On the behavior of quantum systems in a nonmonochromatic external field

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The kinetics of quantum transitions in a two-level system is considered in a radiation field with small and fast frequency fluctuations. A decoupling method is used to obtain an integral equation that describes the dynamics of the variation of the level populations in a wide range of parameters of the problem. The probability of populating the upper level, averaged over the realizations, is obtained as a function of time. The results are used to consider multiphoton transitions in two-level and multilevel systems in a monochromatic-radiation field, and to analyze the changeover from noncoherent multiphoton resonant transitions to coherent multiphoton transitions with decreasing width of the emission spectrum.

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1. INTRODUCTION

1.1 Theoretical investigations of atomic and molecular systems in high-power electromagnetic fields have recently attracted great interest because of the experimental observation of a large number of new interesting phenomena (it suffices to mention, for example, multiphonon ionization of atoms[1] or radiative dissociation of polyatomic molecules[2,3]). Even though quite complicated systems are used in the experimental studies, their theoretical interpretation can in many cases be successfully made within the framework of simple models, which reduce in one sense or another to a two-level system.[4]

In this paper, on the basis of a model of a two-level system, we first consider the dynamics of population of the upper level by a one-photon transition in an external resonant electromagnetic field; we take into account the nonmonochromaticity of the radiation. Then, using the derived equations, we consider, outside the framework of perturbation theory, multiphonon transitions in a nonmonochromatic external field, which occur in a two-level system as well as in some multilevel systems. It will be shown, in particular, that the influence of nonmonochromaticity on the dynamics of an n-photon transition increases rapidly with increasing n and can become decisive already at $n=3-5$.

To our knowledge, the dynamics of multiphoton transitions in a nonmonochromatic radiation field has not been previously considered outside the framework of perturbation theory.

As to allowance for the nonmonochromaticity for one-photon transitions in a two-level system or for the linear susceptibility of a three-level system, the following remarks are in order. This question has been the subject of a large number of studies[5–12] (other works are cited, for example, in Ref. 13). The case of interest to us, that of an external field with fast and small fluctuations of the frequency, was considered, in particular, in Refs. 5–8. In the present paper we consider the dynamics of population of the upper level in such a field and obtain results that are valid in a much larger region of variation of the parameters of the problem than in Refs. 5–8.

1.2. By way of example we indicate the region of applicability of our results and of the results of Refs. 5–8 for the case when the electric field intensity $E(t)$ at a given point is equal to

$$E(t)=E_0\cos[\omega_l t+\varphi(t)],\quad \varphi(t)=\int_0^t\Omega(t_1)\,dt_1+\varphi_0, \tag{1}$$

where E_0 is the constant amplitude of the field, $\Omega(t)$ is the deviation of the radiation frequency, which is a normal process, φ_0 is a quantity randomly distributed in the interval $(0, 2, \pi)$, and ω_l is the carrier frequency of the (laser) radiation field. Let, for example, the fluctuations of the frequency be exponentially correlated:

$$\langle\Omega(t)\rangle=0,\quad \langle\Omega(t)\Omega(t+\tau)\rangle=\langle\Omega^2\rangle\exp(-\gamma|\tau|), \tag{2}$$

with a parameter

$$\varepsilon=\langle\Omega^2\rangle/\gamma^2\ll1. \tag{3}$$

The inequality (3) corresponds to small and fast frequency fluctuations. The expression for the emission line shape then takes the form

$$g(\omega)=\frac{g(0)}{(1+\delta\omega^2/\varepsilon^2\gamma^2)(1+\delta\omega^2/\gamma^2)}, \tag{4}$$

where $\delta\omega=\omega_l-\omega_{01}$ is the deviation of the laser frequency from the resonant frequency ω_{01} of the system. The second-order correlator for the electric field $E(t)$ is equal to

$$\langle E(t)E(t+\tau)\rangle=\frac{E_0^2}{2}\exp\{-\varepsilon\gamma|\tau|-\varepsilon(e^{-\gamma|\tau|}-1)\}\cos\omega_l\tau. \tag{5}$$

We introduce the field broadening $f=E_0d_{01}/\hbar$, where d_{01} is the dipole moment of the $0\to1$ transition. We can now formulate a criterion for the applicability of Refs. 5–8. The results obtained in Refs. 5–7 are valid for our model in the region

$$f\ll\gamma,\quad |\delta\omega|\ll\gamma. \tag{6}$$

This statement will be proved in Sec. 3 below. In the region (6), we can neglect the term $\varepsilon(e^{-\gamma|\tau|}-1)$, of (5), and the problem of the population of the upper level reduces to a solution of an ordinary differential equation of third order with constant coefficients.[5]

Outside the region (6), the condition $\varepsilon\ll1$ is no longer sufficient for neglecting the indicated term in (5). Neglect of this circumstance leads to errors. In this paper

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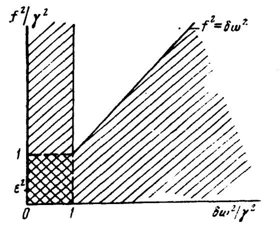

FIG. 1. The hatched part, the cross-hatched part, and the thick line show respectively the regions of applicability of the results of the present paper, the results of Refs. 5–7, and the results of Ref. 8. The inequality signs in Eqs. (6)–(8) are replaced in the figure by an equal sign.

we construct a theoretical scheme that takes into account the parameter ε and is valid in the region

$$|\delta\omega|\ll\gamma \quad \text{or} \quad |\delta\omega|\gg f \tag{7}$$

(see Fig. 1). In the region (7) the problem reduces to a determination of the zeros of a polynomial that, generally speaking, is of infinite order (see Sec. 3). When the field broadening f and the frequency detuning $\delta\omega$ are small enough and satisfy the conditions (6), our formulas lead to the results obtained in Refs. 5–7. The results of Ref. 8 are obtained as a limiting case from our results only at exact resonance $\delta\omega=0$ provided the following condition is satisfied:

$$f^2/\varepsilon^2\gamma^2\gg 1. \tag{8}$$

In regions satisfying conditions (7) but not included in (6) and (8), our present result differs substantially from those obtained in Refs. 5–8.

Section 4 deals with multiphoton transitions in a field with fluctuating frequency in various quantum systems. It contains also a comparison of the results of the calculation of a three-photon transition in the Xe atom with the experimental data.

1.3. To conclude this section, we note that a nonmonochromatic field is taken in the present paper to mean a field with randomly fluctuating parameters, namely with fluctuating frequency. Problems in which the field parameters vary but remain determined functions of the time are considered, for example, in Ref. 14 (see also Refs. 15 and 16 and the detailed bibliography in Ref. 17). The results of the cited papers may not agree with ours, since they deal with differently formulated problems.

We note in addition that the same line shape (4) can be obtained for various types of nonmonochromaticity (statistics) of the field. The results can in this case likewise differ substantially. The particular field statistics in concrete experiments depends on the experimental conditions, on the type of laser, and on other factors. We assume that the model of a field with constant amplitude and with fast and small fluctuations of the frequency describes adequately the radiation at least for some types of lasers.

2. TWO-LEVEL SYSTEM IN AN EXTERNAL FIELD WITH A FLUCTUATING FREQUENCY

2.1. We consider a two-level system in an external field of laser radiation of intensity (1). Under the conditions

$$\omega_l=\text{const};\quad \dot{\varphi},\ f,\ |\delta\omega|,\ 1/T_1,\ 1/T_2\ll\omega_l,$$

where T_1 and T_2 are respectively the longitudinal and transverse relaxation times, the resonance approximation can be used. For the level population difference $\rho=\rho_{11}-\rho_{00}$ under the condition that at the instant $t=0$ when the field is turned on the system is in an equilibrium state $\rho=\rho_0$, $\rho_{01}=\rho_{10}^*=0$, we obtain from the equation for the density matrix a well known integral equation for ρ (see, e.g., Ref. 13)

$$\rho=\rho_0-f^2\int\limits_0^t\int\limits_0^{t_1}\exp\left\{-\frac{t}{T_1}-t_1\left(\frac{1}{T_2}-\frac{1}{T_1}\right)+\frac{t_2}{T_2}\right\}$$
$$\times\cos[\varphi(t_1)-\varphi(t_2)-\delta\omega(t_1-t_2)]\rho(t_2)\,dt_1\,dt_2. \tag{9}$$

In the general case it is impossible to write down a solution of (9) in a final analytic form.

In this paper we confine ourselves to the case when the frequency deviation $\Omega(t)=\dot{\varphi}(t)$ is satisfactory normal process:

$$\langle\Omega(t)\rangle=0,\quad \langle\Omega(t)\Omega(t+\tau)\rangle=\langle\Omega^2\rangle\Psi(|\tau|), \tag{10}$$

where $\Psi(0)=1\geq\Psi(|\tau|)$ and $\Psi(|\tau|)$ satisfy all the properties of the correlation function of a random stationary quantity (see Ref. 18); Eq. (2) is a particular case of (10).

The solution of (9), obtained by iterations, can be represented in the form of a series

$$\rho=\rho_0\sum_{k=0}^{\infty}(-1)^k f^{2k}C_k, \tag{11}$$

where

$$C_k=\int\limits_0^t\dots\int\limits_0^{t_{2k-1}}\exp\left\{-\frac{1}{T_1}(t-t_1+\dots)-\frac{1}{T_2}(t_1-t_2+\dots-t_{2k})\right\}$$
$$\times\cos[\varphi(t_1)-\varphi(t_2)-\delta\omega(t_1-t_2)]\dots\cos[\varphi(t_{2k-1})$$
$$-\varphi(t_{2k})-\delta\omega(t_{2k-1}-t_{2k})]\,dt_1\dots dt_{2k}. \tag{12}$$

The phase shift $\chi(t+\Delta t,t)=\varphi(t+\Delta t)-\varphi(t)$ is a normal random process. Under the condition $t_{i-1}\geq t_i\geq t_{j-1}\geq t_j$ its variance and covariance are given by

$$\lambda_{ii}=\langle\chi^2(t_{i-1},t_i)\rangle=\pi g_\Omega(0)(t_{i-1}-t_i)-2\varepsilon$$
$$+\pi i\sum_k\exp[-\omega_k(t_{i-1}-t_i)]\operatorname{Res}[g_2(i\omega_k)],$$

$$\lambda_{ij}(i\neq j)=\langle\chi(t_{i-1},t_i)\chi(t_{j-1},t_j)\rangle=\pi i\sum_k\{\exp[-\omega_k(t_{i-1}-t_j)] \tag{13}$$
$$+\exp[-\omega_k(t_i-t_{j-1})]-\exp[-\omega_k(t_{i-1}-t_{j-1})]-\exp[-\omega_k(t_i-t_j)]\}\operatorname{Res}[g_2(i\omega_k)],$$

where

$$g_\Omega(\omega)=g_\Omega(0)+\omega^2g_2(\omega)=\frac{2}{\pi}\int\limits_0^\infty\Psi(\tau)\cos\omega\tau\,d\tau$$

is the spectral density of the process $\Omega(t)$ over the positive frequencies and is an even function of ω;

$$\varepsilon=\frac{1}{2}\int\limits_0^\infty g_2(\omega)\,d\omega=\frac{\pi}{2}i\sum_k\operatorname{Res}[g_2(i\omega_k)],$$

where $\{i\omega_k\}$ are the poles of the function $g_2(\omega)$ in the upper half-plane. We confine ourselves to the case when

$$\varepsilon\ll 1,$$
$$\pi i\sum_k\exp[-\omega_k(t_{i-1}-t_i)]\operatorname{Res}[g_2(i\omega_k)]\ll 1 \tag{14}$$

for all $t_{i-1} - t_i \geq 0$.

The validity of the inequality (14) is ensured by satisfaction of the condition

$$\langle\Omega^2\rangle g_\Omega^{max} \ll \gamma,$$

where g_Ω^{max} is the maximum of the function $g_\Omega(\omega)$, and γ is the characteristic width of $g_\Omega(\omega)$. This case is called the case of small and fast fluctuations and the line near the center has a Lorentz shape with width $\pi\langle\Omega^2\rangle g^{max}$.

To find the solution of (9) averaged over the realizations we must average (12) and then sum the series (11). To solve this problem we note that when the conditions (14) are satisfied it follows from (13) that $\lambda_{ij}(i\neq j) \ll 1$ for all instants of time $t_{i-1} \geq t_i \geq t_{j-1} \geq t_j$, and the central second-order moments of the functions of interest to us take the form $(i\neq j)$

$$\langle\cos\chi(t_{i-1},t_i)\cos\chi(t_{j-1},t_j)\rangle = \exp\left\{-\frac{\lambda_{ii}+\lambda_{jj}}{2}\right\}\operatorname{ch}\lambda_{ij} \approx \exp\left\{-\frac{\lambda_{ii}+\lambda_{jj}}{2}\right\},$$

$$\langle\sin\chi(t_{i-1},t_i)\sin\chi(t_{j-1},t_j)\rangle = \exp\left\{-\frac{\lambda_{ii}+\lambda_{jj}}{2}\right\}\operatorname{sh}\lambda_{ij} \sim \varepsilon,$$

$$\langle\sin\chi(t_{i-1},t_i)\cos\chi(t_{j-1},t_j)\rangle = 0.$$

Hence

$$\langle\cos[\varphi(t_1)-\varphi(t_2)]C_k(t_2)\rangle = \langle\cos[\varphi(t_1)-\varphi(t_2)]\rangle\int_0^{t_2}\ldots\int_0^{t_{2k+1}} dt_3\ldots dt_{2k+2}$$
$$\times\exp\left\{-\left(\frac{1}{T_2}-\frac{1}{T_1}\right)(t_3-t_4\ldots)-\frac{1}{T_1}t_2+\frac{1}{T_2}t_{2k+2}\right\}$$
$$\times\prod_{p=1}^{k}\cos[\varphi(t_{2p+1})-\varphi(t_{2p+2})-\delta\omega(t_{2p+1}-t_{2p+2})][1+F(t_3,\ldots,t_{2k+2})],$$

where $|F| < \varepsilon^2$, and the terms $\langle\sin[\varphi(t_1)-\varphi(t_2)]C_k(t_2)\rangle$ are small in the parameter ε.

It is clear therefore than when averaging (12) we can confine ourselves for $\langle C_k\rangle$, in a wide range of the parameters of the problem, to the expression

$$\langle C_k\rangle = \int_0^t\ldots\int_0^{t_{2k-1}}\exp\left\{-\left(\frac{1}{T_2}-\frac{1}{T_1}\right)(t_1-t_2+\ldots)\right.$$
$$\left.-\frac{1}{T_1}t+\frac{1}{T_2}t_{2k}-\frac{1}{2}\lambda_{11}-\ldots-\frac{1}{2}\lambda_{kk}\right\}$$
$$\times\cos[\delta\omega(t_1-t_2)]\ldots\cos[\delta\omega(t_{2k}-t_{2k-1})]dt_1\ldots dt_{2k}. \qquad (15)$$

The operation (15) is usually called uncoupling.[19] It was for discontinuous Markov processes or for the solution of problems within the framework of the transition model. This operation can be used, however, also for radiation with small and fast frequency fluctuations in a wide range of the problem parameters.

We conclude from (15) that the process of the search for an averaged solution of (9) can be greatly simplified. Instead of finding the solution by summing the iteration series (11) with the coefficients (15), we can solve directly the equation

$$\langle\rho\rangle = \rho_0 - f^2\int_0^t\int_0^{t_1}\exp\left\{-\frac{t}{T_1}-t_1\left(\frac{1}{T_2}-\frac{1}{T_1}\right)+\frac{t_2}{T_2}-\frac{\lambda_{11}}{2}\right\}$$
$$\times\cos[\delta\omega(t_1-t_2)]\langle\rho(t_2)\rangle dt_1\,dt_2, \qquad (16)$$

since the iterations of (16) yield exactly the solution of (11) with the coefficients (15).

3. EXPONENTIALLY DAMPED CORRELATIONS OF THE FREQUENCY DEVIATION

We consider the concrete case (10) in the form (2)

$$g_\Omega(\omega) = \frac{2}{\gamma\pi}\frac{1}{1+\delta\omega^2/\gamma^2},$$
$$\lambda_{ii} = 2\varepsilon\gamma(t_{i-1}-t_i)+2\varepsilon[\exp\{-\gamma(t_{i-1}-t_i)\}-1], \qquad (17)$$
$$\lambda_{ij}(i\neq j) = 4\varepsilon\operatorname{sh}\frac{t_{i-1}-t_i}{2}\gamma\operatorname{sh}\frac{t_{j-1}-t_j}{2}\gamma\exp\left\{-\frac{1}{2}\gamma(t_i+t_{i-1}-t_j-t_{j-1})\right\}.$$

The equation (16) takes the form

$$\langle\rho(t)\rangle = \rho_0 - f^2\int_0^t\int_0^{t_1}\exp\left\{-\frac{t}{T_1}-t_1\left(\frac{1}{T_2}-\frac{1}{T_1}\right)+\frac{t_2}{T_2}-\varepsilon\gamma(t_1-t_2)\right. \qquad (18)$$
$$\left.-\varepsilon[e^{-(t_1-t_2)\gamma}-1]\right\}\cos[\delta\omega(t_1-t_2)]\langle\rho(t_2)\rangle dt_1\,dt_2.$$

To obtain the information of interest to us we introduce the Laplace transform of $\langle\rho(t)\rangle$:

$$\langle L(p)\rangle = \int_0^\infty\langle\rho(t)\rangle e^{-pt}dt.$$

Expanding $\exp\{-\varepsilon e^{-(t_1-t_2)\gamma}\}$ in a series and taking the Laplace transform of (18), we get

$$\langle L(p)\rangle = \frac{\rho_0(1+1/T_1p)}{1/T_1+p+f^2e^\varepsilon K(p)}, \qquad (19)$$

where

$$K(p) = \sum_{n=0}^{\infty}\frac{\varepsilon^n}{n!}(-1)^n\frac{p+1/T_2+(\varepsilon+n)\gamma}{[p+1/T_2+(\varepsilon+n)\gamma]^2+\delta\omega^2}.$$

The function $\langle\rho(t)\rangle$ is obtained in standard fashion by taking the inverse Laplace transform.

The poles of the function $\langle L(p)\rangle$ can be determined with the necessary degree of accuracy because of the presence of the small parameter ε. The set of poles of $\langle L(p)\rangle$ can be arbitrarily divided into three groups.

(1) The pole $p=0$. Since all the poles of the functions $\langle L(p)\rangle$ with the exception of the indicated pole have negative real parts, this pole gives the population difference $\langle\rho(t)\rangle$ as $t\to\infty$:

$$\rho_\infty = \frac{\rho_0}{1+T_1f^2e^\varepsilon K(0)} \approx \rho_0\Big/\left[1+\frac{\xi}{\eta_1}\frac{\eta_2y^2+\delta(\delta+1)^2}{(y^2+\delta^2)(y^2+(1+\delta)^2)}\right],$$

where

$$\xi = \frac{f^2}{\gamma^2},\quad \eta_1 = \frac{1}{T_1\gamma},\quad \eta_2 = \frac{1}{T_2\gamma},\quad \delta = \varepsilon+\eta_2,\quad y = \frac{\delta\omega}{\gamma}.$$

The value of ρ_∞ at large detunings $|y| \gtrsim 1$ can substantially differ from the value obtained without allowance for the deviation of the wings of the radiation line from a Lorentz shape.

(2) The poles

$$p_{2n+2} = -(n+\varepsilon)\gamma-\frac{1}{T_2}+\gamma\delta_{2n+2},\qquad p_{2n+3} = -(n+\varepsilon)\gamma-\frac{1}{T_2}+\gamma\delta_{2n+3},$$
$$n = 1, 2, \ldots,$$

where δ_{2n+2} and δ_{2n+3} are corrections that are small in terms of ε. In particular, under the condition

$$|\delta_{2n+2,3}\mp iy| \ll A_{2n+2,3},$$

where the upper and lower signs correspond to the subscripts $2n+2$ and $2n+3$, respectively, and

$$A_{2n+2,3}=-n-\varepsilon-\eta_2+\eta_1\pm iy-\xi\frac{n\mp iy}{n^2\mp 2niy},$$

$$\delta_{2n+2,3}=-\frac{\xi\varepsilon^n(-1)^n}{2A_{2n+2,3}n!}\pm\theta_{1,2}(|B|_{2n+2,3})^{1/2}\exp\left\{i\,\mathrm{arctg}\frac{\mathrm{Im}\,B_{2n+2,3}}{\mathrm{Re}\,B_{2n+2,3}}\right\},$$

$$B_{2n+2,3}=\frac{\varepsilon^{2n}\xi^2}{4A^2_{2n+2,3}n!^2}-y^2,$$

$$\theta_{1,2}=\begin{cases}(-1)^{n+1} & \text{at}\quad \mathrm{Re}\,B_{2n+2,3}>0\\ 1 & \text{at}\quad \mathrm{Re}\,B_{2n+2,3}<0,\end{cases}$$

these poles make small contributions to $\langle\rho(t)\rangle$, namely, the pole $p_{2n+2,3}$ makes a contribution proportional to ε^n.

(3) The poles p_1, p_2, and p_3, which play the decisive role in the variation of $\langle\rho(t)\rangle$. The dimensionless quantities

$$x_1=p_1/\gamma,\quad x_2=p_2/\gamma,\quad x_3=p_3/\gamma$$

are the roots of the cubic equation

$$x^3+d_1x^2+d_2x+d_3=0, \tag{20}$$

where

$$\begin{gathered}d_1=2\delta+\eta_1+\delta_4+\delta_5,\\ d_2=2y^2+\delta^2+\xi(1-\varepsilon)+2\eta_1\delta+(\delta_4+\delta_5)(\eta_1-1+\delta)-\delta_4\delta_5,\\ d_3=\delta^2\eta_1+(\varepsilon+\delta)\xi-2y^2(\eta_1-1)+\delta_4\delta_5(2-\eta)\\ +(\delta_4+\delta_5)[1+2y^2+\xi+\eta_1(\delta-1)-2\delta_4\delta_5].\end{gathered}$$

They can be determined with the aid of Cardan's formulas.

By virtue of the foregoing, the solution of (18) takes the form

$$\begin{aligned}\langle\rho(t)\rangle\approx\rho_\infty&+\frac{(\rho_0-\rho_\infty)x_2x_3-\xi\rho_0}{(x_1-x_2)(x_3-x_1)}e^{x_1\gamma t}+\frac{(\rho_0-\rho_\infty)x_1x_3-\xi\rho_0}{(x_1-x_2)(x_2-x_3)}e^{x_2\gamma t}\\ &+\frac{(\rho_0-\rho_\infty)x_1x_2-\xi\rho_0}{(x_2-x_3)(x_3-x_1)}e^{\gamma x_3 t}.\end{aligned} \tag{21}$$

We have already mentioned that the problem of the population dynamics in a two-level systems had been repeatedly considered for various radiation models.

It is clear that wherever the poles belonging to the third group play the decisive role the solution method used by us is equivalent to a solution of a differential equation for $\langle\rho(t)\rangle$ with constant coefficients, with a characteristic polynomial (20), and with corresponding initial conditions. In particular, when the conditions (6) are satisfied and $\eta_1, \eta_2 \ll 1$ we have

$$\delta_{4,5}\approx-\frac{\xi\varepsilon}{2}\pm\left(\frac{\xi^2\varepsilon^2}{4}-y^2\right)^{1/2}$$

and (20) reduces to the characteristic polynomial obtained in Refs. 5–7 for a third-order differential equation. If (6) is not satisfied, the equation is of third order, but the roots differ greatly.

The solution of (21) in general form is too cumbersome to write out here, and we present the results for the most interesting particular cases.

A) Exact resonance, $\delta\omega = 0$

In this case only two roots of (20) are significant, since the term with $x_3 = -\delta$ makes no contribution to (21). At

$$(1+\delta+\xi-\eta_1)^2\gg\xi\varepsilon|1-\xi|$$

the roots of interest to us take the form

$$x_{1,2}\approx-\frac{1}{2}\left(\eta_1+\eta_2+\frac{\varepsilon(1+\eta_2-\eta_1)}{1+\xi+\eta_2-\eta_1}\right)\pm\left[\frac{1}{4}\left(\eta_1+\eta_2+\frac{\varepsilon(1+\eta_2-\eta_1)}{1+\xi+\eta_2-\eta_1}\right)^2-\xi-\eta_1\delta\right]^{1/2}.$$

In particular, under the condition $\xi \ll 1$ the results agree with those of Refs. 5–7, whereas at $\xi \gg \varepsilon^2$ and $\eta_1 = \eta_2 = 0$ we obtain the result of Ref. 8:

$$\langle\rho\rangle\approx-\cos ft\exp[-\varepsilon\gamma t/2(1+\xi)],$$

which is an additional confirmation of the applicability of the employed uncoupling procedure, for it is precisely in the region of strong fields that its use might not be valid.

Interest attaches also to the case when

$$|\eta_1-\delta-1-\xi|^2\ll 4\xi\varepsilon|1-\xi|\ll(1-\xi)^4/\xi^4$$

and a substantial role in the change of $\langle\rho(t)\rangle$ is played by one of the poles of the second group.

Under the additional condition

$$\xi=\Delta+1,\quad \Delta\neq 1, 2, 3, \ldots,\quad \Delta\gg\varepsilon^{1/2}$$

we obtain

$$\begin{aligned}\langle\rho(t)\rangle=&-\frac{1}{1+(1+\Delta)/\eta_2(2+\Delta+\eta_2)}+\frac{1+\Delta}{\Delta(1+\Delta+\eta_2)}e^{-\gamma(1+\Delta+\eta_2)t}\\ &-\frac{(1+\Delta)}{\Delta(1+\eta_2)}e^{-\gamma(1+\eta_2)t}\cos\left[\gamma\left(\frac{\varepsilon}{\Delta}(1+\Delta)\right)^{1/2}t\right].\end{aligned}$$

This result cannot be obtained within the framework of Refs. 5–8. The population difference can have such a time dependence in the relatively rare case when the time of the longitudinal relaxation is shorter than the time of the transverse relaxation.

B) Large detuning $|\delta\omega| \gg f$

In this case the solution (21) can be represented in the form

$$\langle\rho(t)\rangle\approx(\rho_0-\rho_\infty)e^{-(d_3/d_4)\gamma t}+\rho_\infty+\Delta\rho_{osc}, \tag{22}$$

where $\Delta\rho_{osc}$ are small rapidly oscillating terms that attenuate rapidly compared with the first term of the expression;

$$\frac{1}{\tau_0}=\frac{d_3}{d_4}=-\eta_1-\frac{\xi[(\delta-\eta_1)(1+\eta_2-\eta_1)^2+y^2(\eta_2-\eta_1)]}{[y^2+(\delta-\eta_1)^2][y^2+(1+\delta-\eta_1)^2]}. \tag{23}$$

In the particular case when there is no intrinsic relaxation, the character of the onset of saturation is determined by the shape of the emission line wing at the resonant frequency of the system.

As for the applicability of the approximations (15) and (16) which were the basis for all the results, a direct estimate of the discarded terms of the expansion (11), taken each separately, would yield an applicability condition

$$\frac{f^2\delta\omega^4}{(\delta\omega^2+\varepsilon^2\gamma^2)(1+\delta\omega^2/\gamma^2)}\ll\gamma^4,$$

but a more detailed analysis of the entire sum of the terms not taken into account in (15) shows that the criterion of the applicability of the employed approximation is provided by the inequalities (7).

In the region (7) it is easy to trace with the aid of the results the transformation of the character of the population of the upper level from a coherent character (the Rabi precession) to a noncoherent one (the transition model) with increasing spectral width of the radiation with the fluctuating frequency.

If the field-intensity amplitude $E_0(t)$ is a definite function of the time, then near the line center, i.e., in the region where the line can be regarded as having a Lorentz shape, Eq. (16) can be solved for radiation with a fluctuating frequency in those situations when the equation has a solution for a system with unequal relaxation times in a monochromatic field.

Returning to the general case (13), we point out that the solution of Eq. (16) also reduces to finding the three most significant poles of the corresponding function $\langle L(p)\rangle$. The coefficients of the cubic equation contain in this case two k functions $\delta_4, \delta_5, \ldots, \delta_{2k+3}$ [k is the number of the poles of the function $g_2(i\omega)$ at $\omega > 0$]. The criterion of the applicability of the decoupling in this case is likewise given by the inequalities (7).

4. MULTIPHOTON TRANSITIONS

4.1. We extend our analysis to include multiphoton resonant transitions in multilevel systems. We consider a molecule or an atom with an energy spectrum that has a level $\mathscr{E}_n$ such that

$$\omega_{0n}=(\mathscr{E}_n-\mathscr{E}_0)/\hbar\approx n\omega_l,$$

where $\mathscr{E}_0$ is the energy of the ground state and ω_l is the frequency of the external field. It is assumed that the system is in the ground state at the instant when the field is turned on.

In addition, we assume that there are no levels that might have a resonance of any order with the levels $|0\rangle$ and $|n\rangle$ (see Fig. 2). In the derivation of the equations for the density-matrix elements it is possible to take into account exactly only the stimulated transitions between the levels $|0\rangle$ and $|n\rangle$, and use perturbation theory for all the remaining levels. The last levels serve only as intermediate virtual states and have small populations. In addition, we assume that a direct transition between the levels $|0\rangle$ and $|n\rangle$ is forbidden ($d_{0n}=0$) and the "shortest upward way" from $|0\rangle$ to $|n\rangle$ consists of n steps, so that the probability of populating the state $|n\rangle$ appears only in n-th order of perturbation theory. Similar transitions ("multiphoton coherent transitions") were previously considered for the case of a monochromatic external field and without allowance for the relaxation time.[20-24]

Assume that the conditions

$$|\omega_{0p}-m\omega_l|\gg f(t)_{pk},\ p|\dot{\varphi}|,\ 1/T_{pk}$$

are satisfied for all $p\neq 0,n$ and for all m and k, where $\hbar\omega_{0p}=\varepsilon_p-\varepsilon_0$, $f(t)_{pk}$ is the field-induced broadening for the transition $|p\rangle\to|k\rangle$ and can depend on the time, and T_{pk} are the proper relaxation times.

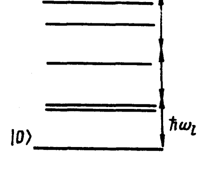

FIG. 2. Level scheme considered in the problem.

Taking into account all the off-diagonal elements of the density matrix with the exception of ρ_{0n}, we obtain by perturbation theory for the slow variables $\rho_{00}, \rho_{nn}, \rho_{0n}'$ the system of equations

$$\frac{d\rho}{dt}=-\frac{\rho-\rho_0}{T_{1,0n}}-i\beta E_0^n(t)\,(e^{in\varphi}\rho_{n0}'-e^{-in\varphi}\rho_{0n}'),$$
$$\frac{id\rho_{0n}'}{dt}=-\frac{\beta E_0^n(t)}{2}e^{in\varphi}\rho+\left[n\omega_l-\mathscr{E}_n+(a_0-a_n)E_0^2(t)-\frac{i}{T_{2,0n}}\right]\rho_{0n}', \tag{24}$$

where

$$\rho=\rho_{nn}-\rho_{00},\qquad \rho_{0n}'=\rho_{0n}e^{in\omega_l},$$
$$\beta=\frac{2}{(2\hbar)^n}\sum_{m_1,\ldots m_{n-1}}\frac{d_{0m_1}\ldots d_{m_{n-1}n}}{(\omega_{m_10}-\omega_l)\ldots(\omega_{m_{n-1}0}-\omega_l(n-1))},$$
$$a_0=-\frac{1}{2}\sum_{m\neq 0,n}\frac{|d_{0m}|^2\omega_{m0}}{(\omega_{m0}^2-\omega_l^2)},\tag{25}$$
$$a_n=-\frac{1}{2}\sum_{m\neq 0,n}\frac{|d_{nm}|^2\omega_{mn}}{(\omega_{mn}^2-\omega_l^2)}.$$

The quantity $(a_0-a_n)E_0^2(t)$ yields the dynamic Stark shift; if it vanishes ($a_0=a_n$), then the shift must be sought in higher order in $E_0(t)$. If $\beta=0$, this means that a resonant n-photon transition is impossible in the system.

From (23) we can easily see that if we establish the correspondence

$$\delta\omega\leftrightarrow n\omega_l-\varepsilon_n+(a_0-a_n)E_0^2(t)=\Delta\omega,$$
$$f(t)\leftrightarrow\beta E_0^n(t)=\Delta_{0n},$$
$$\varphi\leftrightarrow n\varphi,$$

then the problem reduces exactly to the same problem considered above for a two-level system, with the only difference that the frequency detuning depends on the time if $E_0(t)$ is not constant, and we obtain Eq. (16) for n-photon resonance. As can be readily determined from (17), the moments λ_{ij} increase by a factor n^2. Consequently $\varepsilon\gamma$ is replaced by $n^2\varepsilon\gamma$, and the criterion for the applicability of the employed approximation of fast and small approximations becomes more stringent:

$$n^2\varepsilon\ll 1. \tag{26}$$

It should be noted that if the sign of the inequality (26) is reversed when n is large enough, then the multilevel system behaves in a Lorentz-shaped radiation field like a two-level system in a field with a Gaussian spectrum, with a line half-width $n\langle\Omega^2\rangle^{1/2}<n^2\varepsilon\gamma$.

The presence of a time-dependent Stark shift cannot be ignored, since it is at any rate not smaller than Δ_{0n}, but in the case

$$n^2\varepsilon\gamma>|a_0-a_n|E_0^2(t)$$

allowance for this shift should not necessitate large corrections to the results obtained with constant $\Delta\omega$.

Whereas in sufficiently strong fields the nonmonochromaticity of the radiation does not play a substantial role for a two-level system, inasmuch as $f/\varepsilon\gamma\gg 1$, a finite laser line width is of prime significance for multiphoton processes, for even in the case of narrow lines ($\Delta\nu_1\approx 0.03\ \mathrm{cm}^{-1}$) in strong fields ($I\geqslant 10^5\ \mathrm{W/cm}^2$) the following inequality is well satisfied for many systems:

$$\Delta_{0n}\ll n^2\gamma\varepsilon.$$

Thus, whereas for a two-level system the field can frequently be regarded as monochromatic under the ordinary experimental conditions, the situation is reversed for multilevel systems, a fact pointed out already in Ref. 20.

With the aid of the results obtained in Sec. 3 it is easy to trace the change from coherent multiphoton transitions in a nonmonochromatic field. In particular, if $\Delta_{0n}/n^2\varepsilon\gamma \gg 1$ and if the intrinsic relaxation in the system is disregarded, we obtain the results of the theory of coherent multiphoton transitions[20-24]; at $\Delta_{0n}/n^2\varepsilon\gamma \ll 1$ and $t \ll n^2\varepsilon\gamma/\Delta_{0n}^2$ we obtain the perturbation-theory results.

The two-level-system approximation used by us is valid if the levels intermediate between $|0\rangle$ and $|n\rangle$ have small populations. However, they can acquire a substantial population because of the presence of the emission line wings, inasmuch as the order of the transitions into these states is lower than for $|0\rangle \to |n\rangle$.

The criterion for the applicability of the approximation (24) is the requirement that the time of equalization of the populations of the ground and nth level be short compared with the time τ_0 given in (23). In particular, at $\Delta_{0n}/n^2\varepsilon\gamma \gg 1$ we must have

$$\left[1+\frac{(\omega_{01}-\omega_l)^2}{\gamma^2}\right][(\omega_{01}-\omega_l)^2+\varepsilon^2\gamma^2+f^2] \gg \frac{n^2\varepsilon^2\gamma^2 f^2}{\Delta_{0n}^2}. \tag{27}$$

On the other hand, if the inequality (27) does not hold, then the decisive role can be assumed not by multiphoton but by multistep excitation of the system from level to level, despite the nonresonant character of the excitation.

4.2. We consider now several examples of quantum systems for which our results are valid.

A) Nonlinear quantum oscillator

The energy spectrum of a quantum oscillator with weak nonlinearity is described by the formula

$$\varepsilon(k)=\hbar(\omega_{01}+{}^1/_2\alpha)k-{}^1/_2\hbar\alpha k^2, \quad k=0,1,\ldots,$$

where α is the anharmonicity constant.

Assuming that the dipole moments of the transitions are the same as for the harmonic oscillator, we obtain in this case[20] for n-photon resonance

$$|\Delta_{0n}|=\alpha\left(\frac{f}{\alpha}\right)^n\frac{n^2}{n!^{3/2}}, \quad a_0=a_n.$$

The Stark shift is then quadratic in the field intensity, and the condition (27) takes the form

$$\left[1+\frac{\alpha}{4\gamma^2}(n-1)^2\right]\left[\frac{\alpha^2}{4}(n-1)^2+\varepsilon^2\gamma^2+f^2\right] \gg \frac{\varepsilon^2\gamma^2(n!)^3}{n^2(f/\alpha)^{2(n-1)}}. \tag{28}$$

For example, for the typical experimental values $\alpha = 5\ \text{cm}^{-1}$, $d=0.3\text{D}$, $I=10^7\ \text{W/cm}^2$, and $\Delta\nu_1 = 0.05\ \text{cm}^{-1}$ we have

$$|\Delta_{0n}|=0.07\,\text{cm}^{-1} \text{ for } n=2, \quad |\Delta_{0n}|=3\cdot10^{-3}\,\text{cm}^{-1} \text{ for } n=3.$$

We see therefore that, regardless of the character of the radiation fluctuations, allowance for the nonmonochromaticity of the radiation becomes necessary when the excitation of molecules in a strong external field is analyzed outside the framework of perturbation theory.

In particular, if we specify the model of small and fast fluctuations of the frequency, then an analysis of the inequality (28) at $n=3$ shows that at $\varepsilon = 0.1$ the third level is significantly populated via a three-photon resonance transition. On the other hand, if $\varepsilon = 0.01$, then multistep excitation from level to level can become the principal channel of population of the third level.

If the model of the nonlinear oscillator is used to consider the molecule excitation processes, it must be recognized that, because of intermediate transitions with change of rotational numbers, the number of trajectories in expression (25) for β can become large if a multiphoton transition with $n \gg 1$ is realized.

This leads, in particular, to an increase of the region in which the inequality (28) is satisfied.

B) Multiphoton transitions in a two-level system

It is possible to reduce to this problem also multiphoton transitions induced in a two-level system by one laser line. It is easy to show that substantial population of the upper level is possible if

$$\omega_{01}\approx(2n+1)\omega_l, \quad n=1,2,\ldots.$$

In this case we obtain for the density-matrix elements the system of equations

$$\begin{aligned}\frac{d\rho}{dt}&=-\frac{\rho-\rho_0}{T_1}-if\left(\frac{f}{4\omega_l}\right)^{2n}\frac{1}{(n!)^2}\left(e^{i(2n+1)\varphi}\rho_{01}'-e^{-i(2n+1)\varphi}\rho_{10}'\right),\\ i\frac{d\rho_{01}'}{dt}&=-\frac{f}{2}\left(\frac{f}{4\omega_l}\right)^{2n}\frac{1}{(n!)^2}e^{i(2n+1)\varphi}\rho\\ &+\left[(2n+1)\omega_l-\omega_{01}-\frac{f^2(2n+1)}{4\omega_l n(n+1)}-\frac{i}{T_2}\right]\rho_{01}',\end{aligned} \tag{29}$$

where $\rho = \rho_{11}-\rho_{00}, \rho_{01}'=\rho_{01}e^{i(2n+1)\omega_l t}$. This system, subject to some transformations, coincides with (24). Then (27) goes over into

$$\left(1+\frac{4n^2\omega_l^2}{\gamma^2}\right)(4n^2\omega_l^2+\varepsilon^2\gamma^2+f^2) \gg \frac{(2n+1)^2\varepsilon^2\gamma^2(n!)^4}{(f/4\omega_l)^{4n}}.$$

From (29) we obtain for a monochromatic field the results of Ref. 25.

C) Multiphoton transitions in a system of two vibrational levels with rotational substructure

The energy spectrum of such a system is of the form

$$\mathscr{E}(k,J)=\hbar\omega_{01}k+BJ(J+1), \quad k=0,1,$$

where B is the rotational constant (see Refs. 26 and 27 in this connection).

Whereas in the dipole approximation only single-photon transitions $|0,J\rangle \to |1,J\pm1\rangle$ are allowed, multiphoton resonance makes possible effective population of the upper levels with change of the rotational number $J \to 1+2n+1$ (see Fig. 3). Let $2BJ/\hbar \gg f, kT \gtrsim BJ^2$ (where T is the temperature), and

$$-(J+1)J+(J+2n+1)(J+2n+2)\approx\hbar(\omega_l-\omega_{01})/B.$$

The problem of the change of the populations of the levels $|0,J\rangle$ and $|1,J+2n+1\rangle$ reduces then to the previously considered problem of the two-level system subject to the condition

$$\left[1+\frac{B^2n^2(4J+6+4n)^2}{\gamma^2}\right][B^2n^2(4J+6+4n)^2+\varepsilon^2\gamma^2+f^2] \gg f^2\varepsilon\gamma\max\left[\frac{\varepsilon\gamma}{\Delta_{0n}^2},\frac{1}{\Delta_{0n}}\right].$$

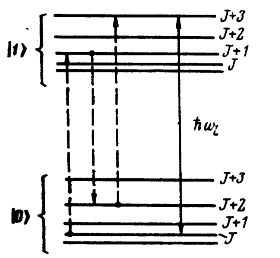

FIG. 3. Sequence of virtual transitions that lead to the multiphoton transition $|0,J\rangle \to |1,J+3\rangle$.

The effective field broadening Δ_{0n} becomes equal to

$$\Delta_{0n} = \frac{E_0^{2n+1}}{2^{2(n+1)}} d_{J,J+1} d^*_{J+1,J+2} \ldots d_{J+2n,J+2n+1}$$

$$\times \left[B^{n-1} \prod_{l=1}^{(n-1)/2} (2J+2l+1)(2J+2n+2l+1) \right]^{-1},$$

where $d_{J,J+1}$ is the dipole matrix element of the $|0,J\rangle \to |1,J+1\rangle$ transition, and the effective width of the emission spectrum for the $|0,J\rangle \to |1,J+2n+1\rangle$ transition is equal to the true width $\varepsilon\gamma$, since a single emission quantum is really absorbed in the transition. We note that the populations of the intermediate states $|0,J+2k\rangle$ can be comparable with the population of the $|0,J\rangle$ state. This, however, does not prevent us from using the proposed scheme; it is easy to verify that allowance for this fact introduces only corrections that are small in f/BJ into the solutions obtained within the framework of the two-level system.

4.3. In conclusion, we consider resonant fluorescence induced in a rarefied gas of two-level atoms by radiation of frequency $\omega_1 \approx \omega_{01}/2n+1$. Let the population of the upper level reach after a time equal to the duration of the laser pulse a value much larger than unity. Let furthermore the duration of the pulse be shorter than the collision time and let the rates of the intrinsic relaxation in the system be determined only by the spontaneous emission.

Then, recognizing that as $t \to \infty$ the system returns to the equilibrium state, we obtain from (29) an estimate for the number of fluorescence protons at the frequency ω_{01}:

$$N_0 = \frac{8PV\omega_l^2}{kT} \int_0^t \int_0^{t_1} \exp\left[-\frac{A}{2}(t_1-t_2) - \frac{\omega_0^2}{2}(t_1-t_2)^2 \right]$$

$$\times \Delta(t_1)\Delta(t_2) \cos\left[(2n+1)[\varphi(t_1)-\varphi(t_2)] - \delta\omega(t_1-t_2) \right.$$

$$\left. - \int_{t_2}^{t_1} \frac{f^2(t_3)(2n+1)}{4n(n+1)\omega_l} dt_3 \right] dt_1 dt_2, \tag{30}$$

where $\Delta(t) = [f(t)/4\omega_l]^{2n+1}$, V is the irradiated volume, P is the gas pressure, T is the gas temperature, k is the Boltzmann constant, A is the probability of the spontaneous $1 \to 0$ transition, and $\delta\omega = (2n+1)\omega_l - \omega_{01}$. In the derivation of (30) we summed over the particles and took into account the Doppler scatter the resonant frequencies to the atoms with half-width $1.67\omega_0$.

It is possible to take into account in (30) both the fluctuations of the phase of the radiation and the changes of its amplitude with time, by averaging the corresponding terms in the integrand over the realizations. The changes of the amplitude must be necessity lead to the presence of an alternating Stark shift of the resonant frequency in (30). Allowance for the influence of this factor on the fluorescence requires a separate analysis. On the other hand, if I in the radiation is constant and its phase fluctuates, then the Stark shift is of no significance. The fluctuations of the phase can then play an important role in the phenomenon in question, since they influence both the quantity N_0 and the width of the fluorescence line. Namely, N_0 will depend substantially on the fluctuations of the phase if $(A \ll \Delta\omega_L)$

$$(2n+1)^2(\Delta\omega_L - 2\pi/t_p) \gg \omega_0 \quad \text{at} \quad \varepsilon \ll 1. \tag{31}$$

As for the fluorescence line width, it can be shown that it depends on the fluctuation spectrums when the inequality sign in (31) is reversed:

$$\Delta\omega_{fl} \approx \frac{2}{3}\pi(2n+1)^2\left(\Delta\omega_L - \frac{2\pi}{t_p}\right) + \frac{3}{t_p}$$

$$\text{at} \quad \frac{1}{t_p} \gg \frac{\pi}{3}\left(\Delta\omega_L - \frac{2\pi}{t_p}\right) \gg \frac{1}{(2n+1)^2 t_p}. \tag{32}$$

We consider now a concrete experiment. A three-photon resonant transition Xe($5p^6\,{}^1S_0 - 5p^5 6s\,{}^3P_1$) was revealed in Ref. 28 by fluorescence of wavelength $\lambda = 1470$ Å due to prior excitation with pulsed radiation of wavelength $\lambda = 4400$ Å. The parameters were $V = 1.2 \cdot 10^{-7}$ cm^3, $P = 8 \cdot 10^{-3}$ Torr, $T = 300K$, $A = 2.2 \cdot 10^8$ sec^{-1}; $3.3\omega_0 = 2.5 \cdot 10^{10}$ sec^{-1}, $d_{0,1} = 2.5$ D [the transition oscillator strength was $f_0 = 0.21$ (Ref. 29)], the radiation intensity at the maximum was $I = 4 \cdot 10^9$ W/cm^2, the width of the laser-emission spectrum was $\Delta\omega_L = 1.5 \cdot 10^9$ sec^{-1} and $\Delta\omega_{fl} = 2.5 \cdot 10^9$ sec^{-1}. The experimental estimate cited in Ref. 28 is $N_0 = 850$.

There were apparently no amplitude fluctuations in the experiment, since variation of the radiation intensity in a wide range near 4×10^9 W/cm^2 did not cause a change change of ω_{fl}. Such a change would be the direct consequence of the presence of fluctuations of this kind, since the Stark shift is large: $3f^2/8\omega_l \approx 2 \cdot 10^{10}$ sec^{-1}.

We confine ourselves to the assumption that whatever fluctuations exist are those of the phase. We note immediately that, on the basis of the presented numerical data, the phase fluctuations do not influence significantly the value of N_0, and allowance for the Doppler scatter is important here (this was not taken into account in Ref. 28). We therefore obtain from (30)

$$N_0 \approx \left(\frac{f}{4\omega_l}\right)^3 \frac{8VP\omega_l^2}{kT} \frac{1}{\omega_0} \left(\frac{\pi}{2}\right)^{1/2} t_p .$$

The duration of the pulse is unfortunately not cited in Ref. 28. It can be determined, however, from the other experimental data, by assuming the model of the radiation fluctuations. Namely, since $\Delta\omega_{fl} > \Delta\omega_L$, we get from (32) at $\varepsilon \ll 1$

$$\varepsilon\gamma \approx \frac{1}{18}\left(\Delta\omega_{fl} - \frac{3}{2\pi}\Delta\omega_L\right) = 10^8 \text{ sec}^{-1}.$$

In this case $t_p = 5$ nsec. If we use this value to calculate N_0, we get $N_0 \approx 1500$, in fair agreement with the experimental value. We note that the estimates obtained in Ref. 28 were based on the results of Ref. 30, which are not applicable in this case. It is therefore not surprising that to obtain a reasonable value of N_0 it was necessary to use a rather arbitrary estimate of d_{01}, instead of using all the necessary experimental data.

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