a) relative density of photons of the second pulse f_{20}/N_0 for case $\Theta_0=\pi/2$ at various values of relative density of molecules N_0/n_0: 0.03 (1), 0.07 (2), 0.1 (3); (b) the initial phase difference Θ_0 at various values of relative density of molecules N_0/n_0: 0.1 (1), 0.14 (2), 0.2 (3), 0.3 (4) in the approximation $n_0>>f_{10}, f_{20}, N_0$.

It should be noted that formulas (56)- (58) are also valid for $f_{10}=f_{20}$. If, however, these parameters satisfy the inverse inequality $f_{20}>f_{10}+f_{10}f_{20}/N_0$, it turns out that $N_+<N_0$. In this case, the solution has the form

$$N = N_0 + \left(N_+ - N_0\right)\cdot sn^2\left(gn_0\sqrt{N_0 - N_-}\,t\right), \tag{59}$$

where

$$k^2 = \left(N_0 - N_+\right)/\left(N_0 - N_-\right),\ T = 2K(k)/\left(gn_0\sqrt{N_0 - N_-}\right). \tag{60}$$

Finally, for $f_{20}=f_{10}+f_{10}f_{20}/N_0$, it turns out that $N_+=N_0$; in this case, the solution has a simple form,

$$N=N_0, \tag{61}$$

i.e., the system is at rest (Figs. 8a, 8d). The same result also follows from expressions (57) and (59) in the limit $N_+=N_0$. In this case, the amplitude oscillations is defined by the formula

$$A = \left|N_0 - f_{10} + f_{10} - \sqrt{\left(N_0 + f_{10} - f_{20}\right)^2 + 4f_{10}f_{20}}\right|\Big/2. \tag{62}$$

These solutions imply that the forms of evolution of the system under initial conditions $\Theta_0=0, \pm\pi$ and $\Theta_0=\pm\pi/2$ are substantially different. It is interesting to consider the solution $N=N_0$, when the system prepared in a certain way does not evolve in time (Figs. 8a, 8d).

Analyzing the general case of the initial condition for the phase difference Θ_0, i.e., assuming that $\Theta_0\neq k\pi$ and $\Theta_0\neq\pm(2k+1)\pi/2$ (k=0, 1, 2,...), we arrive at expressions (15) and (16) for potential energy $W(N)$and total energy E_0. The condition $W=E_0$ leads to the cubic equation

$$N^3 - N^2\left(2N_0+f_{10}-f_{20}\right) + N\left(N_0+f_{10}\right)\left(N_0-f_{20}\right) + N_0 f_{10} f_{20}\cos^2\Theta_0 = 0, \tag{63}$$

which has three real roots $N_{max}>N_{min}>0>N_-$. Root N_- is always negative. Roots N_{max} and N_{min} define the maximal and minimal values of function $N(t)$ in the course of evolution (these values are such that $N_{min}<N_0<N_{max}$). For fixed values of parameters N_0, n_0, f_{10} and f_{20}, the variation of Θ_0 from zero to $\pi/2$ leads to a substantial change in all three roots. As the value of Θ_0 increases from zero to $\pi/2$, root N_{max} monotonically increases, approaching the value N_0+f_{10} at $\Theta_0=\pi/2$. On the contrary, root N_{min} decreases with increasing $\Theta_0=\pi/2$ so that it becomes equal to N_0-f_{20} for $N_0>f_{20}$ or zero for $N_0\leq f_{20}$ for $\Theta_0=\pi/2$. Remaining negative, root N_- decreases in magnitude to zero for $N_0\geq f_{20}$ upon an increase in Θ_0 or approaches the value N_0-f_{20} for $N_0\leq f_{20}$. For $\Theta_0=\pm k\pi$ (k=0, 1, 2, ...), root N_- coincides with expression (56) for N_-, while the values of N_{max} and N_{min} coincide with N_+ from (56) and N_0.

The evolution mode in this case is oscillatory (Fig. 10). The solution for function $N(t)$ has the form

$$N = N_- + (N_0 - N_-)\cdot\left(\frac{1-\frac{N_0-N_{min}}{N_0-N_-}sn^2u}{dnu \pm \sqrt{\frac{(N_0-N_{min})(N_{max}-N_0)}{(N_0-N_-)(N_{max}-N_-)}}\cdot snu\cdot cnu}\right)^2, \quad (64)$$

where variable u, modulus k, and oscillation period T are defined by the formulas

$$u = gn_0\sqrt{N_{max}-N_-}\,t,\ k^2 = (N_{max}-N_{min})/(N_{max}-N_-), T = 2K(k)/\left(gn_0\sqrt{N_{max}-N_-}\right). \quad (65)$$

Fig. 9b shows the curves for the normalized oscillation period T/τ_0 as a function of the initial phase difference Θ_0 for various initial values of concentration N_0/n_0. It can be seen that the oscillation period increases with the initial phase difference and decreases upon an increase in the density of molecules.

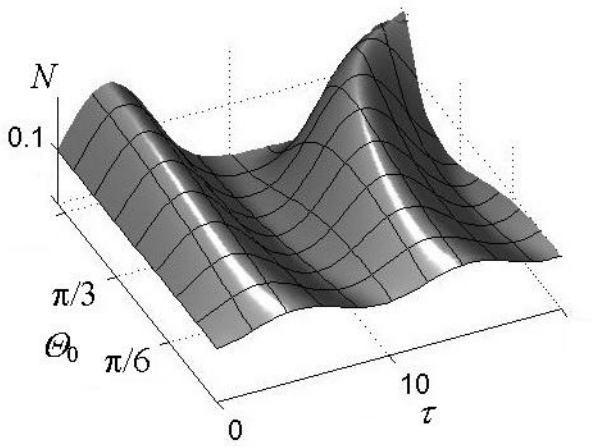

Fig. 10: Time evolution of normalized density of molecules $N(\tau)$ depending on the initial phase difference Θ_0 at initial densities of photons of the first and second pulses $f_{10}=f_{20}=0.1$ in the approximation $n_0>>f_{10}, f_{20}, N_0$.

4.5. Approximation of a preset density of molecules

Let us briefly consider the results obtained in the approximation of a preset density of molecules $N_0>>f_{10}, f_{20}, n_0$ for the initial phase difference $\Theta_0=\pm\pi/2$. If $f_{10}<n_0/2$, the evolution of the molecular density is described by the formula

$$N = N_0 + (f_{10}-f_{20})sin^2\left(gn_0\sqrt{N_0}t\right) \pm \sqrt{f_{10}f_{20}}\,cos\left(2gn_0\sqrt{N_0}t\right). \quad (66)$$

If $f_{10}=n_0/2$, we obtain

$$N = N_0 + f_{10} - \frac{f_{10}+f_{20}}{1+\left(\sqrt{f_{20}/f_{10}} \pm 2g(f_{10}+f_{20})\sqrt{N_0}t\right)^2}. \quad (67)$$

Finally, $f_{10}>n_0/2$, we obtain

$$N = N_0 - f_{20} + \left(f_{20}+\frac{n_0}{2}\right)\cdot\left(\frac{\sqrt{f_{20}+n_0/2}\cdot thx \pm \sqrt{f_{20}}}{\sqrt{f_{20}+n_0/2} \pm \sqrt{f_{20}}\cdot thx}\right)^2, \quad (68)$$

where

$$x = \sqrt{N_0 f_{10}(f_{20}+n_0/2)}\,t. \quad (69)$$

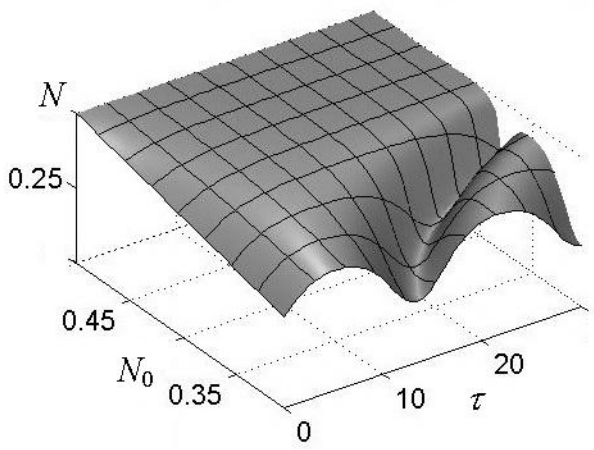

Fig. 11: Time evolution of normalized density of molecules $N(\tau)$ depending on the initial density of molecules N_0 at $f_{10}=f_{20}=0.1$ in the approximation $N_0>>f_{10}, f_{20}, n_0$. The initial phase difference is $\Theta_0=\pi/2$.

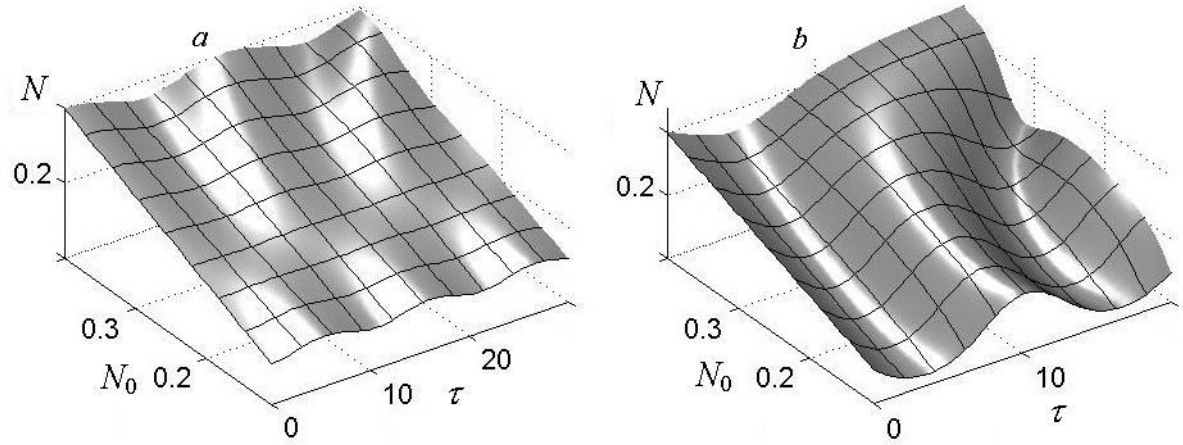

Fig. 12: Time evolution of normalized density of molecules $N(\tau)$ depending on the initial density of molecules N_0 at initial densities of photons of the first and second pulses $f_{10}=0.08$, $f_{20}=0.07$ in the approximation n_0, $N_0>f_{10}, f_{20}$. The initial phase difference Θ_0 are: (a) 0, (b) $\pi/2$.

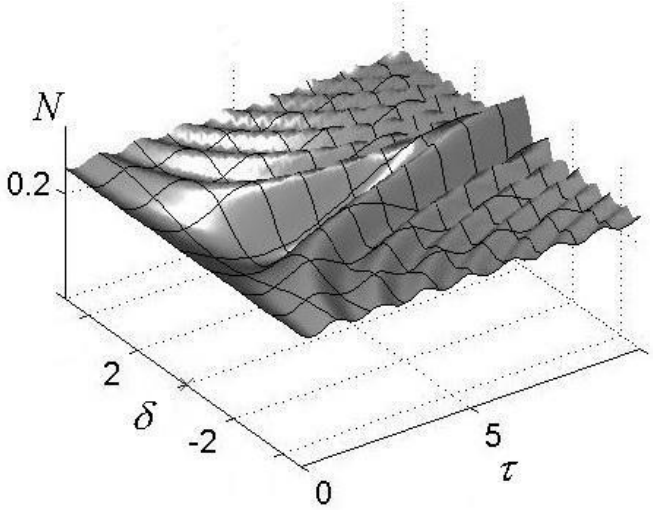

Fig. 13: Time evolution of normalized density of molecules $N(\tau)$ depending on the resonance detuning $\delta=\Delta/g$ at initial densities of photons of the first and second pulses $f_{10}=0.08$, $f_{20}=0.07$ in the approximation n_0, $N_0>f_{10}, f_{20}$. The initial phase difference is $\Theta_0=\pi/2$.

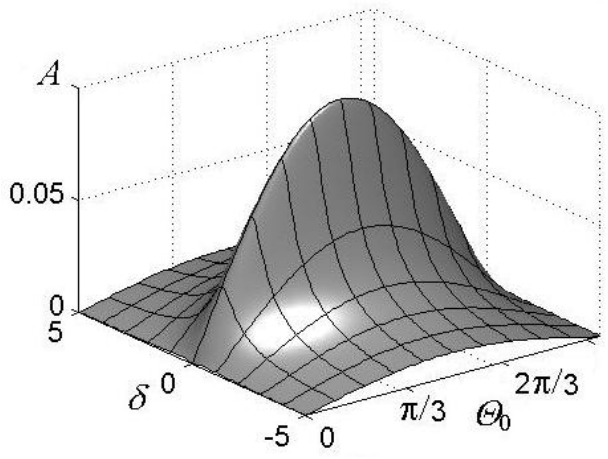

Fig. 14: Normalized amplitude of the molecule density A depending on the normalized resonance detuning $\delta=\Delta/g$ and the initial phase difference Θ_0 in the approximation n_0, $N_0>>f_{10}, f_{20}$.

Hence, it follows that the density of molecules for $f_{10}<n_0/2$ oscillates in time (Fig. 11), varying in the limits $N_0-f_{20}\le N\le N_0+f_{10}$, while for $f_{10}\ge n_0/2$, the molecular density monotonically increases from the value N_0 to $N_0+n_0/2$ at $\Theta_0=-\pi/2$ or first decreases, attains its minimal value N_0-f_{20}, and then increases, asymptotically approaching the value $N_0+n_0/2$ for $\Theta_0=\pi/2$ (Fig. 11). It should be noted that, under the initial condition $\Theta_0=\pm(2k+1)\pi/2$ (k=0, 1, 2,...), the oscillatory mode of evolution of the number density of molecules with a small amplitude is observed for any relation between number densities f_{10} and n_0. In the approximation of the preset density of molecules the oscillation amplitude is approximately equal to $f_{10}+f_{20}$, which is much lower than N_0. For this reason, the evolution of the density of molecules in this approximation has the form of small amplitude oscillations against the background of a high initial density N_0.

4.6. Approximation of a preset density of atoms and molecules

The simplest solutions of the problem can be obtained in the approximation of a preset density of atoms and molecules: n_0, $N_0 >> f_{10}, f_{20}$. Since the problem becomes linear in this approximation, it is convenient to simultaneously take into account the effect of resonance detuning Δ and the initial phase difference Θ_0 on the system dynamics. Using Eq.(12), we can easily derive the following expression for the density $N(t)$ of molecules

$$N = N_0 + \frac{1}{2}(x_+ + x_-) - \frac{1}{2}(x_+ + x_-)\cdot sin\left(\pm\sqrt{\Delta^2 + 4g^2 n_0^2 N_0}\; t + C\right), \tag{70}$$

where

$$x_\pm = \frac{1}{2\cdot\left(1+\dfrac{\Delta^2}{4g^2 n_0^2 N_0}\right)}\left[f_{10} - f_{20} + \frac{\Delta\cdot\sqrt{f_{10}f_{20}}}{4gn_0\sqrt{N_0}}\cos\Theta_0 \pm \right.$$
$$\left. \pm\sqrt{\left(f_{10} - f_{20} + \frac{\Delta\cdot\sqrt{f_{10}f_{20}}}{4gn_0\sqrt{N_0}}\cos\Theta_0\right)^2 + 4\cdot\left(1+\frac{\Delta^2}{4g^2 n_0^2 N_0}\right)\cdot f_{10}f_{20}\cdot\sin^2\Theta_0}\right]. \tag{71}$$

The plus and minus signs in the argument of the sine are determined by the initial phase difference: the minus sign corresponds to $0<\Theta_0<\pi$ and the plus corresponds to $\pi<\Theta_0<2\pi$ (with a period of 2π). The solutions with the plus and minus signs are displaced relative to one another by

$$C = arcsin\left(\left(x_+ + x_-\right)/\left(x_+ - x_-\right)\right). \tag{72}$$

It can be seen that only the oscillatory mode of evolution exists for any value of the initial phase difference. Figure 12 presents the time evolution of the density of molecules in the case when Θ_0=0 (a) and Θ_0=π/2 (b). The oscillation period

$$T = 2\pi \Big/ \sqrt{\Delta^2 + 4g^2 n_0^2 N_0} \tag{73}$$

is independent of the initial number densities f_{10} and f_{20} of photons and the initial phase difference Θ_0, while the oscillation amplitude

$$A = \frac{\sqrt{\left(f_{10} - f_{20} + \dfrac{\Delta \cdot \sqrt{f_{10} f_{20}}}{4gn_0\sqrt{N_0}} cos\,\Theta_0\right)^2 + 4 \cdot \left(1 + \dfrac{\Delta^2}{4g^2 n_0^2 N_0}\right) \cdot f_{10} f_{20} \cdot sin^2\,\Theta_0}}{2 \cdot \left(1 + \dfrac{\Delta^2}{4g^2 n_0^2 N_0}\right)} \tag{74}$$

is determined to a considerable extent by these parameters. This makes it possible to control the dynamics of the system using the initial phase difference Θ_0 and resonance detuning Δ (Figs. 13, 14). The oscillations of the molecular density have the form of fine ripples over the huge background density N_0. It can be seen from Fig. 14 that the oscillation amplitude A has the form resembling the Lorentzian surface depending on resonance detuning Δ and is a periodic function of the initial phase difference Θ_0. The oscillation period T monotonically decreases upon an increase in resonance detuning Δ; an increase in the initial number densities n_0 of atoms and N_0 of molecules leads to a decrease in the oscillation period.

5. Conclusions

The results described above show that the time evolution of the density of atoms, molecules, and photons in the course of stimulated atomic-molecular Raman conversion is determined by the initial number densities N_0, n_0, f_{10} and f_{20} of the particles and by the initial phase difference Θ_0. For Θ_0=$\pm$(2k+1)π/2 (k=0, 1, 2,...), the density of molecules varies with time both aperiodically and periodically, while for $\Theta_0 \neq \pm(2k+1)\pi/2$, the processes of formation and dissociation of molecules can only be periodic. The incident pulses of coherent laser radiation are periodically enhanced or suppressed. By varying the initial phase difference Θ_0 for fixed values of the initial number densities of particles, we can perform phase control of induced Raman atomic-molecular conversion.

The results also show that Bose stimulation of chemical dynamics (binding of Bose-condensate atoms into molecules) is an extremely important circumstance. Collective oscillations of the number densities of atoms and molecules indicate coherence of the system. In a Bose condensate, the chemical conversion considered above is stimulated owing to macroscopic filling of not only the initial atomic system, but also of the final molecular state of a chemical reaction. This feature substantially differs from conventional chemical kinetics, in which the rate of a chemical reaction does not depend on the number of particles in the reaction product and tends to zero at low temperatures in accordance with the Arrhenius law. Raman atomic-molecular conversion in a thermodynamically equilibrium system of atoms at T=0 cannot lead to collective oscillations of this type since the phases of individual atomic-molecular processes are random. In this sense, the processes studied here belong to new chemistry (superchemistry), in which coherent stimulation of chemical reactions takes place. Stimulated quantum dynamics may replace conventional dynamics at ultralow temperatures, providing a new type of collective behavior of the system. The strong dependence of chemical

kinetics on the phase difference indicates the possibility of stabilization of superchemical phase dynamics. In recent years, the problem of phase control of various nonlinear-optics processes has acquired special importance and urgency [63-68]. Specific features of Bose-stimulated chemical dynamics might pave the way for new type of quantum-controlled chemical reactions.

The possibility of the existence of a special evolution mode is predicted, in which the system is at rest in spite of the fact that the number densities of all particles differ from zero since the processes of binding of atoms into molecules and of dissociation of molecules are balanced. This is due to the fact that the total energy of a nonlinear oscillator coincides with the minimum of its potential energy. At the initial instant, the oscillator is at the bottom of the potential well and its initial velocity is zero; in this case, oscillations or any other movements of the oscillator are ruled out.

To estimate the numerical values of oscillation frequencies, it is necessary to know the value of constant g. The values of some parameters for rubidium atoms and molecules are given in [51]. Comparing Hamiltonian (1) with Hamiltonian (2) from [51] to estimate the value of constant g, we can obtain the relation $g = \chi/\hbar f_0$, where constant $\chi/\hbar$ is approximately equal to $7.6\cdot10^{7}\ m^{3/2}/sec$. Assuming that the mean density of photons is $f_{10}=10^{14} cm^{-3}$, 0p we can estimate constant g as $g\approx7.6\cdot10^{18}\ m^{9/2}/sec$. We can now calculate the minimal period $T_0 \approx \pi/g\, n_0\sqrt{f_0}$. For the density $n_0\approx10^{14}$ cm^{-3} of atoms, we obtain $T_0\approx4\cdot10^{-4}$sec, while the oscillation frequency is found to be approximately $1.5\cdot10^{4}\ sec^{-1}$. This result matches the result of numerical experiment [51]. For $n_0=f_0$, we obtain an estimate of parameter τ_0 which was used above $\tau_0=(gn_0^{3/2})^{-1}\approx1.3\cdot10^{-4} sec$.

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