STIMULATED RAMAN ADIABATIC PASSAGE IN PHASE-FLUCTUATING FIELDS

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The phenomenon of stimulated Raman adiabatic passage (STIRAP) in the field of laser pulses with fluctuating phases is studied. The phase fluctuation is described by the Wiener stochastic process. The effect of spontaneous transitions from the excited state on the limiting value of population transfer from the ground state of an atom into the metastable one (or between two metastable states) is discussed. It has been demonstrated that phase fluctuations, besides the restriction on the population transfer efficiency, lead to a shift of the population transfer maximum with respect to the exact two-photon resonance. This shift is proportional to the arithmetic mean of the Stokes and pump field detunings from the single-photon resonance and does not vanish in the limit of high-intensity laser fields.

1. Introduction

The STIRAP phenomenon in atoms and molecules that interact with two laser pulses partially overlapping in time has been intensively studied for last two decades, starting from works [1, 2], where this phenomenon was predicted and experimentally observed for the first time. Its application in various branches of physics and chemistry is associated with the opportunity to populate a certain state of an atom or a molecule (thereinafter, speaking about atoms, we mean molecules as well) with a high efficiency, the probability being close to unity. Nowadays, STIRAP is used for populating high vibrational states of molecules, studying the chemical reactions of vibrationally excited molecules in molecular beams, and in atomic optics [3, 4].

In the elementary case of the three-level Λ-scheme of interaction between an atom and the radiation of two lasers, the possibility of population transfer between atomic states in the course of STIRAP is closely connected with the existence of a trapped or "dark" state which arises provided the two-photon resonance [5–7]. If the frequencies of electromagnetic waves acting upon the atom are close to the frequencies of transitions between states $|1\rangle$, $|2\rangle$ (the pump field) and $|3\rangle$, $|2\rangle$ (the Stokes field), and the frequency difference coincides with the frequency of the transition $|1\rangle \leftrightarrow |3\rangle$, the probability to find the atom in the excited state $|2\rangle$ is close to zero; its state is described by a linear superposition of the basic, $|1\rangle$, and metastable, $|3\rangle$, states, the populations of which are determined by the ratio between the intensities of the Stokes and pump fields. If one changes this ratio slowly, so that the process of atom–field interaction is close to adiabatic, it is possible to transfer the population from state $|1\rangle$ into state $|3\rangle$. For this purpose, it is necessary that the Stokes pulse be the first to act on the atom. Then the pump pulse partially overlapping with the Stokes one in time will act. It is essential that the population of state $|2\rangle$ is low within the whole time interval of atom–field interaction, so that the population losses that are induced by the spontaneous emission from the excited state are close to zero.

The pivot in attaining the high efficiency of the population transfer is the support of the two-photon resonance during all the time of interaction between the atom and the radiation of both lasers. A number of works was devoted to studying the influence of the detuning of the laser pulse carrier frequencies from the one- and two-photon resonances [8–15]. The population transfer efficiency depends much more on the detuning from the two-photon resonance than from the single-photon one [4]. The influence of the uncontrollable detuning from the two-photon resonance, which is caused by phase fluctuations of laser radiation, on the population transfer during STIRAP was studied using both numerical [16, 17] and analytical [18] methods. In essence, the results concerning the influence of the relaxation of off-diagonal elements of the density matrix on the population transfer, which were obtained in work [19], also describe STIRAP in fluctuating-phase fields. In that work, the spontaneous emission from the excited state was neglected, and only the case of two-photon resonance was examined. Whereas in work [18], the spontaneous emission of an atom from the excited state was considered to be accompanied by its transition only into states distinct from $|1\rangle$ and $|3\rangle$ ones, so that the further interaction with the field becomes terminated.

In this work, we consider a more general model studied in work [15] for nonfluctuating fields, which also makes allowance for spontaneous transitions into states $|1\rangle$ and $|3\rangle$, and take advantage of the method developed there for the solution of the Liouville equation for the density matrix. The method is applicable in the case where the period of atom–field interaction is more prolonged than the atom lifetime in the excited state. It will be demonstrated that the availability of those additional relaxation channels changes the fundamental limit of population transfer, to which the probability of population transfer tends as the intensities of the fields grow [18], and results in a new feature, the fundamental shift of the dependence of the population transfer probability on the two-photon detuning from the exact two-photon resonance.

2. Basic Equations

Let a three-level atom be in the field of two light pulses that partially overlap in time (Fig. 1), namely, a pump pulse with the carrier frequency ω_P close to the frequency of the transition between states $|1\rangle$ and $|2\rangle$, and a Stokes pulse with the carrier frequency ω_S close to the frequency of the transition between states $|3\rangle$ and $|2\rangle$:

$$\mathbf{E} = \tfrac{1}{2}\mathbf{E}_P(t)e^{-i\omega_P t - i\varphi_P(t)} + \tfrac{1}{2}\mathbf{E}_S(t)e^{-i\omega_S t - i\varphi_S(t)} + \text{c.c.}$$

The field amplitudes of the Stokes, $\mathbf{E}_P(t)$, and pump, $\mathbf{E}_S(t)$, pulses are supposed to vary smoothly in time with a characteristic time of the order of the pulse duration τ; the phases are assumed to fluctuate. The fluctuations of phases are described by the Wiener process [20], and the time of correlation between phase derivatives τ_{corr} is supposed short in comparison with other time-dimensional quantities in the problem concerned. In this case, the correlation functions for $\xi_P(t) = \dot{\varphi}_P(t)$ and $\xi_S(t) = \dot{\varphi}_S(t)$ look like

$$\langle \xi_P(t)\xi_P(t')\rangle = 2D_P\delta(t-t')$$

and

$$\langle \xi_S(t)\xi_S(t')\rangle = 2D_S\delta(t-t'),$$

respectively. Here, $\delta(t)$ is the Dirac delta-function, the angle brackets $\langle \ldots \rangle$ mean the averaging over the ensemble, and D_P and D_S are the corresponding phase diffusion constants. We are interested in both independent and coinciding-in-time phase fluctuations of the Stokes and pump pulses.

The evolution of the density matrix $\rho(t)$ is described by the quantum-mechanical Liouville equation

$$i\hbar\frac{\partial\rho(t)}{\partial t} = [\mathrm{H}(t), \rho(t)] + \mathrm{R}(t). \quad (1)$$

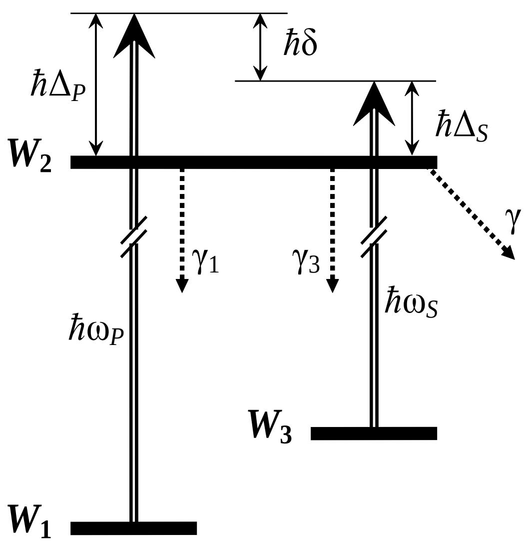

Fig. 1. Scheme of the atom–field interaction. First, the Stokes pulse with the carrier frequency ω_S starts to act upon the atom. Then, the pump pulse with the carrier frequency ω_P and partially overlapped in time with the Stokes pulse starts to act. Notations: W_1, W_2, and W_3 stand for the energies of states $|1\rangle$, $|2\rangle$, and $|3\rangle$, respectively; Δ_P and Δ_S are the detunings from the single-photon resonance; and $\delta = \Delta_P - \Delta_S$ is the detuning from the two-photon resonance

Here, $\mathrm{H}(t)$ is the atomic Hamiltonian, and $\mathrm{R}(t)$ describes relaxation processes.

In the rotating-wave approximation [21] and the dipole approximation with respect to the electric field strength, the Hamiltonian of atom–field interaction looks like

$$\mathrm{H}(t) = \frac{\hbar}{2}\begin{bmatrix} \delta + 2\xi_P(t) & \Omega_P(t) & 0 \\ \Omega_P(t) & -2\Delta & \Omega_S(t) \\ 0 & \Omega_S(t) & -\delta + 2\xi_S(t) \end{bmatrix}, \quad (2)$$

where the Rabi frequencies $\Omega_P(t) = -\mathbf{d}_{12}\mathbf{E}_P(t)/\hbar$ and $\Omega_S(t) = -\mathbf{d}_{32}\mathbf{E}_S(t)/\hbar$ of the pump and Stokes pulses, respectively, are assumed real, which does not violate the generality of consideration; and $\mathbf{d}$ is the operator of the atomic dipole moment. In Eq. (2), we used the notation $\Delta = \frac{1}{2}(\Delta_P + \Delta_S)$ for the average detuning from the single-photon resonance and $\delta = \Delta_P - \Delta_S$ for the detuning from the two-photon one, which are coupled with the detunings of the pump pulse, Δ_P, and the Stokes pulse, Δ_S, from the corresponding transition frequencies:

$$\Delta_P = \omega_P - (W_2 - W_1)/\hbar, \qquad \Delta_S = \omega_S - (W_2 - W_3)/\hbar,$$

and W_j being the energy of state $|j\rangle$ ($j = 1, 2, 3$). The relaxation term R in Eq. (1) looks like

$$\mathrm{R}(t) = -i\frac{\hbar}{2}\begin{bmatrix} -2\gamma_1\rho_{22}(t) & \Gamma\rho_{12}(t) & 0 \\ \Gamma\rho_{21}(t) & 2\Gamma\rho_{22}(t) & \Gamma\rho_{23}(t) \\ 0 & \Gamma\rho_{32}(t) & -2\gamma_3\rho_{22}(t) \end{bmatrix},$$

where $\Gamma = \gamma_1 + \gamma_3 + \gamma$. We neglect the collisions between atoms during their interaction with the field, so that the relaxation of the off-diagonal elements of the density matrix is caused only by spontaneous transitions of the atom from state $|2\rangle$ into states $|1\rangle$, $|3\rangle$, and others — distinct from $|1\rangle$ and $|3\rangle$ – with the rates γ_1, γ_3, and γ, respectively.

It is evident that the delta-correlated noises $\xi_P(t)$ and $\xi_S(t)$ enter multiplicatively into Eq. (1). This allows us to take advantage of the theory of multiplicative processes [22,23] in order to obtain the Liouville equation for the density matrix averaged over the ensemble:

$$i\hbar\frac{\partial\langle\rho\rangle}{\partial t} = [\mathcal{H}, \langle\rho\rangle] + \mathcal{R}, \tag{3}$$

where

$$\mathcal{H} = \frac{\hbar}{2}\begin{bmatrix} \delta & \Omega_P & 0 \\ \Omega_P & -2\Delta & \Omega_S \\ 0 & \Omega_S & -\delta \end{bmatrix}, \tag{4}$$

and the relaxation term looks like

$$\mathcal{R} = -i\frac{\hbar}{2}\begin{bmatrix} -2\gamma_1\langle\rho_{22}\rangle & (\Gamma + 2D_P)\langle\rho_{12}\rangle & 2(D_P + D_S)\langle\rho_{13}\rangle \\ (\Gamma + 2D_P)\langle\rho_{21}\rangle & 2\Gamma\langle\rho_{22}\rangle & (\Gamma + 2D_S)\langle\rho_{23}\rangle \\ 2(D_P + D_S)\langle\rho_{31}\rangle & (\Gamma + 2D_S)\langle\rho_{32}\rangle & -2\gamma_3\langle\rho_{22}\rangle \end{bmatrix} \tag{5}$$

for independent phase fluctuations of the Stokes and pump pulses and like

$$\mathcal{R} = -i\frac{\hbar}{2}\begin{bmatrix} -2\gamma_1\langle\rho_{22}\rangle & (\Gamma + 2D)\langle\rho_{12}\rangle & 0 \\ (\Gamma + 2D)\langle\rho_{21}\rangle & 2\Gamma\langle\rho_{22}\rangle & (\Gamma + 2D)\langle\rho_{23}\rangle \\ 0 & (\Gamma + 2D)\langle\rho_{32}\rangle & -2\gamma_3\langle\rho_{22}\rangle \end{bmatrix} \tag{6}$$

in the case of synchronous fluctuations ($\xi_P(t) = \xi_S(t)$ and $D_P = D_S = D$). We note the basic difference between Eqs. (5) and (6): the zero value of the components $\mathcal{R}_{13}$ and $\mathcal{R}_{31}$ in the latter. It is those components that are responsible, first of all, for a reduction of the population transfer from state $|1\rangle$ into state $|3\rangle$ [19]. One can see that the phase fluctuations of light pulses lead to an increase of the relaxation rate of the off-diagonal elements of the density matrix averaged over the ensemble of light pulses, as compared with the case of the absence of phase-fluctuations. This circumstance allows us, in order to solve Eq. (3), to take advantage of the method applied in [15] for the solution of the Liouville equation in the case of nonfluctuating fields, which differs from Eq. (3) only in the off-diagonal components of the relaxation matrix.

The calculation procedure for the population transfer is as follows. In the Liouville equation, we convert to the basis composed of the "bright", $|\psi_b\rangle$, excited, $|\psi_e\rangle$, and "dark", $|\psi_d\rangle$, states [24]:

$$\begin{aligned} |\psi_b\rangle &= \sin\vartheta(t)|\psi_1\rangle + \cos\vartheta(t)|\psi_3\rangle, \\ |\psi_e\rangle &= |\psi_2\rangle, \\ |\psi_d\rangle &= \cos\vartheta(t)|\psi_1\rangle - \sin\vartheta(t)|\psi_3\rangle, \end{aligned} \tag{7}$$

where $|\psi_j\rangle$ ($j = 1, 2, 3$) are the basis wave functions of the rotating reference frame, in which Hamiltonian 2 is written down. The functions $|\psi_j\rangle$ differ from $|j\rangle$ only in time-dependent phases. In Eq. (7), we introduced the notation $\Omega(t) = \sqrt{\Omega_P(t)^2 + \Omega_S(t)^2}$ and the mixing angle $\vartheta(t)$ which determines the relation between $\Omega_P(t)$ and $\Omega_S(t)$:

$$\Omega_P(t) = \Omega(t)\sin\vartheta(t), \qquad \Omega_S(t) = \Omega(t)\cos\vartheta(t).$$

In the course of STIRAP under examination, $\vartheta(t)$ changes from zero (when only the Stokes pulse acts upon the atom) to $\pi/2$ (when only the pump pulse does it) within the period of atom–field interaction. We assume that the condition for the adiabatic interaction between the atom and the field [3,4], on which STIRAP is based, namely,

$$\left|\frac{\partial\vartheta(t)}{\partial t}\right| \ll \Omega(t), \qquad t_1 \le t \le t_2, \tag{8}$$

where t_1 and t_2 are the times when the atom starts and terminates, respectively, its simultaneous interaction with both fields, holds true. In essence, Eq. (8) means that $\Omega_m\tau \gg 1$, where $\Omega_m = [\max\Omega(t)]_{t_1<t<t_2}$. Concerning detunings, we assume that $|\delta| \ll \Omega_m$, taking into account that the width of the two-photon resonance in nonfluctuating fields is of the order of $\Omega_m/\sqrt{\gamma\tau}$ [9]. The value of Δ is taken as having the same order of magnitude. In order that the method for the solution of the Liouville equations expounded in work [15] could be made use of, the condition $\gamma\tau \gg 1$ must be fulfilled. The other constants of spontaneous emission, γ_1 and γ_3, can either be small or of the order of γ. Independent phase fluctuations of the laser fields give rise to the uncontrollable detuning from the two-photon resonance; and we consider that $D_P \sim D_S \sim \delta$. But if fluctuations are synchronous, only the detuning from the single-photon resonance varies with time, and the sensitivity of the population transfer to such fluctuations is considerably lower. In this case, we consider that $D \sim \Omega_m$. In the reference frame (7), we seek the components of the density matrix, averaged over the ensemble, in the form

$$\widetilde{\rho}_{pq}(t) = \eta_{pq}(t)\exp\left(\int\limits_{t_1}^{t}\Phi(t')dt'\right),$$

where the subscripts p and q acquire the values of b, e, or d. Without any loss of generality, we put $\eta_{dd}(t) = 1$. It is obvious that $\Phi(t)$ is a real quantity and $\eta_{pq} = \eta_{qp}^*$.

We aim at finding the population of the "dark" state $|\psi_d\rangle$ (it coincides with state $|\psi_3\rangle$ at $t > t_2$) at the end of the interaction between the atom and the field,

$$n_3 = \exp\left(\int\limits_{t_1}^{t_2}\Phi(t)dt\right). \tag{9}$$

According to our assumptions, $\gamma\tau \gg 1$, $\Omega_m\tau \gg 1$, and, in the case of synchronous fluctuations, $D\tau \gg 1$. Therefore, in order to construct a perturbation theory with a small parameter $\epsilon = (\Omega_m\tau)^{-1}$ and without introducing dimensionless variables like γ/Ω_m, we make the substitutions $\gamma \to \gamma/\epsilon$, $\gamma_1 \to \gamma_1/\epsilon$, $\gamma_3 \to \gamma_3/\epsilon$, $\Omega \to \Omega/\epsilon$, and, in the case of synchronous fluctuations, $D \to D/\epsilon$ in the Liouville equation for $\widetilde{\rho}_{pq}(t)$; the latter can be easily obtained from Eq. (3). Then, we seek $\eta_{pq}(t)$ and $\Phi(t)$ as the power series in ϵ. At the end of calculations, we put $\epsilon = 1$. Not exposing the details of calculations, which are similar to those given in work [15], we report, in Sections 4. and 5., the results obtained for the cases of independent and synchronous fluctuations, respectively. Meantime, in the following Section 3., we consider the envelope shapes of the light pulses, which will serve as examples to illustrate the expressions obtained for n_3.

3. Time Dependences of Light Pulse Envelopes

Consider that the time dependence of the pump pulse repeats that of the Stokes pulse with a time delay t_d,

$$\begin{aligned}\Omega_P(t) &= \Omega_0 F_n(t - t_d/2),\\ \Omega_S(t) &= \Omega_0 F_n(t + t_d/2),\end{aligned} \tag{10}$$

where $n = 1, 2, 3, \ldots$ enumerates the sequence of functions

$$F_n(t) = \begin{cases}\cos^n(\pi t/\tau), & \text{if } |t| < \tau/2;\\ 0, & \text{if } |t| \ge \tau/2.\end{cases} \tag{11}$$

As n grows, those functions tend to the Gaussian

$$F_{\mathrm{G}}(t) = \exp\left(-t^2/\tau_{\mathrm{G}}^2\right),$$

if τ is defined by the equation

$$\tau = \pi\tau_{\mathrm{G}}\sqrt{n/2}$$

(see Fig. 2). We shall use two members, with $n = 1$ and $n = 4$, of the whole family of light pulses (10). The first model with $n = 1$ and the time delay $t_d = \tau/2$,

$$\begin{aligned}\Omega_P(t) &= \begin{cases}\Omega_0\cos(\pi t/\tau - \pi/4), & \text{якщо } |t - \tau/4| < \tau/2;\\ 0, & \text{якщо } |t - \tau/4| \ge \tau/2,\end{cases}\\ \Omega_S(t) &= \begin{cases}\Omega_0\cos(\pi t/\tau + \pi/4), & \text{якщо } |t + \tau/4| < \tau/2;\\ 0, & \text{якщо } |t + \tau/4| \ge \tau/2,\end{cases}\end{aligned} \tag{12}$$

is remarkable owing to the fact that, within the time interval $-\tau/4 \le t \le \tau/4$, when the atom interacts simultaneously with both pulses, the Rabi frequency $\Omega(t) = \Omega_0$ is independent and the mixing angle $\vartheta(t) = \pi t/\tau + \pi/4$ is a linear function of time [25]. This feature

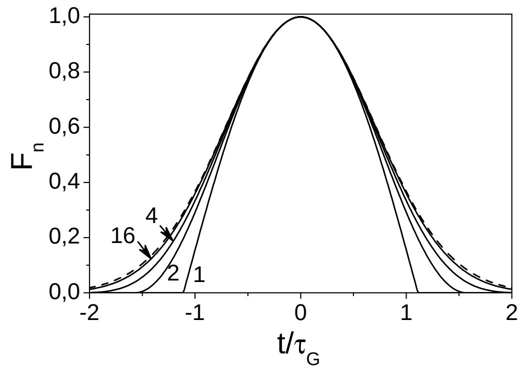

Fig. 2. Comparison of the Gaussian $F_G(t)$ (dashed curve) with the approximation functions $F_n(t)$ for various $n = 1$, 2, 4, and 16

of the model allows one to obtain the analytic expressions for integrals included into the theory and to illustrate the obtained results using a simple example. The other model ($n = 4$), which was applied earlier — e.g., in works [10, 26] — is close to the Gaussian-like shape of pulses. We use it to illustrate the accuracy of the expression found for the population of the target state $|3\rangle$ in the field of pulses with a smooth envelope of an arbitrary shape. For this purpose, we compare the obtained dependences with the results of numerical integration of the Liouville equation.

4. Independent Phase Fluctuations of the Stokes and Pump Fields

Consider first the influence of independent fluctuations of the field phases on the population transfer. In this case, the population of the target state is given by expression (9), where

$$\Phi = \Phi_0 + \frac{\delta}{\gamma}\Phi_1 + \frac{\delta^2}{\gamma^2}\Phi_2 + \ldots \quad (13)$$

Here,

$$\Phi_0 = -\frac{1}{2}\gamma\mathcal{T}\,(D_P + D_S)\sin^2 2\vartheta - \frac{4\gamma\Gamma\,\mathcal{T}}{\Omega^2}\left(\frac{\partial\vartheta}{\partial t}\right)^2 \quad (14)$$

describes the influence of phase fluctuations and the nonadiabaticity of the atom—field interaction on the population transfer, $\mathcal{T} = \left(\gamma + \gamma_1\cos^2\vartheta + \gamma_3\sin^2\vartheta\right)^{-1}$,

$$\Phi_2 = -\frac{\gamma^3}{\Omega^2}\,\Gamma\,\mathcal{T}\sin^2 2\vartheta \quad (15)$$

is responsible for the width of the two-photon resonance, and the term

$$\Phi_1 = -\frac{4\Delta\gamma^2\mathcal{T}^2}{\Omega^2}\sin^2 2\vartheta\,(D_P + D_S)\times$$

$$\times\left(\gamma_1\cos^4\vartheta - \gamma_3\sin^4\vartheta + \gamma\cos 2\vartheta\right) +$$

$$+\frac{16\Delta\gamma^2\Gamma\mathcal{T}}{\Omega^4}\sin 2\vartheta\frac{\partial^2\vartheta}{\partial^2 t} - \frac{64\Delta\gamma^3\Gamma\mathcal{T}^2}{\Omega^4}\times$$

$$\times\cos 2\vartheta\left(\frac{\partial\vartheta}{\partial t}\right)^2 - \frac{16\Delta\gamma^2\Gamma\mathcal{T}^2}{\Omega^4}\left(\frac{\partial\vartheta}{\partial t}\right)^2\times$$

$$\times\left(\gamma_1\cos^2\vartheta\,(1+3\cos 2\vartheta) - \gamma_3\sin^2\vartheta\,(1-3\cos 2\vartheta)\right) \quad (16)$$

gives rise to a shift of the population transfer (9) maximum with respect to the two-photon resonance by

$$\delta_s = -\frac{\gamma\int_{t_1}^{t_2}\Phi_1(t)\,dt}{2\int_{t_1}^{t_2}\Phi_2(t)\,dt}\,. \quad (17)$$

As the field strength increases, the value of δ_s, owing to the identical asymptotic dependences of Φ_1 and Φ_2 on Ω, approaches the limit that is independent of the field and proportional to $\Delta\,(D_P + D_S)$.

In the expressions for Φ_1 and Φ_2 quoted above, we made allowance for only the principal terms describing the dependence of the "dark" state population on the detuning δ from the two-photon resonance. In particular, a small influence of the phase fluctuations of light pulses on the two-photon resonance width was not taken into account there.

Strictly speaking, expression (9) differs from the population of state $|3\rangle$ at $t \to \infty$ because of spontaneous transitions from the poorly populated state $|2\rangle$ after the Stokes pulse terminates. Since the magnitudes of the populations of the "bright" and excited states are of the order of ϵ times the population of the "dark" state, Eq. (9) describes the population of state $|3\rangle$ with the same accuracy.

As the amplitudes of light pulses grow, all terms in Eqs. (14), (15), and (16) — except for the first one in Eq. (14) — tend to zero, and we come to the fundamental limit of the population transfer caused by phase fluctuations of the laser radiation

$$n_f = \exp\left(-\int_{t_1}^{t_2}\frac{\gamma\,(D_P + D_S)\sin^2 2\vartheta}{2\left(\gamma + \gamma_1\cos^2\vartheta + \gamma_3\sin^2\vartheta\right)}\,dt\right), \quad (18)$$

where n_f is the population of state $|3\rangle$. In the case $D_S = D_P$, $\gamma_1 \ll \gamma$, and $\gamma_3 \ll \gamma$, where n_f does not depend on the atom lifetime in the excited state, this expression coincides with the result of work [18]. One can see that the spontaneous transitions from the excited state into states $|1\rangle$ and $|3\rangle$ improve the population transfer into state $|3\rangle$.

4.1. Illustration of the results obtained

For the illustration of the results obtained, consider, as an example, pulses (12), for which the integrals of $\Phi_0(t)$, $\Phi_1(t)$, and $\Phi_2(t)$ can be expressed analytically. First of all, consider the case $\gamma_3 = \gamma_1$ where expression (9) for the population of state $|3\rangle$ after the atom—field interaction having terminated has a simple form:

$$n_3 = \exp\left(-\frac{\gamma\tau\,(D_P + D_S)}{8\,(\gamma + \gamma_1)} - \delta^2 \frac{\gamma\tau\,(\gamma + 2\gamma_1)}{4\Omega_0^2\,(\gamma + \gamma_1)} - \frac{2\pi^2\gamma\,(\gamma + 2\gamma_1)}{\Omega_0^2\tau\,(\gamma + \gamma_1)}\right). \quad (19)$$

The first term in the exponent of the exponential gives the fundamental limit of the population transfer, and the second the dependence of n_3 on the detuning from the two-photon resonance. The spontaneous transitions from the excited state into states $|1\rangle$ and $|3\rangle$ lead to a small reduction of the two-photon resonance width in comparison with the case $\gamma_3 = \gamma_1 = 0$ [9, 15]. The last term in the exponent of the exponential in Eq. (19) results from the term in Eq. (14) that contains $\frac{\partial}{\partial t}\vartheta(t)$ and describes the influence of the atom–field interaction nonadiabaticity on the population transfer from state $|1\rangle$ into state $|3\rangle$. In the case $\gamma_3 = \gamma_1$, the term proportional to δ is absent from the exponent in Eq. (19). Therefore, a shift of the maximum of the dependence n_3 versus δ with respect to the two-photon resonance is also absent.

From Eq. (19), it follows that the effects of spontaneous transitions from state $|2\rangle$ into states $|1\rangle$ and $|3\rangle$ on the population transfer for a quickly fluctuating detuning from the two-photon resonance (the first term in the exponent) and a static two-photon detuning (the second term) differ qualitatively. As γ_1 increases, the population of the target state $|3\rangle$ grows in the former case and falls down in the latter one (for a fixed two-photon detuning), which gives rise to a small narrowing of the two-photon resonance (no more than by a factor of $\sqrt{2}$ in comparison with the case $\gamma_1 = 0$).

In the general case $\gamma_3 \neq \gamma_1$, the population of state $|3\rangle$ for pulses (12) is equal to

$$n_3 = \exp\left(K_0 + \frac{\delta}{\gamma}K_1 + \frac{\delta^2}{\gamma^2}K_2\right),$$

where

$$K_0 = \frac{\gamma\,(\gamma\Gamma + \gamma_1\gamma_3)\,(D_P + D_S)\,\tau}{(\gamma_1 - \gamma_3)^2\sqrt{(\gamma + \gamma_1)\,(\gamma + \gamma_3)}} - \frac{\gamma\,(\gamma + \Gamma)\,(D_P + D_S)\,\tau}{2\,(\gamma_1 - \gamma_3)^2} - \frac{2\pi^2\gamma\Gamma}{\Omega_0^2\tau\sqrt{(\gamma + \gamma_1)\,(\gamma + \gamma_3)}},$$

$$K_1 = \frac{\gamma^2\Delta\,(\gamma + \gamma_1)\,(\gamma + \gamma_3)\,(\gamma + \Gamma)\,(D_P + D_S)\,\tau}{\Omega_0^2\,(\gamma_1 - \gamma_3)^3\sqrt{(\gamma + \gamma_1)\,(\gamma + \gamma_3)}} - \frac{\gamma^2\Delta\,(D_P + D_S)\,\tau}{\Omega_0^2\,(\gamma_1 - \gamma_3)^3}\left[8\,(\gamma + \gamma_1)\,(\gamma + \gamma_3) + (\gamma_1 - \gamma_3)^2\right] - \frac{24\pi^2\gamma^2\Delta\Gamma}{\Omega_0^4\tau\,(\gamma_1 - \gamma_3)}\left(\frac{\gamma + \Gamma}{\sqrt{(\gamma + \gamma_1)\,(\gamma + \gamma_3)}} - 2\right),$$

$$K_2 = \frac{\gamma^3\Gamma\tau}{\Omega_0^2\,(\gamma_1 - \gamma_3)^2}\left(\frac{2\,(\gamma\Gamma + \gamma_1\gamma_3)}{\sqrt{(\gamma + \gamma_1)\,(\gamma + \gamma_3)}} - (\gamma + \Gamma)\right).$$

According to Eq. (17), the ratio of K_1 and K_2 governs the shift of the population transfer maximum

$$\delta_s = -\frac{\gamma K_1}{2K_2} \quad (20)$$

with respect to the two-photon resonance.

As is seen from Fig. 3, where the examples of the dependences of n_3 after the atom—field interaction having terminated on the two-photon detuning for pulses (12) are shown, the results of calculations by formula (12) practically coincide with those of the numerical integration of the Liouville equation (3). The dependences given for the case $\gamma_1 = \gamma_3 = \gamma$ illustrate the appreciable narrowing of the two-photon resonance and the increase of the population transfer maximum for fluctuating fields in comparison with the case $\gamma_1 = \gamma_3 = 0$. At the same time, it is evident from the comparison of curves *1,2* and *3,4* that phase fluctuations practically do not affect the width of the two-photon resonance.

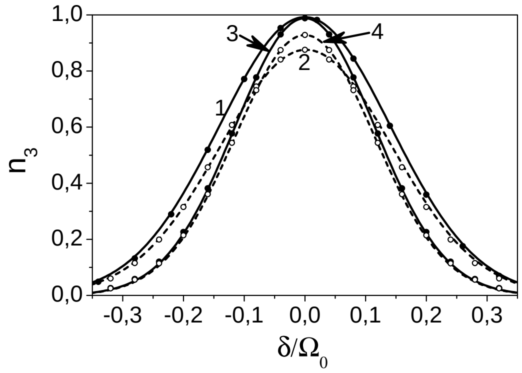

Fig. 3. Dependences of the state-$|3\rangle$ population on the two-photon detuning δ in terms of Ω_0 units in the absence (solid curves) or presence (dashed curves) of phase fluctuations, calculated by expression (19). Parameters: $\Omega_0\tau = 500$, $\gamma\tau = 100$, and $\Delta\tau = 50$ (for all curves); $D_P = D_S = 0$ (*1* and *3*); $D_P\tau = D_S\tau = 0.5$ (*2* and *4*); $\gamma_1 = \gamma_3 = 0$ (*1* and *2*); and $\gamma_1\tau = \gamma_3\tau = 100$ (*3* and *4*). Circles denote similar dependences found by the numerical integration of the Liouville equation (3) with the relaxation matrix (5)

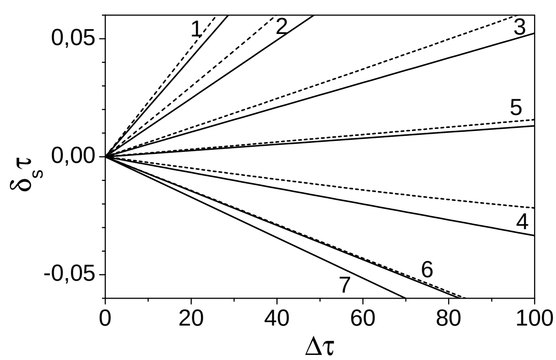

Fig. 4. Dependences of the shift δ_s of the state-$|3\rangle$ population maximum, calculated by expression (20) (solid lines) and found numerically by solving the Liouville equation (3) (dashed lines), with respect to the two-photon resonance on the average single-photon detuning Δ from the resonance in terms of Ω_0 units. Parameters: $\gamma\tau = \gamma_1\tau = 50$, and $\gamma_3 = 0$ (for all curves); $D\tau = 0$ (*1*, *3*, and *5*) and 0.5 (*2*, *4*, and *6*); $\Omega_0\tau = 200$ (*1* and *2*), 400 (*3* and *4*), 800 (*5* and *6*), and ∞ (*7*)

The dependences $\delta_s(\Delta)$, which were calculated either by Eq. (20) or by the numerical integration of the Liouville equation (3), are compared in Fig. 4. As would be expected, the dependences obtained by different methods agree better with each other if the strength of the laser pulse field is higher. Note that the quantity K_1 includes two kinds of terms: those originated from phase fluctuations and independent of them. The contributions of those terms to the shift of the maximum are opposite by sign. In the case $\gamma_3 = 0$ and $\gamma_1 = \gamma$ shown in the figure, the portion of the shift that is proportional to $D_P + D_S$,

$$\delta_{s\,\mathrm{fl}} = \Delta\left(D_P + D_S\right)\left(2\sqrt{2} - 3\right) / \left(4\gamma\right) \qquad (21)$$

does not depend on the field strength of laser pulses, while the fluctuation-independent portion δ_s is proportional to $(\Omega_0\tau)^{-2}$. At $\Omega_0\tau = 312$, the contributions of the terms of both kinds to δ_s become compensated. The straight line *7*, which is described by expression (21), marks the limit, which the shift of the maximum approaches at $\Omega_0\tau \to \infty$.

5. Synchronous Phase Fluctuations of the Stokes and Pump Fields

Provided synchronous phase fluctuations of the Stokes and pump fields, one may expect for a significant reduction of the influence of fluctuations on the population transfer, since, in this case, the detuning of field frequencies from the two-photon resonance does not fluctuate. In work [18], this case was analyzed provided that $\gamma_1 = \gamma_3 = 0$ and the diffusion coefficient of the phase D was low in comparison with Ω_0, when fluctuations did not influence the two-photon resonance width. Now, we consider the case $D \leq \Omega_0$.

The function $\Phi(t)$, which determines the population transfer probability (9), is determined, in its turn, by expression (13), where

$$\Phi_0 = -\frac{4\gamma\mathcal{T}}{\Omega^2}\left(\Gamma + 2D\right)\left(\frac{\partial\vartheta}{\partial t}\right)^2 \qquad (22)$$

describes the influence of the atom–field interaction nonadiabaticity on the population transfer,

$$\Phi_2 = -\frac{\gamma^3\mathcal{T}}{\Omega^2}\left(\Gamma + 2D\right)\sin^2 2\vartheta \qquad (23)$$

is responsible for the two-photon resonance width, and the term

$$\Phi_1 = \frac{16\Delta\gamma^2\mathcal{T}}{\Omega^4}\left(\Gamma + 2D\right)\left[\sin 2\vartheta\frac{\partial^2\vartheta}{\partial^2 t} - \left(\frac{\partial\vartheta}{\partial t}\right)^2 \times\right.$$

$$\times\Big(4\gamma\mathcal{T}\cos 2\vartheta - \mathcal{T}\left[(1 + 3\cos 2\vartheta)\times\right.$$

$$\left.\left.\times\gamma_1\cos^2\vartheta - \gamma_3\sin^2\vartheta\left(1 - 3\cos 2\vartheta\right)\right]\Big)\right] \qquad (24)$$

results in a shift of the maximum of $n_3(\delta)$ by magnitude (17) with respect to the two-photon resonance. One can see that, as Ω grows, Φ_1 decreases much faster than Φ_2, and $\delta_s \to 0$. Note also the absence of the fundamental limit associated with phase fluctuations: as Ω grows, Φ vanishes.

To illustrate the results obtained, consider first the pulses of shape (12), when $\Phi(t)$ can be integrated analytically. In the case of symmetric relaxation, $\gamma_3 = \gamma_1$, the probability of the population transfer n_3 is determined by the expression

$$n_3 = \exp\left(-\delta^2 \frac{\gamma\tau\,(\gamma + 2\gamma_1 + 2D)}{4\Omega_0^2\,(\gamma+\gamma_1)} - \right.$$

$$\left. - \frac{2\pi^2\gamma\,(\gamma + 2\gamma_1 + 2D)}{\Omega_0^2\tau\,(\gamma+\gamma_1)} \right). \qquad (25)$$

As is seen from the first term in the exponent of the exponential in expression (25), phase fluctuation leads to the appreciable reduction of the two-photon resonance width if the value of $2D$ is of the order of the inverse lifetime of the atom in the excited state. The second term testifies that fluctuations, synchronous by phase, can considerably reduce the maximal value of n_3 attainable at $\delta = 0$ only in the case $D \gg \Omega_0$ (beyond the limits where the theory is eligible).

To illustrate the accuracy of the population transfer calculations by formulas (9), (13), (22), (23), and (24), eligible for pulses of any shape with a smooth envelope, consider the interaction of an atom with Gaussian-like pulses [Eqs. (10) and (11), with $n = 4$]. Figure 5 shows that the calculation of n_3 by the formulas indicated produces the results that coordinate well with those obtained by the numerical solution of the Liouville equation (3) with the relaxation matrix (6), if $D \ll \Omega_0$. If $D = \Omega_0$, the results of both the methods for calculating the dependence $n_3(\delta)$ almost coincide up to a value of the detuning from the two-photon resonance a little larger than its halfwidth; the difference between the results of calculations at large δ's evidences for an important contribution made to Φ by terms with the orders higher than δ^2, which were not taken into account in series (13).

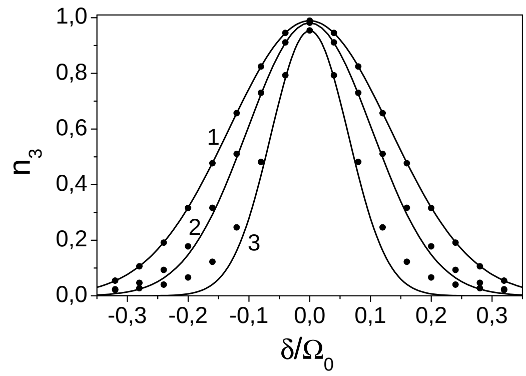

Fig. 5. Dependences of the state-$|3\rangle$ population in the case of synchronous phase fluctuations of the Stokes and pump pulses on the two-photon detuning δ in terms of Ω_0 units, found by Eqs. (9), (13), (22), (23), and (24). Parameters: $\Omega\tau = 500$, $\gamma\tau = \gamma_1\tau = \gamma_3\tau = 100$, and $\Delta\tau = 50$ (for all curves); $D\tau = 0$ (*1*), 100 (*2*), and 500 (*3*). Circles denote similar dependences found by the numerical integration of the Liouville equations (3) with the relaxation matrix (6). Time-dependences of the light pulse envelopes are described by expressions (10) and (11), where $n = 4$ and $t_d = 0.2\tau$

6. Conclusions

We have studied the influence of the phase fluctuations of light fields on the population transfer in the course of the stimulated Raman adiabatic passage, using, as an example, a three-level Λ-scheme of the interaction between light pulses and an atom, in the case where the phase fluctuations are described by the Wiener process. In contrast to the previous researches, we have considered now spontaneous transitions from the excited state into the ground and metastable ones, between which the population is transferred. The results presented are eligible in the case where the rate of spontaneous transitions from the excited atomic state into others that are not coupled by the field with the excited one considerably exceeds the period of atom–field interaction, which, as a rule, occurs in experiments with atomic beams. The independent phase fluctuations of light pulses lead to a reduction of the population transfer maximum and, practically, do not affect the two-photon resonance width. If the fluctuations are synchronous, the width of the two-photon resonance decreases more than the maximal value of the population transfer does.

For independent phase fluctuations, if the intensity of light pulses grows, the maximum of the population transfer tends to a limit that is determined by the amplitude of the phase fluctuations of laser pulses and the ratio of the rates of spontaneous transitions through various relaxation channels. In the case of synchronous phase fluctuations, the population transfer probability in its maximum approaches unity if the intensity of laser pulses grows.

We have found a new feature of the population transfer, the shift of its maximum with respect to the two-photon resonance, which is proportional to the arithmetic mean of single-photon detunings. This shift arises owing to the probability difference between the spontaneous transitions from the excited state into the ground and metastable ones. It does not disappear with increase in the intensity of laser pulses, if the phase fluctuations of the Stokes and pump fields are independent.

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СТИМУЛЬОВАНЕ РАМАНІВСЬКЕ АДІАБАТИЧНЕ ПРОХОДЖЕННЯ В ПОЛЯХ З ФЛУКТУЮЧОЮ ФАЗОЮ

В.І. Романенко

Р е з ю м е

Теоретично досліджується явище стимульованого раманівського адіабатичного проходження в полі імпульсів з флуктуючими фазами, що описуються вінерівським стохастичним процесом. Обговорюється вплив спонтанних переходів зі збудженого стану на гранично можливе перенесення населеності між основним та метастабільним (чи двома метастабільними) станами атома. Показано, що крім обмеження на максимальну величину перенесення населеності флуктуації фази приводять до зсуву максимуму відносно двофотонного резонансу на величину, пропорційну середньому арифметичному значень відстроювань стоксового поля і поля накачки від однофотонного резонансу, який не усувається у граничному випадку високих інтенсивностей лазерних полів.