

Population distribution over energy states of a three-level system interacting with three strong resonance fields

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Received February 14, 2000

We describe the steady-state distribution of population over energy states of a three-level atom interacting with the fields of three monochromatic waves of arbitrary intensities. In so doing spontaneous and collisional relaxation of energy levels and transitions is taken into account. Using the exact algebraic solution of the problem, obtained for the first time in the approximation of a rotating wave, and the relaxation constants, we analyze, for all the possible configurations of the system, the behavior of the energy level populations as functions of the field amplitudes, frequency detuning, and the resultant phase of the fields. It is found that populations depend on the resultant phase of the fields even within the limits of high intensities that results in an irremovable noise in the contours of absorption and spontaneous emission lines due to radiation fluctuations.

Introduction

A three-level model of quantum systems resonantly interacting with radiation is one of the most widespread models for atoms, molecules, and solid bodies. In accordance with the selection rules for the electric dipole interaction of atoms with radiation, we distinguish three possible configurations of a three-level system, namely, Λ -system, in which the transitions between the upper and two lower states are allowed, while the dipole transition between the two lower states being forbidden, V-system with the allowed transitions between the lower and the two upper states, and Ξ -system in which the step transitions are allowed from the lower to the mid state and from the mid one to the upper state (ladder-type system).

The exact solution of problems on the resonance interaction of the three-level systems in different configurations with one and two strong fields is known practically for all situations (see, for examples, Refs. 1–4). The exact solution provides a quantitative explanation of a series of experimentally observed nontrivial effects. In the case of two and more fields, the nonlinear interference effects (NIE)^{1–6} due to interference of polarizations induced by the fields on coupled transitions must be added to such effects. In a Λ -system, interacting resonantly with two fields, under certain conditions the effect of coherent capture of population (CCP) occurs^{7,8} that results in the clearing up of the medium due to formation of the superposition state in which the entire population is shared by two lower levels and no radiation absorption takes place.

The consequences of NIE and CCP are in drastic transformations of shapes of the absorption and spontaneous emission lines, redistribution of the level population and variation of the absorption cross section resulting in the medium clearing up. As the condition for the above nonlinear field effects to occur is high radiation intensity, enough, for example, for the absorption saturation, to predict and analyze the consequences of the nonlinear effects the exact solution of the problem on the steady-state interaction of the system with the fields, which is not limited by the scope of the perturbation

theory, is needed. Such a solution, being derived with regard for spontaneous and collisional relaxation, is rather cumbersome. Moreover, no such a solution has been derived for the most general case of ring scheme of interaction between the three-level system with three resonance fields, when one of these acts at the frequency of a forbidden transition.

The third strong field, interacting with a forbidden transition, essentially affects the NIE and CCP and, besides, introduces a new qualitative factor – the relations among the field phases. As shown in Ref. 9, in the absence of radiative and collisional relaxation the population distribution and the absorption in three- and four-level systems are very sensitive to the resultant field phase. This made a basis for the method of atomic interferometry proposed in that paper. An alternative example using the phase relations is the problem on deep-freezing of atoms modeled by W -scheme of levels (double Λ -system) under conditions of ring field summation.¹⁰ In this case, owing to a special selection of phases, it was possible to propose such an experiment that would enable obtaining previously inaccessible degrees of cooling.

The goal of the paper is the exact (within the limits of resonance approximation) solution of the problem on the population distribution over energy states of a three-level system of arbitrary configuration, given its radiative and collisional relaxation, which resonantly interacts with three strong fields. We also aimed at analyzing the effect of a strong field at the forbidden transition of the system and the resultant field phase on the NIE and CCP.

1. Statement of the problem and its general solution

Now consider a closed three-level system being in the field of three monochromatic waves. We consider that each of the waves interacts only with one of the system transitions, and that the frequency detuning of the waves ω_i from the natural frequencies ω_j of the resonant transitions

$$\Omega_1 = -\omega_1 + \omega_{31}, \quad \Omega_2 = \omega_2 - \omega_{32}, \quad \Omega_3 = \omega_3 - \omega_{21} \quad (1)$$

are arbitrary in value and satisfy the conditions of a cyclic resonance (ring interaction of the fields with the system)

$$\Omega_1 + \Omega_2 + \Omega_3 = 0. \quad (2)$$

The quantum system, consisting of the ground state (1), medium state (2), and the upper state (3), is described by the density matrix $\hat{\rho}$, whose diagonal elements $\rho_{ii} \Rightarrow \rho_i$ are the level populations. For the off-diagonal elements of the density matrix ρ_{ij} , which are, accurate to a constant factors, the polarizations of the corresponding transitions, we introduce the following designations:

$$R_1 = \rho_{31}, \quad R_2 = \rho_{32}, \quad R_3 = \rho_{21}. \quad (3)$$

Here and below, when applied to all configurations of the system, the transition between the lower (1) and upper (3) level is called the first transition and it is designated by the index 1, the transition between the mid level (2) and the upper (3) level by 2, and between the lower (1) and the mid level (2) by 3.

We use the standard quantum kinetic equation for the density matrix of the medium in the model of relaxation constants for the case of homogeneous broadening.¹ In the representation of interaction and resonance approximation of a rotating wave the dynamics of the considered model of the three-level system is fully determined by six differential equations. Taking into account condition (2) one can transform these equations to the stationary case:

$$\begin{aligned} & -2 \operatorname{Im}(V_1^* R_1) - 2 \operatorname{Im}(V_3^* R_3) - (\gamma_1 + \gamma_2) \rho_1 + \\ & + (A_3 + \gamma_1) \rho_2 + (A_1 + \gamma_2) \rho_3 = 0, \\ & -2 \operatorname{Im}(V_2^* R_2) + 2 \operatorname{Im}(V_3^* R_3) + \gamma_1 \rho_1 - \\ & - (A_3 + \gamma_1 + \gamma_3) \rho_2 + (A_2 + \gamma_3) \rho_3 = 0, \\ & R_1(\Gamma_1 - i(\Omega_2 + \Omega_3)) + iR_2V_3 - iR_3V_2 = iV_1(\rho_1 - \rho_3); \quad (4) \\ & R_2(\Gamma_2 - i\Omega_2) + iR_1V_3^* - iR_3^*V_1 = iV_2(\rho_2 - \rho_3), \\ & R_3(\Gamma_3 - i\Omega_3) - iR_1V_2^* + iR_2^*V_1 = iV_3(\rho_1 - \rho_2). \end{aligned}$$

Here $V_j = d_j E_j / 2\eta$ is the Raby frequency of the transition j ; d_j and E_j are the matrix element of the dipole moment and the electric field strength matched with the transition j ; A_j is the first Einstein coefficient of the corresponding transition; γ_j denotes the constants of collisional relaxation of the level; Γ_j denotes the constants of transition relaxation including the radiative decay determined by the Einstein coefficients and the collisional decay characterized by the constants $\tilde{\gamma}_j$:

$$\begin{aligned} \Gamma_1 &= (A_1 + A_2) / 2 + \tilde{\gamma}_1, \quad \Gamma_2 = (A_1 + A_2 + A_3) / 2 + \tilde{\gamma}_2, \\ \Gamma_3 &= A_3 / 2 + \tilde{\gamma}_3. \end{aligned} \quad (5)$$

In the case of a forbidden transition the field and the system interaction is considered to be of magnetic dipole type, so that $V_j = \mu_j H_j / 2\eta$, where μ_j is the matrix element of the magnetic dipole moment and H_j is the magnetic field strength.

The set of equations (4) should be completed with the normalization condition for the diagonal elements of the density matrix:

$$\rho_1 + \rho_2 + \rho_3 = 1. \quad (6)$$

Let us distinguish the phase factors in the Raby frequencies and determine the resultant field phase Φ :

$$V_j = v_j e^{i\varphi_j}, \quad v_j = v_j^*, \quad j = 1, 2, 3; \quad \Phi = \varphi_1 - \varphi_2 - \varphi_3. \quad (7)$$

The quantities sought in solving the problem (4)–(6) are the populations of the upper and mid levels (ρ_3 and ρ_2) as functions of the frequency detuning Ω_j , modules of the Raby frequencies v_j , the resultant phase Φ , the Einstein coefficients A_j , and of the constants of collisional relaxation $\gamma_j, \tilde{\gamma}_j$.

The exact solution of the set of equations (4) relative to ρ_2 and ρ_3 is given in the Annex.

Further, in considering the solutions, we have restricted ourselves to case without collisions, since the occurrence of NIE in its full measure is only possible in the absence of collisions.^{5,6,11,12} Assuming, in the complete solution, that $\gamma_j = 0$ and $\tilde{\gamma}_j = 0$, one can represent the population of levels as follows:

$$\begin{aligned} \rho_2 &= 2\zeta(2\zeta + A_1\beta_1 + A_2\beta_2) / d_0, \\ \rho_3 &= 2\zeta[2\zeta + A_2(\beta_2 - \beta_1)] / d_0; \\ d_0 &= 12\zeta^2 + 2\zeta[(A_1 + A_3)\alpha_1 + (A_2 - A_3)\alpha_2 + \\ & + (A_1 - 2A_3)\beta_1 + (A_2 + 2A_3)\beta_2] + \\ & + A_3(A_1 + A_2)(\alpha_1\beta_2 - \alpha_2\beta_1). \end{aligned} \quad (8)$$

The magnitudes of ζ , α_j , and β_j are given in the Annex. In this case, no collisional relaxation is included in the constants of the transition relaxation (5).

Since the total number of parameters of a solution to the problem (8), with the allowance for condition (2), equals 9 ($A_1, A_2, A_3, v_1, v_2, v_3, \Phi, \Omega_2, \Omega_3$), below we analyze only most interesting cases for all possible configurations of the system, considering the dependence of population of the two upper levels from detuning, field amplitudes and their resultant phase.

All analytical expressions for the population of levels, presented in the text and the Annex, were checked up by comparing their numerical values at some arbitrary values of the above parameters with the values obtained for the same sets of parameters using numerical solution of Eqs. (4) and (6).

2. The case of equal Raby frequencies

As the case of equal Raby frequencies for all the transitions is particular for a limiting transition to strong fields and also because the algebraic expressions for the populations in this case become quite comprehensible, we shall consider this situation in a more detail assuming the resultant field phase to be equal to zero

$$v_1 = v_2 = v_3 = v; \quad \Phi = 0. \quad (9)$$

Let us consider separately the Ξ -, V -, and Λ -system, for which the first, second and third transitions can be recognized to be of magnetic dipole type, i.e., A_1, A_2 , and $A_3 = 0$, respectively, and the Raby frequency of the forbidden transition is not equal to zero and is determined by the value of the magnetic dipole moment and the magnetic field strength of a light wave. Having substituted Eq. (9) in the solution (8) and using explicit expressions for all the coefficients of this solution, taken from the

Annex, we can obtain an analytical solution for each configuration of the system. The simplest expressions for population are obtained for the Λ -system when normalizing all the quantities having the frequency dimensionality to the Einstein coefficient of the first transition ($A_3 = 0, A_1 \equiv 1$):

$$\begin{aligned} \rho_2 = & \left[A_2^3 (2v^2 - 2v\Omega_3 + \Omega_3^2) + 2A_2^2 (3v^2 - 2v\Omega_3 + \Omega_3^2) + \right. \\ & + 2v^2 (1 + 4\Omega_2^2 + 4\Omega_2\Omega_3 + 2\Omega_3^2) + \\ & + A_2 (6v^2 - 2v\Omega_3 + (1 + 4v^2)\Omega_3^2) + \\ & \left. + 4\Omega_2^2 (2v^2 - 2v\Omega_3 + \Omega_3^2) \right] / d_1, \\ \rho_3 = & 4v^2 (1 + A_2)\Omega_3^2 / d_1; \tag{10} \\ d_1 = & 4v^2 + 2v\Omega_3 + \Omega_3^2 + 20v^2\Omega_3^2 + 8v\Omega_3^3 + 4\Omega_3^4 + \\ & + A_2^3 (4v^2 - 2v\Omega_3 + \Omega_3^2) + 8\Omega_2\Omega_3 (3v^2 + 2v\Omega_3 + \Omega_3^2) + \\ & + 4\Omega_2^2 (4v^2 + 2v\Omega_3 + \Omega_3^2) + A_2^2 (12v^2 - 2v\Omega_3 + 3\Omega_3^2) + \\ & + A_2 [12v^2 + 2v\Omega_3 + 8v^2\Omega_2\Omega_3 + \\ & + 3(1 + 4v^2)\Omega_3^2 + 4\Omega_2^2 (4v^2 - 2v\Omega_3 + \Omega_3^2)]. \end{aligned}$$

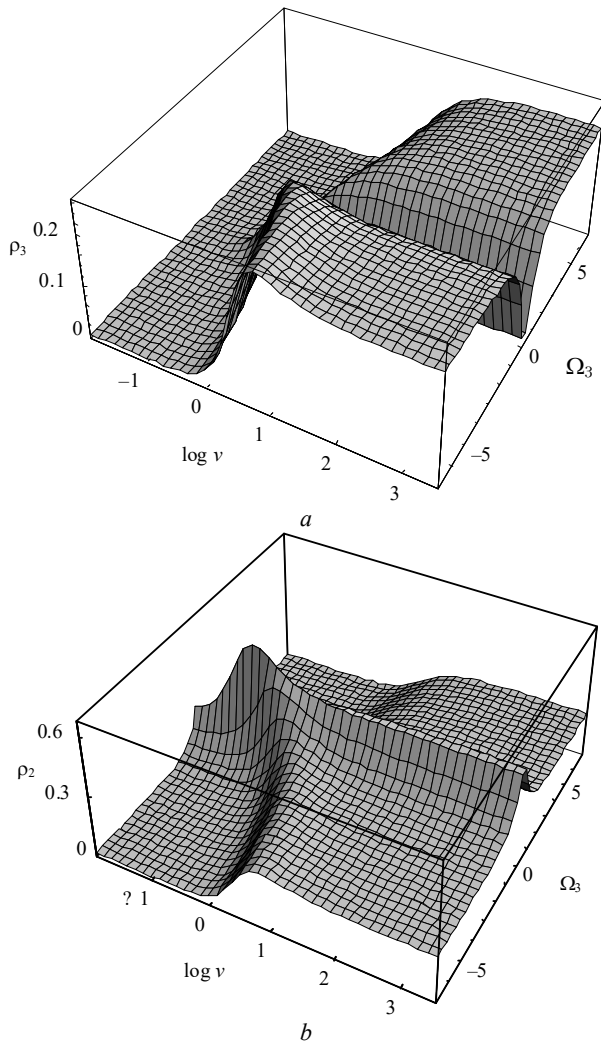


Fig. 1. The dependence of population of the third (a) and second (b) level of Λ -system on the frequency detuning and Raby frequency of the same absolute value for all transitions at $\Phi = 0, \Omega_3 = \Omega_1 (\Omega_2 = 0)$.

Equations (10) are valid for arbitrary though equal in absolute value amplitudes and any detuning of the field frequencies. Note that the population ρ_3 of the upper level of a Λ -system is directly proportional to the frequency detuning from the forbidden transition, and at the exact resonance of the field V_3 with the forbidden transition ($\Omega_3 = 0$) the effect of coherent capture of population occurs when $\rho_3 = 0$ and $\rho_1 = \rho_2 = 1/2$ independent of the field intensity and coefficients of spontaneous relaxation (Fig. 1a). A narrow dip in the upper level population close to zero frequency detuning from the forbidden transition having the width of the order of $A_1 = 1$, corresponds to CCP. Thus, in the presence of a strong field matched with the forbidden transition, when absolute values of amplitudes of all the fields are the same, CCP is possible only under conditions of exact or approximate resonance $\Omega_3 \approx 0$.

Expressions for the populations in the cases of Ξ - and V -configurations are more cumbersome being the fractions with polynomials of the fourth power of field amplitudes in the numerators and the denominators. Numerical values of the populations (8) for these cases are shown in Figs. 2 and 3, respectively, in the form of the population dependences on Raby frequency and frequency detuning at the allowed third transition given $A_1 = 0, A_2 = 1$, and $A_3 = 1/4$ for Ξ -system and $A_1 = 1/4, A_2 = 0, A_3 = 1$ for V -system under conditions of the exact field resonance with the forbidden transition.

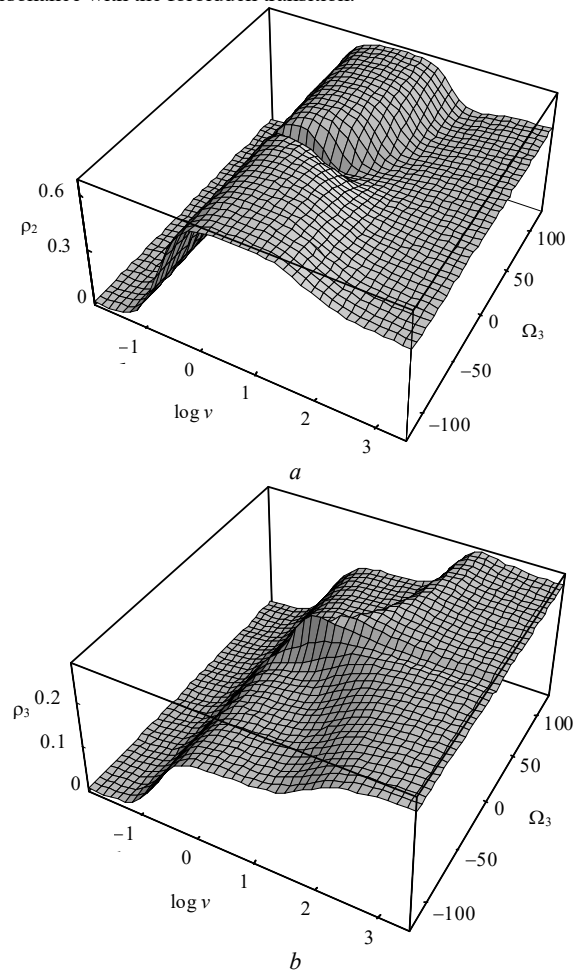


Fig. 2. The dependence of population of the second (a) and third (b) level of Ξ -system on the frequency detuning from the allowed transitions $\Omega_3 = -\Omega_2 (\Omega_1 = 0)$ and Raby frequency, equal for all the transitions, at $A_3/A_2 = 1/4$.

As is seen from Fig. 2, for Ξ -system at $A_1 = 0$, $A_2 = 1$, and $A_3 = 1/4$ the dependence of population of the mid level on the field intensity reaches maximum in the region of moderately strong fields and vanishes near $\Omega_3 = 0$. The field amplitude increase results in a slight decrease in ρ_2 and then in its stabilization. The frequency detuning from the third transition Ω_3 in the case of exact resonance $\Omega_1 = 0$ with the forbidden transition, when $\Omega_2 = -\Omega_3$ does not affect qualitatively the population of the second level in the range of small and large Raby frequencies, but in the range of moderately strong fields at a resonance with the allowed transitions ($\Omega_2 = \Omega_3 = 0$) the second level population has a narrow dip. In this case the resonance transfer of population to the third level occurs: ρ_3 reaches its highest value in the range of moderate fields, and ρ_2 goes into the range of saturation. In the range of strong fields, the population of each level is close to $1/3$, but no exact equality is attained in this case, as is seen from Fig. 2.

Population of the second and third level of a V -system for a specific set of parameters $A_1 = 1/4$, $A_2 = 0$, $A_3 = 1$ depends qualitatively on the Raby frequency and frequency detuning from the first and third transitions. Therefore Fig. 3 shows only the dependence of the third level population ρ_3 on the Raby frequency and detuning $\Omega_3 = \Omega_1$. The relative difference of populations of two upper levels $(\rho_3 - \rho_2)/\rho_2$ is less than 1% and reaches maximum at $\Omega_3 = 0$. As is seen from the relationship given below, the population of the second and third level is exactly the same if $A_1 = 1$, i.e., the Einstein coefficients of the allowed transitions are equal to:

$$\rho_3 - \rho_2 = 4v^2(8v^2 + A_1)(1 - A_1^2) / [A_1^3(1 + 8v^2 + 4\Omega_3^2) + 4v^2A_1^2(5 + 16v^2 + 8v\Omega_3 + 4\Omega_3^2) + 4A_1(2v^2 + 48v^4 - 24v^3\Omega_3 + \Omega_3^2 + 16v^2\Omega_3^2 + 4\Omega_3^4) + 16v^2(4v^2 + 2v\Omega_3 + \Omega_3^2 + 20v^2\Omega_3^2 + 8v\Omega_3^3 + 4\Omega_3^4)]. \quad (11)$$

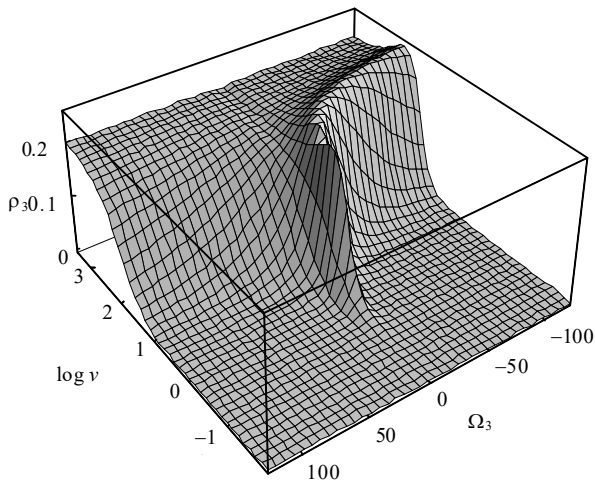


Fig. 3. The dependence of the V -system second level population on the frequency detuning from the allowed transitions $\Omega_3 = \Omega_1$ ($\Omega_2 = 0$) and Raby frequency equal for all transitions, at $A_1/A_3 = 1/4$.

Comparing the behavior of population for Ξ - and V -systems (Figs. 2 and 3), one can note that in the range of strong fields in both cases the upper level population reaches its maximum. Although in the case of a Ξ -system, the middle and upper levels

have different field dependences but they depend symmetrically on the frequency detuning and in the case of V -system the upper-level populations are practically equal but have a marked asymmetry of the dependence on the frequency detuning. On the diagram of Fig. 3, with the increase of the Raby frequency, the resonance peak of the second level population, increasing somewhat in width, shifts to the range of negative frequency detuning, and then, because of essential broadening, covers the range of positive detuning. The dependences of the second and third level populations of Ξ -system on the frequency detuning (Fig. 1a and b) are also asymmetric as in the case with V -system (Fig. 3), but ρ_2 has a small peak at the center, reaching $1/2$, and ρ_3 has a gap typical of the population capture. In the range outside the resonance, the upper and middle level populations of Λ -system show qualitatively same dependences on the field strength and frequency detuning.

Note that within the limits of strong fields and equal modules of Raby frequencies and zero resultant phase of the field the dependence of level populations on frequency detuning retains:

$$\begin{aligned} \rho_{2\text{lim}} = & [A_1^4 + A_2^4 + 4A_2^3A_3 + A_1^3(4A_2 + 3A_3) + \\ & + 2A_3^2(\Omega_2 + \Omega_3)^2 + A_2^2(5A_3^2 + 4\Omega_2^2 + 2\Omega_3^2) + \\ & + 2A_2A_3(A_3^2 + 4\Omega_2^2 + 2\Omega_2\Omega_3 + 2\Omega_3^2) + \\ & + A_1^2(6A_2^2 + 10A_2A_3 + 3A_3^2 + 4\Omega_2^2 + 4\Omega_2\Omega_3 + 2\Omega_3^2) + \\ & + A_1(4A_2^3 + 11A_2^2A_3 + 4A_2(2A_3^2 + 2\Omega_2^2 + \Omega_2\Omega_3 + \Omega_3^2) + \\ & + A_3(A_3^2 + 4(2\Omega_2^2 + 2\Omega_2\Omega_3 + \Omega_3^2)))] / d_{\text{lim}}; \\ \rho_{3\text{lim}} = & [A_1^3A_3 + A_2^3A_3 + A_2^2(3A_3^2 + 2\Omega_3^2) + \\ & + A_1^2(3A_2A_3 + 3A_3^2 + 2\Omega_3^2) + A_3^2(A_3^2 + 2(\Omega_2^2 + 2\Omega_3^2)) + \\ & + A_2A_3(3A_3^2 + 4(\Omega_2^2 + \Omega_3^2)) + \\ & + A_1(3A_2^2A_3 + A_2(6A_3^2 + 4\Omega_2^2) + \\ & + A_3(3A_3^2 + 4(\Omega_2^2 + 2\Omega_3^2)))] / d_{\text{lim}}; \\ d_{\text{lim}} = & 2A_1^4 + 2A_2^4 + 10A_2^3A_3 + 2A_1^3(4A_2 + 5A_3) + \\ & + 2A_3^2(A_3^2 + 3\Omega_2^2 + 4\Omega_2\Omega_3 + 5\Omega_3^2) + \\ & + 2A_1^2(6A_2^2 + 15A_2A_3 + 8A_3^2 + 4\Omega_2^2 + 6\Omega_2\Omega_3 + 5\Omega_3^2) + \\ & + A_2^2(17A_3^2 + 8\Omega_2^2 + 4\Omega_2\Omega_3 + 6\Omega_3^2) + \\ & + A_2A_3(11A_3^2 + 4(5\Omega_2^2 + 3\Omega_2\Omega_3 + 2\Omega_3^2)) + \\ & + A_1(4A_2 + 5A_3)(2A_2^2 + 5A_2A_3 + \\ & + 2(A_3^2 + 2(\Omega_2^2 + \Omega_2\Omega_3 + \Omega_3^2))). \end{aligned} \quad (12)$$

Expressions (12) are valid for any configuration of the considered three-level system and actually have more simple form because only one of the Einstein coefficients must be equal to zero, namely, the coefficient, which corresponds in this configuration to the forbidden transition.

3. The dependence on phase in the case of Raby frequencies of equal moduli

In this section we consider the dependence of populations on the resultant phase of fields Φ .

When Raby frequencies differ only in phase for all transitions, the solution of Eq. (8) includes trigonometric functions with the resultant phase, and in this case the numerators and denominators of expressions for populations are polynomials of 12th power of the field amplitudes. As in the previous section, we consider numerical solutions separately for each individual configuration of a system.

Figure 4 shows the dependence of the second and third level populations of Λ -system for $\nu = 2, A_1 = 1, A_2 = 1/4, A_3 = 0, \Omega_1 = \Omega_2$ on the frequency detuning Ω_2 and the resultant phase Φ . The population ρ_3 has the Lorentz dependence on the frequency detuning (that enters the denominator squared) and reaches its maximum at the exact resonance $\Omega_2 = 0$ and in the range $\Phi = \pi(2n - 1)/2$ (here and below n is the integer number). The maximum value decreases rapidly to zero at $\Phi = \pi n$.

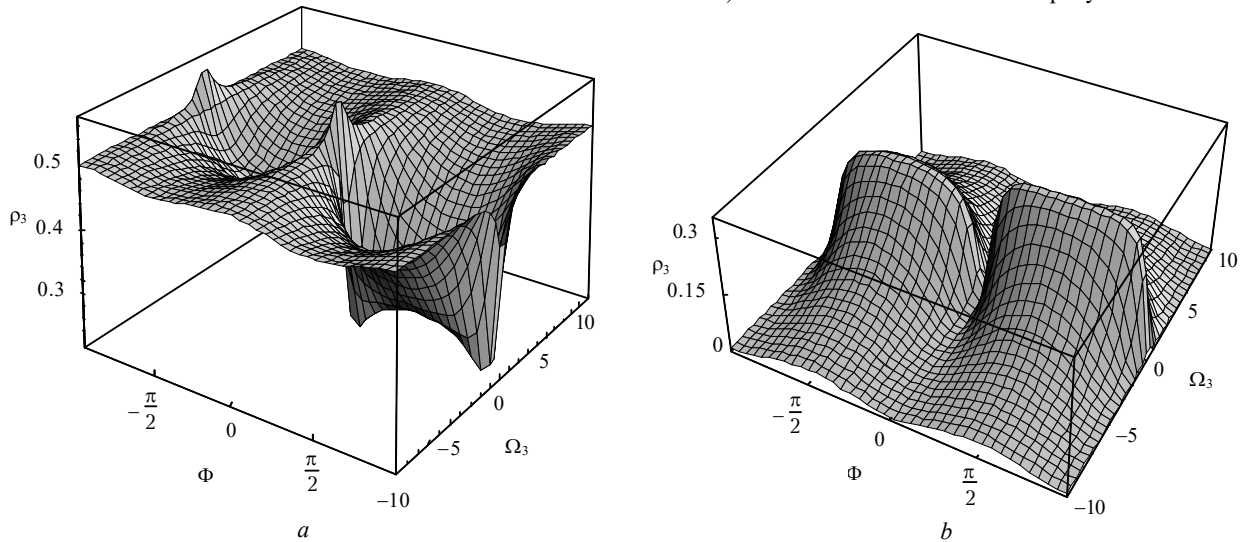


Fig. 4. The dependence of the Λ -system second (a) and third (b) level populations on the resultant phase and the frequency detuning $\Omega_2 = -\Omega_1$ ($\Omega_3 = 0$) at $\nu = 2$ and $A_2/A_1 = 1/4$.

Figures 5 and 6 show the dependences of populations on the Raby frequency and resultant phase for Ξ -system at $A_1 = 0, A_2 = 1, A_3 = 1/4$ and for V -system at $A_1 = 1/4, A_2 = 0, A_3 = 1$. As can be seen from Figs. 5 and 6, the phase dependence is essential only in the range of mean values of the Raby frequency. Within the limit of strong field all populations are equal to $1/3$ that means that no NIE occurs.

Qualitatively the phase dependence of the third level population in a Ξ -system is similar to that for the second level of a V -system: in the range of zero values of Φ the limiting level of population can be obtained at a stronger field than in the range $\Phi = \pi(2n - 1)/2$. In the range of moderate fields the population of third level of a V -system has its maximum at $\Phi = \pi/2$ and minimum at $\Phi = -\pi/2$.

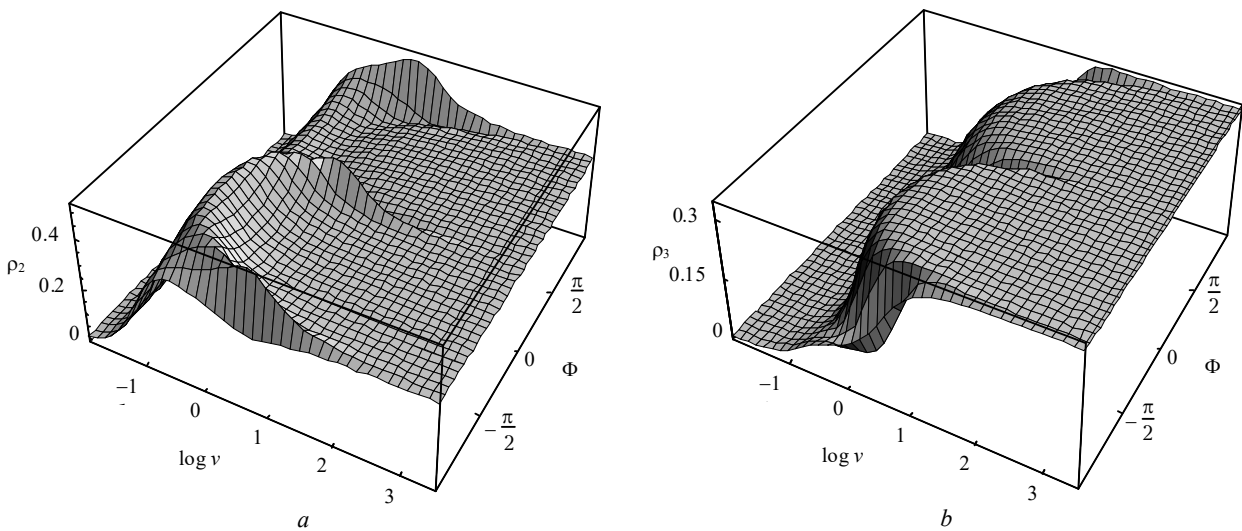


Fig. 5. The dependence of population of the second (a) and third (b) level in a Ξ -system on the resultant phase and Raby frequency equal in modulus for all transitions, at $\Omega_1 = \Omega_2 = \Omega_3 = 0$ and $A_3/A_2 = 1/4$.

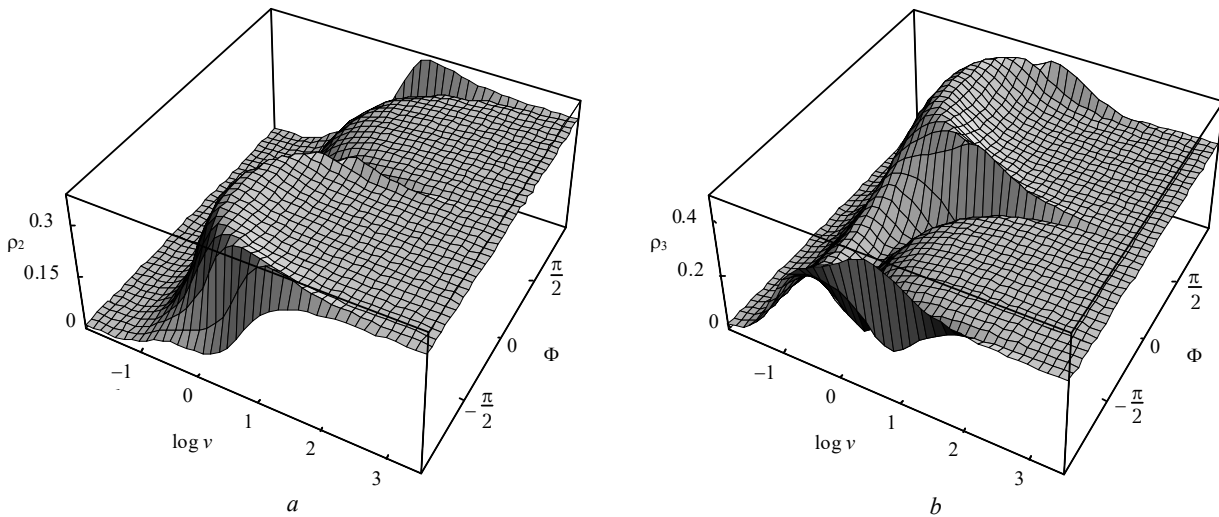


Fig. 6. The dependence of population of the second (a) and third (b) level in a V-system on the resultant phase and Raby frequency equal in modulus for all transitions, at $\Omega_1 = \Omega_2 = \Omega_3 = 0$ and $A_3/A_2 = 1/4$.

4. Solution for the general case in the range of strong fields

In this section, we consider the population distribution in the case of large, but not equal in value, Raby frequencies in the absence of the collisional relaxation. The exact solutions of Eq. (8) for the populations, in this case, are the ratios of polynomials of the twelfth power of Raby frequencies that reduce, in the limit of $\nu_j \rightarrow \infty$, to:

$$\rho_j = \frac{1}{3} + \frac{a_j}{b - c \cdot \cos^2 \Phi}, \tag{13}$$

where the coefficients a_j , b , and c depend on the coefficients of spontaneous relaxation and the ratios $\theta_2 = (\nu_2/\nu_1)^2$, $\theta_3 = (\nu_3/\nu_1)^2$ of modules of Raby frequencies as follows:

$$\begin{aligned} a_1 &= Z(2\theta_2 - \theta_3 - 1), \quad a_2 = Z(2 - \theta_2 - \theta_3), \\ a_3 &= Z(2\theta_3 - \theta_2 - 1); \\ Z &= [A_1(\theta_2 - \theta_3) + A_2(1 - \theta_3) + A_3(\theta_2 - 1)] \times \\ &\quad \times [A_1 + A_2 + (A_1 + A_2 + A_3)\theta_2 + A_3\theta_3] / 9; \\ 3b &= (A_1 + A_2)(A_2 + 2A_3) + \\ &+ \theta_2 [A_1^2 + A_2A_3 + 2(A_2 + A_3)^2 + A_1(3A_2 + 2A_3)] + \\ &+ \theta_3 [2A_1^2 + 2A_1(A_2 + A_3) + A_3(3A_2 + 2A_3)] + \\ &+ 2\theta_2\theta_3 [A_1^2 + (A_2 + A_3)^2 + A_1(2A_2 + 3A_3)] + \\ &+ \theta_2^2 [(A_1 + A_2)(2A_1 + A_2 + 2A_3)] + \\ &+ 2\theta_3^2 [A_1^2 + A_2^2 + A_3^2 + A_1(2A_2 + A_3)] + \\ &+ \theta_2^2\theta_3 [2(A_2 + A_3)(A_1 + A_2 + A_3) - A_2A_3] + \\ &+ \theta_2\theta_3^2 [2(A_1 + A_2)^2 + A_3(2A_1 + 4A_2 + A_3)] + \\ &+ \theta_3^3 [(A_1 + A_3)(A_1 + A_2 + A_3)] + 2\theta_3^3 A_3(A_1 + A_2); \\ c &= 4(A_1 + A_2 + A_3)^2 \theta_2 \theta_3. \end{aligned} \tag{14}$$

The components b and c are always positive. In this case, the numerical investigations indicated that in the range of allowed positive values of Einstein coefficients and squares of relative Raby frequencies θ_2 , θ_3 the inequality $b > c$ always holds, and therefore the populations (13), as was to be expected, have no any singularity.

We assume the factor Z in Eq. (14) to be zero, and the conditions can easily be found under which for all j levels the equation $a_j = 0$ is fulfilled, and, as a consequence, all populations become equal to 1/3 and are independent of the resultant phase (the condition of an efficient suppression of interference):

$$\theta_2(A_1 + A_2) = \theta_3(A_1 + A_2) - A_2 + A_3. \tag{15}$$

Now we set the factor in brackets after Z equal to zero in expressions (14), for the coefficients a_j , and for each j additional to (15) condition can easily be constructed under which the population of a given level also equals the equilibrium value 1/3. In this case, the population of two other levels depends on the ratios between the field intensity and Einstein coefficients and are different than 1/3. Now we follow the variability limits of two other populations $\rho_{m,n}$, which, as follows from (6) and (13), can be represented as $\rho_{m,n} = 1/3 \pm \Delta$, where the sign before the second term depends on the values of the indices $m, n \neq j$. The results of numerical determination of the value of nonequilibrium addition to the population Δ are given in the table.

In the 4th, 6th and 8th columns of the table the ranges of variation of the nonequilibrium addition Δ are given relative to the level population for each of the possible configurations of the system, for the cases, when the population of one of the levels, shown in the first column, equals 1/3. The variation limits of the resultant phase at the numerical search of Δ were from $-\pi$ to π . The results, given in the table, correspond to the maximum in absolute value of Δ at changes of θ_3 within the limits from 0 to 10 and given positive values of θ_2 presented in the first column. To specify the sign of Δ , the second column gives the levels, for which the addition has the indicated sign. The sign of the addition for the remaining level, which is not

given in the first and second columns of the table, is, in this case, opposite to that shown in the table.

Table 1 shows that, in some configurations of the system and certain relations between the parameters θ_2 and θ_3 , the nonequilibrium addition to the population of the level indicated in the second column of the table has one and the same sign for all values of Einstein coefficients, while in the other configurations and relations between the intensities the sign of the addition depends also on the relation between the coefficients of spontaneous relaxation. Therefore, in the 3rd, 5th, and 7th columns of the table the conditions are given these coefficients must satisfy.

Note that calculations showed that the squared value of the ratios of the field amplitudes θ_3 does not affect the sign of nonequilibrium addition Δ , but it is critical for its absolute

value. In other words, the relative value of the field matched with the forbidden transition in the cases of Ξ - and Λ -systems is the factor, which strongly determines the population distribution among the energy levels within the strong field approximation.

As follows from the table, the maximum of absolute value caused by NIE of the nonequilibrium part of the population Δ in most cases is comparable with the equilibrium value of the population 1/3, and in some cases the above maximum exactly equals this value. The latter means that by choosing the ratio between the field intensities, within the limits of high intensities, one can completely devastate certain levels of Λ - and Ξ -systems. For the V -system complete devastation of one of the levels cannot be reached but the populations in this case also vary within a wide range.

Table 1. The variation ranges of nonequilibrium addition to the level populations for three-level systems of different configurations

Ratio of intensities	Level	Limits of variation of nonequilibrium addition within the range of $\Phi [-\pi, \pi]$ variation					
		$\Xi (A_1 = 0, A_2 = 1)$		$V (A_2 = 0, A_3 = 1)$		$\Lambda (A_3 = 0, A_1 = 1)$	
$2\theta_2 = 1 + \theta_3$ ($\rho_1 = 1/3$) $\theta_2 = 2 - \theta_3$ ($\rho_2 = 1/3$)	2	$A_3 < 2$	[0; 1/3]	$A_1 < 1$	[0; 0.313]	$10 > A_2 > 0$	[0; 1/3]
	1	$10 > A_3 > 2$	[-0.181; 0]	$10 > A_1 > 1$	[-3/13; 0]	$10 > A_2 > 0$	[0; 1/3]
$\theta_2 = 2\theta_3 - 1$ ($\rho_3 = 1/3$)	1	$10 > A_3 > 0$	[0; 1/3]	$10 > A_1 > 0$	[0; 7/22]	$10 > A_2 > 0$	[0; 1/3]
	1	$A_3 < 0.5$	[0; 0.167]	$10 > A_1 > 0$	[0.023; 0.327]	$A_2 < 1$	[0; 1.667]
		$10 > A_3 > 0.5$	[-0.283; 0]			$10 > A_2 > 1$	[-0.136; 0]

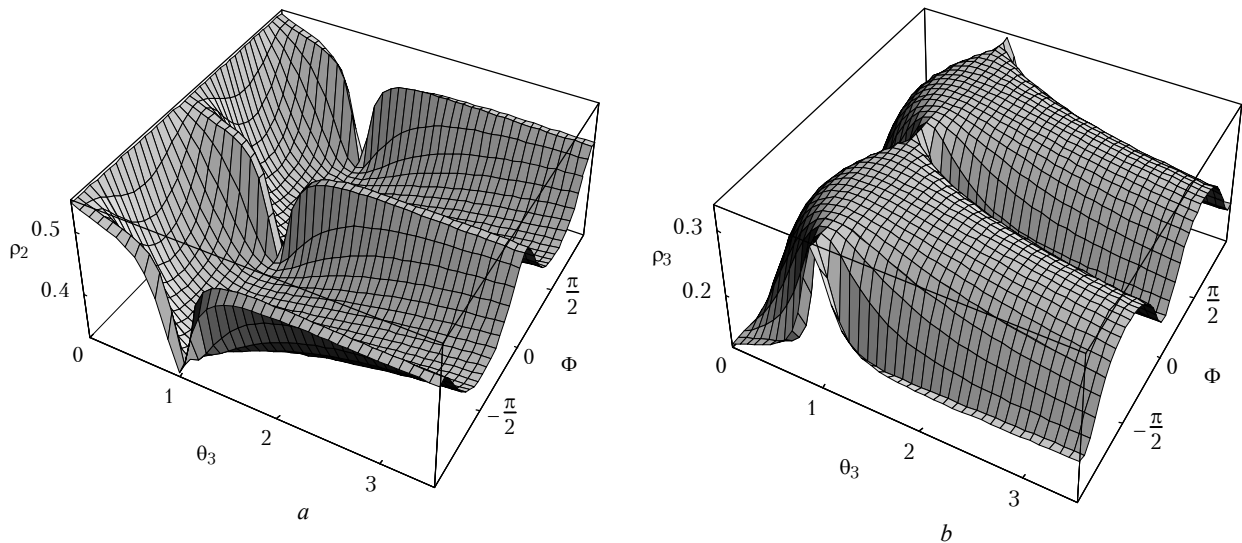


Fig. 7. The dependence of population of the second (a) and third (b) level of a Ξ -system on the resultant phase and the relative field intensity at allowed transitions in the range of strong fields at $A_3/A_2 = 1/4$, and $\theta_2 = (1 + \theta_3)/2$ ($\rho_1 = 1/3$).

The dependences of the levels' population on the ratio between the field intensities and their resultant phase within the strong field approach are presented in Fig. 7 for the Ξ -system, as an example. It should be noted that at the ratio of the squared Raby frequencies $\theta_3 \approx 1$, corresponding approximately to the condition (15), the dependence of populations of all levels on the phase practically disappears while at the other values of this parameter such a dependence is more pronounced.

5. Phase fluctuations

The above-mentioned dependence of the energy state population on the field phase ratio can manifest itself, due to their random fluctuations, in the experiments as an irremovable noise in shape of the absorption and spontaneous emission lines, which does not disappear even at strong fields. Assuming the resultant phase Φ to be uniformly distributed over the interval from 0 to 2π , within

the limits of strong fields (13), one can calculate the mean value of the population $\langle \rho_j \rangle$ of the level j and the root-mean-square deviation σ_j :

$$\langle \rho_j \rangle = \frac{1}{2\pi} \int_0^{2\pi} \rho_j(\Phi) d\Phi = \frac{1}{3} + \frac{a_j}{\sqrt{b^2 - c^2}}; \quad (16)$$

$$\sigma_j^2 = \int_0^{2\pi} (\rho_j - \langle \rho_j \rangle)^2 d\Phi = \frac{a_j^2}{b^2 - c^2} \left(\frac{b}{\sqrt{b^2 - c^2}} - 1 \right). \quad (17)$$

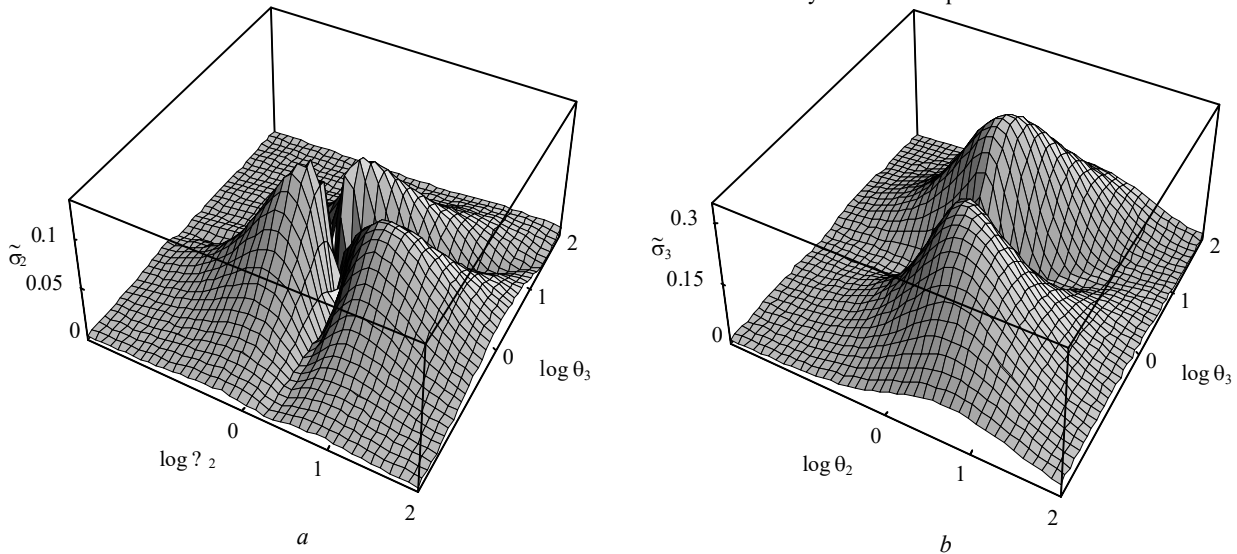


Fig. 8. The dependence of the relative rms deviation $\tilde{\sigma}_j = \sigma_j / \langle \rho_j \rangle$ for the second (a) and third (b) levels of Ξ -system on the logarithms of relative field intensities in the range of high intensities at $A_3/A_2 = 1/4$.

Conclusion

Precise algebraic expressions for population of energy states in a closed three-level system interacting with three fields of arbitrary intensities were first obtained allowing for the spontaneous and collisional relaxation in the framework of the resonance approximation. The above expressions also describe the result of interaction of the three-level system of an arbitrary configuration with one or two fields acting at any allowed and forbidden transitions. To choose the required type of the system, it is sufficient to assume, in the general solution, the individual field amplitudes and phases, which do not participate in the interaction with the system, to be equal to zero as well as the Einstein coefficients of any dipole-forbidden transitions.

We have considered in detail the behavior of population for all three possible configurations of the system depending on the amplitudes, frequency detuning, and the resultant phase of the fields. We have revealed the cases interesting from the viewpoint of many applications when the preferred population and devastation of system levels in different configurations are possible due to nonlinear interference effects. We have considered the influence of the field matched with the dipole-forbidden transition on the coherent capture of populations in the Λ -system.

Now we numerically determine the value of the relative root-mean-square deviation $\sigma_j / \langle \rho_j \rangle$. Let us make calculations for populations in the Ξ -system, as an example, and represent the results in the form of diagrams of the dependence of rms deviation of the second and third level populations on the ratio of squared Raby frequencies of the forbidden and allowed transitions (Fig. 8). As is seen from Fig. 8, the ranges of the maximum fluctuations of the middle and upper level populations do not coincide, and the relative rms deviation reaches 30%. Thus, the noise in the line contour due to nonlinear interference effects and random phase radiation modulation under conditions of ring combination of fields in the three-level system can be quite considerable.

The dependence of population on the resultant phase of fields was found to be essential not only in the range of moderate, but also in the limit of high radiation intensities. In the presence of a random phase radiation modulation, this results in the noise in the shape of absorption and spontaneous emission lines. The maximum relative value of the noise reaches 30% and it is not removed with the increase of radiation intensity.

Annex

The exact solution of equations (4) and (6) for the populations of the middle (2) and upper (3) levels of the three-level system interacting with three laser fields in the ring diagram of their combination with the account for the radiative and collisional relaxation has the form:

$$\rho_2 = \{4\zeta^2 + 2\zeta [A_1 \beta_1 - (\alpha_2 + \beta_1)\gamma_1 + \alpha_1 (\gamma_1 + \gamma_2) + \beta_2(A_2 + \gamma_1 + \gamma_3)] - (\alpha_2\beta_1 - \alpha_1\beta_2) \times (A_1\gamma_1 + A_2\gamma_1 + A_2\gamma_2 + \gamma_1\gamma_2 + \gamma_1\gamma_3 + \gamma_2\gamma_3)\} / d;$$

$$\rho_3 = \{4\zeta^2 - 2\zeta [A_3(\beta_1 - \beta_2) - (\alpha_1 - \alpha_2 - \beta_1 + \beta_2)\gamma_1 - \alpha_1\gamma_2 - \beta_2\gamma_3] - (\alpha_2\beta_1 - \alpha_1\beta_2) \times (A_3\gamma_2 + \gamma_1\gamma_2 + \gamma_1\gamma_3 + \gamma_2\gamma_3)\} / d;$$

$$\begin{aligned}
 d = & 12\zeta^2 + 2\zeta\{A_2\alpha_2 + A_1(\alpha_1 + \beta_1) + \\
 & + A_2\beta_2 + A_3(\alpha_1 - \alpha_2 - 2\beta_1 + 2\beta_2) - \\
 & - 3(\alpha_2 + \beta_1 - \beta_2)\gamma_1 + 3\alpha_1(\gamma_1 + \gamma_2) + 3\beta_2\gamma_3\} - \\
 & - (\alpha_2\beta_1 - \alpha_1\beta_2)\{(2A_3 + 3\gamma_1)\gamma_2 + A_2(A_3 + 2\gamma_1 + \gamma_2) + \\
 & + [A_3 + 3(\gamma_1 + \gamma_2)]\gamma_3 + A_1(A_3 + 2\gamma_1 + \gamma_3)\}; \\
 \zeta = & v_1^2 v_2^2 v_3^2 (\Gamma_1^2 + \Gamma_2^2 + \Gamma_3^2) + v_1^2 v_2^4 \Gamma_2 \Gamma_3 + \\
 & + v_1^4 \Gamma_1 (v_3^2 \Gamma_2 + v_2^2 \Gamma_3) + v_1^2 v_3^2 \Gamma_3 [v_3^2 \Gamma_2 + \Gamma_1 (\Gamma_2^2 + \Omega_2^2)] + \\
 & + v_1^2 v_2^2 \Gamma_1 \Gamma_2 (\Gamma_3^2 + \Omega_3^2) + \\
 & + v_1^2 v_2^2 \{v_2^2 \Gamma_1 \Gamma_2 + \Gamma_3 [v_3^2 \Gamma_1 + \Gamma_2 (\Gamma_1^2 + (\Omega_2 + \Omega_3)^2)]\} + \\
 & + 2\cos \Phi v_1 v_2 v_3 \{v_3^2 \Gamma_3 [(\Gamma_1 + \Gamma_2)\Omega_2 + \Gamma_2 \Omega_3] + \\
 & + v_1^2 \Gamma_1 (-\Gamma_3 \Omega_2 + \Gamma_2 \Omega_3) - v_2^2 \Gamma_2 [\Gamma_3 \Omega_2 + (\Gamma_1 + \Gamma_3)\Omega_3]\} - \\
 & - \cos^2 \Phi v_1^2 v_2^2 v_3^2 (\Gamma_1 + \Gamma_2 + \Gamma_3)^2; \\
 \alpha_1 = & v_2^6 \Gamma_2 + v_2^4 \{v_3^2 (-2\Gamma_2 + \Gamma_3) + \\
 & + 2\Gamma_2 [\Gamma_1 \Gamma_3 - \Omega_3 (\Omega_2 + \Omega_3)]\} + \\
 & + v_3^2 \Gamma_3 \{v_3^4 + 2v_3^2 [\Gamma_1 \Gamma_2 - \Omega_2 (\Omega_2 + \Omega_3)] + \\
 & + (\Gamma_2^2 + \Omega_2^2) [\Gamma_1^2 + (\Omega_2 + \Omega_3)^2]\} + \\
 & + v_1^2 \{v_2^4 \Gamma_1 + v_2^2 [v_3^2 \Gamma_1 + \Gamma_2 (\Gamma_1^2 + (\Omega_2 + \Omega_3)^2)] + \\
 & + v_2^2 [-2v_3^2 \Gamma_1 + \Gamma_3 (\Gamma_1^2 + (\Omega_2 + \Omega_3)^2)]\} + \\
 & + v_2^2 \{v_3^4 (\Gamma_2 - 2\Gamma_3) + \Gamma_2 (\Gamma_3^2 + \Omega_3^2) [\Gamma_1^2 + (\Omega_2 + \Omega_3)^2] + \\
 & + v_3^2 [2(\Omega_2 + \Omega_3)(\Gamma_3 \Omega_2 + \Gamma_2 \Omega_3) + \\
 & + \Gamma_1 (\Gamma_2^2 - 2\Gamma_2 \Gamma_3 + \Gamma_3^2 + (\Omega_2 + \Omega_3)^2)]\} + \\
 & + 2\cos \Phi v_1 v_2 v_3 \{(v_3^2 - v_2^2)(\Gamma_1 + \Gamma_2 + \Gamma_3)(\Omega_2 + \Omega_3) - \\
 & - (\Gamma_3 \Omega_2 - \Gamma_2 \Omega_3) [\Gamma_1^2 + (\Omega_2 + \Omega_3)^2]\}; \\
 \alpha_2 = & v_1^4 (v_2^2 - v_3^2) \Gamma_1 + v_1^2 \{v_2^4 \Gamma_2 + v_2^2 [\Gamma_1^2 \Gamma_2 + v_3^2 (\Gamma_1 - \Gamma_3) + \\
 & + (\Gamma_2 + \Gamma_3)\Omega_2 (\Omega_2 + \Omega_3) - \Gamma_1 (\Gamma_2 \Gamma_3 + \Omega_2 \Omega_3)] + \\
 & + v_2^2 \{-v_3^2 (\Gamma_1 + \Gamma_2 - \Gamma_3) + (\Omega_2 + \Omega_3)(\Gamma_3 \Omega_2 - \Gamma_2 \Omega_3) + \\
 & + \Gamma_1 (\Gamma_2 \Gamma_3 + \Omega_2 \Omega_3)\} + v_3^2 \{-v_2^4 \Gamma_2 + \\
 & + v_2^2 [v_3^2 (\Gamma_2 - \Gamma_3) + (\Omega_2 + \Omega_3)(\Gamma_3 \Omega_2 + \Gamma_2 \Omega_3) + \\
 & + \Gamma_1 (\Gamma_2 (\Gamma_2 - \Gamma_3) + \Omega_2 (\Omega_2 + \Omega_3))] + \Gamma_3 [v_3^4 + \\
 & + 2v_3^2 (\Gamma_1 \Gamma_2 - \Omega_2 (\Omega_2 + \Omega_3)) + \\
 & + (\Gamma_2^2 + \Omega_2^2) (\Gamma_1^2 + (\Omega_2 + \Omega_3)^2)]\} + \\
 & + \sin \Phi v_1 v_2 v_3 \{v_1^2 \Gamma_1 (\Gamma_1 + \Gamma_2 + \Gamma_3) + \\
 & + v_2^2 \Gamma_2 (\Gamma_1 + \Gamma_2 + \Gamma_3) + \\
 & + \Gamma_3 (v_3^2 + \Gamma_1 \Gamma_2) (\Gamma_1 + \Gamma_2 + \Gamma_3) + (\Gamma_1 + \Gamma_2 - \Gamma_3) \Omega_2^2 + \\
 & + (\Gamma_1 + \Gamma_2 - \Gamma_3) (\Gamma_1 - \Gamma_2 + \Gamma_3) \Omega_2 \Omega_3 + \\
 & + \Gamma_2 (\Gamma_1 - \Gamma_2 + \Gamma_3) \Omega_3^2\} + \\
 & + \cos \Phi v_1 v_2 v_3 \{-\Omega_2 [-2v_3^2 \Gamma_1 - 2v_3^2 \Gamma_2 - 2v_3^2 \Gamma_3 + \Gamma_1^2 \Gamma_3 + \\
 & + \Gamma_2^2 \Gamma_3 + \Gamma_1 \Gamma_3^2 + \Gamma_2 \Gamma_3^2 + v_2^2 (\Gamma_1 + \Gamma_2 + \Gamma_3) + 2\Gamma_3 \Omega_2^2] + \\
 & + \Omega_3 [-\Gamma_2 (\Gamma_1 + \Gamma_3) (-\Gamma_1 + \Gamma_2 + \Gamma_3) + \\
 & + v_3^2 (\Gamma_1 + \Gamma_2 + \Gamma_3) + (-\Gamma_1 + \Gamma_2 - 3\Gamma_3) \Omega_2^2] - \\
 & - (\Gamma_1 - \Gamma_2 + \Gamma_3) \Omega_2 \Omega_3 - v_1^2 (\Gamma_1 + \Gamma_2 + \Gamma_3) (\Omega_2 + \Omega_3)\}; \\
 \beta_1 = & v_1^4 (v_2^2 - v_3^2) \Gamma_1 - v_1^2 \{-v_2^4 \Gamma_2 + v_2^2 [v_3^2 (\Gamma_1 + \Gamma_2 - \Gamma_3) - \\
 & - (\Omega_2 + \Omega_3)(\Gamma_3 \Omega_2 - \Gamma_2 \Omega_3) - \Gamma_1 (\Gamma_2 \Gamma_3 + \Omega_2 \Omega_3)] + \\
 & + v_2^2 [-\Gamma_1^2 \Gamma_2 + v_3^2 (-\Gamma_1 + \Gamma_3) - \\
 & - (\Gamma_2 + \Gamma_3) \Omega_2 (\Omega_2 + \Omega_3) + \Gamma_1 (\Gamma_2 \Gamma_3 + \Omega_2 \Omega_3)]\} +
 \end{aligned}$$

$$\begin{aligned}
 & + v_3^2 \{-v_2^4 \Gamma_2 + v_2^2 [v_3^2 (\Gamma_2 - \Gamma_3) + \\
 & + (\Omega_2 + \Omega_3)(\Gamma_3 \Omega_2 + \Gamma_2 \Omega_3) + \\
 & + \Gamma_1 (\Gamma_2^2 - \Gamma_2 \Gamma_3 + \Omega_2 (\Omega_2 + \Omega_3))] + \Gamma_3 [v_3^4 + \\
 & + 2v_3^2 (\Gamma_1 \Gamma_2 - \Omega_2 (\Omega_2 + \Omega_3)) + \\
 & + (\Gamma_2^2 + \Omega_2^2) (\Gamma_1^2 + (\Omega_2 + \Omega_3)^2)]\} + \\
 & + \sin \Phi v_1 v_2 v_3 \{-v_1^2 \Gamma_1 (\Gamma_1 + \Gamma_2 + \Gamma_3) - \\
 & - v_2^2 \Gamma_2 (\Gamma_1 + \Gamma_2 + \Gamma_3) + \\
 & + \Gamma_3 [- (v_3^2 + \Gamma_1 \Gamma_2) (\Gamma_1 + \Gamma_2 + \Gamma_3) - (\Gamma_1 + \Gamma_2 - \Gamma_3) \Omega_2^2 + \\
 & + (-\Gamma_1^2 + (\Gamma_2 - \Gamma_3)^2) \Omega_2 \Omega_3 + \Gamma_2 (-\Gamma_1 + \Gamma_2 - \Gamma_3) \Omega_3^2\} + \\
 & + \cos \Phi v_1 v_2 v_3 \{-\Omega_2 [-2v_3^2 \Gamma_1 - 2v_3^2 \Gamma_2 - 2v_3^2 \Gamma_3 + \Gamma_1^2 \Gamma_3 + \\
 & + \Gamma_2^2 \Gamma_3 + \Gamma_1 \Gamma_3^2 + \Gamma_2 \Gamma_3^2 + v_2^2 (\Gamma_1 + \Gamma_2 + \Gamma_3) + 2\Gamma_3 \Omega_2^2] + \\
 & + \Omega_3 \{-\Gamma_2 (\Gamma_1 + \Gamma_3) (-\Gamma_1 + \Gamma_2 + \Gamma_3) + \\
 & + v_3^2 (\Gamma_1 + \Gamma_2 + \Gamma_3) + (-\Gamma_1 + \Gamma_2 - 3\Gamma_3) \Omega_2^2\} - \\
 & - (\Gamma_1 - \Gamma_2 + \Gamma_3) \Omega_2 \Omega_3 - v_1^2 (\Gamma_1 + \Gamma_2 + \Gamma_3) (\Omega_2 + \Omega_3)\}; \\
 \beta_2 = & v_1^6 \Gamma_1 + v_1^4 [v_2^2 \Gamma_2 + v_2^2 (-2\Gamma_1 + \Gamma_3) + \\
 & + 2\Gamma_1 (\Gamma_2 \Gamma_3 + \Omega_2 \Omega_3)] + \\
 & + v_1^2 \{v_3^4 (\Gamma_1 - 2\Gamma_3) + v_3^2 [-2v_2^2 \Gamma_2 + \Gamma_1^2 \Gamma_2 + \Gamma_2 \Gamma_3^2 + \Gamma_2 \Omega_2^2 + \\
 & + 2\Gamma_3 \Omega_2^2 + 2\Gamma_3 \Omega_2 \Omega_3 - 2\Gamma_1 (\Gamma_2 \Gamma_3 + \Omega_2 \Omega_3)] + \\
 & + (\Gamma_2^2 + \Omega_2^2) [v_2^2 \Gamma_3 + \Gamma_1 (\Gamma_3^2 + \Omega_3^2)]\} + \\
 & + v_3^2 \{v_2^2 [v_3^2 \Gamma_2 + \Gamma_1 (\Gamma_2^2 + \Omega_2^2)] + \\
 & + \Gamma_3 [v_3^4 + 2v_3^2 (\Gamma_1 \Gamma_2 - \Omega_2 (\Omega_2 + \Omega_3)) + \\
 & + (\Gamma_2^2 + \Omega_2^2) (\Gamma_1^2 + (\Omega_2 + \Omega_3)^2)]\} + \\
 & + 2\cos \Phi v_1 v_2 v_3 \{(v_3^2 - v_1^2) (\Gamma_1 + \Gamma_2 + \Gamma_3) \Omega_2 - \\
 & - (\Gamma_2^2 + \Omega_2^2) [\Gamma_3 \Omega_2 + (\Gamma_1 + \Gamma_3) \Omega_3]\}.
 \end{aligned}$$

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