A two-level system in a random field

P. V. Elyutin

Moscow State University
(Submitted March 25, 1975)
Zh. Eksp. Teor. Fiz. 69, 1156–1162 (October 1975)

The behavior of the population difference of a two-level system in a strong quasimonochromatic random field is considered. It is shown that the fluctuations of the field lead to a damping of the oscillation of the population difference. In the case when the amplitude of the field is a stationary normal process the effect of the lack of coherence can be taken into account by introducing an effective relaxation time. In the case of a field of constant amplitude, the fluctuations in phase bring about in addition to damping also a change in the frequency of oscillation. The damping rate in this case is not directly related to the field spectrum.

PACS numbers: 42.50.+q

An investigation of the interaction of a two-level system with a strong field is of interest for a wide circle of problems. As an example we refer to the theory of propagation of short light pulses in a resonant medium. An investigation of the problem in which both the amplitude and the phase of the field are deterministic functions of the time has been carried out in sufficient detail (references concerning this problem are given in Kaplan's papers[1,2]). However, in a number of cases, including the example mentioned above, the external field is not always coherent. The coherence time of the field can be comparable to the characteristic time for the system: the period of oscillation of the populations or the relaxation times. Naturally the evolution of the system in a noncoherent field will be determined not only by the characteristics of the system and the average value of the field, but will also depend on the coherence properties of the field. It is therefore of interest to investigate the interaction of a two-level system with a strong noncoherent field and to determine the influence of the field parameters on the behavior of the population difference of a two-level system in those cases when the amplitude or the phase of the external field is a random function.

1. The density matrix for a two-level system σ_{ik} which is required to be Hermitean and to satisfy the condition $\mathrm{Tr}\sigma = 1$ is uniquely determined if three real parameters are given. As such parameters we choose the difference in the diagonal elements (populations)

$$y=\sigma_{22}-\sigma_{11}, \quad (1)$$

the value of x which is twice the modulus and the phase ψ of an off-diagonal element:

$$\sigma_{12}=(x/2)e^{i\psi}. \quad (2)$$

In future we chall choose the phase ψ to lie in the interval $(-\pi/2, \pi/2)$, and we shall record the quantity x together with its sign. The equations of motion for the components of the density matrix of the system coupled to an external quasimonochromatic field

$$V(t)=E(t)\cos(\Omega t+\varphi(t)), \quad (3)$$

where E(t) and $\varphi(t)$ are the amplitude and the phase, which vary slowly compared with Ω, and are well known. In the case of exact resonance they are of the form[3,4]

$$x'+\alpha x=\omega(t)y\sin\theta, \quad (4)$$

$$y'+\beta(y-y_0)=-\omega(t)x\sin\theta, \quad (5)$$

$$\psi' x=\omega(t)y\cos\theta, \quad (6)$$

where the notation $\theta = \Psi - \varphi$ has been introduced for the difference in the phases of the field and of the off-diagonal element. Here y_0 is the equilibrium value of the population difference,

$$\alpha=T_2^{-1}, \quad \beta=T_1^{-1} \quad (7)$$

are the reciprocals of the transverse and longitudinal relaxation times. The Rabi frequency is given by

$$\omega(t)=\mu\hbar^{-1}E(t), \quad (8)$$

where μ is the matrix element for the transition.

We consider a monochromatic field (the amplitude and the phase are constant). The dependence of the population difference on the time is determined by the following formula

$$y=\exp\left\{-\frac{\alpha+\beta}{2}t\right\}(A\sin at+B\cos at)+\frac{\alpha\beta}{\omega^2+\alpha\beta}y_0, \quad (9)$$

$$a=\left[\omega^2-\frac{(\alpha-\beta)^2}{4}\right]^{1/2}. \quad (10)$$

The solution (9) depends only on the two constants which are determined by the initial values of the variables y and $x\sin\theta$, and does not depend on the value of the integral of the system:

$$Z=x\cos\theta e^{\alpha t}. \quad (11)$$

The dependence of the phase on the time has the form

$$\mathrm{tg}\,\theta=Z^{-1}\left\{e^{(\alpha-\beta)t/2}(P\sin at+Q\cos at)+\frac{\omega\beta y_0}{\omega^2+\alpha\beta}e^{\alpha t}\right\}, \quad (12)$$

where the constants P,Q and Z are determined by the initial values of all three variables appearing in the system (4)–(6). We note that in the case of arbitrary initial conditions ($P, Q, y_0 \sim 1$) the phase begins to be established in the practically important case $\alpha \gg \beta$ at the time when the second term in (12) becomes the dominant one. This occurs at times much greater than τ:

$$\tau=\frac{1}{\alpha}\ln\frac{\omega^2+\alpha\beta}{\omega\beta}. \quad (13)$$

The time for the establishment of the phase τ in order of magnitude is equal to the transverse relaxation time ($T_2 \approx 10^{-10}-10^{-11}$ sec for atoms in solids) and depends only weakly on the Rabi frequency, and therefore on the magnitude of the field. The assumption of a rapid establishment of phase[3] is not always valid in problems concerning the interaction of a two-level system with a field. The fact that the interaction with a coherent field at small values of the time is not coordinated in phase can result in a deformation of the leading front of the pulse propagating in a resonant medium.

2. We consider the change in the population difference of a two-level system being acted upon by a resonant

quasimonochromatic field whose amplitude E(t) represents a random stationary normal process with a known correlation function. Such a model is applicable to the description of a field with large amplitude fluctuations—spontaneous radiation of thermal or gas discharge sources and radiation from superluminescent or multimode lasers.

The solution of the system (4)–(6) in the case of an arbitrary form of dependence of the amplitude on the time is possible only when the relaxation times are equal to each other: $\alpha = \beta$. With the initial conditions

$$y(0)=y_0=1,\quad Z=0 \tag{14}$$

it is of the form [1]

$$y=e^{-\alpha t}\cos\xi(t)+\alpha e^{-\alpha t}\left[\sin\xi(t)\int_0^t e^{\alpha\tau}\sin\xi(\tau)\,d\tau+\cos\xi(t)\int_0^t e^{\alpha\tau}\cos\xi(\tau)\,d\tau\right]. \tag{15}$$

Here we have introduced the notation $\xi(t)$ for the dimensionless quantity:

$$\xi(t)=\mu\hbar^{-1}\int_0^t E(\tau)\,d\tau. \tag{16}$$

The quantity $\xi(t)$ is also distributed according to the normal law with the dispersion given by D(t) the expression for which depends on the form of the correlation function for the field E(t). For the sake of definiteness we consider an exponentially correlated process:

$$\begin{gathered}\mu\hbar^{-1}\langle E(t)\rangle=b,\\ \mu^2\hbar^{-2}\langle E(t+\tau)E(t)\rangle=d^2\exp\{-\eta|\tau|\}+b^2.\end{gathered} \tag{17}$$

The quantities b and d defined in such a manner have the dimensions of reciprocal time. The quantity η can be interpreted as the reciprocal of the coherence time of the field. Under the assumptions (17) the dispersion of the process $\xi(t)$ will be equal to ([5], Sec. 33)

$$D(t)=2d^2\eta^{-2}[\eta t-1+\exp\{-\eta t\}]. \tag{18}$$

The value of the average population difference will be determined by averaging the general solution (15) over the distribution. It is of the form

$$\bar{y}(t)=\cos\bar{\xi}(t)\exp\left\{-\alpha t-\frac{D(t)}{2}\right\}+\alpha\int_0^t dz\cos\bar{\xi}(z)\exp\left\{-\alpha z-\frac{D(z)}{2}\right\}. \tag{19}$$

From formula (18) it can be seen that for times large compared to the coherence time η^{-1}, the dispersion D grows linearly. In this case an approximate evaluation of the integral in (19) is possible:

$$\bar{y}(t)\approx e^{-\zeta t}\left\{\cos bt\left(1-\frac{\alpha\zeta}{b^2+\zeta^2}\right)+\frac{\alpha b}{b^2+\zeta^2}\sin bt\right\}+\frac{\alpha\zeta}{b^2+\zeta^2}, \tag{20}$$

$$\zeta=\alpha+d^2/\eta. \tag{21}$$

From a comparison with (9) it can be seen that the effect of the amplitude fluctuations on the behavior of the population difference reduces to the appearance of an additional relaxation proportional to the intensity of the noise component of the field. The relaxation introduced by the field can be compared with the relaxation characteristic of the system itself. Thus, for transitions between the ground state and a metastable state the relaxation of which is due to spontaneous radiation ($\mu^2\hbar^{-2}\approx 10^{14}$ g^{-1} cm $\alpha\approx 10^3$ sec^{-1}), with coherence times $\eta^{-1}\approx 10^{-9}$ sec typical for spontaneous radiation the relaxation being introduced becomes comparable to that characterizing the system when the average intensity of the noise field is of the order of magnitude of several W · cm^{-2}.

We consider the special case of a system without relaxation and a field with an average amplitude equal to zero. Then for small times and a weak field (for $D(t)\ll 1$) we have

$$\bar{y}(t)\approx 1-D(t)/2. \tag{22}$$

The probability of transition

$$W\approx D(t)/4 \tag{23}$$

depends quadratically on the time for small t. For $t\gtrsim\eta^{-1}$ the dependence on the time becomes linear, and formula (23) reduces to the Fermi golden rule for transitions in a discrete spectrum under the action of broadband radiation, since the value of $d^2\eta^{-1}$ is proportional to the spectral intensity of the field at the frequency of the transition.

3. We consider the variation of the population difference of a two-level system without relaxation coupled to an external field of constant amplitude and with a random phase

$$V(t)=E\cos(\Omega t+\varphi(t)). \tag{24}$$

Such a model can be applied to the description of the radiation field from single mode lasers. We denote the deviation of the frequency φ' by f and go over to dimensionless variables having chosen for the scale of reciprocal time the Rabi frequency $\omega=\mu\hbar^{-1}E$. Then the system (4)–(6) can be reduced to a third order differential equation

$$y'''-y''\frac{f'}{f}+y'(1+f^2)-y\frac{f'}{f}=0 \tag{25}$$

or to the integro-differential equation which is equivalent to it

$$y''+y=-f(t)\int_0^t y'(\tau)f(\tau)\,d\tau+\frac{y''(0)+y(0)}{f(0)}f(t). \tag{26}$$

We seek solutions of (26) in the form of iteration series, whose zero order terms are solutions of (26) without the right-hand side:

$$A_0(t)=A\cos t,\quad B_0(t)=B\sin t. \tag{27}$$

Since (26) is linear the iterated solution can be carried out independently for each of the functions A(t), B(t). The solution obtained from the first function is of the form

$$A(t)=A\cos t\left[1+\hat{p}\sum_{n=0}^{\infty}(-1)^n(\hat{p}^2+\hat{q}^2)^n\hat{p}\right]-A\sin t\left[\hat{q}\sum_{n=0}^{\infty}(-1)^n(\hat{p}^2+\hat{q}^2)^n\hat{p}\right], \tag{28}$$

where $\hat{p}$ and $\hat{q}$ are operators defined by the relations

$$\hat{p}g(t)=\int_0^t f(x)\sin(x)g(x)\,dx,\quad \hat{p}=\hat{p}\cdot 1, \tag{29}$$

$$\hat{q}g(t)=\int_0^t f(x)\cos(x)g(x)\,dx,\quad \hat{q}=\hat{q}\cdot 1. \tag{30}$$

The solution which is obtained from the second function has a somewhat more complicated structure:

$$\begin{aligned}B(t)=B\sin t&\left[1+\hat{q}\sum_{n=0}^{\infty}(-1)^n\prod_{m=0}^{n}(\hat{p}^2+(-1)^m\hat{q}^2)^{(1-\delta_{m0})}\hat{q}\right]\\ -B\cos t&\left[\hat{p}\sum_{n=0}^{\infty}(-1)^n\prod_{m=0}^{n}(\hat{p}^2+(-1)^m\hat{q}^2)^{(1-\delta_{m0})}\hat{q}\right],\end{aligned} \tag{31}$$

where δ_{m0} in the power index is the Kronecker symbol.

It can be easily seen that the functions A(t) and B(t) are exact solutions of (25). In future we shall restrict ourselves to an investigation of the solution A(t) which satisfies the initial conditions

$$A(0)=A,\quad A'(0)=0. \tag{32}$$

We assume that the deviation of the frequency f(t) is a random stationary normal process with an average value equal to zero and with the correlation function

$$C(\tau)=\delta^2\exp(-\gamma|\tau|). \tag{33}$$

We average expression (28) over the distribution. From the structure of the iterated solution it is clear that the n-th term of the expansion represents a sum of 2^n 2-fold integrals of products of functions of an ordered sequence of arguments. As a result of averaging terms of the form $\langle f(x_1)\ldots f(x_{2n})\rangle$ will appear in the integrand together with trigonometric functions. Such averages in the case of a normal stationary process can be expressed in terms of pair correlation functions

$$F_{2n}=\langle f(x_1)\ldots f(x_{2n})\rangle=\sum\prod_{i=1}^{n}C_i(x_j,x_k), \tag{34}$$

where on the right hand side there appears the sum of $(2n-1)!!$ terms corresponding to possible decompositions of 2n arguments into pairs. We note that the iteration series corresponding to the third linearly independent solution of (25), the zero order term of which is the solution of (26) without the integral term on the right hand side, can be expressed in terms of products of functions $f(x_i)$ of an odd number of arguments. Since we have set the average deviation of the frequency to be equal to zero then for arbitrary initial conditions such an iteration series will vanish when averaged over the distribution.

It is convenient to represent the different terms in (34) by numbering the initial arguments of pairs in decreasing order and by denoting by the same numbers the final arguments of the corresponding pairs. For example,

$$F_2=(1.\ 1), \tag{35}$$

$$F_4=(1.\ 1.\ 2.\ 2)+(1.\ 2.\ 1.\ 2)+(1.\ 2.\ 2.\ 1). \tag{36}$$

The use of the correlation function (33) enables one to evaluate in an elementary manner all the integrals that are encountered. We examine the different terms corresponding to F_2:

$$\begin{aligned}
\overline{p^2}&=\varepsilon\left[\frac{\gamma}{2}(t-\sin t\cos t)-\frac{1}{2}\sin^2 t+\frac{1}{1+\gamma^2}\{1-(\gamma\sin t+\cos t)e^{-\gamma t}\}\right],\\
\overline{pq}&=\varepsilon\left[\frac{1}{2}(t-\sin t\cos t)+\frac{\gamma}{2}\sin^2 t-\frac{1}{1+\gamma^2}\{1-(\gamma\sin t+\cos t)e^{-\gamma t}\}\right],\\
\overline{qp}&=\varepsilon\left[-\frac{1}{2}(t+\sin t\cos t)+\frac{\gamma}{2}\sin^2 t+\frac{1}{1+\gamma^2}\{\gamma-(\gamma\cos t-\sin t)e^{-\gamma t}\}\right],\\
\overline{q^2}&=\varepsilon\left[\frac{\gamma}{2}(t+\sin t\cos t)+\frac{1}{2}\sin^2 t-\frac{1}{1+\gamma^2}\{\gamma-(\gamma\cos t-\sin t)e^{-\gamma t}\}\right].
\end{aligned} \tag{37}$$

Here we have introduced the notation

$$\varepsilon=\delta^2/(1+\gamma^2). \tag{38}$$

The averages of more complicated combinations of the operators $\hat{p}$ and $\hat{q}$ also have an analogous structure. We note the presence of terms of three types: those increasing with an increase in t (power terms), those bounded for any value of t (trigonometric and constant terms) and those exponentially decreasing with increasing t. Considering the case $t\gg 1$ we restrict ourselves to taking into account only the power terms.

Elementary but rather awkward calculations show the following. All terms of order 2^n contain the factor ϵ^n. In the term of order 2n the contribution of the n-th power yields only a term in which the neighbouring arguments are paired:

$$F_{2n}{}'=(1,1,2,2,\ldots,n,n). \tag{39}$$

All the other terms give a contribution with a lower power of t. The coefficient in front of $t^n/n!$ is a polynomial in γ. Regrouping the power terms, one can represent the coefficients in front of $A\cos t$ in the form

$$1+g_1(\varepsilon,\gamma)\frac{\varepsilon t}{2}+g_2(\varepsilon,\gamma)\frac{1}{2!}\left(\frac{\varepsilon t}{2}\right)^2+\ldots, \tag{40}$$

$$g_n(\varepsilon,\gamma)=(-1)^n\operatorname{Re}(\gamma+i)^n+\varepsilon G_n^{(2)}(\gamma)+\varepsilon^2G_n^{(3)}(\gamma)+\ldots \tag{41}$$

$G_n^{(i)}$ is a rational function of γ without any singularities for real values of γ.

Assuming the inequality $\epsilon\ll 1$ to be valid, we shall restrict ourselves in each of the coefficients in g_n to the term of zero order in ϵ. To justify such an approximation we must show that the expansion (41) does not diverge for any finite value of ϵ. Although the result should not depend strongly on the specific form of the correlation function, with our choice of $C(\tau)$ the investigation of the functions $g_n(\epsilon,\gamma)$ is considerably simplified. The magnitude of the contribution of a given term in F_{2n} for each of the products $p^kq^mp^n\ldots$ is uniquely determined by the placing of the initial arguments of pairs in the sequence of 2n arguments and does not depend on the position of the final arguments. For example, in the fourth order we have

$$(1,2,1,2)=(1,2,2,1). \tag{42}$$

Instead of $(2n-1)!!$ terms we shall have to deal with a significantly smaller number of classes of equivalent terms. As a representative of each class we choose the decomposition with nonintersecting pairings. It is important to note that the number of terms K in each class increases with an increase in the length of pairings: From K = 1 for the leading term Fl_{2n} to K = n! for the term with the imbedded pairings

$$F_{2n}^{(m)}=(1,2,\ldots,n,n,\ldots,2,1). \tag{43}$$

As an example we give the expansion of the term of the sixth order:

$$\begin{aligned}F_6=&(1,1,2,2,3,3)+2(1,2,2,1,3,3)\\&+2(1,1,2,3,3,2)+4(1,2,2,3,3,1)+6(1,2,3,3,2,1).\end{aligned} \tag{44}$$

In calculating the values of the terms with long pairings compensation of large numerical coefficients occurs by factors of the type of n^{-1}, $(n^2+m^2\gamma^2)^{-1}$ which appear when exponentials with large exponents are integrated. As the number of embedded pairings increases the index of the power in the principal increasing term also decreases.

The summation of the terms of zero order in formula (40) and in the analogous relationship for the coefficient in front of A sin t is not complicated. Returning to dimensional variables we arrive at the following expression for the average population difference:

$$\bar{y}(t)=A\exp\left\{-\frac{\delta^2}{\omega^2+\gamma^2}\gamma\frac{t}{2}\right\}\cos\left[\omega t\left(1+\frac{\delta^2}{2(\omega^2+\gamma^2)}\right)\right]. \tag{45}$$

Thus, in an external field of constant amplitude with a fluctuating phase the population difference relaxes with time towards zero value. Evidently, in contrast to the case considered in Sec. 2, the effect of the fluctuations of the phase cannot be taken into account by introducing effective relaxation constants in Eqs. (4)–(6) since the frequency of oscillations is not smaller than the Rabi frequency in contrast to a.

We consider different limiting cases of formula (45). In the case of large and slow reductions in frequency ($\delta \gg \delta$, technical line shape) the condition for the applicability of iterational calculations

$$\delta^2 \ll \gamma^2 + \omega^2 \tag{46}$$

forces us to restrict ourselves to the case of strong field ($\omega^2 \gg \delta^2$). The rate of effective relaxation

$$\alpha_S \approx \delta^2\gamma/2\omega^2 \tag{47}$$

turns out to be quite small. For typical values of $\delta \sim 10^3$ sec^{-1} the condition (46) leads to the estimate $\alpha_S \lesssim 10^1$ sec^{-1}. It is of interest to note that the spectrum of the field in this case depends only on δ, but not on γ([5], Sec. 37), and a direct coupling between the spectrum of the field acting on the system and the relaxation in the oscillation of the populations is absent. In the limit $\gamma \to 0$ formula (45) goes over into

$$\bar{y}(t) = A \cos[\omega t(1 + \delta^2/2\omega^2)]. \tag{48}$$

In this expression it is not difficult to regocnize an expansion in terms of the small detuning of the exact solution of (25) in the case $f = \delta = \text{const}$:

$$y(t) = A \cos\sqrt{\omega^2 + \delta^2}\, t. \tag{49}$$

In the opposite limiting case of small and rapid reductions of frequency ($\delta \ll \gamma$, natural line shape), Eq. (46) does not impose any restrictions on the value of ω. The relaxation introduced by the fluctuations of phase is greatest in relatively weak fields, for $\omega \lesssim \gamma$. We then have

$$\bar{y}(t) = A \exp\left\{-\frac{\delta^2}{2\gamma} t\right\} \cos\omega t. \tag{50}$$

The rate of damping in this case is equal to one half of the width Δ of the Lorentz line

$$\alpha_F = \delta^2/2\gamma = \Delta/2. \tag{51}$$

In the case of a sufficiently great line width (for example, in the case of a semiconductor lasers Δ can attain the value of 10^4 sec^{-1}) the rate of relaxation α_F can exceed the rates of relaxation characteristic of metastable levels.

The author expresses his thanks to V. I. Grigor'ev, L. V. Keldysh and R. V. Khokhlov for useful discussions.

[1]A. E. Kaplan, Zh. Eksp. Teor. Fiz. 65, 1416 (1973) [Sov. Phys.-JETP 38, 705 (1974)].
[2]A. E. Kaplan, Zh. Eksp. Teor. Fiz. 68, 823 (1975) [Sov. Phys.-JETP 41, 409 (1975)].
[3]T. M. Il'inova and R. V. Khokhlov, Izv. vuzov, Radiofizika 8, 899 (1965).
[4]P. G. Kryukov and V. S. Letokhov, Usp. Fiz. Nauk 99, 169 (1969) [Sov. Phys.-Usp 12, 641 (1970)].
[5]S. M. Rytov, Vvedenie v statisticheskuyu radiofiziku (Introduction to Statistical Radiophysics), Nauka, 1966.

Translated by G. Volkoff
126