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DYNAMICS OF TWO-PHOTON PROCESS IN THREE-LEVEL SYSTEM

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There are a number of reasons causing the importance of the examination of three-level systems (emitters) pictured in fig.1. Such a system contains three energy levels $\hbar\Omega_j$ $(j = 1,2,3)$, where the upper level |3> is connected with the levels |1> and |2> by dipole transitions whereas the transition |1> - |2> is a forbidden one. This system is widely investigated in a theory of a two-mode laser, in a theory of resonance Raman scattering, in connection with the problem of light by light control and also in the theory of superradiation effect (see /1-4/ and the references given therein). In our previous paper some exact results for the three-level system of a type described above were obtained. Here on their basis we shall examine a two-photon-process dynamics for a number of concrete initial states of the system.

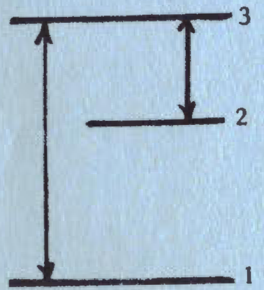


Fig.1. Two-photon process in a three-level system with a common upper level.

A Hamiltonian describing such a process can be presented in the following form (see /4/)

$$\hat{H} = \sum_{j=1}^{3} \hbar\Omega_j \hat{R}_{jj} + \sum_{a=1}^{2} \hbar\omega_a \hat{a}_a^+ \hat{a}_a - i\hbar \sum_{a=1}^{2} g_a (\hat{a}_a \hat{R}_{3a} - \hat{a}^+ \hat{R}_{a3}). \qquad (1)$$

Here the operator $\hat{R}_{jj} \equiv |j\rangle\langle j|$ describes the population of a j-th level. Then $\hat{R}_{kj} \equiv |k\rangle\langle j|$ $(k \neq j)$ corresponds to the transition from the state $|j\rangle$ to the state $|k\rangle$. The operators $\hat{R}_{kj}$ are generators of SU(3) group and obey:

$$\hat{R}_{kj}\hat{R}_{\ell m} = \hat{R}_{km}\delta_{\ell j},$$

$$[\hat{R}_{kj}, \hat{R}_{\ell m}] = \hat{R}_{km}\delta_{\ell j} - \hat{R}_{\ell j}\delta_{km}.$$

The photon operators $\hat{a}_a$, $\hat{a}_a^+$ describe two modes with the resonance frequencies $\omega_a = \Omega_3 - \Omega_a$, and g_a $(a = 1,2)$ are the field-emitter coupling parameters.

In paper /4/ the operator equations of motion for $\hat{R}_{jj}(t)$ together with the $\hat{N}_a(t) \equiv \hat{a}_a^+(t)\,\hat{a}_a(t)$ were integrated exactly. Their solution can be represented in the following manner

$$\hat{R}_{11}(t) = \hat{\mu}_1(\cos\hat{\lambda}t - 1) + \hat{\beta}_1 \sin\hat{\lambda}t + \hat{\lambda}_1^2\{\hat{\mu}_2(\cos 2\hat{\lambda}t - 1) + \hat{\beta}_2 \sin 2\hat{\lambda}t\} + \hat{R}_{11}(0),$$

$$\hat{R}_{22}(t) = -\hat{\mu}_1(\cos\hat{\lambda}t - 1) - \hat{\beta}_1 \sin\hat{\lambda}t + \hat{\lambda}_2^2\{\hat{\mu}_2(\cos 2\hat{\lambda}t - 1) + \hat{\beta}_2 \sin 2\hat{\lambda}t\} + \hat{R}_{22}(0),$$

$$\hat{R}_{33}(t) = -\hat{\lambda}^2\{\hat{\mu}_2(\cos 2\hat{\lambda}t - 1) + \hat{\beta}_2 \sin 2\hat{\lambda}t\} + \hat{R}_{33}(0), \qquad (2)$$

$$\hat{N}_1(t) = \hat{R}_{11}(t) + \hat{M}_1, \quad \hat{N}_2(t) = \hat{R}_{22}(t) + \hat{M}_2.$$

Here $\hat{M}_a$ are time-independent operators defining the "Rabi-frequency operators" $\hat{\lambda}_a$ and the nonlinear oscillation frequency operator $\hat{\lambda}$ /4/

$$\hat{\lambda}_a = g_a\sqrt{\hat{M}_a + 1}, \quad \hat{\lambda} = \sqrt{\hat{\lambda}_1^2 + \hat{\lambda}_2^2}. \qquad (3)$$

The "amplitudes operators" $\hat{\mu}_a$, $\hat{\beta}_a$ are defined by the initial conditions as follows

$$\hat{\mu}_1 = \{\hat{\lambda}^2[\hat{\lambda}_2^2\hat{R}_{11}(0) - \hat{\lambda}_1^2\hat{R}_{22}(0)] + [\hat{\lambda}_2^2 - \hat{\lambda}_1^2]\hat{K}\}\hat{\lambda}^{-4},$$

$$\hat{\mu}_2 = \{\hat{\lambda}^2[1 - 2\hat{R}_{33}(0)] + \hat{K}\}/(2\hat{\lambda}^4),$$

$$\hat{\beta}_1 = \{\hat{\lambda}_2^2\dot{\hat{R}}_{11}(0) - \hat{\lambda}_1^2\dot{\hat{R}}_{22}(0)\}(\hat{\lambda})^{-3}, \quad \hat{\beta}_2 = \{\dot{\hat{R}}_{11}(0) + \dot{\hat{R}}_{22}(0)\}/(2\hat{\lambda}^3).$$

Here $\hat{K}$ is the integral of motion /4/

$$\hat{K} \equiv g_1 g_2\{\hat{a}_1(t)\hat{a}_2^+(t)\hat{R}_{21}(t) + \hat{a}_1^+(t)\hat{a}_2(t)\hat{R}_{12}(t)\} - \hat{\lambda}_1^2\hat{R}_{22}(t) - \hat{\lambda}_2^2\hat{R}_{11}(t) = \text{const}$$

and the first derivatives $\dot{\hat{R}}_{aa}(0)$ obey the equations of a form /4/

$$\dot{\hat{R}}_{aa}(0) = g_a[\hat{a}_a(0)\hat{R}_{3a}(0) + \hat{a}_a^+(0)\hat{R}_{a3}(0)], \quad a = 1,2.$$

Let $\hat{\rho}(0)$ be a density matrix for some initial state of a total system "emitter + field". Then the time behaviour for the observable mean values of the level populations and photon-mode occupation can be defined as

$$\langle\hat{C}(t)\rangle = \mathrm{Tr}\,\hat{C}(t)\hat{\rho}(0), \qquad (4)$$

where $\hat{C}$ is $\hat{R}_{jj}$ or N_a.

First of all let us consider a simple but interesting case when at the initial moment $t = 0$ the emitter is in a state $|j\rangle$ and the field is in a quantum state with definite occupation numbers $|n_1, n_2\rangle$. Then

$$\hat{\rho}(0) = |\{m_0\}\rangle\langle\{m_0\}|, \quad |\{m_0\}\rangle \equiv |j;\, n_1, n_2\rangle. \qquad (5)$$

One can easily see that an initial state $|\{m_0\}\rangle$ is one of the basis states of the total system. In this basis the density matrix $\hat{\rho}(0)$ has only one nonzero element

$$\rho_{\{m'\},\{m''\}} = \langle\{m'\}|\hat{\rho}(0)|\{m''\}\rangle = \delta_{\{m'\},\{m_0\}}\,\delta_{\{m''\},\{m_0\}} .$$

The operators $\hat{\lambda}_a$ are diagonal in this representation. So $\forall\hat{\mathcal{O}}$

$$\langle\hat{\mathcal{O}} f(\hat{\lambda}_a)\rangle = \langle\{m_0\}|\,\hat{\mathcal{O}} f(\hat{\lambda}_a)|\{m_0\}\rangle = \tag{6}$$
$$= \langle\{m_0\}|\,\hat{\mathcal{O}}|\{m_0\}\rangle f(\langle\{m_0\}|\hat{\lambda}_a|\{m_0\}\rangle) = \langle\hat{\mathcal{O}}\rangle f(\langle\hat{\lambda}_a\rangle),$$

where $f(\hat{\lambda}_a)$ can be any function of the operator $\hat{\lambda}_a$. Below we shall use the following notation: $\langle\hat{\mathcal{O}}\rangle \equiv \mathcal{O}$, $\forall\hat{\mathcal{O}}$. Now from (2) in compliance with the expression (6) we obtain for any time moment t that

$$R_{11}(t) = -2\mu_1 \sin^2(\frac{\lambda t}{2}) - 2\lambda_1^2\mu_2 \sin^2\lambda t + R_{11}(0),$$

$$R_{22}(t) = 2\mu_1 \sin^2(\frac{\lambda t}{2}) - 2\lambda_2^2\mu_2 \sin^2\lambda t + R_{22}(0),$$

$$R_{33}(t) = 2\lambda^2\mu_2 \sin^2\lambda t + R_{33}(0), \tag{7}$$

$$N_1(t) = -2\mu_1 \sin^2(\frac{\lambda t}{2}) - 2\lambda_1^2\mu_2 \sin^2\lambda t + N_1(0),$$

$$N_2(t) = 2\mu_1 \sin^2(\frac{\lambda t}{2}) - 2\lambda_2^2\mu_2 \sin^2\lambda t + N_2(0).$$

Here λ_a, λ define the nonlinear oscillation frequencies in the system under consideration:

$$\lambda_a = g_a\sqrt{N_a(0) - R_{aa}(0) + 1}\,,\quad a = 1,2;\quad \lambda = \sqrt{\lambda_1^2 + \lambda_2^2}\,. \tag{8}$$

Amplitudes of the above-mentioned oscillations are defined as

$$\mu_1 = 2\lambda_1^2\lambda_2^2\{R_{11}(0) - R_{22}(0)\}\lambda^{-4},\quad \mu_2 = \{\lambda_1^2 R_{11}(0) + \lambda_2^2 R_{22}(0) - \lambda^2 R_{33}(0)\}/(2\lambda^4). \tag{9}$$

Let us now concretize the initial condition (5).

Case 1. At $t = 0$ the emitter is in the unexcited state $|1\rangle$ with the energy $\hbar\Omega_1$ and thus $|\{m_0\}\rangle = |1; n_1, n_2\rangle$. Then $R_{11}(0) = 1$, $R_{22}(0) = R_{33}(0) = 0$ and from the expression (8) it follows that

$$\lambda_1 = g_1\sqrt{n_1},\quad \lambda_2 = g_2\sqrt{n_2 + 1},\quad \lambda = \sqrt{g_1^2 n_1 + g_2^2(n_2 + 1)}\,. \tag{10}$$

Now instead of (7), we obtain for the level populations and photon-mode occupations

$$R_{11}(t) = N_1(t) + 1 - n_1 = 1 - 4\lambda_1^2\lambda_2^2\lambda^{-4}\sin^2(\frac{\lambda t}{2}) - \lambda_1^4\lambda^{-4}\sin^2\lambda t,$$

$$R_{22}(t) = N_2(t) - n_2 = 4\lambda_1^2\lambda_2^2\lambda^{-4}\sin^4(\frac{\lambda t}{2}), \tag{11}$$

$$R_{33}(t) = \lambda_1^2\lambda^{-2}\sin^2\lambda t.$$

For a small time $t \ll \lambda^{-1}$ we have

$$R_{11}(t) = 1 - \lambda_1^2 t^2 + \frac{1}{12}(4\lambda_1^2 + \lambda_2^2)\lambda_1^2 t^4 + \dots,$$

$$R_{22}(t) = \frac{1}{4}\lambda_1^2\lambda_2^2 t^4 + \dots, \tag{12}$$

$$R_{33}(t) = \lambda_1^2 t^2 - \frac{1}{3}(\lambda_1^2 + \lambda_2^2)\lambda_1^2 t^4 + \dots .$$

To determine the transition probabilities, let us introduce the Schrödinger representation with a wave-function of the total system $|\psi(t)\rangle$, where $|\psi(0)\rangle = |1; n_1, n_2\rangle$. Then the probability for one-photon transition 1-3 in the system is described by a standard way

$$P_1(t; 1\to 3) = \sum_{n_1', n_2'} |\langle\psi(t)|3; n_1', n_2'\rangle|^2 = \langle\psi(t)|\hat{R}_{33}|\psi(t)\rangle = R_{33}(t).$$

By analogy for the probability of a two-photon process of a Raman scattering type (transitions $1\to 3\to 2$) one can obtain $P_2(t; 1\to 3\to 2) = R_{22}(t)$. So from (10)-(11) we have

$$P_1(t; 1\to 3) = \frac{g_1^2 n_1}{g_1^2 n_1 + g_2^2(n_2+1)} \sin^2\{t\sqrt{g_1^2 n_1 + g_2^2(n_2+1)}\},$$

$$P_2(t; 1\to 3\to 2) = \frac{4g_1^2 g_2^2 n_1(n_2+1)}{\{g_1^2 n_1 + g_2^2(n_2+1)\}^2} \sin^4\{\frac{t}{2}\cdot\sqrt{g_1^2 n_1 + g_2^2(n_2+1)}\}. \tag{13}$$

This partial conclusion (13) coincides with the result of paper /1/ in which the kinetic equation for the two-mode laser has been obtained.

Case 2. At $t = 0$ the emitter is in the excited state $|3\rangle$ with the energy $\hbar\Omega_3$ and $|\{m_0\}\rangle = |3; n_1, n_2\rangle$. In this case $R_{11}(0) = R_{22}(0) = 0$, $R_{33}(0) = 1$ and (8) leads to the following expressions

$$\lambda_a = g_a\sqrt{n_a+1}\,,\quad a = 1,2;\quad \lambda = \sqrt{\sum_a g_a^2(n_a+1)}\,. \tag{14}$$

From expressions (7) one can now receive

$$R_{11}(t) = N_1(t) - n_1 = \left(\frac{\lambda_1 \sin\lambda t}{\lambda}\right)^2,$$
$$R_{22}(t) = N_2(t) - n_2 = \left(\frac{\lambda_2 \sin\lambda t}{\lambda}\right)^2,\qquad R_{33}(t) = \cos^2\lambda t\,. \tag{15}$$

Drawing a parallel with the previous case one can obtain now that the probability $P_1(t; 3\to a)$ of the transition $3-a$ ($a = 1,2$) coincides with $R_{aa}(t)$. So from (14) we receive

$$P_1(t; 3\to a) = \frac{g_a^2(n_a+1)}{g_1^2(n_1+1)+g_2^2(n_2+1)}\sin^2\{t\sqrt{g_1^2(n_1+1)+g_2^2(n_2+1)}\}\,. \tag{16}$$

This expression also is in confirmity with the results of paper /1/.

Case 3. At $t = 0$ the emitter is in the state $|2\rangle$ and $|\{m_0\}\rangle = |2; n_1, n_2\rangle$. This case is described by formulae (11)-(13) with the substitution of index 2 instead of 1 and 1 instead of 2.

Let us consider below the case of the initially coherent state of a field. For a two-level system such a condition can lead to the collapse and revival of the oscillations of the dynamical parameters /5/. Here we show that a similar phenomenon may also happen in the three-level system. For simplicity we propose here that only coherent mode 1 is present in the system at $t = 0$. The coherent state of mode 1 is described by

$$|z\rangle_1 = \exp(-|z|^2/2)\sum_{n_1=0}^{\infty}\frac{z^{n_1}}{\sqrt{n_1!}}|n_1\rangle_1\,. \tag{17}$$

Let the emitter be in the state $|1\rangle$ with the energy $\hbar\Omega_1$ at $t = 0$. Then the initial state of the total system emitter + field is

$$|\psi_0\rangle = |1\rangle\otimes|z\rangle_1\otimes|0\rangle_2 = \sum_{n_1=0}^{\infty} e^{-\frac{1}{2}|z|^2}\frac{z^{n_1}}{\sqrt{n_1!}}|1; n_1, 0\rangle\,. \tag{18}$$

The density matrix for such a pure state is $\hat\rho(0) = |\psi_0\rangle\langle\psi_0|$. We consider a special case of the operator $\hat{\mathcal{O}}$ which obeys the following condition

$$\langle 1; n_1', 0|\hat{\mathcal{O}}|1; n_1'', 0\rangle \sim \delta_{n_1', n_1''}\,. \tag{19}$$

Then

$$\langle\hat{\mathcal{O}}\rangle \equiv \langle\psi_0|\hat{\mathcal{O}}|\psi_0\rangle = \sum_{n_1=0}^{\infty} e^{-\bar n_1}\frac{\bar n_1^{\,n_1}}{n_1!}\langle\hat{\mathcal{O}}\rangle_{n_1}\,. \tag{20}$$

Here $|z|^2 \equiv \bar n_1$ is the mean number of the photons in the coherent mode 1 at $t = 0$ and $\langle\dots\rangle_{n_1}$ denotes the average with the composite basis state $|1; n_1, 0\rangle$: $\langle\hat{\mathcal{O}}\rangle_{n_1} \equiv \langle 1; n_1, 0|\hat{\mathcal{O}}|1; n_1, 0\rangle$.

It should be noted that the operators $\hat\mu_a$, $\hat\beta_a$ and thus the operators $\hat R_{jj}(t)$, $\hat N_a(t)$ obey condition (19). Thus, the mean values of these operators over state (18) can be expanded in compliance with (20) into a sum over all n_1 of the averages $\langle\hat{\mathcal{O}}\rangle_{n_1}$ which is calculated in confirmity with expressions (10), (11). So, we get

$$R_{11}(t) = N_1(t) + 1 - \bar n_1 = 1 - \sum_{n_1=0}^{\infty}\frac{4g_1^2g_2^2n_1}{(g_1^2n_1+g_2^2)^2}\cdot\sin^2\left(\frac{t}{2}\sqrt{g_1^2n_1+g_2^2}\right)\cdot P(n_1) -$$
$$- \sum_{n_1=0}^{\infty}\frac{g_1^4n_1^2}{(g_1^2n_1+g_2^2)^2}\cdot\sin^2\left(t\sqrt{g_1^2n_1+g_2^2}\right)\cdot P(n_1)\,,$$
$$R_{22}(t) = N_2(t) = \sum_{n_1=0}^{\infty}\frac{4g_1^2g_2^2n_1}{(g_1^2n_1+g_2^2)^2}\cdot\sin^4\left(\frac{t}{2}\sqrt{g_1^2n_1+g_2^2}\right)\cdot P(n_1)\,, \tag{21}$$
$$R_{33}(t) = \sum_{n_1=0}^{\infty}\frac{g_1^2n_1}{g_1^2n_1+g_2^2}\cdot\sin^2\left(t\sqrt{g_1^2n_1+g_2^2}\right)\cdot P(n_1)\,.$$

Here the weight factor $P(n_1)$ is the Poisson distribution

$$P(n_1) = e^{-\bar n_1}\cdot\frac{\bar n_1^{\,n_1}}{n_1!}\,.$$

Let us now consider a special case of a strong enough initial pumping, i.e., $\bar n_1 \gg 1$. Then instead of (21) we obtain

$$R_{11}(t) = N_1(t) + 1 - \bar n_1 \cong 1 - \frac{32g_1^2g_2^2\bar n_1}{W^4}F\left(\frac{t}{2}\right) - \frac{8g_1^4\bar n_1^2}{W^4}F(t)\,,$$
$$R_{22}(t) = N_2(t) \cong \frac{8g_1^2g_2^2\bar n_1}{W^4}\left\{4F\left(\frac{t}{2}\right) - F(t)\right\},\quad R_{33}(t) \cong \frac{2g_1^2\bar n_1}{W^2}F(t)\,. \tag{22}$$

Here $W \equiv 2\sqrt{g_1^2\bar{n}_1 + g_2^2}$, $F(t) \equiv 1 - \sum_{n_1=0}^{\infty} P(n_1)\cos(2t\sqrt{g_1^2 n_1 + g_2^2}) \cong$
$\cong 1 - f(t)\exp[-\psi(t)]\cos\phi(t)$ and

$$f(t) = \left(1 + \frac{16 g_1^8 \bar{n}_1^2 t^2}{W^6}\right)^{-1/4},$$

$$\psi(t) = 2\bar{n}_1 \sin^2(g_1^2 t/W)\left(1 + \frac{16 g_1^8 \bar{n}_1^2 t^2}{W^6}\right)^{-1},$$

$$\phi(t) = Wt + \bar{n}_1 \sin(2g_1^2 t/W) - 2g_1^2\bar{n}_1 t/W - \frac{1}{2}\arctan[4g_1^4\bar{n}_1 t/W^3].$$

Here we substitute $\bar{n}_1$ instead of n_1 in the fractional factors in (21) and use the approximation of paper /5/ for the functions $F(t)$. One can see that the time behaviour of $F(t)$ and thus of $R_{jj}(t)$, $N_a(t)$ has a character of fast oscillations which collapse and again revive in a time period

$$T_R = \pi g_1^{-2} W = \frac{2\pi}{g_1}\bar{n}_1^{-1/2}\left(1 + \frac{g_2^2}{g_1^2\bar{n}_1}\right)^{1/2}. \qquad (23)$$

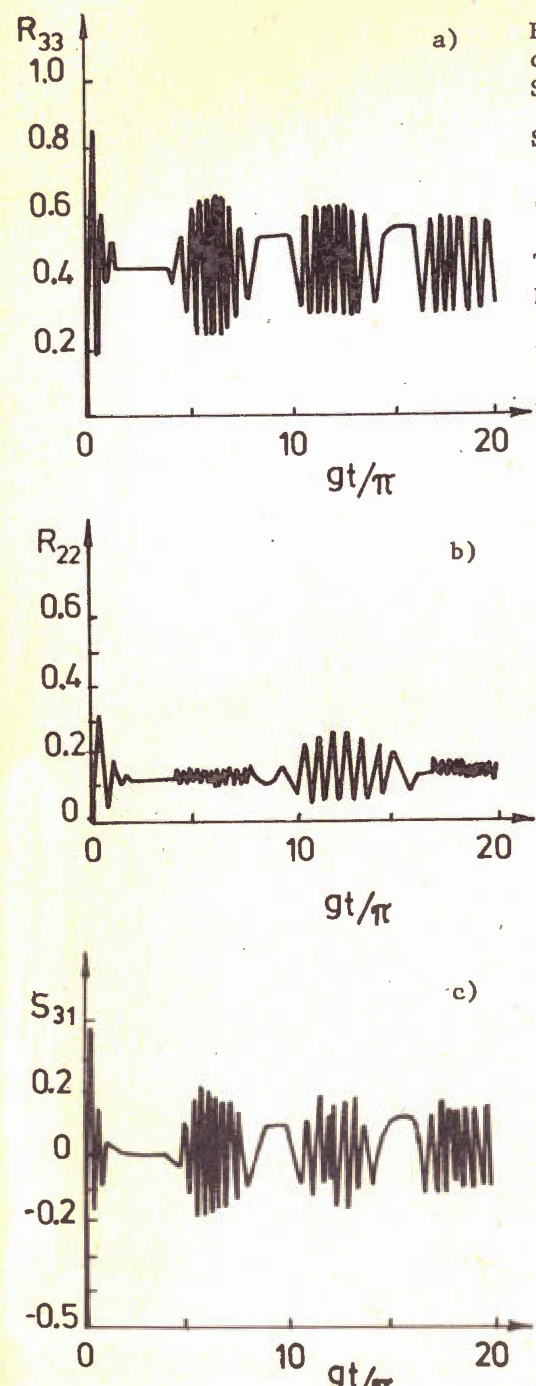
For $t \ll T_R$ these oscillations collapse during the time

$$T_c = \frac{W}{g_1^2\sqrt{2\bar{n}_1}} = \frac{\sqrt{2}}{g_1}\sqrt{1 + g_2^2/(g_1^2\bar{n}_1)}$$

(see fig.2). Subsequent revivals ought to vanish when $t \to \infty$. We shall call the phenomenon of the revival of fast oscillations in a time period T_R "autoecho" to distinguish it from the echo induced by two classical coherent pulses with the areas $\pi/2$ and π. The latter case is connected with the dephasing because of the inhomogeneous broadening /6/, whereas the former case is a pure quantum nonlinear phenomenon. The most interesting and surprising is the revival of the oscillations of the second level population $R_{22}(t)$ (see fig.2b). The oscillations which are resumed in a period $2T_R$ have a much larger amplitude than the same in a period T_R. Such behaviour can be explained with the aid of the second expression in (22). Really, the resuming connected with the term $4F(t/2)$ has a period $2T_R$ and an amplitude which is four times as large as the one due to the term $F(t)$ with a period T_R.

From our formulas (22) we can obtain the results of paper /5/ for the two-level system. For this aim let us introduce the quantity S_{31} defining the population difference of levels 3 and 1 $S_{31} = (R_{33} - R_{11})/2$. Then from (22) we have

$$S_{31} \cong -\frac{1}{2} + \frac{16 g_1^2 g_2^2 \bar{n}_1}{W^4} F\left(\frac{t}{2}\right) + \frac{g_1^2\bar{n}_1}{W^2}\left(1 + \frac{4g_1^2\bar{n}_1}{W^2}\right) F(t). \qquad (24)$$



For the two-level case we ought to put here $g_2 = 0$. So, from (23) we obtain

$$S_{31} \cong -\frac{1}{2} + \frac{1}{2}F(t) =$$

$$= -\frac{1}{2} f(t)\exp[-\psi(t)]\cos\phi(t).$$

This is the result of paper /5/.

Thus, in the present paper the dynamics of the three-level two-mode system was rigorously examined on the basis of our exact operator solution /4/. Some initial states were considered and the autoecho phenomemon was described. It should be emphasized that other initial states can also be examined for the problem above considered. It can be a subject for further investigations.

Fig.2. The autoecho or the revival of the fast oscillations in the system with the coherent initial pumping (17). Time is in units of π/g and $g_1 = g_2 = g$, $\bar{n}_1 = 9$.

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Динамика двухфотонного процесса в трехуровневой системе

Исследовано динамическое поведение двухфотонного процесса в трехуровневой системе с общим верхним уровнем. Описано явление автоэха. Это квантовое нелинейное явление состоит в восстановлении и ослаблении быстрых осцилляций динамических величин при начальном когерентном состоянии поля.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

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Dynamics of Two-Photon Process in Three-Level System

Dynamical behaviour of a two-photon process in a three-level system with a common upper level is examined. The auto-echo phenomenon is described. This quantum nonlinear phenomenon consists of the revival and collapse of the fast oscillations for dynamical parameters due to the coherence of an initial pumping.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

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