Exact theory of relaxation of two-level systems in a strong nonmonochromatic field

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An equation of motion is derived for the population of two-level systems in a strong field with amplitude and frequency that vary arbitrarily with time. Exact solutions of the equation for an arbitrary amplitude envelope of the field are obtained in the case of equal relaxation times when the field and transition frequencies are identical. Exact solutions are found for a number of envelopes of particular shape in the case of unequal times.

INTRODUCTION

The interaction of a two-level system with a monochromatic resonant field has by now been investigated quite fully. Among the problems studied are saturation of strong-field absorption in the stationary regime[1] 1), periodic oscillations of the transition probabilities (in systems without relaxation) with frequency determined by the field amplitude[3], the interaction of a relaxing system with a weak field in the presence of a strong monochromatic field[4–6], nonlinear polarizability of matter in resonant absorption[7,8] and the self-focusing associated with it[9], nonlinear susceptibility in an inhomogeneously-broadened line[10], etc.

There have been much fewer investigations of the interaction of a two-level system with a strong monochromatic field, the amplitude and frequency of which vary arbitrarily with time. At the same time, this is a timely question for many problems, e.g., the interaction of ultrashort pulses with a nonlinear resonant absorber or amplifier in a laser, the change in the line shape in the field of strong spontaneous emission, the theory of nonlinear susceptibility due to resonant nonstationary absorption and of the corresponding self-action of the light, the measurement of the relaxation characteristics of transitions of the nonstationary-field parameters, etc. With rare exceptions, there are no exact results in this region. From among the exact results we can point to the solutions (see[11,12]) that describe the behavior of the populations of an idealized two-level system without relaxation, in the presence of a field that is arbitrarily amplitude-modulated at exact frequency resonance. In essence, a field of this type is also one of the results[4] in which relaxation (with equal times) is taken into account, but zero population in the system is assumed in the absence of the field. Equations for the density matrix in the case of a nonmonochromatic field were apparently obtained for the first time in[13,14]. Investigations of the behavior of quantum systems in strong spontaneous fields with a wide spectrum have been recently initiated, both experimental[15], including self-action effects[16], and theoretical[17–19]. In[17], in particular, they consider an "empty" system (there is no population for the working transition) characterized only by a transverse relaxation time, and the time variation of the field is simulated by segments of an oscillation whose parameters (amplitude, frequency, phase) remain constant inside each segment and vary from segment to segment; the results do not make it possible to describe the population relaxation. Attempt to construct a solution with the aid of perturbation theory[20] or its modifications[18] lead to iteration series that converge only at field powers lower than a certain critical value.

The purpose of the present paper is the development of an exact theory of the behavior of a relaxing two-level system with nonzero equilibrium population in a strong monochromatic field, the magnitude and rate of whose parameters (amplitudes and frequencies) can be arbitrary in comparison with the relaxation times. It is shown (Sec. 1) that from the known equations for the density matrix it is possible to obtain, for the population difference, by "truncation," a third-order differential equation with variable coefficients that are determined by the instantaneous values of the frequency and amplitude of the field. In most cases of physical interest it is possible to obtain a solution for this equation.

In particular, at exact frequency resonance and at an arbitrary field-envelope amplitude, the obtained equation reduces to a second order equation, the exact solution of which for different types of envelopes are given in Sec. 2 (case of equal relaxation times) and in Sec. 3 (case of unequal times).

1. EQUATION OF MOTION

1. The kinetic equations for the density-matrix elements σ_{ik} of a two-level system having molecules with a dipole moment and interacting with an electric field $\mathbf{E}(t)$ can be written in the form (see, e.g.,[11])

$$\begin{gathered}\dot{n}+(n-n_0)/\tau=2i\mathbf{d}\mathbf{E}(t)(\sigma_{21}-\sigma_{12})/\hbar,\\ \dot{\sigma}_{21}+(i\omega_0+1/T)\sigma_{21}=i\mathbf{d}\mathbf{E}(t)n/\hbar,\\ n=\sigma_{11}-\sigma_{22},\quad \sigma_{12}=\sigma_{21}^*,\quad \sigma_{11}+\sigma_{22}=1,\end{gathered}\tag{1.1}$$

where n is the population difference between the ground and excited levels (per molecule), n_0 is the equilibrium population difference at a given temperature in the absence of the field, T and τ are respectively the transverse and longitudinal relaxation times, and ω_0 is the transition probability. Equations (1.1) were obtained under the usual assumption that the interaction is weak:

$$|V/\hbar|\ll\omega_0,\quad V(t)=-\mathbf{d}\mathbf{E}(t),\tag{1.2}$$

where V(t) is the energy of the interaction of the molecule with the field. It is assumed, in addition, that the molecules and the field are linearly polarized. The field $\mathbf{E}(t)$ is assumed to be a given function of the time. We are interested here only in the case of a quasiresonant field, i.e., we assume that the field frequency $\omega(t)$ and the width of the spectrum $\Delta\omega$ satisfy the following conditions (besides the condition that the system be narrowband):

$$|\omega(t)-\omega_0|\ll\omega_0,\quad \Delta\omega\ll\omega_0,\quad 1/T,\ 1/\tau\ll\omega_0\tag{1.3}$$

(no limitations are imposed, however, on the ratio of the spectrum width $\Delta\omega$ to the width 1/T of the resonance line

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of the medium). The field can therefore be represented in the form

$$E(t)=R(t)\cos[\omega_0 t+\varphi(t)],\quad \dot{\varphi}=\nu(t)=\omega(t)-\omega_0,$$

where, by virtue of the condition (1.3), the amplitude R(t) and the instantaneous frequency detuning $\nu(t)$ are slow functions of the time in comparison with cos $\omega_0 t$.

2. We derive the equation of motion for the population difference n(t). We first renormalize Eq. (1.1), putting

$$x(t)=n(t)/n_0,\quad y=2i\sigma_{21}(t)e^{i\omega_0 t}/n_0$$
$$A=dE(t)/\hbar=-V/\hbar,\quad r=dR/\hbar,\quad a=re^{i\varphi}. \tag{1.4}$$

The relative field A and its amplitude r have now the dimension of frequency. This has a direct physical meaning; for example, in definite cases r(t) is exactly equal to the frequency of the population oscillations (see Sec. 2). We have introduced also the "slow" function y instead of the high-frequency off-diagonal element σ_{21}. Representing now A(t) in the form $A(t) = \frac{1}{2}[a(t)e^{i\omega_0 t} + \text{c.c.}]$ (a(t) is a "slow" function by virtue of condition (1.3), and expressing (1.1) in terms of the notation in (1.4), we follow the usual "truncation" procedure, i.e., we retain in the equations only the slow terms and discard all the rapidly-varying ones; this is possible because of the conditions (1.2) and (1.3). As a result we obtain the following system of equations:

$$\hat{D}_\tau(x-1)={}^1/_2(ay+a^*y^*), \tag{1.5}$$
$$\hat{D}_T(y)=-xa^*, \tag{1.6}$$

where $\hat{D}_\tau$ and $\hat{D}_T$ are linear differential operators:

$$\hat{D}_\tau=\frac{d}{dt}+\delta_\tau,\quad \hat{D}_T=\frac{d}{dt}+\delta_T\quad\left(\delta_\tau=\frac{1}{\tau},\ \delta_T=\frac{1}{T}\right).$$

An equation for x(t) only can now be obtained, for example, in the following manner. Applying the operator D_T to both sides of (1.5) and transforming then the right-hand side with allowance for Eq. (1.6) and for the definition (1.4) of a, we get

$$\hat{D}_T[\hat{D}_\tau(x-1)]={}^1/_2(\dot{a}y+\dot{a}^*y^*)-r^2x. \tag{1.7}$$

Performing the same operation on (1.7), we obtain

$$\hat{D}_T^2[\hat{D}_\tau(x-1)]={}^1/_2(\ddot{a}y+\ddot{a}^*y^*)-r\dot{r}x-\hat{D}_T(r^2x) \tag{1.8}$$

Eliminating now y and y* from the system of equations (1.5), (1.7) and (1.8), we obtain an equation for x(t) only, whence, with allowance for the definition (1.4) of a, we obtain ultimately after a number of transformations

$$\frac{r}{\nu}\hat{D}_T\left\{\frac{r}{\nu}\left[\frac{1}{r}\hat{D}_T\left(\frac{1}{r}\hat{D}_\tau(x-1)\right)+x\right]\right\}+\hat{D}_\tau(x-1)=0. \tag{1.9}$$

This is a linear inhomogeneous third-order equation with variable coefficients, but in individual cases it can degenerate into an equation of lower order. For example, in the absence of a field ($r \equiv 0$), Eq. (1.9) reduces to a first-order equation:

$$\hat{D}_\tau(x-1)=0. \tag{1.10}$$

3. An important particular case that lends itself to a most detailed investigation is that of an arbitrary envelope of the field envelope at exact frequency resonance ($\nu \equiv 0$). Then (1.9) reduces to the second-order equation

$$\frac{1}{r(t)}\hat{D}_T\left[\frac{1}{r(t)}\hat{D}_\tau(x-1)\right]+x=0. \tag{1.11}$$

The initial conditions for (1.11), corresponding to the fact that at t_0, the instant when the field is turned on, the population has the equilibrium value ($n(t_0) = n_0$) and the polarization is equal to zero ($\sigma_{21} = \sigma_{12} = 0$), which is the most typical situation, are given by the equations

$$x(t_0)=1,\quad \dot{x}(t_0)=0. \tag{1.12}$$

We consider now the main particular cases, when exact solutions of (1.11) can be obtained.

2. ARBITRARY FIELD AMPLITUDE ENVELOPE AT EXACT FREQUENCY RESONANCE ($\nu \equiv 0$). CASE OF EQUAL RELAXATION TIMES ($T = \tau$)

At equal relaxation times, Eq. (1.11) can be integrated for an arbitrary form of r(t). At $T = \tau$, putting $\delta_T = \delta_\tau = \delta$ and introducing a new unknown $v = (x-1)e^{\delta t}$ and a new independent variable $\xi = \int r\,dt$, we reduce (1.11) to the form

$$v_\xi''+v=-e^{\delta t}, \tag{2.1}$$

where $e^{\delta t}$ must be expressed in terms of ξ. Solving this equation and returning to x, we have

$$x=u^{-1}\left(\sin\xi\int\sin\xi\,du+\cos\xi\int\cos\xi\,du\right),\quad u=e^{\delta t}. \tag{2.2}$$

The initial conditions (1.12) correspond to the solution

$$x=\delta u^{-1}\left[\sin\xi\int_{t_0}^{t}u\sin\xi\,dt+\cos\xi\left(\int_{t_0}^{t}u\cos\xi\,dt+u(t_0)/\delta\right)\right],\quad \xi=\int_{t_0}^{t}r\,dt, \tag{2.3}$$

where t_0 is the instant when the field is turned on.

Let us consider a few principal cases when the integrals in (2.3) can be calculated in final form.

1. <u>There is no relaxation</u> ($1/T = 1/\tau = \delta = 0$). From (2.3) we have:

$$x=\cos\left(\int_{t_0}^{t}r\,dt\right) \tag{2.4}$$

meaning undamped population oscillations with an instantaneous frequency equal to the relative field amplitude r(t); this corresponds to the result of[11] (or to the result of[3] at r = const).

2. <u>Abrupt step</u> ($r = 0$ at $t < 0$ and $r = r_0$ at $t \geq 0$). It follows from (2.3) that

$$x=\frac{1}{1+k^2}[1+ke^{-\delta t}(\sin r_0t+k\cos r_0t)],\quad k=\frac{r_0}{\delta}, \tag{2.5}$$

i.e., x(t) represents oscillating relaxation to a new equilibrium value $x_{sat} = 1/(1+k^2)$; the ratio r_0^2/δ^2 determines both the saturation of the system and the oscillation intensity, viz., at $r_0^2 \gg \delta^2$ the amplitude of the oscillations at the start of the relaxation process is close to unity (Fig. 1a).

3. <u>Smooth step with specified slope and amplitude</u>: $r(t) = r_0(1+\tanh(\delta t/q))/2$ (q – natural). At q = 1 and $t_0 \to -\infty$ (the field increases from zero) we get from (2.3)

$$x=[1+e^{-\delta t}(1+k\sin\xi-\cos\xi)]/(1+k^2),\quad k=r_0/\delta, \tag{2.6}$$

where $\xi = k\ln(1+e^{\delta t})$. Comparing this expression with (2.5) we see that at equal saturation the oscillations are already smaller here, owing to the presence of a finite time within which the field is turned on (~T); at $r_0^2 \gg \delta^2$ their maximum amplitude is $\sim 2/3\pi$ (Fig. 1b).

At q = 2 (more gently sloping step) we have

$$x=\frac{1}{1+k^2}\left\{1+\frac{1}{u(1+4k^2)}[3k(2k\sqrt{u}-\sin\xi)+(2k^2-1)(1-\cos\xi)]\right\}, \tag{2.7}$$

where $k = r_0/\delta$, $u = e^{\delta t}$, $\xi = 2k\ln(1+\sqrt{u})$. The oscillations are even smaller, and their maximum at $r_0^2 \gg \delta^2$ is $\sim 2/(3\pi)^2$ (Fig. 1c). Naturally, the amplitude of the

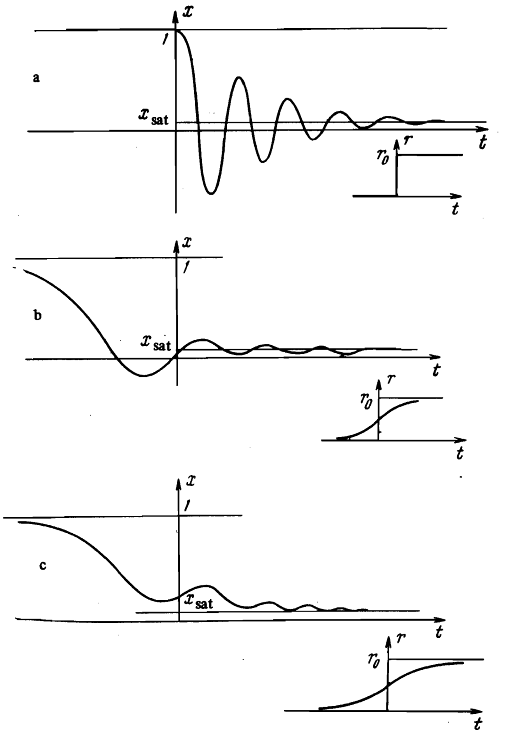

FIG. 1. Relaxation of the population in a field having the form of an abrupt (a) or smooth (b, c) step.

oscillations in the relaxation process decreases with increasing q, i.e., with increasing time required to turn on the field.

4. Radiation pulse with fixed duration $\Delta t_p \sim T$ and arbitrary amplitude: $r = r_0/\cosh^2(\delta t/2)$. From (2.3) we obtain as $t_0 \to -\infty$ (the field increases from zero)

$$x=ke^{-\delta t}\left\{\frac{1}{k-\xi}-\frac{\cos\xi}{k}-\sin(k-\xi)[\mathrm{ci}(k-\xi)-\mathrm{ci}(k)]+\cos(k-\xi)[\mathrm{si}(k-\xi)-\mathrm{si}(k)]\right\},\quad k=4r_0/\delta, \tag{2.8}$$

where $\xi = \frac{1}{2}k(1 + \tanh(\delta t/2))$, and $\mathrm{si}(\xi)$ and $\mathrm{ci}(\xi)$ are the integral sine and cosine:

$$\mathrm{si}(\xi)=\int_\infty^\xi \frac{\sin z}{z}dz,\quad \mathrm{ci}(\xi)=\int_\infty^\xi \frac{\cos z}{z}dz. \tag{2.9}$$

The number of oscillations during the entire time of action of the pulse is

$$N=\frac{1}{2\pi}\int_{-\infty}^{\infty}|r|\,dt. \tag{2.10}$$

In this case we have $N = 2r_0/\delta\pi$ and to observe at least one pulsation it is necessary to satisfy the condition $r_0 > \delta\pi/2$.

5. Radiation pulse of arbitrary duration and amplitude:

$$r(t)=r_0e^{\delta t}/\mathrm{ch}^2[g(e^{\delta t}-1)]. \tag{2.11}$$

The pulse duration is specified by the value of g; in particular, when $g \gg 1$ we have $r \approx r_0/\cosh^2(g\delta t)$, i.e., formula (2.11) yields in this case an ultrashort pulse of duration $\Delta t_p \sim 2T/g \ll T$ at approximately half the intensity level. The exact solution of (2.3), corresponding to the pulse (2.11) as $t_0 \to -\infty$, is

$$x=(2ge^{\delta t})^{-1}\{\sin(\xi+p_1)[\mathrm{si}(\xi+p_1)-\mathrm{si}(p_1)]+\cos(\xi+p_1)[\mathrm{ci}(\xi+p_1)-\mathrm{ci}(p_1)]-\sin(p_2-\xi)[\mathrm{si}(p_2-\xi)-\mathrm{si}(p_2)]-\cos(p_2-\xi)[\mathrm{ci}(p_2-\xi)-\mathrm{ci}(p_2)]\}, \tag{2.12}$$

where

$$\xi=\frac{r_0}{\delta g}[\mathrm{th}\,g(e^{\delta t}-1)+\mathrm{th}\,g],$$

$$p_1=\frac{r_0}{\delta g}(1-\mathrm{th}\,g),\quad p_2=\frac{r_0}{\delta g}(1+\mathrm{th}\,g).$$

The case of greatest interest is that of a high-power short pulse, when strong oscillations can be observed. Their number during the time of the pulse is given by (2.10), so that to excite them we must satisfy here the condition $p_2 > 2\pi$ or else, if $g \gg 1$, the condition $r_0 > \pi\delta g$. Let us trace, starting with the solution (2.12), the evolution of the behavior of the system with increasing amplitude r_0 of a pulse of fixed duration $\Delta t_p \ll T$ (Fig. 2). The first oscillations appear at $r_0 > \pi\delta g$ and it turns out that during the pulse the motion of the population obeys approximately the relation (2.4); the system behaves as if there were no relaxation. On the other hand, after the end of the field pulse the population relaxes exponentially to the stationary state $x = 1$ in accordance with (1.10); the initial value of the exponential is the value of the population reached in the system at the end of the pulse, i.e., in this case $x \approx 1 - [1 - \cos(\xi_\infty - \xi_{-\infty})]e^{-\delta t}$. Thus, if the pulse is short enough, then the oscillation amplitude is large (~ 1) and the relaxation tail that follows the pulse is a periodic function of the field integral (2.10).

With further increase of the amplitude r_0, the oscillations have time to attenuate during the time of the pulse (Fig. 2). This occurs at $r_0/\delta \gtrsim \cosh^2 g \sim \cosh^2 \delta\Delta t_p$. This spells out the condition for the "shortness" of the pulse, a condition necessary for the excitation of the oscillations during the entire pulse, followed by a tail that depends periodically on the field integral:

$$(\Delta t_p)_{\mathrm{sat}}\equiv|\Delta t_2(r=\delta)-\Delta t_1(r=\delta)|\ll T, \tag{2.13}$$

where $(\Delta t_p)_{\mathrm{sat}}$ is the duration pulse at the level of the saturation intensity (i.e., at the level $r = \delta$), and not at half intensity.

6. Exponential field pulses: $r = r_0\exp(\delta_E t)$. Here δ_E is the specified field increment (or decrement). From (2.2) we have

$$x=1-\xi^{-q}[I_q(\xi)+C_1\sin\xi+C_2\cos\xi], \tag{2.14}$$

where $\xi = r_0e^{\delta_E t}/\delta_E$, $q = \delta/\delta_E$, and we have introduced the function $I_q(\xi)$

$$I_q(\xi)=\sin\xi\int\xi^q\cos\xi\,d\xi-\cos\xi\int\xi^q\sin\xi\,d\xi, \tag{2.15}$$

which satisfies the recurrence formula

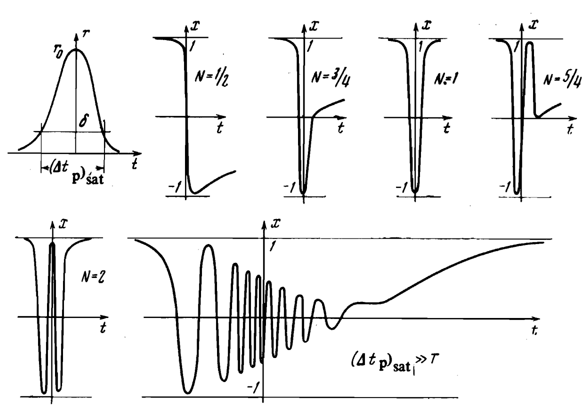

FIG. 2. Relaxation of the population in the field of a short pulse ($\Delta t_{\min} \ll 1/\delta$).

$$I_q(\xi)=\xi^q-(q-1)qI_{q-2}. \quad (2.16)$$

We see therefore that the solution (2.14) can be expressed in terms of a finite power series in ξ (as well as $\sin\xi$ or $\cos\xi$) when q is an integer and positive, in terms of the integral sine and cosine functions when q is an integer and negative, and in terms of Fresnel integrals when q is a half-integer. For example, at $\delta_E = \delta$ (q = 1) we have $x = (\sin\xi)/\xi$ (Fig. 3b, $t_0 \to -\infty$), at $\delta_E = 2\delta$ (q = 1/2) we have

$$x=\sqrt{\pi/2\xi}\,[\sin\xi S(\xi)+\cos\xi C(\xi)]$$

(Fig. 3a, $t \to -\infty$), where $S(\xi)$ and $C(\xi)$ are Fresnel integrals (see (2.18) below)), and at $\delta_E = -\delta$ (q = −1) we have

$$x=1-\xi\{\sin\xi[\operatorname{ci}(\xi)-\operatorname{ci}(\xi_0)]-\cos\xi[\operatorname{si}(\xi)-\operatorname{si}(\xi_0)]\},$$

where $\xi_0 = \xi(t_0)$. Figure 3 shows also the cases $\delta_E = \delta/2$ ($x = (\sin(\xi/2))^2/(\xi/2)^2$, q = 2) and $\delta_E = \delta/3$ (q = 3, $x = 6(1-\sin\xi/\xi)/\xi^2$). An analysis of the relaxation under the influence of short decreasing or increasing pulses ($|\delta_E| \gg \delta$) leads to the same results as in Subsec. 5.

7. Passage of field amplitude through zero with arbitrary value of the derivative at zero: $r = r_0e^{\delta t}(e^{\delta t}-1)$. The solution of (2.3) under the initial conditions (1.12) and as $t_0 \to -\infty$ yields (Fig. 4a)

$$x=e^{-\delta t}|\sqrt{\pi\delta/r_0}|\{\sin\xi[S(|r_0/2\delta|)+\operatorname{sign}(t)S(\xi)]+\cos\xi[C(|r_0/2\delta|)+\operatorname{sign}(t)C(\xi)]\},\quad \xi=|r_0/2\delta|(e^{\delta t}-1)^2, \quad (2.17)$$

where sign(t) is equal to $t/|t|$ at $t \neq 0$ and to zero at t = 0, while $S(\xi)$ and $C(\xi)$ are the Fresnel integrals

$$S(\xi)=\frac{1}{(2\pi)^{1/2}}\int_0^\xi\frac{\sin z}{z^{1/2}}dz,\qquad C(z)=\frac{1}{(2\pi)^{1/2}}\int_0^\xi\frac{\cos z}{z^{1/2}}dz. \quad (2.18)$$

Thus, if the field amplitude reverses sign rapidly ($|\dot r| \gg \delta^2$ at r = 0), assuming large values on both sides of zero ($r^2 \gg \delta^2$), then the population has no time to return to the equilibrium value x = 1, and rather strong oscillations develop, the amplitude of which at the first instance after passing through zero is

$$a_m\sim(2\pi/|r_0|)^{1/2}=\delta(2\pi/|\dot r|)^{1/2}|_{t=0}, \quad (2.19)$$

after which it decreases like $\sim e^{-\delta t}$. At the point where the field amplitude passes through zero (t = 0) we have for $r_0^2 \gg \delta^2$

$$x(t=0)\sim{}^1\!/_2|\sqrt{\pi\delta/r_0}|=a_m/2\sqrt2,\quad \dot x(t=0)\sim\delta,\qquad \ddot x(t=0)\sim-|r_0\delta/2|. \quad (2.20)$$

It can be shown that the values of a_m, x, $\dot x$, and $\ddot x$ are determined by the same formulas for any function r(t) that behaves in analogous fashion near zero. Incidentally, for

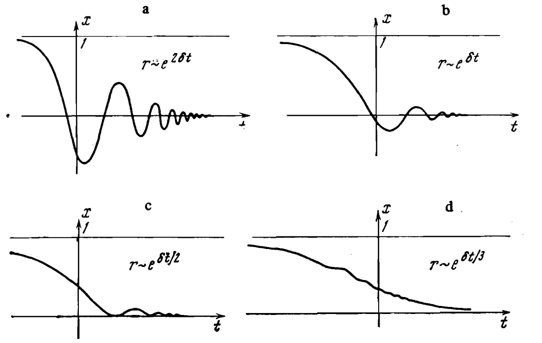

FIG. 3. Relaxation of population in an exponentially growing field.

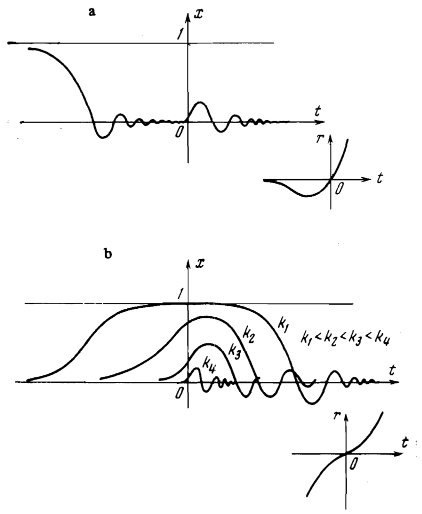

FIG. 4. Population relaxation when the field passes through zero (a) and evolution of the relaxation when the rate of growth of the field at zero is varied (b).

the function $r = r_0e^{\delta t}(e^{\delta t}-1)$, if the field is turned on not at $t_0 \to -\infty$ but at $t_0 = 0$, i.e., the field starts with zero and never reverses sign, then the form of the relaxation changes:

$$x=e^{-\delta t}\{|\sqrt{\pi\delta/r_0}|[\sin\xi S(\xi)+\cos\xi C(\xi)]+\cos\xi\}. \quad (2.21)$$

Here the amplitude of the oscillations is initially close to unity at $r_0^2 \gg \delta^2$.

8. Amplitude varies monotonically and reverses sign: $r = r_0\sinh 2\delta t$. If the field is turned on at $t_0 \to -\infty$, then the initial condition should be chosen to be $x \to 0$ as $t \to -\infty$ instead of (1.12). Calculating the solution (2.2) under this condition, we obtain (see Fig. 4b)

$$x=e^{-\delta t}|\sqrt{\pi/2k}|\{\sin(k\operatorname{sh}^2\delta t)[{}^1\!/_2+\operatorname{sign}(t)S(k\operatorname{sh}^2\delta t)]+\cos(k\operatorname{sh}^2\delta t)[{}^1\!/_2+\operatorname{sign}(t)C(k\operatorname{sh}^2\delta t)]+\sin(k\operatorname{ch}^2\delta t)[S(k\operatorname{ch}^2\delta t)-{}^1\!/_2]+\cos(k\operatorname{ch}^2\delta t)[C(k\operatorname{ch}^2\delta t)-{}^1\!/_2]\},\quad k=|r_0/\delta|; \quad (2.22)$$

the amplitudes of the oscillations at $k^2 \gg 1$ and the values of x, $\dot x$, and $\ddot x$ at t = 0 are given in (2.19) and (2.20).

9. Periodic modulation of the amplitude is discussed in Sec. 3, Subsec. 5.

3. AMPLITUDE MODULATION OF THE FIELD AT EXACT FREQUENCY RESONANCE ($\nu \equiv 0$). CASE OF UNEQUAL RELAXATION TIMES ($T \neq \tau$)

In this case it is possible to obtain exact solutions for only a few physically interesting forms of the function r(t).

1. Abrupt step, r = 0 at t < 0 and $r = r_0$ at $t \geq 0$. This field corresponds to the solution of (1.11) at r = const $= r_0$, satisfying the condition (1.12) at $t_0 = 0$, i.e.,

$$x=x_{sat}+(1-x_{sat})\exp(-\delta_\Sigma t/2)\left(\cos\varkappa t+\frac{\delta_\Sigma}{2\varkappa}\sin\varkappa t\right), \quad (3.1)$$

$$x_{sat}=(1+r_0^2/\delta_T\delta_\tau)^{-1},\quad \varkappa=(r_0^2-\delta_\Gamma^2/4)^{1/2},\quad \delta_\Gamma=\delta_T-\delta_\tau,\quad \delta_\Sigma=\delta_T+\delta_\tau. \quad (3.2)$$

The relaxation thus has an oscillatory character only if $r_0^2 > \delta_\Gamma^2/4$. We note that this condition does not coincide in the general case with the saturation condition $r_0^2 > \delta_T\delta_\tau$. At $\delta_T \lesssim 4\delta_\tau$ the oscillations appear before the saturation does (this is true, in particular, for the limiting case of a rarefied gas, when $\delta_T = \delta_\tau/2$, and all the more when $\delta_T = \delta_\tau$). In the other limiting case $\delta_T \gg \delta_\tau$ (the relaxation time within the line is much shorter than

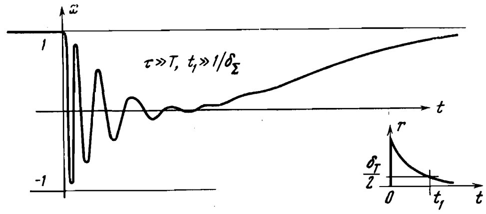

FIG. 5. Relaxation of the population in the field of an exponentially decreasing short pulse ($\Delta t_p \ll 1/\delta_\Sigma$) in the case of non-coinciding relaxation times ($T \neq \tau$).

the lifetime of the excited state) the oscillations are excited only in the case of strong saturation.

2. Smooth step, rising from zero like $\sim\exp(\delta_r t/2)$, with arbitrary steady-state amplitude $r_\infty = B\delta_r/2$:

$$r(t)=B\delta_r/2(1+B\exp(-\delta_r t))^{1/2}. \quad (3.3)$$

It is useful here to introduce new variables:

$$\eta=\int r\exp(-\delta_r t)\,dt, \quad v(\eta)=(x-1)\exp(\delta_\tau t), \quad \delta_r=\delta_T-\delta_\tau, \quad (3.4)$$

after which (1.11) takes the form

$$v_\eta''+\exp\{2\delta_r t(\eta)\}v=-\exp\{\delta_\perp t(\eta)\}, \quad \delta_\perp=2\delta_T-\delta_\tau. \quad (3.5)$$

For the function (3.3) we have $\eta = (1 + B\exp(-\delta_r t))^{1/2}$ and (3.5) takes the form

$$v_\eta''+\left(\frac{B}{\eta^2-1}\right)^2 v=-\left(\frac{B}{\eta^2-1}\right)^{\delta_\perp/\delta_r}. \quad (3.6)$$

The solution of the corresponding homogeneous equation is known (see[21]). Solving (3.6) with its aid and then changing over to x, we obtain

$$x=1+\frac{B^2}{b\,\mathrm{sh}^q\psi}\left(\cos b\psi\int\sin b\psi\,\mathrm{sh}^q\psi\,d\psi-\sin b\psi\int\cos b\psi\,\mathrm{sh}^q\psi\,d\psi\right),$$
$$b=(B^2-1)^{1/2}, \quad \psi=\mathrm{Arsh}\,(\exp(\delta_r t)/B)^{1/2}, \quad q=\frac{\delta_\Sigma}{\delta_r}=\frac{\delta_T+\delta_\tau}{\delta_T-\delta_\tau}. \quad (3.7)$$

Thus here, too, the excitations are excited only when $|r_\infty| > |\delta_r|/2$. In particular, at $\delta_T \gg \delta_\tau$ ($q \to 1$) and under the initial conditions (1.12) we have[2)] as $t_0 \to -\infty$

$$x\approx\sin b\psi/b\,\mathrm{sh}\psi=\exp(-\delta_T t/2)\sin b\psi/(|B|-1/|B|)^{1/2} \quad (3.8)$$

and at $\delta_T = 3\delta_\tau$ ($q = 2$)

$$x=1-\frac{B^2}{B^2+3}\left[1-\left(\sin\frac{b\psi}{2}\right)^2\Big/\left(\frac{b}{2}\,\mathrm{sh}\,\psi\right)^2\right]. \quad (3.9)$$

3. Steeper step, increasing at zero like $\exp(3\delta_r t)$

$$r=\frac{\delta_r B\exp(3\delta_r t)}{2(1+e^{2\delta_r t})\sqrt{1+B^2+e^{2\delta_r t}}} \quad (3.10)$$

Equation (3.5) with this r(t) and the variables (3.4) takes the form

$$v_\eta''+\left(\frac{B^2}{\mathrm{sh}^2 2\eta}-1\right)v=-\left(\frac{B^2}{\mathrm{sh}^2 2\eta}-1\right)^{\delta_\perp/2\delta_r}, \quad \delta_\perp=2\delta_T-\delta_\tau, \quad (3.11)$$

where $2\eta = \sinh^{-1}(B/\sqrt{1+\exp(2\delta_r t)})$. In analogy with the two preceding cases, this equation has an oscillatory solution only at $r_\infty^2 > \delta_r^2/4$ ($B^2 > 1$). It is easy to verify that this solution is

$$v=\frac{\sqrt{\mathrm{sh}\,2\eta}}{b}\left[\cos b\psi\int\sin b\psi\sqrt{\mathrm{sh}\,2\eta}\left(\frac{B^2}{\mathrm{sh}^2 2\eta}-1\right)^{\delta_\perp/2\delta_r}d\eta\right.$$
$$\left.-\sin b\psi\int\cos b\psi\sqrt{\mathrm{sh}\,2\eta}\left(\frac{B^2}{\mathrm{sh}^2 2\eta}-1\right)^{\delta_\perp/2\delta_r}d\eta\right], \quad b=\sqrt{B^2-1}, \quad (3.12)$$

where $\psi = \frac{1}{2}\ln|\tanh\eta|$. In particular, for the case $\delta_T \gg \delta_\tau$, converting again to x, we obtain[3)] under the initial conditions (1.12) and as $t_0 \to -\infty$,

$$x\approx\left(\frac{\mathrm{sh}\,2\eta}{\mathrm{sh}\,2\eta_0}\right)^{1/2}\cos b\psi=[1+\exp(2\delta_r t)]^{-1/4}\cos b\psi, \quad \eta_0=\frac{1}{2}\mathrm{Arsh}\,B. \quad (3.13)$$

Comparing (3.13) with (3.8) we can see that a more abrupt turning on of the field (3.10) corresponds also to larger oscillations: at $B^2 \gg 1$ their maximum amplitude in (3.13) is $\sim 1/\sqrt{2\pi+1} \sim 0.37$, as against $\sim 2/3\pi \sim 0.2$ for (3.8). For an abrupt step (see Subsec. 1) the maximum amplitude of the oscillations in (3.2) is equal to ~ 1.

4. Exponential pulses, $r = r_0\exp(\delta_E t)$. Making in (1.11) the change of variables

$$\xi=\int r\,dt=\frac{r_0}{\delta_E}\exp(\delta_E t), \quad v=(1-x)\exp(\delta_\tau t),$$

we reduce this equation to the form

$$v_\xi''+(\beta-\alpha)\frac{v_\xi'}{\xi}+v=-\left(\frac{\delta_E}{r_0}\xi\right)^\alpha, \quad \alpha=\frac{\delta_\tau}{\delta_E}, \quad \beta=\frac{\delta_T}{\delta_E}. \quad (3.14)$$

Solving this equation and reconverting to x, we obtain

$$x=1-\xi^{m-\alpha}[s_{m+\beta,m}(\xi)+C_1J_m(\xi)+C_2Y_m(\xi)], \quad (3.15)$$

where $m = \frac{1}{2}(1+\alpha-\beta) = \frac{1}{2}(1-\delta_p/\delta_E)$, $J_m(\xi)$ and $Y_m(\xi)$ are Bessel functions of the first and second kind, and $s_{\mu,m}(\xi)$ is the Lommel function[32], defined as

$$s_{\mu,m}(\xi)=\frac{\pi}{2}\left[Y_m(\xi)\int_0^\xi z^\mu J_m(z)\,dz-J_m(\xi)\int_0^\xi z^\mu Y_m(z)\,dz\right], \quad (3.16)$$

which satisfies the recurrence formula

$$s_{\mu+2,m}(\xi)=\xi^{\mu+1}-[(\mu+1)^2-m^2]s_{\mu,m}(\xi). \quad (3.17)$$

Using the expansion[4)] of $s_{\mu,m}(\xi)$[22]

$$s_{\mu,m}(\xi)=\xi^{\mu+1}\sum_{n=0}^{\infty}(-1)^n\left(\frac{\xi}{2}\right)^{2n}\frac{\Gamma(\frac{1}{2}(\mu+m+1))\Gamma(\frac{1}{2}(\mu-m+1))}{\Gamma(\frac{1}{2}(\mu+m+2n+3))\Gamma(\frac{1}{2}(\mu-m+2n+3))} \quad (3.18)$$

(i.e., $s_{\mu,m}(\xi)\xi^{-m-1}[(\mu+1)^2-m^2] \to 1$ as $\xi \to 0$), and taking into account the known expansions for $J_m(\xi)$ and $Y_m(\xi)$, we find that at $\delta_E > 0$ (increasing field pulse) and under the initial conditions (1.12) the constants C_1 and C_2 in (3.15) vanish as $t_0 \to -\infty$, i.e., in this case we have

$$x=1-\xi^{m-\alpha}s_{m+\beta,m}(\xi). \quad (3.19)$$

When one of the quantities $\mu \pm m$ (in our case, β or $\alpha+1$) is a negative odd number, the definition (3.18) becomes meaningless, and it is necessary to use another Lommel function $S_{\mu,m}(\xi)$[22] instead of $s(\xi)$. It is very useful if one of the quantities $\mu \pm m$ (i.e., β or $\alpha+1$) is an odd positive number; then $S(\xi)$ is a series with a finite number of terms

$$S_{m+\beta,m}(\xi)=P(\xi)=\xi^{m+\beta-1}-[(m+\beta-1)^2-m^2]\xi^{m+\beta-3}$$
$$+[(m+\beta-1)^2-m^2][(m+\beta-3)^2-m^2]\xi^{m+\beta-5}-\ldots, \quad m=\tfrac{1}{2}(1+\alpha-\beta). \quad (3.20)$$

This yields simple formulas describing the relaxation at integer odd β or $\alpha+1$. We present several particular cases (we assume that the conditions (1.12) are always satisfied for $\delta_E > 0$ as $t_0 \to -\infty$).

a. At $\delta_E = \delta_T$ ($\beta = 1$, $P = \xi^{\alpha/2}$)

$$x=\Gamma(\gamma+1)\left(\frac{2}{\xi}\right)^\gamma J_\gamma(\xi)=\exp\left(\frac{-\delta_\tau t}{2}\right)J_\gamma\left(\frac{r_0}{\delta_T}\exp(\delta_T t)\right)\frac{\Gamma(\gamma+1)}{(r_0/2\delta_T)^\gamma}, \quad (3.21)$$

where $\gamma = \delta_\tau/2\delta_T = T/2\tau \leq 1$; in particular, at $T = 2\tau$ ($\gamma = 1$) (limiting case, rarefied gas) we have

$$x=\frac{\delta_\tau}{r_0}\exp(-\delta_T t)J_1\left(\frac{r_0}{\delta_T}\exp(\delta_T t)\right)$$

and $\delta_T \gg \delta_\tau$ ($\gamma \gg 1$) we have $x \approx J_0(r_0\exp(\delta_T t)/\delta_T)$.

b. At $\delta_E = \delta_T/3$ ($\beta = 3$, $P = \xi^{\alpha/2}(\xi - 2\alpha/\xi)$)

$$x=\frac{12\gamma}{\xi^2}\left[1-\Gamma(3\gamma)\left(\frac{2}{\xi}\right)^{3\gamma-1}J_{3\gamma-1}(\xi)\right], \quad (3.22)$$

in particular, at $\delta_\tau = {}^2\!/_3\delta_T$ ($\gamma = {}^1\!/_3$) we have $x = 4[1 - J_0(\xi)]/\xi^2$, and at $\delta_\tau = \delta_T/3$ ($\gamma = {}^1\!/_6$) we have $x = (\sin(\xi/2))^2/(\xi/2)^2$.

c. The case $\delta_E \gg \delta_\tau$ ($\alpha \to 0$) (the condition $\delta_E \gg \delta_T$ is not obligatory here), with the aid of which we can describe the relaxation in the case of rapid growth of the field amplitude. From (3.20) and at $\alpha = 0$ we obtain $P = \xi(\beta-1)/2$, whence

$$x\approx\Gamma\left(\frac{\beta+1}{2}\right)\left(\frac{2}{\xi}\right)^{(\beta-1)/2} J_{(\beta-1)/2}(\xi). \qquad (3.23)$$

The presented solution can, of course, be used to describe the relaxation in the field of a pulse of finite duration, terminated at some instant of time t_1. After the termination of the pulse, the system is described by Eq. (1.1) with initial condition $x(t_1) = x(t_1 = 0)$, i.e.,

$$x(t\geq t_1)=1-[1-x(t_1)]\exp[-\delta_\tau(t-t_1)]. \qquad (3.24)$$

d. Rapidly decreasing field pulse turned on abruptly at the instant $t = 0$; $\delta_E < 0$, $|\delta_E| \gg \delta_\tau$ ($|\alpha| \ll 1$), but the condition $|\beta| \ll 1$ is not obligatory. Assuming, just as in Subsec. c, that $|\alpha| \to 0$ and $P = \xi(\beta-1)/2$, we obtain in the case when $|r_0| \gg \delta_T/2$ and $t_0 = 0$

$$x\approx\left(\frac{\xi}{\xi_0}\right)^{m-\alpha}\frac{\pi\xi_0}{2}[J_m(\xi)Y_{m-1}(\xi_0)-Y_m(\xi)J_{m-1}(\xi_0)], \qquad (3.25)$$

i.e., $x \approx \exp(-\delta_\Sigma t/2)\cos(\xi - \xi_0)$ at $|\xi| > m$; when the instant t_1 at which $r \sim \delta_T/2$ ($\xi \sim m$) is reached, i.e., at $t_1 \sim (1/\delta_E)\ln|\delta_T/2r_0|$, the population relaxes practically exponentially like $\exp(-\delta_\tau t)$, in accordance with (3.24). This process is illustrated in Fig. 5 for the case $t_1 \gg 1/\delta_\Sigma$.

e. At $\delta_E = \delta_\tau/2$ ($\alpha = 2$, $P = [\xi - 2(\beta-1)/\xi]\,\xi(\beta-1)/2$, see (3.20)), we have

$$x=\frac{2(1-\gamma)}{\gamma\xi^2}\left[1-\Gamma\left(\frac{1-\gamma}{2\gamma}\right)\left(\frac{2}{\xi}\right)^{(1-3\gamma)/2\gamma}J_{(1-2\gamma)/2\gamma}(\xi)\right],\quad \gamma=\delta_\tau/2\delta_T; \qquad (3.26)$$

for example, at $\delta_\tau = \delta_T/2$ ($\gamma = {}^1\!/_4$) we have $x = 6(1 - \sin\xi/\xi)/\xi^2$.

The solutions for $\alpha = 4$, 6, etc. and $\beta = 5$, 7, etc. are obtained in similar fashion.

f. The solution (3.15) takes on a simpler form if $|m|$ is a half-integer and J_m and Y_m can be expressed in terms of trigonometric functions. In particular, at $m = 1/2$ (i.e., $\alpha = \beta$ and consequently $\delta_T = \delta_\tau$), we obtain the solutions indicated in Sec. 2, Subsec. 6. We can obtain analogously the solution in all those cases when the ratio $|\delta_r/\delta_E|$ is an even number. For example, in the case $\delta_E = \delta_r/2$, i.e., $m = -1/2$ (this corresponds at $\delta_T > \delta_\tau$ ($\gamma < {}^1\!/_2$) to a rising pulse and at $\delta_T \ll \delta_\tau$ to a falling pulse), we obtain a relation similar to (2.14), where $q = \delta/\delta_E$ must now be replaced by $q = 1 + \alpha = 1 + \delta_\tau/\delta_E = (1 + 2\gamma)/(1 - 2\gamma)$.

In the case $\delta_E = -\delta_r/2$, $m = {}^3\!/_2$ (this pulse is the mirror image, as a function of the time, of the preceding pulse), taking into account the known relations for $J_{3/2}(\xi)$ and $Y_{3/2}(\xi)$ as well as the recurrence formula (2.16), we obtain in the notation of (2.15)

$$x=1-\frac{1}{\alpha-1}[\alpha\xi^{1-\alpha}I_{\alpha-1}(\xi)-\xi^{-\alpha}I_\alpha(\xi)],\quad \alpha=\frac{\delta_\tau}{\delta_E}=\frac{4\gamma}{2\gamma-1}. \qquad (3.27)$$

5. A small perturbation $\Delta(t)$ of arbitrary form is superimposed on the constant field amplitude r_0. If the perturbation is small ($\Delta^2 \ll r_0^2$), then we obtain from (1.11) an equation for $y = x - x_{sat}$ (see (3.21)):

$$\hat{D}_T\hat{D}_\tau(y)+r_0^2y=(\dot{\Delta}+2\delta_T\Delta)/(r_0/\delta_\tau+\delta_T/r_0), \qquad (3.28)$$

whence, e.g., for a periodic perturbation $\Delta = -r_m\cos\Omega t$ we obtain under the condition $r_0^2 \gg \delta_\Sigma^2/4$ the steady-state value

$$x\approx x_{sat}-\frac{r_m}{r_0}\frac{\delta_\tau}{\delta_\Sigma}\frac{\sin(\Omega t+\varphi)}{[1+(2\Delta\Omega/\delta_\Sigma)^2]^{1/2}},\quad \mathrm{tg}\,\varphi=\frac{\delta_\Sigma}{2\Delta\Omega}, \qquad (3.29)$$

where $\Delta\Omega = \Omega - \kappa$ (κ and x_{sat} are taken from (3.2)), i.e., the system has near the frequency κ a resonance of width $\delta_\Sigma/2$. It is interesting that the amplitude of the population oscillations at resonance, $r_m\delta_\tau/r_0\delta_\Sigma$, can exceed the value of the dc component x_{sat} due to the stationary saturation (i.e., the population can assume negative values), whereas the depth of the initial modulation is small; this calls for the condition $r_m > \delta_T\delta_\Sigma/r_0$.

6. We present in conclusion an approximate solution for the deviation of the population from the equilibrium value $x = 1$ under the condition that $r(t)$ is much smaller than the saturation value, $r^2 \ll \delta_T\delta_\tau$. Since the population deviates little from equilibrium in this case, i.e., $|x - 1| \ll 1$, we obtain, expressing (1.11) in the form $r^{-1}\hat{D}_T[r^{-1}\hat{D}_\tau(1-x)] \approx 1$,

$$1-x\approx{}^1\!/_2\exp(-\delta_\tau t)\left[\left(\int r\exp(\delta_T t)\,dt\right)^2\exp(-\delta_\perp t)+\delta_\perp\int\exp(-\delta_T t)\times\left(\int^t r\exp(\delta_T t')\,dt'\right)^2dt\right],\quad \delta_\perp=2\delta_T-\delta_\tau, \qquad (3.30)$$

and in particular, at $\delta_\tau = 2\delta_T$ and under the initial conditions (1.12), we have

$$1-x\approx{}^1\!/_2\exp(-2\delta_T t)\left(\int_{t_0}^{t} r\exp(\delta_T t)\,dt\right)^2. \qquad (3.31)$$

CONCLUSION

We have obtained for the description of the relaxation of the populations of a two-level system (with two characteristic relaxation times) in a strong nonmonochromatic field a differential equation of third order with variable coefficients, the values of which are determined by the instantaneous values of the field frequency and amplitude. When the frequencies of the field and of the transition are equal, this equation reduces to a second-order equation and can be solved exactly for an arbitrary amplitude of the field envelope at equal relaxation times, and for certain envelope shapes at different relaxation times. The number of situations for which an exact calculation is possible can be greatly increased by joining together several functions $r(t)$ for which the solution is known.

At equal relaxation times, oscillations of the population are excited. Their instantaneous frequency is equal to the reduced field amplitude at the given instant, and their amplitude increases with increasing rate of change of the field. The maximum amplitude of the oscillations is close to the equilibrium value in the case when the characteristic time of field variation is shorter than the relaxation time. For a pulse of finite duration, the total number of oscillations is proportional to the integral of the field amplitude. If the pulse length at the saturation level is small in comparison with the relaxation time, then the exponential tail that follows the oscillatory regime is a periodic function of this integral.

These properties of the relaxation of a two-level system make this system, in a certain sense, an absolute instrument for the measurement of the pulse amplitude integral, and even of the instantaneous values of the amplitudes (inasmuch as the duration of each spike is $\Delta t \sim 2\pi/r(t)$). At the same time, by using radiation pulses of known power we can determine the characteristics of the system (the dipole moment and the relaxation time).

Relatively strong population oscillations are excited also when the field amplitude goes rapidly through zero, in which case the oscillation amplitude is proportional to $|\dot{r}|^{-1/2}|_{r=0}$.

At unequal relaxation times, qualitative differences from the case of equal times appear mainly at small field amplitudes. In particular, if the relative field amplitude is smaller than $\delta_r/2 = (\delta_T - \delta_\tau)/2$, then the oscillatory relaxation regime does not set in at all; when the field amplitude varies slowly, the oscillation frequency equals $(r_0^2 - \delta_r^2/4)^{1/2}$. The population oscillations can be excited in a field weaker than the saturating field $(r_{sat} = \sqrt{\delta_T/\delta_\tau})$ if $\delta_T < 4\delta_\tau$. The oscillation growth increment is given by $\delta_\Sigma/2 = (\delta_T + \delta_\tau)/2$. The same quantity determines the width of the resonance of the system relative to weak oscillations of frequency close to $(r_0^2 - \delta_r^2/4)^{1/2}$ in a strong field with constant amplitude r_0; the population difference can become negative in this case even if the perturbations are relatively weak.

The obtained exact solutions are not only of independent interest, but can serve as a basis for the calculation of approximate relaxation regimes of two-level systems in the general case, and in particular for the determination of the behavior of relaxing systems in a strong spontaneous field.

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[1]As is well known, the first experimental observation of the effect dates back to 1926.

[2]We note that in (3.8) we have $x \to 0$ as $t \to \infty$, although $r(t) < \infty$. The reason is that at $\delta_T \gg \delta_\tau$ the oscillator condition $r_\infty^2 > \delta_r^2/4$ corresponds to very strong saturation ($r_{sat}^2 = \delta_T\delta_\tau \ll \delta_r^2$), so that x_{sat} drops out in the approximation (3.8); its value is determined from (3.2).

[3]See the preceding footnote.

[4]With the exception of the case when one of the numbers $\mu \pm m$ is odd and negative, in which case (3.18) becomes meaningless.

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