Two-level system in a resonant multifrequency field

V. Yu. Finkel'shteĭn

P. N. Lebedev Physics Institute, USSR Academy of Sciences

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The evolution of a two-level system in a multifrequency resonant field with random phased modes is investigated. A method is developed that takes automatic account of the periodicity of such a noise field in time. It is shown that in the case of a broad emission spectrum and sufficiently high intensity a "shift" of the probabilities is observed for the density matrix of the system at instants nT_0 ($n \geq 2$, T_0 is the period of the complex amplitude of the field). At these instants, the averaged density-matrix elements assume values that differ substantially from zero. Also investigated are cases of a weak field and the case of amplitude fluctuations with a periodic correlation function. The results yield the correlation function for the population difference of the system when it is acted upon by a structureless Gaussian noise. The possibility of observing the probability shift are discussed.

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1. INTRODUCTION

Experimental observation of a large number of new interesting phenomena that take place in atomic and molecular systems acted upon by powerful electromagnetic fields (it suffices to mention, e.g., multiphonon ionization of atoms[1] or radiative dissociation of polyatomic molecules[2,3]) has stimulated many theoretical investigations in recent years. In particular, great interest is shown in the study of the behavior of quantum systems in a nonmonochromatic external field, inasmuch as in the description of the quantum processes that occur in the field of real laser radiation one cannot ignore the essentially nonmonochromatic character of the emission of a powerful pulsed multimode laser, which is usually employed in the experiments.[4,5] By nonmonochromatic field is mean in this paper a field with randomly fluctuating parameters (amplitude, phase). Ordinarily one considers the interaction of radiation of this kind with quantum systems that reduce in one sense or another to a two-level system.[6]

The present paper is also devoted to the study of the evolution of the density matrix of a two-level system in a nonmonochromatic external field. We are interested in the density matrix averaged over all the possible realizations of the field. The difference between this paper and the preceding one is the following. The radiation with which the quantum system interacts is usually regarded as a structureless noise with a correlation function that attenuates monotonically at infinity (see, e.g., Refs. 7–15). However, the radiation intensity of a Q-switched multimode laser is periodic with a so-called axial period $T_0 = 1/\Delta\nu_0 = 2L/c$, where L is the resonator length. If the phases of the modes are random, the intensity of the radiation in the axial period is also a random function of the time.[4] The correlation function of the radiation field

$$\lambda_{12} = \langle E(t_1) E^*(t_2) \rangle$$

is in this case periodic in time, accurate to within slow damping at infinity because of the finite widths of the modes and other weak fluctuations. We shall disregard hereafter this slow damping and assume the duration of the radiation pulse to be much shorter than the characteristic times of violation of the periodicity of the correlation function. Usual values for a pulsed laser are

$$t_{\text{pul}} \approx 200 \text{ msec.}, \quad \Delta\nu_0 \approx 10^{-2} \text{ cm}^{-1}, \quad N \gg 30$$

(N is the number of longitudinal modes). During the pulse time t_{pul} the structure of the noise repeats itself about several hundred times. In this paper we take into account this feature of the radiation field and show that it leads to nontrivial consequences. Up to now, no account was taken in the study of the evolution of a two-level system outside the framework of perturbation theory, of the time periodicity of the noise field (with the exception of the case $N=2$; see, e.g., Refs. 16 and 17).

The radiation field $\mathscr{E}(t)$ of a multimode Q-switched laser is very well described by the model[4,18]

$$\mathscr{E}(t) = E(t) \exp\{i\omega_l t\} + \text{c.c.},$$

$$E(t) = \frac{E_0}{2} \sum_{[-N/2]}^{[N/2]} \exp\left\{ i\left[\varphi_n + \frac{\pi t}{T_0}\left(2n - \frac{1+(-1)^N}{2}\operatorname{sign}(n)\right)\right]\right\}. \quad (1)$$

Here ω_l is the radiation carrier frequency, E_0 is the amplitude of one mode, and φ_n are the phases and are randomly distributed in the interval $(0, 2\pi)$. The quantity $E(t)$ constitutes complex noise whose structure repeats in time with a period T, with $T=T_0$ at $N=2k+1$ and $T=2T_0$ at $N=2k$.

We shall assume henceforth that

$$N \gg 1, \quad \alpha \ll N, \quad (2)$$

where $\alpha = E_0^2 d_{01}^2 T_0^2 / 2\hbar^2$, d_{01} is the matrix of the transition $|0\rangle \to |1\rangle$. The condition (2) is usually called the case of a broad radiation spectrum, for in this case the width $\Delta\nu = N\Delta\nu_0$ of the laser-radiation spectrum is much larger than that of the frequencies characterizing the rates of the quantum processes in a two-level system. At the usual value of the intensity of a powerful laser, $I = 3\times 10^6$ W/cm^2 at $d_{01} = 0.3$ D we obtain $\alpha = 10$. Thus, the proposed model describes the interaction of a real laser radiation with a real two-level system. For a multimode laser, Eq. (2) is a good approximation. It is easy to show that if the phases φ_n are independent of one another, then, if condition (3) is satisfied, $E(t)$ can be simulated with a high degree of accuracy by a complex Gaussian noise.

$$\langle (E(t)+E^*(t))^{2k}\rangle=(2k-1)!!\left(\frac{NE_0^2}{2}\right)^k\left(1-\frac{k(k-1)}{4N}+\ldots\right),$$

Re$E(t)$ and Im$E(t)$ can be regarded in this case as stationary normal and mutually independent processes.[19]

In the present paper we develop a method of describing the evolution of a two-level system; this method takes automatically into account the periodicity of the noise (1) in time. We shall demonstrate the direct connection between this method and the quasi-energy description at $T\nu_l=p$, where $p\gg N$ is an integer.

It is well known (see, e.g., Ref. 20, §40) that in the case of a monochromatic field the population difference $\rho_0=\rho_{00}-\rho_{01}$ varies periodically in time at $\rho_{00}(0)=1$ (the Rabi precession):

$$\rho_3=\cos\left[\frac{d_{01}}{\hbar}\left(\frac{8\pi I}{c}\right)^{1/2}t\right]. \quad (3)$$

On the other hand, for a structureless Gaussian noise we obtain (see, e.g., Ref. 17)

$$\langle\rho_3\rangle=\exp\{-4\pi d_{01}^2It/\hbar^2\Delta\nu c\}. \quad (4)$$

In the well-investigated case of small and fast fluctuation of the radiation phase (see, e.g., Ref. 9), the transition from the excitation regime (3) to (4) proceeds with decreasing intensity I at fixed spectrum width $\Delta\nu$.

It will be shown in the present paper that for a two-level system interacting with the radiation (1), if condition (2) is satisfied, the situation is in a certain sense reversed. Namely, in the case of a strong field ($\alpha\gg1$) the regime of excitation of the system coincides with (4) up to the instant $t=T_0$. Next, at $t>T_0$, substantial differences due to the periodicity of the noise (1) appear. Namely, at the instants $t=nT_0$, with $n\geqslant 2$, a "shift" of the probability for the population difference and polarization takes place (see Fig. 2 below). At these instants they take on values substantially different from zero [cf. (4)]. In a weak field $\alpha<1$ at $N=2k+1$ there will be observed a slowly damped Rabi precession in the field of one radiation mode:

$$\langle\rho_3\rangle\approx\cos\left[\frac{d_{01}}{\hbar}\left(\frac{8\pi I}{cN}\right)^{1/2}t\right]\exp\left\{-\frac{\alpha^2}{144}\frac{d_{01}^2\pi I}{\hbar^2cN}t^2\right\},$$

which is reminiscent of (3).

The strong-field case is considered in Sec. 4, where a diagram technique is used to obtain exact solutions for the averaged density matrix at $0\leqslant t\leqslant 2T_0$. Amplitude fluctuations of the radiation are also considered (Sec. 5). In Sec. 6 is analyzed the case of a weak field. The conclusion deals with the possibility of observing the "shift" of the probability for the level-population difference.

2. ROTATIONAL REPRESENTATION OF THE EVOLUTION OF A TWO-LEVEL SYSTEM

We consider the interaction of a two-level system with the radiation field (1). We use the rotating wave (resonant) approximation (see, e.g., Refs. 6 and 20). In the absence of relaxation at resonance $\omega_l=\omega_{01}(\hbar\omega_{01}=E_1-E_0)$ the system of equations for the population level amplitudes is of the form

$$i\hbar\dot{A}_0=-E(t)d_{01}A_1,\quad i\hbar\dot{A}_1=-E^*(t)d_{01}A_0. \quad (5)$$

In the absence of relaxation, description of a quantum system in a pure state in terms of a density matrix is exactly equivalent to a description in terms of amplitudes. On the other hand, the system of equations for ρ_{ij} contains one more equation than (5). This notwithstanding, the analysis will be continued for the density-matrix elements

$$\rho_{00}=|A_0|^2,\quad \rho_{11}=|A_1|^2,\quad \rho_{10}=A_1^*A_0. \quad (6)$$

This is due to two circumstances. First, all the observable physical quantities (absorbed energy, polarization, and others) are expressed directly in terms of ρ_{ij}. Therefore direct interest attaches to averaging of precisely the biquadratic combinations of the amplitudes (6) over all possible realizations of the external field.

Second, for the density matrix there is a very lucid vector model of the energetic spin,[6,21] which we shall use henceforth to obtain concrete results.

From (5) we obtain a system of equations for the elements of the density matrix:

$$\frac{d}{d\tau}\rho_1=-Y\rho_3,\quad \frac{d}{d\tau}\rho_2=X\rho_3,\quad \frac{d}{d\tau}\rho_3=Y\rho_1-X\rho_2$$

or

$$\frac{d}{d\tau}\boldsymbol{\rho}=-[\mathbf{H}\times\boldsymbol{\rho}],\quad \boldsymbol{\rho}=\begin{pmatrix}\rho_1\\ \rho_2\\ \rho_3\end{pmatrix},\quad \mathbf{H}=\begin{pmatrix}X\\ Y\\ 0\end{pmatrix}, \quad (7)$$

where

$$\rho_1=2\mathrm{Re}\,\rho_{10},\quad \rho_2=2\,\mathrm{Im}\,\rho_{10},\quad \rho_3=\rho_{00}-\rho_{11},\quad \tau=t/T_0,$$

$$X=-(2\alpha)^{1/2}\sum^N\cos(2\pi n\tau+\varphi_n),\quad Y=-(2\alpha)^{1/2}\sum^N\sin(2\pi n\tau+\varphi_n)\quad \text{at } N=2k+1;$$

$$X=-(2\alpha)^{1/2}\sum^N\cos\left(2\pi\left(n+\frac{1}{2}\right)\tau+\varphi_n\right),\quad Y=-(2\alpha)^{1/2}\sum^N\sin\left(2\pi\left(n+\frac{1}{2}\right)\tau+\varphi_n\right)\quad \text{at } N=2k; \quad (8)$$

$\alpha=f^2T_0^2/2$, $f=d_{01}E_0/\hbar$ is the field broadening, and d_{01} is the dipole moment of the transition $|0\rangle\to|1\rangle$.

An essential feature of (7) is that it is a system of linear differential equations with periodic coefficients (8). Such systems have special properties[22] which we shall in fact use later on. To this end, we represent the solutions of the system (7) in the form

$$\boldsymbol{\rho}(\tau)=M(\tau)\boldsymbol{\rho}(0). \quad (9)$$

The matrix $M=[m_{ij}]$ $(i,j=1,2,3)$ satisfies the equation

$$dM/d\tau=AM,\quad M(0)=E=[\delta_{ij}], \quad (10)$$

$$A=\begin{pmatrix}0&0&-Y\\0&0&X\\Y&-X&0\end{pmatrix},\quad \det[M(\tau)]=1.$$

It is clear from (9) that $M(\tau)$ contains complete information on the evolution of the density matrix at arbitrary initial conditions. $M(\tau)$ is an orthogonal real matrix that describes the proper rotations in a three-dimensional Euclidean space (the space of the energetic spin). The rotation matrix $M(\tau)$ is completely specified by the angle of rotation about an axis with direction cosines[23] $C_1(\tau)$, $C_2(\tau)$, $C_2(\tau)$.

If we regard the rotation of the three orthogonal unit vectors $\mathbf{m}_1, \mathbf{m}_2, \mathbf{m}_3$ as the rotation of a rigid body with a fixed point, then

$$m_{ij}(\tau)=(\mathbf{m}_i(0)\mathbf{m}_j(\tau)).$$

It is convenient to express m_{ij} in terms of the Euler angles

$$\begin{gathered}m_{11}=\cos\xi\cos\zeta\cos\eta-\sin\xi\sin\zeta,\\ m_{12}=-(\cos\zeta\cos\eta\sin\xi+\sin\zeta\cos\xi),\qquad m_{13}=\cos\zeta\sin\eta,\\ m_{21}=\sin\zeta\cos\xi\cos\eta+\cos\zeta\sin\xi,\\ m_{22}=-\sin\zeta\cos\eta\sin\xi+\cos\zeta\cos\xi,\qquad m_{23}=\sin\xi\sin\eta,\\ m_{31}=-\sin\eta\cos\xi,\qquad m_{32}=\sin\eta\sin\xi,\qquad m_{33}=\cos\eta.\end{gathered}\tag{11}$$

The instantaneous-velocity vector $\mathbf{H}$ lies at all time in the plane of the vectors $\mathbf{m}_1(0)$ and $\mathbf{m}_2(0)$, and

$$\frac{d}{d\tau}\mathbf{m}_i=-[\mathbf{H}\times\mathbf{m}_i].$$

If the period of the coefficients X and Y is equal to T, then $M(\tau)$ has the remarkable property

$$M(\tau+Tn)=M(\tau)M^n(T),\qquad n=0,1,\ldots\,.\tag{12}$$

Relation (12) reflects the fact that if the instantaneous angular velocity of a body with a fixed point is a periodic function of the time with period T, then the rotation of the body is a sequence of rotations through an angle $\theta(T)$ about a certain fixed axis within a time T.

The vector interpretation of the evolution of a two-level system is well known. In principle, by constantly following the precession of the vector ρ we would obtain in the present problem complete information on the evolution of the density matrix. It would be sufficient here to know the time variation of the two angles that specify the position of the vector ρ.

The description of the evolution in terms of a rotation matrix calls for knowledge of the three Euler angles and in the ordinary case yields unnecessary extra information. If, however, the instantaneous angular velocity is periodic in time, then knowledge of $M(\tau)$ only on the segment $(0, T)$ makes it possible, because of the property (12), to obtain complete information of the evolution of the two-level system for any instant of time $\tau > T$. It is this way we use a description in terms of $M(\tau)$ in the present paper. We shall henceforth call this representation of the evolution of a two-level system rotational. It is easy to show that[23]

$$\begin{gathered}1+2\cos n\theta(T)=m_{11}(nT)+m_{22}(nT)+m_{33}(nT),\\ m_{ii}(nT)=C_i^2(T)+(1-C_i^2(T))\cos n\theta(T).\end{gathered}\tag{13}$$

We use these relations to average $M(\tau)$ over the realizations of the external field (1).

At $\omega_{01}=\omega_f$ there exist two wave functions, Ψ_1 and Ψ_2 having in the rotating-wave approximation (5) the form

$$\begin{gathered}\Psi_1=[a_0(t)\,|0\rangle+a_1(t)\exp\{-i\omega_f t\}\,|1\rangle]\exp\{i\Omega t\},\\ \Psi_2=[a_1^*(t)\,|0\rangle-a_0^*(t)\exp\{-i\omega_f t\}\,|1\rangle]\exp\{-i\Omega t\},\end{gathered}\tag{14}$$

where

$$|a_0|^2+|a_1|^2=1,\qquad a_0(t+T)=a_0(t),\qquad a_1(t+T)=a_1(t).$$

It is easy to show that

$$\begin{gathered}C_1(T)=a_{00}^2-a_{10}^2,\qquad C_2(T)=2a_{00}a_{10}\sin(\beta_1-\beta_2),\\ C_3(T)=2a_{00}a_{10}\cos(\beta_1-\beta_2),\\ \Omega={}^1/{}_2\theta(T),\quad\text{or}\quad \pi-{}^1/{}_2\theta(T)\quad(0\leqslant\theta(T)\leqslant2\pi,\quad 0\leqslant\Omega\leqslant\pi)\\ a_0(0)=a_{00}\exp\{i\beta_1\},\qquad a_1(0)=a_{10}\exp\{i\beta_2\}.\end{gathered}\tag{15}$$

Which of the two possible values of Ω is realized depends on the details of the system dynamics on the segment $(0, T)$. If $T\nu_f=p$, where p is an integer, then the field $\mathscr{E}(t)$ is periodic with a period T. Then Ψ_1 and Ψ_2 coincide with two quasi-energy functions,[24-26] and Ω coincides with the quasienergy. The rotational representation is thus directly connected with the quasienergy description of a two-level system in a periodic external field.

We present the solutions (11) in explicit form

$$\begin{gathered}m_{11}=1-\sum_{n=0}^{\infty}(-1)^n\int_0^{\tau}\ldots\int_0^{\tau_{2n+1}}Y_1R_2{}^nY_{2n+2}\prod_1^{2n+2}d\tau_i,\\ m_{33}=\sum_{n=0}^{\infty}(-1)^n\int_0^{\tau}\ldots\int_0^{\tau_{2n-1}}R_1{}^n\prod_1^{2n}d\tau_i,\\ m_{12}=\sum_{n=0}^{\infty}(-1)^n\int_0^{\tau}\ldots\int_0^{\tau_{2n-1}}Y_1R_2{}^nX_{2n+2}\prod_1^{2n+2}d\tau_i,\\ m_{13}=-\sum_{n=0}^{\infty}(-1)^n\int_0^{\tau}\ldots\int_0^{\tau_{2n}}Y_1R_2{}^n\prod_1^{2n+1}d\tau_i,\\ m_{31}=\sum_{n=0}^{\infty}(-1)^n\int_0^{\tau}\ldots\int_0^{\tau_{2n}}R_1{}^nY_{2n+1}\prod_1^{2n+1}d\tau_i.\end{gathered}\tag{16}$$

Here

$$\begin{gathered}R_m{}^n=R^n(\tau_m)=(X_mX_{m+1}+Y_mY_{m+1})\ldots(X_{m+2n-2}X_{m+2n-1}+Y_{m+2n-2}Y_{m+2n-1}),\\ X_k=X(\tau_k),\qquad Y_k=Y(\tau_k),\qquad R^0=1,\qquad m=1,2.\end{gathered}$$

The interchange $Y\rightleftarrows X$ yields

$$m_{11}\to m_{22},\quad m_{12}\to m_{21},\quad m_{13}\to -m_{23},\quad m_{31}\to -m_{32}.$$

3. AMPLITUDE PHASE FLUCTUATIONS OF THE RADIATION. THE CORRELATION FUNCTION

It is easy to ascertain from (8) that if the phases are mutually independent, then $X(\tau)$ and $Y(\tau)$ are at $N\gg1$ normal, stationary, and mutually independent processes with a correlation function

$$\lambda_{12}^{X,Y}=\langle X_1X_2\rangle=\langle Y_1Y_2\rangle=\alpha\sin[\pi N(\tau_1-\tau_2)]/\sin[\pi(\tau_1-\tau_2)],\tag{17}$$

$$\langle X\rangle=\langle Y\rangle=\langle X_1Y_2\rangle=0.\tag{18}$$

This is the case of amplitude-phase fluctuations (APF) of the radiation. In the rotational representation the instantaneous rotation axis changes its direction randomly in the plane of the vectors $\mathbf{m}_1(0)$ and $\mathbf{m}_2(0)$. The angular velocity is also random. The correlation function λ_{12} is periodic in time, with $T=T_0$ at $N=2k+1$ and $T=2T_0$ at $N=2k$. The function λ is characterized by two times, $\tau_1=1/N$ and $\tau_2=1$, with $\tau\gg\tau_2$. Since $\lambda_{12}(\tau_1-\tau_2)$ does not attenuate at infinity ($|\tau_1-\tau_2|\to\infty$), the quantum system "remembers" the noise structure, generally speaking, for an arbitrarily long time (we recall that we consider no fluctuations other than those due to the random distribution of the mode phases).

We shall be interested hereafter in the rotation matrix averaged over all possible realizations of the external field. Let the characteristic time τ_0 of the variation of $\langle M(\tau)\rangle$ be much longer than τ_1. Then $\lambda_{12}^{X,Y}$ can be represented in the form

$$\begin{gathered}\lambda_{12}^{X,Y}=\alpha\sum_{n=0}^{\infty}\delta(|\tau_1-\tau_2|-n)\qquad\text{at}\quad N=2k+1,\\ \lambda_{12}^{X,Y}=\alpha\sum_{n=0}^{\infty}(-1)^n\delta(|\tau_1-\tau_2|-n)\qquad\text{at}\quad N=2k.\end{gathered}\tag{19}$$

It will be shown later that if λ_{12} is used in the form (19) the following inequality is valid:

$$\tau_0 \geqslant 1/\alpha.$$

Thus, the condition of validity of (19) is satisfaction of the inequality

$$\alpha \ll N. \qquad (20)$$

The parameter (α/π^2+1) has a clear-cut physical meaning. This is the number of radiation modes that act resonantly on the quantum system. Thus, (20) means that the "effective" number of modes is much smaller than their real number.

We shall consider hereafter only the case (20), which we name the case of a broad radiation spectrum. In this case α can be larger as well as smaller than unity. We shall consider both cases. We note that in the limit $\alpha\to\infty(T_0\to\infty, f^2T_0=\text{const})$ we obtain the case of a structureless Gaussian noise.

4. CASE OF STRONG FIELD ($\alpha \gg 1$)

We consider first the evolution of the system in the rotating-wave approximation at $\alpha\gg 1$. On the basis of the relations (13) above we obtain concrete results for the instants of time $\tau=n$. If we know the probability densities of $\theta(T)$ and $C_i(T)$, then we can determine the diagonal elements $\langle m_{ii}(nT)\rangle$. From (16), taking (18) into account, it is obvious that only the diagonal elements do not vanish upon averaging over the field realizations.

For the Euler angles η, ζ, and ξ we obtain from (10) and (11)

$$\dot\eta = X\sin\zeta - Y\cos\zeta, \quad \dot\zeta\,\mathrm{tg}\,\eta = X\cos\zeta + Y\sin\zeta, \qquad (21)$$

$$\xi=\xi(0)-\int_0^\tau d\zeta/\cos\eta,$$

$$\zeta(0)=\pi-\mathrm{arctg}(X/Y), \quad \eta(0)=0, \quad \xi(0)=-\zeta(0). \qquad (22)$$

The integral in (22) is taken along the trajectory of the representative point [the end point of the vector $\mathbf{m}_3(\tau)$]. The initial conditions are chosen such that $\zeta(0)$ and $\dot\zeta(0)$ are finite.

We consider the motion of some vector, say $\mathbf{m}_3$. Since (21) contains two random unknown parameters X and Y, it follows that η and ζ will also vary randomly, and the motion of the vector $\mathbf{m}_3$ will be random. During the time $\tau_1=(1/N)$, the vector $\mathbf{m}_3$ deviates from its initial position by some angle $\delta(\tau_1)$, with $\langle\delta^2(\tau_1)\rangle\sim\alpha/N$. The representative point then "fills" with its trajectory an area $\sim\alpha/N$ on the unit sphere, and during the time $\tau_2=1$ it fills an area $\sim\alpha$. The condition $\alpha\gg1$ means that the trajectory of the representative point will cover the unit sphere many times, and by the instant τ_2 the motion of the vector $\mathbf{m}_3$ becomes ergodic.[27] The angles $\eta(1)$ and $\zeta(1)$ are independent at that instant both of each other and of their initial values. Mathematically this is expressed by stating that the probability finding the vector $\mathbf{m}_3$ in the solid-angle element dO becomes constant:

$$P[\eta;\ \zeta;\ \tau=1]=\sin\eta_1 d\eta_1 d\zeta_1/4\pi.$$

We are interested however, in the motion of not only the vector $\mathbf{m}_3$ but of the entire system of vectors $\{\mathbf{m}_i\}$. The third Euler angle is not independent, but is a functional of the angles η and ζ [see (22)]. It depends, for any concrete realization of the random process (8), on the values of η and ζ along the entire trajectory. The condition $\alpha\gg1$ means also that a large number of trajectories, having the same realization probabiliy, lead to the same point (η,ζ). Since the successive rotations about the different axes do not commute, the angle $\xi(\eta)$ can have, at fixed $\eta(\tau)$ and $\zeta(\tau)$, different values that depend on the concrete trajectory, and by the instant $\tau=1$ it is already a random quantity independent of ζ_1 and η_1, and the equipartition law

$$P[\xi;\ \tau=1]=d\xi_1/2\pi.$$

is valid for it, too. Thus, the total probability density is

$$P[\eta,\ \xi,\ \zeta;\ \tau=1]=\sin\eta_1 d\eta_1 d\zeta_1 d\xi_1/8\pi^2. \qquad (23)$$

From (23) we can also obtain the probability distributions for $\theta(1)$ and $C_i(1)$:

$$P(\theta, C_1, C_2, C_3;\ \tau=1)=\pi^{-2}\delta(1-C_1^2-C_2^2-C_3^2)(1-\cos\theta)\,dC_1dC_2dC_3d\theta. \qquad (24)$$

Thus, at $\alpha\gg1$ the axis about which the body must be rotated from the initial position ($\tau=0$) to the final one ($\tau=1$) is quite randomly directed, and the rotation angle does not depend on its direction.

We consider now the case of an odd number of modes $N=2k+1$, with $T=1$. We then obtain from (13) with the aid of (24)

$$\langle m_{ii}(0)\rangle=1, \quad \langle m_{ii}(1)\rangle=0; \quad \langle m_{ii}(n)\rangle={}^1/_3, \quad n\geqslant2. \qquad (25)$$

It is easy to show, using the properties of an orthogonal matrix, that

$$\langle m_{ii}^2(1)\rangle=\langle m_{ii}(2)\rangle={}^1/_3$$

and in general

$$\langle m_{ij}^2(1)\rangle={}^1/_3.$$

This result, incidentally, follows directly from the qualitative reasoning presented above. From (13) and (24) we can obtain the values of the variances of the quantities of interest to us at the instants $\tau=n$. For example,

$$(\langle(m_{33}(n)-\langle m_{33}(n)\rangle)^2\rangle)^{1/2}=\begin{cases}1/\sqrt{3}, & n=1\\ 4/3\sqrt{5}, & n\geqslant2\end{cases}.$$

We see thus that the rotation-matrix elements fluctuate strongly.

If the number of modes is even, then $T=2$. All the results, however, remain in force also in this case, since at $\alpha\gg1$ a change in the number of the modes by unity cannot change the physical picture of the phenomenon. The same applies to all cases

$$\Delta\omega=|\omega_{01}-\omega_l|\ll 2\pi N/T_0.$$

It is important that all the foregoing results are independent of the details of the statistics of X and Y. It suffices that X and Y be independent random quantities with sufficiently broad spectra and with zero mean values.

At $\alpha\lesssim1$, as well as for $\tau\neq n$ at $\alpha\gg1$, relations (25) do not hold and exact averaging of Eqs. (16) is necessary to obtain information on the evolution. It is possible, however, to obtain the result for $\tau=n\to\infty$ directly.

Since at any $\alpha \neq 0$ the probability density for $\theta(T)$, owing to the chatter of the instantaneous rotation axis, has a finite width, it follows that

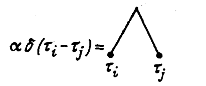
$$\lim_{n\to\infty} \langle (1-C_i^2(1)) \cos n\theta(1) \rangle = 0, \qquad \langle m_{ii}(\infty) \rangle = \langle C_i^2(1) \rangle. \tag{26}$$

The result (26) demonstrate the principal feature of the case of periodic fluctuations. Equation (26) means that at $\alpha \neq 0$ the density matrix averaged over the realizations does not tend to zero as $\tau \to \infty$, as in the usually considered case of a purely random noise.

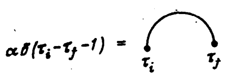
We now obtain exact averaged solutions for the diagonal elements of $M(\tau)$ at $2 \geq \tau \geq 0$. The correlation function is given by (19). To be specific, we consider

$$\langle m_{33}(\tau) \rangle = \sum_{k=0}^{\infty} (-1)^k \int_0^{\tau} \ldots \int_0^{\tau_{2k-1}} \langle R_i^k \rangle \prod_1^{2k} d\tau_i. \tag{27}$$

It is convenient to analyze the series (27) by a diagram technique (see, e.g., Refs. 12 and 13). We introduce the graphic notation

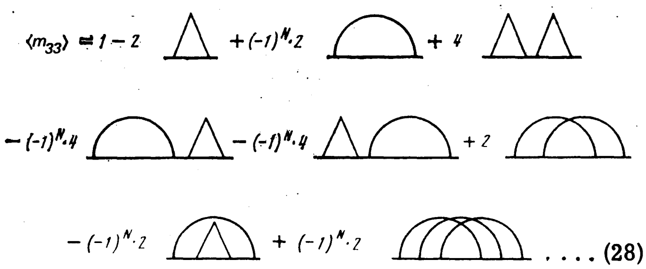


at $\tau_i \geq \tau_j$ — a corner,



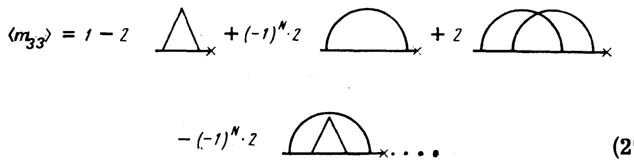
at $\tau_i \geq \tau_j$ — a loop .

A diagram of k-th order is an aggregate of k corners and loops. A diagram is called irreducible if it contains no pairs of neighboring points not connected or not bracketed by a loop



(28)

The expansion (28) can be written in the form of Dyson's equation:



(29)

The cross after the last vertex diagram in the expansion (29) means inclusion of the factor $\langle m_{33}(\tau_{2k}) \rangle$ in the integral with respect to τ_{2k}. A detailed calculation of (29) is contained in Ref. 28. We present here the end results (see Fig. 1).

At $0 \leq \tau \leq 1$ we obtain the well known result

$$\langle m_{33} \rangle = \exp\{-\alpha\tau\}. \tag{30}$$

This result is obtained with the aid of the decoupling operation,[12,16,21,29] which is valid in this case.

At $\tau > 1$, however, the decoupling operation no longer

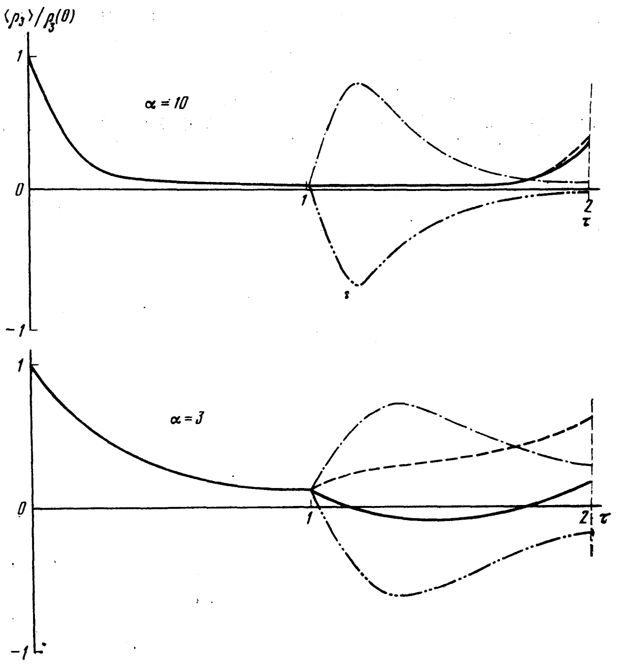


FIG. 1. Dependence of the population difference on the time in the segment $0 \leq \tau \leq 2$, $\langle m_{33}(\tau) \rangle = \langle \rho_3(\tau) \rangle / \rho_3(0)$. Solid line—exact solution in the Gaussian approximation for $N = 2k+1$. Dashed line—exact solution in the Gaussian approximation for $N = 2k$. Dash-dot—solution in the decoupling approximation for $N = 2k$. Double-dot-dash—solution in the decoupling approximation for $N = 2k+1$.

yields correct results, since an important role is assumed by the field correlations $\langle E(\tau)E^*(\tau-1) \rangle$, which are expressed in terms of diagrams with loops that bracket corners. At $1 \leq \tau \leq 2$ we obtain

$$\langle m_{33} \rangle = {}^2/_3 \exp\{\alpha - 2\alpha\tau\} + {}^1/_3 \exp\{\alpha(\tau-2)\} + (-1)^{N-1} \exp\{-\alpha/2\} [\exp\{-2\alpha(\tau-1)\} - 1]. \tag{31}$$

At $\alpha \gg 1$ it is seen from (31) that $\langle m_{33}(2) \rangle \approx 1/3$. For comparison we present the result obtained in the decoupling approximation (the loops in the diagrams do not bracket corners):

$$\langle m_{33} \rangle_{\text{dec}} = \exp\{-\alpha\tau\} + (-1)^N 2\alpha(\tau-1) \exp\{-\alpha(\tau-1)\}, \quad 1 \leq \tau \leq 2. \tag{32}$$

The results (30)–(32) are shown in Fig. 1.

In analogy with $\langle m_{33} \rangle$ we can obtain

$$\begin{aligned} &\langle m_{22} \rangle = \langle m_{11} \rangle = \exp\{-\alpha\tau/2\}, \quad 0 \leq \tau \leq 1, \\ &\langle m_{22} \rangle = \langle m_{11} \rangle = {}^1/_3 \exp\{\alpha(\tau-2)/2\} + {}^1/_6 \exp\{-\alpha(5\tau+1)/2\} \\ &\quad + (-1)^{N-1}\, {}^1/_2 \exp\{-\alpha\} [\exp\{-3\alpha(\tau-1)/2\} \\ &\quad - \exp\{\alpha(\tau-1)/2\}] + {}^1/_2 \exp\{-\alpha\tau/2\}, \quad 1 \leq \tau \leq 2. \end{aligned} \tag{33}$$

For $\tau > 2$ we can also obtain in principle, by introducing new diagrams, exact relations assuming that the noise is Gaussian. To be sure, the ensuing mathematical difficulties increase greatly. Such a detailed calculation, however, is unnecessary. At $\alpha \gg 1$, knowing $\langle m_{ii} \rangle$ on the segment (0, 2) and $\langle m_{ii}(n) \rangle$, we can understand the evolution of the two-level system. In fact, at $\alpha \gg 1$ the quantities $\langle m_{ii}(\tau) \rangle$ are exponentially small everywhere except in the vicinities of the points $\tau = n$, since the rotations corresponding to the matrices $M(n) = M^n(1)$ and $M(\tau')(0 \leq \tau' \leq 1, \tau = n + \tau', 1/\alpha \ll 1/2 - |\tau' - 1/2|)$ are practically independent. Therefore at

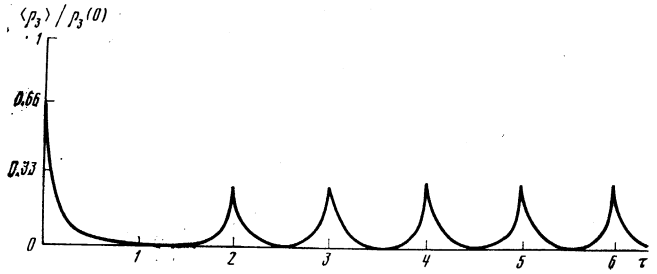


FIG. 2. Dependence of the population difference on the time (in the APF case) at $\alpha \gg 1$. A probability shift takes place.

$n \geq 2$

$$\langle M(n+\tau')\rangle \approx \langle M(\tau')\rangle \langle M^n(1)\rangle = {}^1/_3 \begin{pmatrix} \delta \\ \delta \\ \delta^2 \end{pmatrix},$$
$$\delta = \exp\{-{}^1/_4\alpha(1-|2\tau'-1|)\}.$$

On the other hand in the vicinities of the points $\tau = n$, $n \geq 2$, a shift of the probability takes place (see Fig. 2). Using (30), (31), and (32) we obtain at $\alpha \gg 1$

$$\langle C_3^2\rangle = {}^1/_3(1-\beta\exp(-\alpha/2)),$$
$$\langle C_2^2\rangle = \langle C_1^2\rangle = {}^1/_3(1+{}^1/_2\beta\exp(-\alpha/2)),\quad \beta\sim 1.$$

This makes clear the limits of applicability of the previously obtained results. The "ergodic" approximation corresponding to the distributions (23) and (24) is valid if

$$\exp\{-\alpha/2\} \ll {}^1/_3 \quad \text{or} \quad \alpha \gtrsim 5 \quad (f/\Delta\omega_0 \gtrsim {}^1/_2).$$

These inequalities determine also the region of applicability of the results (31)–(33) obtained under the assumption that the noise is Gaussian. At $\alpha < 5$ the number of radiation modes resonantly acting on the system becomes small and almost unnoticeable deviations of the probability density of the noise from the normal law [see (3)] begin to play a substantial role. We shall dwell on this in detail in Sec. 6.

On the basis of (31)–(33) we can, in particular, obtain additional information on Gaussian complex noise with a broad spectrum and with a correlation function that falls off at infinity. By only slightly modifying the diagrams, we obtain

$$\langle m_{33}(\tau_1)m_{33}(\tau_2)\rangle = {}^1/_3\exp\{-\alpha|\tau_1-\tau_2|\}$$
$$+{}^2/_3\exp\left\{-\frac{\alpha}{2}[3|\tau_1+\tau_2|-|\tau_1-\tau_2|\right\},\quad 0\leq\tau_1,\tau_2\leq 1;$$
$$\langle m_{11}(\tau_1)m_{11}(\tau_2)\rangle = \langle m_{22}(\tau_1)m_{22}(\tau_2)\rangle = {}^1/_3\exp\left\{-\frac{\alpha}{2}|\tau_1-\tau_2|\right\}$$
$$+{}^1/_2\exp\left\{-\frac{\alpha}{2}|\tau_1+\tau_2|\right\}+{}^1/_6\exp\{-{}^3/_2\alpha|\tau_1+\tau_2|+\alpha|\tau_1-\tau_2|\}.$$

We put $\lambda_{12} = \alpha\delta(\tau_1-\tau_2)$. We change to dimensional time and to the limit

$$T_0\to\infty,\quad \frac{f^2T_0}{2} = \frac{\alpha}{T_0} = \frac{4\pi d_{01}^2}{c\hbar^2}\frac{I}{\Delta\nu} = \text{const},$$

where I is the average radiation intensity and $\Delta\nu$ is the width of its spectrum. We then obtain for the population difference ρ_3 under the standard initial conditions $\rho_3(0) = 1$, $\rho_2(0) = \rho_1(0) = 0$

$$\langle\rho_3(t_1)\rho_3(t_2)\rangle = {}^1/_3\exp\{-2\gamma\Delta\nu|t_1-t_2|\}+{}^2/_3\exp\{-\gamma\Delta\nu[3|t_1+t_2|-|t_1-t_2|]\}$$

at

$$\gamma = \frac{2\pi d_{01}^2}{c\hbar^2}\frac{I}{\Delta\nu} \ll 1.$$

To our knowledge, correlators of this type have heretofore not been calculated because of the mathematical difficulties.[1)]

5. AMPLITUDE FLUCTUATIONS OF THE RADIATION

We compare now the results obtained for the amplitude-phase fluctuations (APF) with the results for the amplitude fluctuations. If we connect the phases $\{\varphi_n\}$ by the condition $\varphi_n + \varphi_{-n} = 0$, we obtain $Y = 0$,

$$\langle X\rangle = -(2\alpha)^{1/2},\quad \lambda_{12}^x = 2\alpha\left[\sum_{n=0}^{\infty}\delta(|\tau_1-\tau_2|-n)-1\right] \quad \text{at} \quad N=2k+1;$$
$$\langle X\rangle = 0,\quad \lambda_{12}^x = 2\alpha\sum_{n=0}^{\infty}(-1)^n\delta(|\tau_1-\tau_2|-n) \quad \text{at} \quad N=2k. \tag{34}$$

This is the case of amplitude fluctuations (AF) of the radiation,

$$m_{33} = m_{22} = \cos\left[\int_0^\tau X_1 d\tau_1\right],\quad m_{23} = -m_{32} = \sin\left[\int_0^\tau X_1 d\tau_1\right]. \tag{35}$$

The remaining elements of the rotation matrix are zero. Next,

$$C_3 = C_2 = 0,\quad C_1 = 1,$$
$$\theta(T) = \left|\int_0^T X_1 d\tau_1\right| = \begin{cases}(2\alpha)^{1/2} & \text{at } N=2k+1\\ 0 & \text{at } N=2k\end{cases}.$$

$\theta(T)$ is in this case not a random quantity (cf. the APF case). This singularity is a direct manifestation of the commutativity of rotations about a fixed axis.

The case of AF with λ_{12} falling off at infinity has been well studied (see, e.g., Ref. 16). If $\lambda_{12} = \langle(X_1-\langle X\rangle)(X_2-\langle X\rangle)\rangle = 2\alpha\delta(\tau_1-\tau_2)$, then

$$\langle m_{33}\rangle = \langle m_{22}\rangle = \cos[\langle X\rangle\tau]\exp\{-\alpha\tau\},$$
$$\langle m_{23}\rangle = -\langle m_{32}\rangle = \sin[\langle X\rangle\tau]\exp\{-\alpha\tau\}. \tag{36}$$

For a nonperiodic correlation function we have from (34)

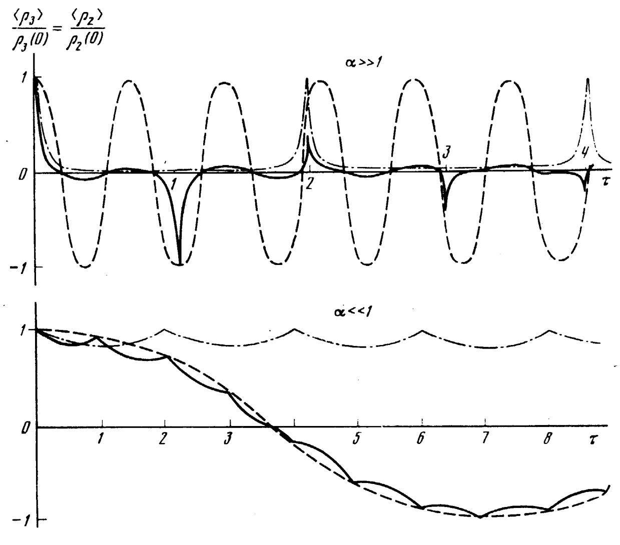


FIG. 3. Time dependences of the population difference and of the polarization (in the AF case), $\langle m_{22}(\tau)\rangle = \langle\rho_2(\tau)\rangle/\rho_2(0)$. Solid line—$N = 2k+1$. Dash-dot line—$N = 2k$.

and (35) (see Fig. 3)

$$\langle m_{33}\rangle=\langle m_{22}\rangle=\cos\left((2\alpha)^{1/2}\tau\right)\exp\{-\alpha\tau'(1-\tau')\},\quad \text{at } N=2k+1,$$
$$\langle m_{23}\rangle=-\langle m_{32}\rangle=-\sin\left((2\alpha)^{1/2}\tau\right)\exp\{-\alpha\tau'(1-\tau')\}$$
$$\tau=n+\tau',\quad 0\leqslant\tau'\leqslant 1,\quad n=0,1,2,\ldots, \tag{37}$$
$$\langle m_{33}\rangle=\langle m_{22}\rangle=\exp\{-\alpha F(\tau)\},\quad \langle m_{23}\rangle=-\langle m_{32}\rangle=0 \quad \text{at } N=2k,$$
$$F(\tau)=\begin{cases}\tau' & \text{at } n=2p\\ 1-\tau' & \text{at } n=2p+1,\end{cases}\qquad p=0,1,\ldots$$

As $T_0\to\infty$ and at constant f^2T_0, Eq. (37) goes over into (36). We see that allowance for the periodicity of λ_{12} leads also in the AF case to a substantial change of the character of the evolution. In addition, even at $\alpha\gg 1$ the results for even and odd numbers of modes differ substantially. This is one more difference between the AF and the *APF* cases. It is a manifestation of the commutativity of rotations about a fixed axis and noncommutativity of two successive rotations about two different axes.

6. CASE OF WEAK FIELD ($\alpha < 1$)

Let the field broadening f be so small that not more than one mode can act resonantly on the system. This is the case $\alpha < 1$. Inasmuch as in this case the rotation matrix changes little after a time $\tau=1$, knowledge of the averaged quantities at the instants $\tau=n$ yields practically all the information on the evolution of the system. Since the field is weak, it follows that $\Omega=\theta(1)/2$ and

$$\langle m_{ii}(n)\rangle\approx\langle C_i^2(1)\rangle+\langle(1-C_i^2(1))\rangle\langle\cos(2\Omega n)\rangle.$$

Using perturbation theory with respect to the parameter $\varepsilon=f/2\Delta\omega_0=(2\alpha)^{1/2}4\pi\ll 1$, we can find the functions Ω, $a_0(\tau)$, and $a_1(\tau)$ [see (14)] directly from the Schrödinger equation (see Ref. 28).

From (15) at $N\gg 1$ we obtain

$$\langle m_{33}(t)\rangle\approx\frac{\alpha}{12}+\left(1-\frac{\alpha}{12}\right)\cos\left[ft\left(1-\frac{\alpha}{24}\right)\right]\exp\left\{-\frac{\alpha^2}{1152}f^2t^2\right\},$$
$$\langle m_{11}\rangle=\langle m_{22}\rangle\approx\frac{1}{2}\left(1-\frac{\alpha}{12}\right) \tag{38}$$
$$+\frac{1}{2}\left(1+\frac{\alpha}{12}\right)\cos\left[ft\left(1-\frac{\alpha}{24}\right)\right]\exp\left\{-\frac{\alpha^2}{1152}f^2t^2\right\}.$$

It is easy to ascertain from (38) that at $\alpha\approx 5$ the results obtained for $\alpha\lesssim 1$ "join up" with the results for $\alpha\gg 1$.

Let us see now what the solution is if we use the decoupling operation. For $\langle m_{33}\rangle$ we get

$$\langle m_{33}\rangle_{\rm dec}=1-\int_0^\tau\int_0^{\tau_1}\lambda_{12}(\tau_1-\tau_2)\langle m_{33}(\tau_2)\rangle_{\rm dec}\,d\tau_1 d\tau_2.$$

At $N=2k+1$

$$L(\rho)=\int\langle m_{33}\rangle_{\rm dec}\,e^{-\rho\tau}d\tau=\frac{1}{\rho+\alpha+2\alpha/(e^\rho-1)}. \tag{39}$$

At $\alpha\ll 1$ we have for the poles $s_n=i\rho_n$

$$\operatorname{tg} s/2=\alpha/s,\quad s_0=\pm(2\alpha)^{1/2}(1-\alpha/12),$$
$$s_n=\pm(\pi n+\alpha/2\pi n),\quad n=1,2,\ldots .$$

Thus

$$\langle m_{33}\rangle_{\rm dec}\approx\cos(2\alpha)^{1/2}\tau+O(\alpha).$$

The decoupling operation yields therefore a solution that is not damped as $\tau\to\infty$. The damping of the true solution is due to the non-decoupling correlations that are not accounted for in (39). In this section we have not used so far the assumption that the noise is Gaussian. If the noise is assumed Gaussian also at $\alpha\ll 1$, then an analysis of the diagrams of k-th order shows that

$$\langle\Omega^{2k}\rangle_{\rm Gauss}=k!\alpha^k/2^k$$

and, for example,

$$\langle m_{33}(n)\rangle\approx F(1,1/2;-\alpha n^2/2), \tag{40}$$

where $F(1,1/2,-\alpha n^2/2)$ is a confluent hypergeometric function. The results (38) and (40) are shown for comparison in Fig. 4.

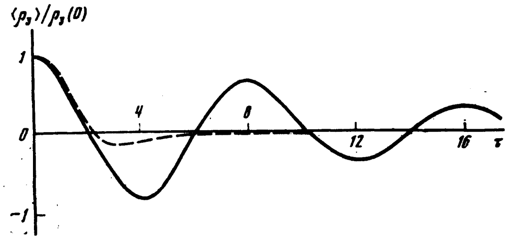


FIG. 4. Time dependence of the population difference (in the APF case) at $\alpha<1$ for $N=2k+1$. Solid line—perturbation-theory approximation in $\varepsilon=f/2\Delta\omega_0$. Dashed line—approximation of complex Gaussian noise with zero mean values.

The solution (40) attenuates much more rapidly than the exact solution (38). The error is due to the fact that in the derivation of (40) no account was taken of the deviation of the noise distribution from a normal distribution. Consider, for example, the term

$$\langle R_1^2\rangle=\langle(X_1X_2+Y_1Y_2)(X_3X_4+Y_3Y_4)\rangle=4\lambda_{12}\lambda_{34}+2\lambda_{13}\lambda_{24}+2\lambda_{14}\lambda_{23}$$
$$-2\alpha\lambda(\tau_1-\tau_2+\tau_3-\tau_4)-2\alpha\lambda(\tau_1-\tau_2-\tau_3+\tau_4). \tag{41}$$

The last two terms of (41) are not Gaussian. If $\alpha\gg 1$, then upon integration in (27) the total contribution of the non-Gaussian terms is small and inessential. If, however, $\alpha<1$, it can be shown that the contribution of the last two terms in (41) compensates for the contributions of the second and third terms, which correspond to nondecoupled correlations. Thus, over a sufficiently long time interval the decoupling operation is applicable and the results obtained with it can be used at $\alpha\lesssim 1$ for not too long times (see Fig. 5). In terms of higher order than (41), the non-Gaussian correlations no longer cancel completely the Gaussian nondecoupled correlations,

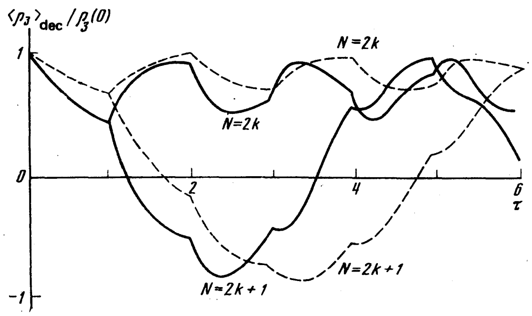


FIG. 5. Time dependence of the population difference, obtained with the aid of the decoupling operation. Solid line—$\alpha=1$, dashed—$\alpha=0.5$.

and it is this which causes the solution to attenuate as $t \to \infty$.

7. CONCLUSION

Thus, in the approximation of a broad radiation spectrum we have two essentially different regimes, $\alpha \gg 1$ and $\alpha < 1$. As already indicated, the parameter α has a clear-cut physical meaning. Namely, $(1 + \alpha/\pi^2)$ is the number of radiation modes that act resonantly on the two-level system.

In the case of a strong field, $\alpha \gg 1$, the number of resonant radiation modes is large and we can use the approximation of a complex Gaussian noise with zero mean values. At the instant of time $t = nT_0$, $n \geq 2$, a shift of the probability takes place in the system for the density-matrix elements ρ_1, ρ_2, ρ_3. At these instants they taken on values substantially different from zero. The probability shift is a direct manifestation that the system "remembers" the periodic structure of the noise (1). We discuss now some possibilities of observing the probability shift.

Assume that noise radiation from a powerful laser and radiation from a weak laser are incident from two different directions on a cell with gas. If the carrier frequencies of both lasers are equal to the frequency of the transition $|0\rangle \to |1\rangle$ of the particles of the irradiated gas, then the noise field causes a shift of the probability for the populations, which could be revealed by the absorption of the radiation of the weak laser operating in the spike regime. If the spike repetition time coincides with the time T_0 and the two lasers are turned on synchronously, then absorption of the weak radiation is observed, and if they are not turned on synchronously, the average weak radiation will be zero, since

$$\langle W \rangle_w \sim \langle \rho_3 \rangle,$$

where W_w is the average absorbed power of the weak radiation.

In addition, the shift of the probability should lead to a strong modulation of the contour of the absorption line of the weak signal that acts near the resonance of the $|0\rangle \to |1\rangle$ transition ($|0\rangle$ is the ground state) of a three-level system in the presence of the strong noise radiation (1) that is at resonance with the frequency of the transition between the upper level (see, e.g., Refs. 11–13). For weak-signal absorption line shape we have

$$\operatorname{Im} \chi(\Delta\omega) \sim \int_0^\infty \cos(\Delta\omega\tau) \langle G(\tau) \rangle d\tau, \qquad (42)$$

where $\chi(\Delta\omega)$ is the linear susceptibility of the three-level system; it can be shown that at $\alpha \gg 1$ a phenomenon similar to the probability shift takes place for $\langle G(\tau) \rangle$. This should lead to a strong modulation, with frequency $\Delta\nu_0$, of the absorption line shape (42).

In the case of a weak field $\alpha < 1$, the approximation of a complex Gaussian field with zero mean values is no longer valid, since the quantum system "perceives" the field acting on it as a Gaussian noise only if the number of resonant modes is large enough. In a weak field this criterion is no longer satisfied and the system is effectively acted upon by only the mode whose frequency coincides with ω_{01}. The role of the remaining modes reduces only to a slow damping of the precession. As $t \to \infty$, the populations of the levels become equal. It is usually assumed that large population inversion can be obtained in a two-level system with a stronger field. This is the situation, for example, in the case of fluctuations of the radiation frequency.[9,10] In this case, however, the situation is just the reverse. At $a \ll N$, with decreasing radiation intensity, the "acting" field becomes more and more monochromatic and an ever increasing population of the upper level is obtained, albeit after a long time. This variety of possibilities emphasizes once more the need for taking a detailed account of the nonmonochromaticity of the radiation in the analysis of concrete experiments.

In conclusion, the author thanks V. L. Ginzburg and V. N. Sazonov for helpful advice and interest, and V. A. Namiot and A. V. Masalov for numerous stimulating discussions.

[1)] In. Ref. 29 was approximately calculated the correlator in the limiting case $t_{1,2}\Gamma \gg 1$, where Γ is the reciprocal time of relaxation in the system.

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