ANALOG OF THE COHERENT POPULATION TRAPPING STATE IN A POLYCHROMATIC FIELD

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The interaction between a three-level atom and a polychromatic field with an equidistant spectrum (Λ -scheme of the atom-field interaction) has been studied theoretically. It is shown that the interaction of an atom with such a field can be reduced to its interaction with a bichromatic field with additional light shifts of transition frequencies and an additional coupling of the lower atomic levels, which is proportional to the field intensity. Owing to this coupling, the idea of the coherent population trapping can be considered only as an approximation, because the dark state is not an eigenstate of the effective Hamiltonian in the general case of arbitrary dipole moments. The analyzed model gives a simple theoretical interpretation for the formation of the atomic state, which is close to the coherent population trapping, in the radiation field of a femtosecond laser.

1. Introduction

The coherent population trapping (CPT) phenomenon [1–5] is a basis for the explanation of the electromagnetically induced transparency [6–9], the population transfer among the states by means of stimulated Raman adiabatic passage (STIRAP) [10–13], and the fabrication of compact quantum laser-based frequency references [14]. It also serves as a basis for one of the methods aimed at the cooling of atoms down to ultralow temperatures [15].

In the simplest case, the CPT manifests itself at the interaction between a three-level atom and a bichromatic radiation, when every spectral component couples one of two metastable atomic states (one of them can be stable) with the excited one (Λ -scheme of the atom-field interaction). Then, while registering the dependence of the fluorescence intensity on the frequency of either of

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spectral components, a narrow dip (a dark resonance) is observed, when the difference between component frequencies becomes close to the frequency of the transition between long-lived atomic states. From the physical viewpoint, the CPT is based on the existence of a "dark state" in the atom, i.e. a superposition of longlived states, which is determined by the ratio between the intensities of spectral components; if the atom is in this state, it does not absorb radiation.

Recently, the interest has been growing to dark resonances in a polychromatic field with equidistant spectral components [16–20]. The dark resonance arises in the case where the frequency of the transition between long-lived atomic states is a multiple of the frequency difference between the neighbor spectral components of the field. Such resonances were observed under the excitation of atoms with radiation emitted by a femtosecond laser [20], whereas the resonance of the electromagnetically induced transparency (which is based on the CPT phenomenon) was registered in the field produced by a sequence of light pulses generated by a mode locked picosecond laser. A dark resonance with a contrast close to 100% was also observed in sodium vapor in the radiation field of a free-running multimode laser [16].

In this work, we analyze the interaction between a three-level system and a low-intensity polychromatic field neglecting relaxation processes and demonstrate that the polychromatic interaction can be reduced to a bichromatic one, but with a certain field-dependent frequency shift for transitions in the atom. In addition, we show that, in the general case of arbitrary transition dipole moments, one may talk only approximately about the CPT, because the dark state is not an eigenstate of



Fig. 1. Diagram of the interaction between a three-level atom and a polychromatic field. The frequency difference between the neighbor spectral components is equal to ϖ

the effective Hamiltonian; namely, an eigenstate of the effective Hamiltonian, which is the nearest to the "dark" one, almost always has a small admixture of an excited state. The theoretical analysis of the resonances in the CPT in a polychromatic laser field with a frequencyshifted feedback [21] has already been carried out by us taking advantage of the perturbation theory for the density matrix and considering the uniform and non-uniform broadening of spectral lines in the radiation emitted by atoms in a buffer-gas environment [22]. Although this research allows one to obtain expressions for the frequency shifts of transitions associated with a large number of spectral components, it disregards, however, the physical essence of the CPT resonance, namely, the formation of a non-absorbing or, more precisely, low-absorbing state. To elucidate this issue, one has to find a form for the effective Hamiltonian, which would describe very weak, but numerous spectral components of the field.

Our analysis is based on the averaging over the reciprocal frequency difference between neighbor spectral components; it is the minimal characteristic time τ of the problem. In other words, we average over the quick motion of the system. The corresponding formalism for a particle moving in a rapidly changing field is given in work [23]. Its modification to the case of a twolevel quantum-mechanical system was used to calculate the light pressure force acting on the atom in a frequency-modulated field [24], as well as an atomic trap based on the interaction between an atom and counterpropagating pulses [25].

In Section 2, the basic equations needed to describe the interaction between an atom and the field are presented. Section 3 contains a derivation of the effective Hamiltonian, which is responsible for a slow – with a characteristic time longer than τ – variation of probability amplitudes for the atom to remain in one of its states. Light shifts of transition frequencies and the coupling between metastable states, which is connected with the field polychromaticity, are considered in Section 4 with the use of a Gaussian-like distribution of the spectral intensity as an example. In Section 5, one of the eigenstates of the effective Hamiltonian is found, which is an analog of the CPT state in the case of polychromatic field. In Section 6, CPT resonances in a polychromatic field are studied making allowance for relaxation processes. The results obtained are briefly summarized in Section 7.

2. Basic Equations

Consider a three-level atom with two metastable states, $|1\rangle$ and $|3\rangle$, and one excited state $|2\rangle$, which interacts with the polychromatic field characterized by equidistant frequencies between its components (see Fig. 1). The frequency difference between neighbor spectral components equals ϖ , and the frequency of the component with the maximum intensity equals ω .

The electric field $\boldsymbol{\mathcal{E}}(t)$ at the atom location point looks like

$$\boldsymbol{\mathcal{E}}(z,t) = \frac{\boldsymbol{\varepsilon}\mathcal{E}_0}{2} \sum_{n=-\infty}^{\infty} a_n \exp\left[-i\left(\omega + n\boldsymbol{\varpi}\right)t - i\varphi_n\right] + \text{c.c.}$$
(1)

Here, ε is the polarization unit vector, \mathcal{E}_0 the amplitude of the spectral component with the maximum intensity, and a_n are the relative amplitudes of spectral components. We normalize the relative amplitudes a_n in such a way that the component with n = 0 has the maximum amplitude, $a_0 = 1$. For model calculations, let us select a realistic distribution of relative amplitudes in the form

$$a_n = \exp\left(-\frac{n^2}{2n_0^2}\right). \tag{2}$$

In the rotating-wave approximation [26], the interaction between the three-level atom and the field is described by the Hamiltonian

$$\mathcal{H} = \frac{\hbar}{2} \begin{bmatrix} -2\delta_1 & \Omega_1^*(t) & 0\\ \Omega_1(t) & 0 & \Omega_3(t)\\ 0 & \Omega_3^*(t) & -2\delta_3 \end{bmatrix}.$$
 (3)

Here, $\delta_1 = \omega_{12} - \omega - n_1 \varpi$ is a detuning of the spectral component of laser radiation with the frequency $\omega + n_1 \varpi$,

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which is the nearest to the frequency ω_{12} of the transition $|1\rangle \rightarrow |2\rangle$, from ω_{12} , and $\delta_3 = \omega_{32} - \omega - n_3 \varpi$ is a detuning of the spectral component of laser radiation with the frequency $\omega + n_3 \varpi$, which is the nearest to the frequency ω_{32} of transition $|3\rangle \rightarrow |2\rangle$, from the frequency ω_{32} . The Rabi frequencies $\Omega_1(t)$ and $\Omega_3(t)$ in Eq. (3) are introduced by the relations

$$\Omega_j(t) = \Omega_{j0} \sum_n a_n \exp\left[i(n_j - n)\varpi t - i\varphi_n\right],\tag{4}$$

where j = 1, 3, $\Omega_{j0} \equiv -(\boldsymbol{\varepsilon} \cdot \boldsymbol{d}_{j2}) \mathcal{E}_0/\hbar$, and \boldsymbol{d}_{j2} is the matrix element of the dipole moment, which is specifically selected to provide $\Omega_{j0} > 0$.

3. Effective Hamiltonian

Let us suppose that

$$\Omega_{10}, \ \Omega_{30}, \ |\delta_1|, \ |\delta_3| \ll \varpi.$$

$$\tag{5}$$

Hence, we consider the case of a so large frequency difference between the neighbor spectral components of the field that the atom can be excited by only two components with the relative amplitudes a_{n_1} and a_{n_3} , the frequencies of which are close to the transition frequencies ω_{12} and ω_{32} , respectively, whereas the other spectral component leads to a light shift of resonance frequencies. The evolution of the wave function is characterized by two time scales. The quick motion occurs with the characteristic time ϖ^{-1} , and the slow one is determined by low characteristic frequencies of the problem (see Eq. (5)). To obtain an equation describing the slow variation of probability amplitudes, let us use the standard averaging procedure described in work [23] for the case of the mechanical motion in a field with a rapidly oscillating force.

Let us write down the Rabi frequencies (4) in the form of sums of two terms; one of those terms is timeindependent, the other oscillates with the frequency ϖ ,

$$\Omega_j(t) = \Omega_{j,s}(t) + \Omega_{j,f}, \qquad j = 1, 3, \tag{6}$$

where

$$\Omega_{j,s}(t) = \Omega_{j0} a_{n_j} e^{-i\varphi_{n_j}},\tag{7}$$

$$\Omega_{j,f}(t) = \Omega_{j0} \sum_{n \neq 0} a_{n+n_j} \exp\left[-in\varpi t - i\varphi_{n+n_j}\right]. \tag{8}$$

The probability amplitudes for atomic states can expectedly be written down as a sum of two terms,

$$C_i = C_i + c_i, \qquad i = 1, 2, 3,$$
(9)

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the second of which is small in comparison with the first one and oscillates. From the Schrödinger equation

$$i\hbar \frac{d}{dt} \begin{pmatrix} C_1 \\ C_2 \\ C_3 \end{pmatrix} = \mathcal{H} \begin{pmatrix} C_1 \\ C_2 \\ C_3 \end{pmatrix}$$
(10)

we obtain

$$i\tilde{\tilde{C}}_{1} + \underline{i\dot{c}_{1}} = -\delta_{1}\tilde{C}_{1} - \underline{\delta_{1}c_{1}} + \frac{1}{2}\Omega_{1,s}^{*}\tilde{C}_{2} + \frac{1}{2}\Omega_{1,f}^{*}c_{2} + \frac{1}{2}\Omega_{1,f}^{*}\tilde{C}_{2} + \frac{1}{2}\Omega_{1,s}^{*}c_{2}, \qquad (11)$$

$$i\tilde{\tilde{C}}_{2} + \underline{i\dot{c}_{2}} = \frac{1}{2}\Omega_{1,s}\tilde{C}_{1} + \frac{1}{2}\Omega_{3,s}\tilde{C}_{3} + \frac{1}{2}\Omega_{1,f}c_{1} + \frac{1}{2}\Omega_{1,f}\tilde{C}_{1} + \frac{1}{2}\Omega_{1,s}c_{1} + \frac{1}{2}\Omega_{1,s}c_{1} + \frac{1}{2}\Omega_{1,f}\tilde{C}_{1} + \frac{1}{2}\Omega_{1,s}c_{1} + \frac{1}{2}\Omega_{1,s}c_{$$

$$+ \underline{\underline{2}}_{3,f} \underline{c_3} + \underline{2}_{3,f} \underline{C_3} + \underline{2}_{3,s} \underline{c_3}, \tag{12}$$

$$i\dot{\tilde{C}}_{3} + \underline{i\dot{c}_{3}} = -\delta_{3}\tilde{C}_{3} - \underline{\delta_{3}c_{3}} + \frac{1}{2}\Omega_{3,s}^{*}\tilde{C}_{2} + \frac{1}{2}\Omega_{3,f}^{*}c_{2} + \frac{1}{2}\Omega_{3,f}^{*}\tilde{C}_{2} + \frac{1}{2}\Omega_{3,s}^{*}c_{2}.$$
(13)

The terms underlined once rapidly oscillate and give zero after their averaging over the oscillation period $2\pi/\varpi$. The terms underlined twice consist of products of oscillating factors. Each of them can be divided into a part that slowly varies in time and an oscillating part with a zero averaged value. To exclude the rapid motion, let us postulate that the corresponding terms on the left- and right-hand sides of Eqs. (11)–(13), which belong to the same type of the temporal dependences, i.e. the rapid-or the slow-varying one, can be equated to each other independently [23]. The oscillating parts of Eqs. (11)–(13) bring about

$$i\dot{c}_1 = \frac{1}{2}\Omega^*_{1,f}\tilde{C}_2,$$
 (14)

$$i\dot{c}_2 = \frac{1}{2}\Omega_{1,f}\tilde{C}_1 + \frac{1}{2}\Omega_{3,f}\tilde{C}_3,\tag{15}$$

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$$i\dot{c}_3 = \frac{1}{2}\Omega^*_{3,f}\tilde{C}_2.$$
 (16)

Here, we took inequalities $|c_i| \ll 1$ and $|\dot{c}_i| \sim \varpi$, as well as inequalities (5), into account and neglected small terms.

The parts of Eqs. (11)-(13), which are slowly varying in time, give

$$\dot{\tilde{C}}_{1} = -\delta_{1}\tilde{C}_{1} + \frac{1}{2}\Omega_{1,s}^{*}\tilde{C}_{2} + \frac{1}{2}\langle\Omega_{1,f}^{*}c_{2}\rangle, \qquad (17)$$

$$i\dot{\tilde{C}}_{2} = \frac{1}{2}\Omega_{1,s}\tilde{C}_{1} + \frac{1}{2}\langle\Omega_{1,f}c_{1}\rangle + \frac{1}{2}\Omega_{3,s}\tilde{C}_{3} + \frac{1}{2}\langle\Omega_{3,f}c_{3}\rangle,$$
(18)

$$i\tilde{\tilde{C}}_3 = -\delta_3\tilde{C}_3 + \frac{1}{2}\Omega^*_{3,s}\tilde{C}_2 + \frac{1}{2}\langle\Omega^*_{3,f}c_2\rangle,$$
 (19)

where $\langle \cdots \rangle$ means time averaging over the interval $2\pi/\varpi$.

The solution of Eqs. (14)–(16) looks like

$$c_1 = -\tilde{C}_2 \Omega_{10} \sum_{n \neq 0} \frac{a_{n+n_1}}{2n\varpi} \exp\left[in\varpi t + i\varphi_{n+n_1}\right], \qquad (20)$$

$$c_2 = \tilde{C}_1 \Omega_{10} \sum_{n \neq 0} \frac{a_{n+n_1}}{2n\varpi} \exp\left[-in\varpi t - i\varphi_{n+n_1}\right] +$$

$$+\tilde{C}_3\Omega_{30}\sum_{n\neq 0}\frac{a_{n+n_3}}{2n\varpi}\exp\left[-in\varpi t - i\varphi_{n+n_3}\right],\tag{21}$$

$$c_3 = -\tilde{C}_2 \Omega_{30} \sum_{n \neq 0} \frac{a_{n+n_3}}{2n\varpi} \exp\left[in\varpi t + i\varphi_{n+n_3}\right], \qquad (22)$$

Substituting the quantities c_i into Eqs. (17)–(19), we arrive at equations for slowly varying amplitudes, which have the form of a Schrödinger equation with the effective Hamiltonian

$$\mathcal{H}_{eff} = \frac{\hbar}{2} \begin{bmatrix} -2\delta_1 + S_1 & \Omega_{1,s}^* & R\\ \Omega_{1,s} & -S_1 - S_3 & \Omega_{3,s}\\ R^* & \Omega_{3,s}^* & -2\delta_3 + S_3 \end{bmatrix}.$$
 (23)

Here, the expressions

$$S_1 = \frac{\Omega_{10}^2}{2\varpi} \sum_{n \neq 0} \frac{|a_{n+n_1}|^2}{n},$$
(24)

$$S_3 = \frac{\Omega_{30}^2}{2\varpi} \sum_{n \neq 0} \frac{|a_{n+n_3}|^2}{n}$$
(25)

are responsible for light frequency shifts of the transitions $|1\rangle \rightarrow |2\rangle$ and $|3\rangle \rightarrow |2\rangle$. These shifts are determined by the spectral components of laser radiation, which are different from n_1 or n_3 for the transition $|1\rangle \rightarrow |2\rangle$ or $|3\rangle \rightarrow |2\rangle$, respectively, and the coupling between states $|1\rangle$ and $|3\rangle$ determined by the coefficient

$$R = \frac{\Omega_{10}\Omega_{30}}{2\varpi} \sum_{n \neq 0} \frac{a_{n+n_1}a_{n+n_3}}{n} \exp(i\varphi_{n+n_1} - i\varphi_{n+n_3}).$$
(26)

The latter has the dimensionality of frequency. The condition for the two-photon resonance is obeyed, if

$$\delta_3 = \delta_1 + \frac{1}{2}(S_3 - S_1). \tag{27}$$

4. Light Shifts and the Coupling Between Metastable States

The calculations of the constants S_1 , S_3 , and R will be carried out for the Gaussian-like field spectrum (Eq. (2)) with a large number of spectral components. In this case, let n_1 and n_3 be also much smaller than n_0 : $n_1 \ll n_0$ and $n_3 \ll n_0$. Then,

$$a_{n+n_j} = \exp\left(-\frac{n^2}{2n_0^2}\right) \left[1 - \frac{nn_j}{n_0^2} - \frac{n_j^2}{2n_0^2} + \frac{n_j^2 n(n+n_j)}{2n_0^4} - \frac{n^3 n_j^3}{6n_0^6}\right] + \dots,$$
(28)

where j = 1, 3 and

$$S_j = -\frac{n_j \sqrt{\pi} \Omega_{j0}^2}{n_0 \varpi} \left(1 - \frac{2n_j^2}{3n_0^2} \right).$$
(29)

It is evident that n_3 and n_1 differ from each other by an integer number N,

$$n_3 = n_1 - N, (30)$$

which is determined by the ratio between the quantities $\omega_{12} - \omega_{32} = \omega_{13}$ and ϖ . Let us introduce the two-photon detuning

$$\delta = \omega_{13} - N\varpi = \delta_1 - \delta_3,\tag{31}$$

where the integer N is selected to minimize $|\delta|.$ Then, the quantity

$$\delta = \frac{1}{2} \left(S_1 - S_3 \right) \tag{32}$$

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demonstrates how much the CPT resonance is detuned from the accurate two-photon one. This shift is minimal, if

$$n_{1} = \left[\frac{\Omega_{30}^{2}N}{\Omega_{30}^{2} - \Omega_{10}^{2}} \left(1 - \frac{2\Omega_{10}^{2}N^{2}\left(\Omega_{10}^{2} + \Omega_{30}^{2}\right)}{3n_{0}^{2}\left(\Omega_{30}^{2} - \Omega_{10}^{2}\right)^{2}}\right)\right].$$
 (33)

In this case,

$$n_3 = \left[\frac{\Omega_{10}^2 N}{\Omega_{30}^2 - \Omega_{10}^2} \left(1 - \frac{2\Omega_{30}^2 N^2 \left(\Omega_{10}^2 + \Omega_{30}^2\right)}{3n_0^2 \left(\Omega_{30}^2 - \Omega_{10}^2\right)^2}\right)\right].$$
 (34)

Here, the square brackets mean the integer part of the expression, because n_1 is an integer number. In the case where the matrix elements of transition dipole moments are identical, i.e., $\Omega_{30}^2 = \Omega_{10}^2$, the light shift of the two-photon resonance is described by the expression

$$\delta_3 - \delta_1 = \frac{N\sqrt{\pi}\Omega_{10}^2}{2n_0^2\varpi}.$$
(35)

For $\varphi_n = 0$, we find, using Eqs. (26) and (28), that

$$R = \frac{\Omega_{10}\Omega_{30}(n_1 + n_3)}{2n_0\varpi}.$$
(36)

Comparing Eqs. (29) and (36), we see that the quantities S_1 , S_3 , and R are of the same order of magnitude.

Since $n_1 + n_3 \ll n_0$ and $\Omega_{10}, \Omega_{30} \ll \varpi$, the coupling between metastable states is very weak in comparison with that between the metastable states, on the one hand, and the excited one, on the other hand. Under the condition $n_1 = -n_3$, i.e. when the frequency of the spectral component with the maximum intensity is equal to $\omega = \frac{1}{2}(\omega_{12} + \omega_{32})$, the coupling between metastable states is absent.

5. Analog of Coherent Population Trapping State in a Polychromatic Field

It is evident from the effective Hamiltonian (23) that, in the general case, states $|1\rangle$ and $|3\rangle$ are coupled with each other by means of the field. If R = 0, the effective Hamiltonian (23) is identical to the Hamiltonian of an atom in a bichromatic field. In this case, if the two-photon resonance is realized, one of the Hamiltonian eigenstates – namely, the state corresponding to the zero eigenvalue – does not include the excited state $|2\rangle$ [12, 13]. In the previous section, it was shown that the quantity R is small in comparison with the Rabi frequencies. Therefore, in the case of the two-photon resonance, one should expect that the population of the excited states belonging to one of the eigenstates of the effective Hamiltonian

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(23) would be low. Let us determine this eigenstate. Let $\delta_1 = S_1/2$, $\delta_3 = S_3/2$, and let the condition $\varphi_n = 0$ be satisfied. Standard calculations give the following equation for the characteristic λ -values of the Hamiltonian:

$$2\lambda(2\lambda - \Omega_0)(2\lambda + \Omega_0) + 4(S_1 + S_3)\lambda^2 - \\ -\Omega_0^2 R\sin 2\theta - R^2(S_1 + S_3 + 2\lambda) = 0.$$
(37)

Here, we introduced the notations

$$\theta = \arctan \frac{\Omega_{1,s}}{\Omega_{3,s}}, \qquad \Omega_0 = \sqrt{\Omega_{1,s}^2 + \Omega_{3,s}}.$$
(38)

From Eq. (37), it follows in the zeroth-order approximation in R and $S_1 + S_3$ that

$$\lambda \approx 0,\tag{39}$$

$$\lambda_{\pm} \approx \pm \frac{1}{2} \Omega_0. \tag{40}$$

The characteristic value (39) corresponds to the state of coherent population trapping. Let us analyze it in more details. Consider the corresponding eigenstate. As the first approximation, using Eq. (37), we find

$$\lambda = -R\sin\theta\cos\theta = -R\frac{\Omega_{1,s}\Omega_{3,s}}{\Omega_{1,s}^2 + \Omega_{3,s}^2}.$$
(41)

The second approximation adds nothing new to expression (41). Although it is easy to obtain the third approximation, we do not present it, because it is not essential for the results obtained here. The eigenstate of Hamiltonian, which corresponds to the characteristic value (41), looks like

$$\psi = \frac{\Omega_0}{\sqrt{\Omega_0^2 + R^2 \cos^2 2\theta}} \begin{pmatrix} -\cos \theta \\ \frac{R}{\Omega_0} \cos 2\theta \\ \sin \theta \end{pmatrix}.$$
 (42)

It is evident that the population of the excited Hamiltonian eigenstate (42) is low and amounts to $(\Omega_0/\varpi)^2$ by the order of magnitude.

6. Resonances of Coherent Population Trapping in a Polychromatic Field

Proceeding from the effective Hamiltonian of an atom in a polychromatic field described by Eq. (23), one may expect, at first sight, that the shift of the atomic fluorescence minimum with respect to the two-photon resonance is determined by the difference between the light shifts of the first and third atomic states. Unlike the ordinary formation of the CPT resonance in a bichromatic field, when the latter couples states $1\rangle$, $|2\rangle$ and $|3\rangle$, $|2\rangle$, Hamiltonian (23) demonstrates that the field also couples states $|1\rangle$, $|3\rangle$. Such a coupling is similar to that between oscillators, which, as is known [23], results in a shift of their characteristic frequencies. Therefore, there is a need to analyze the consequences, which the presence of the term R in the effective Hamiltonian brings about.

For the analysis of the atomic fluorescence dependence on the two-photon detuning, let us take advantage of the equation for the density matrix. Interaction between the atom and the field is taken into account by Hamiltonian (23), so that all we need is to include the influence of relaxation processes on the density matrix evolution into consideration. The application of the effective Hamiltonian formalism is eligible, if the difference ϖ between the neighbor spectral components of the polychromatic field considerably exceeds the rate of relaxation processes.

Suppose that, owing to the spontaneous emission, the atom can transit from excited state $|2\rangle$ into states $|1\rangle$ and $|3\rangle$ at the rates γ_1 and γ_3 , respectively. In addition, the relaxation processes give rise to the relaxation of the coherence ρ_{13} at a low rate γ_0 , which is much slower than $\gamma_1 + \gamma_3$. As a result, the equations for the density matrix look like

$$\begin{split} \frac{d}{dt}\rho_{11} &= \frac{i}{2}\Omega_1\rho_{12} - \frac{i}{2}\Omega_1\rho_{21} + \frac{i}{2}R\left(\rho_{13} - \rho_{31}\right) + \gamma_1\rho_{22},\\ \frac{d}{dt}\rho_{12} &= \frac{i}{2}\left(2\delta_1 - 2S_1 - S_3\right)\rho_{12} + \frac{i}{2}\Omega_1\left(\rho_{11} - \rho_{22}\right) - \\ -\frac{i}{2}R\rho_{32} + \frac{i}{2}\Omega_3\rho_{13} - \frac{1}{2}\left(\gamma_1 + \gamma_3\right)\rho_{12},\\ \frac{d}{dt}\rho_{13} &= \frac{i}{2}\left(2\delta_1 - 2\delta_3 + S_3 - S_1\right)\rho_{13} + \\ +\frac{i}{2}R\left(\rho_{11} - \rho_{33}\right) + \frac{i}{2}\Omega_3\rho_{12} - \frac{i}{2}\Omega_1\rho_{23} - \gamma_0\rho_{13},\\ \frac{d}{dt}\rho_{23} &= \frac{i}{2}\left(S_1 + 2S_3 - 2\delta_3\right)\rho_{23} - \frac{i}{2}\Omega_1\rho_{13} + \\ +\frac{i}{2}\Omega_3\left(\rho_{22} - \rho_{33}\right) - \frac{i}{2}R\rho_{21} - \frac{1}{2}\left(\gamma_1 + \gamma_3\right)\rho_{23},\\ \frac{d}{dt}\rho_{33} &= \frac{i}{2}\Omega_3\left(\rho_{32} - \rho_{23}\right) + \frac{i}{2}R\left(\rho_{31} - \rho_{13}\right) + \gamma_3\rho_{22}, \end{split}$$

$$\rho_{nm} = \rho_{mn}^*, \quad n, m = 1, 2, 3, \quad \rho_{11} + \rho_{22} + \rho_{33} = 1.$$
(43)

To make the notations more compact, we did not mark the Rabi frequencies with the identical additional subscript s.

To estimate the fluorescence signal, it is necessary to find an expression for the population ρ_{22} in excited state. The CPT resonance width is small, provided that the Rabi frequencies Ω_1 and Ω_3 are of the order of γ_0 . As is seen from expressions (29) and (36), the light shifts and the frequency R describing the coupling between states $|1\rangle$ and $|3\rangle$ are small in comparison with the Rabi frequencies. The latter are low in comparison with γ_1 and γ_3 . Therefore, we assume that

$$R/\Omega_j \ll 1, \quad S_j/\Omega_j \ll 1, \quad \Omega_j \ll \gamma_1 + \gamma_3,$$

 $\Omega_j \sim \gamma_0, \quad j = 1, 3.$ (44)

Under those conditions, the shift of the population maximum in the excited state, which we find from the stationary solution of Eqs. (43), is minimal, if

$$\delta = \delta_R + \delta_S,\tag{45}$$

where

$$\delta_{R} = \Omega_{1}\Omega_{3}R\gamma_{0} \left(\Omega_{3}^{4}\gamma_{1} - \Omega_{1}^{4}\gamma_{3}\right) (\gamma_{1} + \gamma_{3}) \times \left[\Omega_{1}^{2}\Omega_{3}^{2} \left(\Omega_{1}^{2} + \Omega_{3}^{2}\right) \left(\Omega_{1}^{2}\gamma_{3} + \Omega_{3}^{2}\gamma_{1}\right) - \left(\Omega_{1}^{2} - \Omega_{3}^{2}\right) \left(\Omega_{1}^{2}\gamma_{3} - \Omega_{3}^{2}\gamma_{1}\right) (\gamma_{1} + \gamma_{3})^{2}R^{2}\right]^{-1}.$$
(46)

Provided that conditions (44) are satisfied for a wide enough spectrum of laser radiation, when $\omega_{13}/n_0 \varpi \ll$ $\varpi/(\gamma_1 + \gamma_3)$, the second term in the square brackets in Eq. (46) is much less than the first one; then, by the order of magnitude, $\delta_R \sim (\gamma_1 + \gamma_3)R/\gamma_0$. In expression (45), the second term plays a dominating role, as a rule, in those partial cases where the first term is small, e.g., if R = 0. Then,

$$\delta_S = \frac{1}{2} \left(S_1 - S_3 \right). \tag{47}$$

The obtained expression for the resonance shift is valid in the case of a cell without buffer gas, when the relaxation rate of atoms in the excited state is governed by the spontaneous emission, and all relaxation rates are

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much slower than ϖ . In the presence of a buffer gas, the relaxation rate for the optical coherences considerably exceeds ϖ . Therefore, the interaction between the atom and the polychromatic field cannot be characterized any more by an effective Hamiltonian. Such a case was analyzed in work [22].

If the radiation spectrum width considerably exceeds ω_{13} , the Rabi frequencies of spectral components, which are resonant to the atomic transition frequency, differ very slightly from the frequency of the Rabi component with the maximal intensity. Taking into account that they are proportional to the dipole moment of the corresponding transition and the probability of the transition from the excited state into state $|1\rangle$ or $|3\rangle$ is proportional to the square of this quantity, we easily obtain the relation

$$\frac{\Omega_1^2}{\gamma_1} = \frac{\Omega_3^2}{\gamma_3},\tag{48}$$

so that Eq. (46) takes the form

$$\delta_R = \frac{\gamma_3 - \gamma_1}{2\Omega_1 \Omega_3} \gamma_0 R. \tag{49}$$

Hence, $\delta_R = 0$, if $\gamma_3 = \gamma_1$. In this case, the resonance shift (45) is determined by the term δ_S .

In Fig. 2, the dependences of population ρ_{22} in the excited state on the two-photon detuning δ from the resonance are depicted. They were obtained by solving Eqs. (43) for the density matrix under condition (48). As is evident from the figure, the position of the excited state population minimum at $R \neq 0$ obtained from the solution of the equations for the density matrix agrees well with the result of calculations by formula (49). The minimum of the curve ($\delta_S = -0.005\gamma_0$) obtained for the case R = 0 also agrees well with the result of calculations by formula (47), which is valid in this case.

7. Conclusions

To summarize, we showed that, if a three-level atom interacts with a polychromatic field with equidistant frequencies, there emerges a state, which is similar to that of coherent population trapping in a bichromatic field. In contrast to the case of bichromatic field, this state, besides metastable states, also includes an insignificant admixture of the excited state, the amplitude of which is smaller, the wider is the spectra of laser radiation. The effective Hamiltonian, which describes a slow, in comparison with the field period, variation of the probability amplitudes for atomic state populations, contains





Fig. 2. Dependences of population ρ_{22} in the excited state on the two-photon detuning δ from the resonance obtained from the solution of Eqs. (43) for the density matrix under conditions (48). The calculation parameters are $\Omega_1 = \gamma_0$, $\Omega_3 = 2\gamma_0$, $\gamma_1 + \gamma_3 =$ $100\gamma_0$, $S_1 = 0.01\gamma_0$, $S_3 = 0.02\gamma_0$, and $\delta_1 = 0$. The solid curve corresponds to $R = 0.01\gamma_0$, the dashed one to R = 0. The vertical line denotes the position of the excited state population minimum $(\delta_R = 0.15\gamma_0)$ calculated by formula (49)

a coupling between the metastable states. Such a coupling gives rise, generally speaking, to a resonance shift in the coherent population trapping.

An application of the developed theory to the creation of a frequency reference on the basis of an ensemble of cold ²²⁹Th atoms or ions [27] could be of interest. The corresponding split in the nuclear ground state is about 7.6 eV [28] (vacuum ultraviolet), and the lifetime of the nuclear excited state (an hour by the order of magnitude) [29] testifies in favor of the creation of a high-precision clock on the basis of the transition in this nucleus.

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- E. Arimondo and G. Orriols, Nuovo Cim. Lett. 17, 333 (1976).
- G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, Nuovo Cim. B Ser. 2 36, 5 (1976).
- H.R. Gray, R.W. Whitley, and C.R. Stroud, jr., Opt. Lett. 3, 218 (1978).
- 4. G. Orriols, Nuovo Cim. B Ser. 2 53, 1 (1979).
- G. Alzetta, L. Moi, and G. Orriols, Nuovo Cim. B Ser. 2 52, 209 (1979).
- O.A. Kocharovskaya and Ya.I. Khanin, Sov. Phys. JETP 63, 945 (1986).
- M.B. Gornyĭ, B.G. Matisov, and Yu.V. Rozhdestvenskiĭ, Sov. Phys. JETP 68, 728 (1989).

- K.-J. Boller, A. Imamoglu, and S.E. Harris, Phys. Rev. Lett. 66, 2593 (1991).
- 9. S.E. Harris, Phys. Today 50, 36 (1997).
- J. Oreg, F.T. Hioe, and J.H. Eberly, Phys. Rev. A 29, 690 (1984).
- U. Gaubatz, P. Rudecki, M. Becker, S. Schiemann, M. Kulz, and K. Bergmann, Chem. Phys. Lett. **149**, 463 (1988).
- K. Bergmann, H. Theur, and B.W. Shore, Rev. Mod. Phys. 70, 1003 (1998).
- N.V. Vitanov, T. Halfmann, B.W. Shore, and K. Bergmann, Ann. Rev. Phys. Chem. 52, 763 (2001).
- S. Knappe, R. Wynands, J. Kitching, H.G. Robinson, and L. Hollberg, J. Opt. Soc. Am. B 18, 1545 (2001).
- A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste, and C. Cohen-Tannoudji, Phys. Rev. Lett. **61**, 826 (1988).
- G. Alzetta, S. Gozzini, A. Lucchesini, S. Cartaleva, T. Karaulanov, C. Marinelli, and L. Moi, Phys. Rev. A 69, 063815 (2004).
- V.A. Sautenkov, Yu.V. Rostovtsev, C.Y. Ye, G.R. Welch, O. Kocharovskaya, and M.O. Scully, Phys. Rev. A 71, 063804 (2005).
- L. Arissian and J.-C. Diels, Opt. Commun. 264, 169 (2006).
- Yu.V. Vladimirova, B.A. Grishanin, V.N. Zadkov, V. Biancalana, G. Bevilacqua, Y. Dancheva, and L. Moi, Zh. Èksp. Teor. Fiz. **103**, 528 (2006).
- M. Auzinsh, R.A. Malitskiy, I.V. Matsnev, A.M. Negriyko, V.I. Romanenko, V.M. Khodakovskyy, and L.P. Yatsenko, Ukr. Fiz. Zh. 54, 975 (2009).
- L. P. Yatsenko, B. W. Shore, and K. Bergmann, Opt. Commun. 236, 183 (2004).
- V.I. Romanenko, A.V. Romanenko, L.P. Yatsenko, G.A. Kazakov, A.N. Litvinov, B.G. Matisov, and Yu.V. Rozhdestvensky, J. Phys. B 43, 215402 (2010).

- L.D. Landau and E.M. Lifshitz, *Mechanics* (Pergamon Press, Oxford, 1976).
- 24. M. Cashen, O. Rivoire, V. Romanenko, L. Yatsenko, and H. Metcalf, Phys. Rev. A 64, 063411 (2001).
- V.I. Romanenko, A.V. Romanenko, and L.P. Yatsenko, J. Phys. B 44, 215402 (2011).
- B.W. Shore, The Theory of Coherent Atomic Excitation (Wiley, New York, 1990).
- 27. E. Peik and Chr. Tamm, Europhys. Lett. 61, 181 (2003).
- 28. B.R. Beck, J.A. Becker, P. Beiersdorfer, G.V. Brown, K.J. Moody, J.B, Wilhelmy, F.S. Porter, C.A. Kilbourne, and R.L. Kelly, Phys. Rev. Lett. 98, 142501 (2007).
- E.V. Tkalya, A.N. Zherykhin, and V.I. Zhudov, Phys. Rev. C 61, 064380 (2000).

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АНАЛОГ СТАНУ КОГЕРЕНТНОГО ПОЛОНЕННЯ НАСЕЛЕНОСТЕЙ У ПОЛІХРОМАТИЧНОМУ ПОЛІ

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Резюме

Теоретично досліджено взаємодію трирівневого атома з поліхроматичним полем з еквідістантними спектральними компонентами (Λ-схема взаємодії з полем). Показано, що взаємодія атома з таким полем зводиться до взаємодії з біхроматичним полем з додатковими світловими зсувами частот переходів і додатковим, пропорційним інтенсивності поля, зв'язком нижніх рівнів атома. Наявність цього зв'язку дозволяє говорити про когерентне полонення населеності лише наближено, оскільки у загальному випадку довільних дипольних моментів переходів темний стан не є власним станом ефективного гамільтоніана. Розглянута модель дає просту теоретичну інтерпретацію формування близького до когерентного полонення населеностей стану атома у полі випромінювання фемтосекундного лазера.