

# Light-induced relaxation in an intense Rayleigh field

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The instantaneous modulation theory is extended to a system subjected to the action of a Rayleigh radiation field, which is a Gauss-Markov random process. Analytic solutions are obtained which describe light-induced relaxation in the balance approximation, for which the transition probability between levels,  $w$ , fluctuates as a result of amplitude modulation of the field. The results are used for the calculation of the degree of saturation in a system of inhomogeneously broadened atoms produced by incoherent radiation of various structure and arbitrary intensity.

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## 1. INTRODUCTION

The stochastic description of quantum transitions that arise in a two-level system under the action of light assumes that the transition probability  $\bar{w}$  is constant in time if the radiation spectrum is sufficiently broad. The constancy of the probability guarantees in particular an exponential increase in the populations  $\rho_{11}$  and  $\rho_{22}$  of both levels of the free atom in the process of light-induced relaxation:

$$n = \rho_{11} - \rho_{22} = n(0) \exp(-2\bar{w}t). \quad (1.1)$$

The probabilistic description of relaxation is so well established that, as a rule, it is sufficient to obtain it only in the initial stage, where

$$n \approx n(0)(1 - 2\bar{w}t) \quad (t \ll (2\bar{w})^{-1}), \quad (1.2)$$

without being concerned as to how the process develops further. Such a statement of the problem goes back to Dirac, who resorted to time-dependent perturbation theory to calculate the probability by this method.

However, it was noted many times<sup>[1-3]</sup> that a derivation of (1.2) does not mean the confirmation of the long-time kinetics of the relaxation (1.1). In a powerful narrow band of radiation, i.e., outside the limits of perturbation theory, light-induced relaxation always takes place nonexponentially, and even if (1.2) is derived at first, this amounts at most only to an imitation of the stochastic development of the process. The actual kinetics that obtains can be decided only if a more rigorous method is available for the solution of the problem than time-dependent perturbation theory. It is necessary here to have much more complete information on the structure of the radiation, for if its effect on the atom is strong, then the reaction of the latter depends not only on the second moment of the field (in terms of which the probability of transition is determined), but also on all the moments of higher order. Thanks to this, a concrete model of radiation appears in the kinetics of light-induced relaxation, and, in particular, the cause of its broadening—phase, frequency, or amplitude modulation—is determined.

However, identification of the radiation by the reaction on it is possible only in the case in which the latter is sufficiently reliably determined. From this view-

point, the radiation, which is stable in frequency and amplitude, but broadened because of phase modulation (radiation of a single-mode laser in the generation regime),<sup>[4]</sup> has been studied rigorously and completely.<sup>[3, 5-7]</sup> Under certain assumptions, the problem is also solved in the case in which the broadening of the radiation is due to Doppler shift of the light frequency,<sup>[3, 8]</sup> and also for mixed frequency-phase modulation.<sup>[9]</sup> However, there has been little progress in the study of amplitude-phase modulation, and the present work has as its goal to fill this gap.

The rigorous analysis, carried out below, confirms the fact that in irradiation by a broad and weak amplitude-modulated light the long-time relaxation of the populations is exponential, and the damping decrement is equal to the transition probability determined from perturbation theory. When the Rayleigh radiation becomes powerful or narrow-band, then the equalization of the populations takes place at the same rate, but entirely according to some other law which imitates (1.1) only initially. In this respect, the considered example is very instructive. It clearly demonstrates how inconsistent can be the prediction of long-time kinetics of the process on the basis of its initial part. Moreover, the value of  $\bar{w}$ , although it characterizes as before the mean time of equalization of the populations under the action of the light, not only does not make this process stochastic, but is no longer even the probability that characterizes the stationary light absorption or the degree of saturation of the system in the presence of an energy sink. This circumstance must be kept in mind for the interpretation of the nonlinear dependence of the population of inhomogeneously broadened two-level systems on the power of incoherent pumping.

## 2. RAYLEIGH RADIATION

It is usually assumed that the amplitude of radiation broadened by amplitude-phase modulation is real ( $E \geq 0$ ) and is either a normal process<sup>[6, 10-13]</sup> or a purely discontinuous Markov noise without correlation.<sup>[13, 14]</sup> However, in actuality, there is no basis for ignoring the complex character of the field component that is at resonance with the atom and appears in the interaction matrix element

$$\langle 1 | \hat{F} | 2 \rangle = DE e^{i\omega t}, \quad (2.1)$$

in which  $D = \langle 1 | \hat{D} | 2 \rangle$  is the corresponding element of the dipole moment and  $E = u + iv$ . The restriction to a real field component ( $E(t) = u(t)$ ), dictated exclusively by methodological considerations, greatly simplifies the solution of the problem but deprives it of meaning, since the complex and real processes are characterized by different distributions of  $|E| = E_0$ , and their actions on the atom are not comparable.

If  $E(t)$  is a complex normal process, then the modulus of the field and its phase  $\psi$  have a Rayleigh distribution at any time of the process:

$$dW(E) = \varphi_0(E) d^2E = \exp\left(-\frac{E_0^2}{\mathcal{E}^2}\right) \frac{2E_0 dE_0}{\mathcal{E}^2} \frac{d\psi}{2\pi}. \quad (2.2)$$

In the classical theory of a single-mode layer, it has been established that just such a field distribution exists below the generation threshold, and its change with time represents a normal and moreover a Gauss-Markov process.<sup>[4]</sup> The latter is completely determined by the conditional probability that the field at the instant  $t$  has the value  $E$  if it was equal to  $E'$  at the time  $t'$ :

$$\varphi(E', t'; E, t) = \frac{1}{\pi \mathcal{E}^2 (1 - \exp[-\nu(t-t')])} \exp\left\{-\frac{|E-E' \exp[-\nu(t-t')]|^2}{\mathcal{E}^2 (1 - \exp[-\nu(t-t')])}\right\}; \quad (2.3)$$

the correlation function of the field amplitude being

$$k(t, t') = \overline{E^*(t) E(t')} = \iiint E' E' \varphi_0(E') \varphi(E', t'; E, t) d^2E d^2E' = \mathcal{E}^2 \exp[-\nu(t-t')]. \quad (2.4)$$

It is evident that the variance of the field  $\mathcal{E}^2$  characterizes the intensity of the radiation and  $\nu$  is the width of its spectrum, which is Lorentzian in shape.

Irradiation of exactly the same structure can also be thermal. An ordinary rarefied gas emits it if the dominant source of line broadening is adiabatic loss of phase stability. The complete field is composed of fields emitted by all the atoms, which in this case are naturally assumed to differ only in phase:

$$E = \sum_{k=1}^N e_k = e_0 \sum_{k=1}^N e^{i\alpha_k}.$$

For sufficiently large  $N$ , the total field has a Rayleigh distribution by virtue of the central limit theorem,<sup>[15]</sup> and  $\overline{E^2} = \mathcal{E}^2 = Ne_0^2$ . Along with this, each phase  $\alpha_k$  is a purely discontinuous Markov variable, which changes in an uncorrelated way for strong collisions, taking after the collision any possible value from 0 to  $2\pi$  with equal probability. The field as a whole is also a pure Markov quantity: however, it changes  $N$  times more frequently, but by a smaller amount, due to the jump in the  $k$ -th component, i.e., in correlated fashion (see Fig. 1). The conditional probability of such a process is determined by the Feller equation<sup>[3, 16]</sup>:

$$\frac{\partial \varphi(E', t'; E, t)}{\partial t} = -\frac{1}{\tau_0} \left[ \varphi(E', t'; E, t) - \iint f(E'', E) \varphi(E', t'; E'', t) d^2E'' \right]. \quad (2.5)$$

The mean time between phase discontinuities of a single atom is  $\tilde{\tau} = N\tau_0$ , and

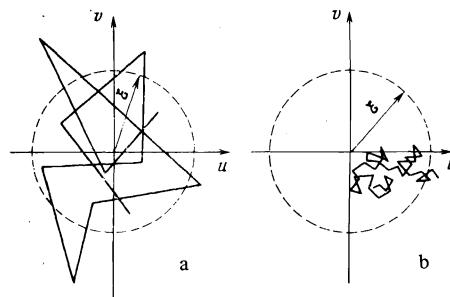


FIG. 1. One of the realizations of the complex random process  $E(t) = u(t) + iv(t)$ : a) uncorrelated process, b) correlated process.

$$f(E', E) = \frac{1}{\pi(1-\gamma^2)\mathcal{E}^2} \exp\left[-\frac{|E-\gamma E'|^2}{(1-\gamma^2)\mathcal{E}^2}\right] \quad (0 \leq \gamma < 1) \quad (2.6)$$

is the distribution of the total field after the discontinuity about its preceding value  $E'$ , which satisfies the condition of stationarity.<sup>[17]</sup> For large  $N$  we have  $\gamma^2 \rightarrow 1 - N^{-1}$  and (2.5) reduces, by the standard expansion<sup>[3]</sup>, to the Fokker-Planck differential equation:

$$\varphi(E', t'; E, t) = \sum_{\alpha=u, v} \nu \left( 1 + \alpha \frac{\partial}{\partial \alpha} + \frac{\mathcal{E}^2}{2} \frac{\partial^2}{\partial \alpha^2} \right) \varphi(E', t'; E, t), \quad (2.7)$$

the solution of which is (2.3) (with  $\nu = (1-\gamma)/\tau_0 \sim 1/\tilde{\tau}$ ).<sup>[3]</sup> This confirms that the process of interest to us is simultaneously normal and Markovian. In the latter quality, it can be classified as "strongly correlated" ( $1-\gamma \ll 1$ ), since an isochronous random change in the field changes its value but little.

From the mathematical viewpoint, consideration of the reaction of the system to a correlated random action represents a much more complicated problem than that arising in the uncorrelated variant ( $\gamma = 0$ ). However, since the synchronization of collisions is physically absurd, the uncorrelated process has nothing in common with reality and bears little resemblance to the real (correlated) process on which we shall focus our attention. If we do not consider a wave modulated only in phase, the action of which has been computed for any  $\gamma$ ,<sup>[5-7]</sup> then such a problem is first raised here. Because of the more complicated structure of the field, modulated both in amplitude and in phase it is not possible at the present time to solve the problem over the entire volume. However, in the most widespread situation, which achieves simplification of the quantum kinetic equations to the balance scheme, both processes, the noncorrelated and the correlated, can be treated quite rigorously.

### 3. KINETIC EQUATIONS

The reaction of a quantum system to a strong external action is nonlinear. In second-order perturbation theory it is quadratic in the interaction and therefore it is universally expressed in terms of the second moment (the correlation function) of the noise (2.4). In the next orders, higher moments appear, which reduce to multiplications of the second moments only for normal processes. But even for a normal process, the reaction to a strong non-adiabatic perturbation has never been

calculated,<sup>1)</sup> since this requires summation of all orders of perturbation theory. Purely discontinuous Markov processes possess indisputable advantages over the normal in that the reaction of quantum systems to such perturbations lends itself to rigorous averaging. The procedure of averaging leads to the equations of the theory of sudden modulation, developed by one of the authors,<sup>[3,5-7]</sup> which, in application to the present problem, have the form

$$\frac{\partial n(E, t)}{\partial t} = -4D \operatorname{Im} E \sigma(E, t) - \frac{1}{\tau_0} \left[ n(E, t) - \iint f(E', E) n(E', t) d^2 E' \right] - \frac{n(E, t) - n_0 \varphi_0(E)}{\tau_1}, \quad (3.1a)$$

$$\frac{\partial \sigma(E, t)}{\partial t} = (i\Omega - T_2^{-1}) \sigma(E, t) + iD E n(E, t) - \frac{1}{\tau_0} \left[ \sigma(E, t) - \iint f(E', E) \sigma(E', t) d^2 E' \right]. \quad (3.1b)$$

Here  $n = \rho_{11} - \rho_{22}$ ;  $\sigma = \rho_{12} e^{-i\omega t}$ ;  $\rho_{ik}$  ( $i, k = 1, 2$ ) are the elements of the density matrix of a two-level system:

$$\Omega = \langle 2 | \hat{H}_0 | 2 \rangle - \langle 1 | \hat{H}_0 | 1 \rangle - \omega = \omega_0 - \omega$$

are the frequency deviations reckoned from the resonance frequency;  $n_0$  is the equilibrium difference of populations;  $T_1$  and  $T_2$  are the times of longitudinal and transverse relaxations, respectively. The final averaged value of the density matrix element is

$$\bar{\rho}_{ik} = \iint \rho_{ik}(E, t) d^2 E. \quad (3.2)$$

In order to preserve continuity with the Dirac formulation of the problem, we shall assume that the system was not polarized up to the turning-on of the field, i.e.,

$$n(E, 0) = n(0) \varphi_0(E), \quad \sigma(E, 0) = \sigma(0) \varphi_0(E) = 0. \quad (3.3)$$

Moreover, if  $n(0) = 1$ , then at the moment of switching-on the atom was located on the lower level, but if  $n(0) = n_0$ , then it is in equilibrium, but these differences are unimportant for the evolution of the relaxation, if only  $n(0) \neq 0$ .

Since the equations given above are entirely rigorous, their solution, at least in principle, opens the way to a strong interaction: it allows us to consider all orders of perturbation theory and correlation moments of the field of any degree, without turning to them directly. This is important, since the reaction of the quantum system to strong action is unique and can be regarded as the means of revealing the structure of the perturbing field.<sup>[18,19]</sup> In contrast to a system of counters, which gives information on the correlation of the amplitudes or intensities,<sup>[20]</sup> i.e., on low moments of the field, the kinetics of the relaxation make it possible to cast light on its more complete characteristics of the field, such as the distribution function of the amplitudes in terms of which, as will be shown, the kinetics is universally expressed.

Limiting our consideration to relaxation of the populations from the initial state (3.3), we must note that

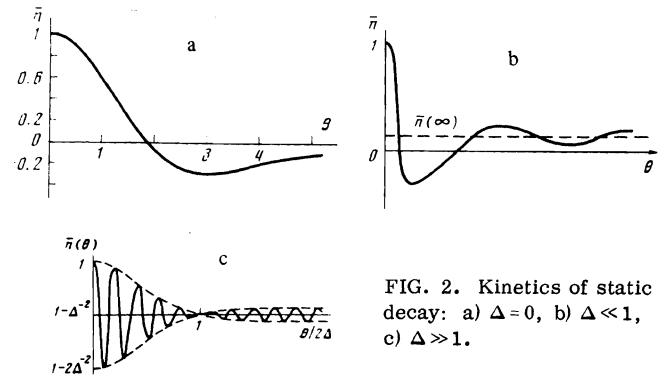


FIG. 2. Kinetics of static decay: a)  $\Delta = 0$ , b)  $\Delta \ll 1$ , c)  $\Delta \gg 1$ .

it is not possible to study it in the general case. In the present work, we discuss certain important limiting cases, which allow us to put together a rather complete picture of the kinetics of the process.

#### 4. STATISTICAL DECAY

In the time interval  $0 < t \ll (\nu + T_2^{-1})^{-1}$ , the amplitude of the light does not change significantly and therefore the action of the radiation on the atom bears to a certain degree a static character. When all the relaxation processes are turned off ( $\nu = T_1^{-1} = T_2^{-1} = 0$ ), the evolution of the populations, calculated from (3.1) is purely dynamic.<sup>[3,21]</sup> However, even in the static limit, the element of chance is not completely excluded. If the characteristic dimension  $L$  of the irradiated surface of the sample greatly exceeds the coherence length of the light,  $L \gg c/\nu$ , then the dynamic solution must be averaged over the Rayleigh distribution of the fields in the sample. This gives

$$\frac{\bar{n}(\theta)}{n(0)} = 1 - 2 \int_0^\infty \frac{xe^{-x}}{x + \Delta^2} \sin^2 \frac{\sqrt{x + \Delta^2}}{2} \theta dx, \quad (4.1)$$

where  $\theta = 2D\mathcal{E}t$  is the dimensionless time, and  $\Delta = |\Omega| / 2D\mathcal{E}$  the measure of the resonance in units of the interaction.

The evolution of the system in the static limit is interesting as an example of a nonexponential development of the process. It can be traced both in a situation close to resonance ( $\Delta \ll 1$ ) and far from it ( $\Delta \gg 1$ ). As is seen from Fig. 2, the traces of the dynamic process (mutation) are clearer the greater the departure from resonance, populations are never equalized in the process, as they would be if the process actually had an irreversible character. In essence, this process is similar to the spreading out of a wave packet the components of which are propagated with a different velocity, and it differs from the decay of a free induction signal, due to the inhomogeneity of the field<sup>[11]</sup> only in that it is stimulated.

#### 5. PERTURBATION THEORY

A direct alternative to the static limit is the reaction of the system to broadband weak radiation. As shown

in<sup>[3]</sup>, in this case the kinetics of the relaxation of populations satisfies the simple balance equation

$$\dot{n} = -2\bar{w}\bar{n} - (\bar{n} - n_0)T_1^{-1} \quad (t \gg (v + T_2^{-1})^{-1}), \quad (5.1)$$

if the initial stage of the process that is excluded from it does not last long in comparison with  $T_1$ , i.e.,  $T_1^{-1} \ll v + T_2^{-1}$ . In Eq. (5.1), the probability of a light-induced transition

$$\bar{w} = 2(D\mathcal{E})^2 \frac{v + T_2^{-1}}{\Omega^2 + (v + T_2^{-1})^2} \quad (5.2)$$

is identical with the Dirac definition (1.1) at  $T_2 = \infty$ .

The basic condition of applicability of perturbation theory, which justifies the splitting, has the form<sup>[3]</sup>

$$2\bar{w} \ll v. \quad (5.3)$$

It limits from below the width of the spectrum capable of producing exponential relaxation of the population. The kinetics of the process can be traced from beginning to end, for any power and width of the actual radiation, within the framework of the balance approximation, to the consideration of which we now turn.

## 6. THE BALANCE APPROXIMATION

If the transverse relaxation takes place sufficiently rapidly ( $2\bar{w} + v + T_1^{-1} \ll T_2^{-1}$ ), then at times  $t \gg T_2$  we can neglect the time derivation in (3.1b) and the components proportional to  $\tau_0^{-1}$  in comparison with the other terms (the approximation that is "quasistationary in the phase elements of the density matrix" or "balance" approximation). Eliminating the phase element  $\sigma$  from the set of equations (3.1), we get the balance approximation for one equation that is closed relative to  $n$ . The solution of this equation cannot be compared directly with Eq. (1.1), since the latter describes the light-induced relaxation in a free atom, and in the specified equation, just as in (3.1a), the connection of the atom with the medium is taken into account by means of  $T_1$ . It is therefore convenient to represent the sought-for function in the form

$$n(E, t) = n(0)m(E, t)\exp\left(-\frac{t}{T_1}\right) + n_0 \int_0^t m(E, t')\exp\left(-\frac{t'}{T_1}\right) \frac{dt'}{T_1}. \quad (6.1)$$

The new variable  $m(E, t)$  obeys the balance equation

$$\frac{\partial m(E, t)}{\partial t} = -2w(E)m(E, t) - \frac{1}{\tau_0} \left[ m(E, t) - \iint f(E', E)m(E', t)d^2E' \right] \quad (6.2)$$

with the initial condition  $m(E, 0) = \varphi_0(E)$ , except that the transition probability

$$w(E) = 2(D\mathcal{E}_0)^2 T_2 / [1 + (\Omega T_2)^2], \quad (6.3)$$

which controls the evolution of the population, is here a random Markov variable.

The function

$$\bar{m}(t) = \iint m(E, t)d^2E,$$

which determines the kinetics of the process in the high-temperature limit ( $n_0 \approx 0$ ) corresponds to the Dirac statement of the problem. Under the condition (5.3):

$$\bar{m}(t) = \exp(-2\bar{w}t), \quad (6.4)$$

as is also  $n(t)/n(0)$  in (1.1). If the inequality (5.3) is reversed, then the concept of the probability as a kinetic characteristic loses meaning. In particular, in the static limit ( $\tau_0 \rightarrow \infty$ ), when one can neglect the term proportional to  $\tau_0^{-1}$  in (6.2), we have

$$\bar{m}(t) = 1/(1 + 2\bar{w}t). \quad (6.5)$$

The kinetics of the decay (6.5) in no way resembles (6.4), although the rate of both processes is the same:  $2\bar{w}$ . The decay (6.5) imitates the exponential process only at the beginning of its stage, where  $\bar{m}(t) \approx 1 - 2\bar{w}t$ , i.e., at the limits of the time-dependent perturbation theory ( $2\bar{w}t \ll 1$ ).

We now proceed to the determination of the exact solution of Eq. (6.2) in the case of a strongly correlated process.

## 7. THE CORRELATED PROCESS

If  $1 - \gamma \ll 1$ , then Eq. (6.2) reduces to the following:

$$\frac{\partial m(E, t)}{\partial t} = \sum_{\alpha=u,v} \left[ -k\alpha^2 + v \left( 1 + \alpha \frac{\partial}{\partial \alpha} + \frac{\mathcal{E}^2}{2} \frac{\partial^2}{\partial \alpha^2} \right) \right] m(E, t) \quad (7.1)$$

with the initial condition  $m(E, 0) = \varphi_0(E) = \varphi_0(u)\varphi_0(v)$ . Here

$$k = \frac{(2D)^2 T_2}{1 + (\Omega T_2)^2}, \quad \varphi_0(\alpha) = \frac{1}{\sqrt{\pi\mathcal{E}}} \exp\left(-\frac{\alpha^2}{\mathcal{E}^2}\right). \quad (7.2)$$

The reaction of the quantum system to a Gauss-Markov-Rayleigh process is rigorously determined by this equation.

Separating the variables by the substitution

$$m(E, t) = X(u, t)X(v, t), \quad (7.3)$$

we find that the function  $X(\alpha, t)$  obeys the equation

$$\frac{\partial X}{\partial t} = -k\alpha^2 X + v \left( X + \alpha \frac{\partial X}{\partial \alpha} + \frac{\mathcal{E}^2}{2} \frac{\partial^2 X}{\partial \alpha^2} \right) \quad (7.4)$$

with the initial condition  $X(\alpha, 0) = \varphi_0(\alpha)$ . Equation (7.4), with a change of variable which cancels the first derivative with respect to  $\alpha$ , reduces to the Schrödinger equation for the harmonic oscillator with a purely imaginary energy. The solution of this equation gives

$$\bar{m}(t) = \left[ \int_{-\infty}^{\infty} X(\alpha, t)d\alpha \right]^2 = \frac{4\beta \exp[-(\beta-1)v t]}{(\beta+1)^2 - (\beta-1)^2 \exp(-2\beta v t)}, \quad (7.5)$$

$$\beta = (1 + 4\bar{w}/v)^{1/2}.$$

Since (2.7) follows from (2.5) at  $t \gg \tau_0$ , the validity of (7.5) is limited by the condition  $2\bar{w}\tau_0 \sim \bar{w}/Nv \ll 1$ .

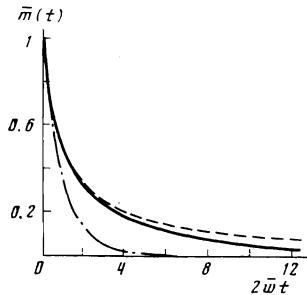


FIG. 3. Kinetics of the relaxation of population in the case of correlated process ( $\bar{w}/\nu = 20$ )—continuous curve; dashed curve—purely static decay; dot-dash curve—results of perturbation theory.

In these limits, the result (7.5) gives a complete representation of the time development of the process. At the beginning and at the end, it takes place in entirely different fashion; in the beginning, the process is static, and at the end, stochastic. In fact (see Fig. 3),

$$\bar{m}(t) = \begin{cases} \frac{1}{1+2\bar{w}t} & 2\beta\nu t \ll 1 \\ \frac{4\beta}{(\beta+1)^2} e^{-(\beta-1)\nu t}, & e^{2\beta\nu t} \gg 1 \end{cases} \quad (7.6a)$$

$$e^{2\beta\nu t} \gg 1 \quad (7.6b)$$

It is then seen that if  $2\bar{w} \ll \nu$ , then  $1 - \beta \approx 2\bar{w}/\nu \ll 1$ , and the relaxation, in accord with (7.6b), is exponential from the very beginning, in correspondence with the results of perturbation theory.

On the other hand, for a strong interaction,  $\sqrt{\bar{w}} \gg \sqrt{\nu}$ , the basic part of the decay (up to  $t_{\tau} = (2\beta\nu)^{-1} \approx [4(\bar{w}\nu)^{1/2}]^{-1} \gg (2\bar{w})^{-1}$ ) takes place in accord with the static formula (6.5), and only the remote asymptote of the process is exponential. It is important to note that the correlated process differs from the uncorrelated only in the parameter of the exponential decay. In other words, the latter simulates well only the static stage of the process, while the actual picture of relaxation of the populations is more complicated, an explicit representation of it is given in Fig. 3. It should be emphasized once again that even there, where the decay becomes exponential, its rate is nowhere determined only by the average transition probability, but also by the spectral width of the active light, i.e., the asymptotic rate of decay under the action of the Gauss-Markov process is  $2(\bar{w}\nu)^{1/2}$ .

## 8. STATIONARY SATURATION

For the calculation of the stationary characteristics of light absorption, in particular the stationary difference of populations  $\bar{n}_s = \lim \bar{m}(t)$ ,  $t \rightarrow \infty$ , it is appropriate to turn directly to Eq. (3.1). Setting the derivatives in the latter equal to zero and eliminating  $\sigma$  from them, we can obtain an equation relative to  $n_s(E) = \lim n_s(E, t)$ ,  $t \rightarrow \infty$ :

$$(2D)^2 \text{Re} \iint E' E' \Phi(E', E; p_0) n_s(E') d^2 E' + \frac{1}{\tau_0} \left[ n_s(E) - \iint f(E', E) n_s(E') d^2 E' \right] + \frac{n_s(E) - n_0 \Phi_0(E)}{T_1} = 0. \quad (8.1)$$

Here

$$p_0 = T_2^{-1} - i\Omega, \quad \Phi(E', E; p_0) = \int_0^\infty \varphi(E', 0; E, \tau) e^{-p_0 \tau} d\tau.$$

It has not been possible to solve Eq. (8.1) in general form. However, the problem is materially simplified if the inequality  $\nu T_2 \ll 1$  is satisfied. In this case,  $\Phi(E', E; p_0) \approx \delta(E' - E)/p_0$  and, as is not difficult to show, by comparing Eqs. (6.2), (8.1).

$$\bar{n}_s = n_0 \int_0^\infty \bar{m}(t) e^{-t/T_1} \frac{dt}{T_1}, \quad (8.2)$$

where the function  $m(t)$  is the result of averaging over  $E$  the solution of the balance equation (6.2). We thus see that the result for  $n_s$ , which can be obtained in the balance approximation, is nevertheless valid, as has frequently been the case,<sup>[22]</sup> over much wider limits than the latter, since the range of its applicability is limited by the condition  $\nu T_2 \ll 1$ .

Taking these considerations into account, we calculate the saturation of an assembly of inhomogeneous averaged two-level systems pumped by noncoherent radiation, which was realized recently in experiment.<sup>[23]</sup>

In the optical range,  $n_0 \approx 1$  and the population of the excited state, in correspondence with (8.2), has the form

$$\bar{n}_2 = \frac{1}{2} (1 - \bar{n}_s) \approx \frac{1}{2} \left[ 1 - \int_0^\infty \bar{m}(t) e^{-t/T_1} \frac{dt}{T_1} \right]. \quad (8.3)$$

The integrated population of the upper level is

$$N_2 = \int_{-\infty}^\infty \bar{n}_2(\omega - \omega_0) \varphi(\omega_0) d\omega_0 \approx \varphi(\omega) \int_{-\infty}^\infty \bar{n}_2(\omega - \omega_0) d\omega_0, \quad (8.4)$$

where  $\varphi(\omega_0)$  is the distribution function of the ensemble of two-level systems over their frequencies; this function is assumed to be broad enough.

The analysis carried out with the use of Eqs. (7.5), (8.3), and (8.4), showing that the following limiting results are valid for the population of the excited level:

$$N_2 = \begin{cases} \frac{a}{(1+a)^{1/2}}, & \nu T_1 \gg 1 \\ \frac{\sqrt{\pi}}{2} \frac{a-2}{a^{1/2}} \exp(a^{-1}) \text{Erfc}(a^{-1/2}), & \nu T_1 \ll 1 \end{cases} \quad (8.5a)$$

$$a = 2\bar{w}(0)T_1 = \frac{(2D)^2 T_1}{T_2^{-1} + \nu}, \quad \text{Erfc}(x) = 1 - \Phi(x), \quad (8.5b)$$

Here we have used the notation

$$a = 2\bar{w}(0)T_1 = \frac{(2D)^2 T_1}{T_2^{-1} + \nu}, \quad \text{Erfc}(x) = 1 - \Phi(x),$$

$\Phi(x)$  is the probability integral.<sup>[24]</sup> Equation (8.5a) can be obtained formally if we use the result of perturbation theory (6.4) in (8.3) and (8.4), and (8.5b) follows from (8.3) and (8.4) if we use Eq. (6.5), obtained in the static limit.

Although the population increases with the power of the radiation in both cases—linearly for weak illumination ( $a \ll 1$ ), and as the square root for strong illumination

tion ( $\alpha \gg 1$ )—the coefficients of proportionality in the latter case are different, and the ratio of the expression (8.5b) to (8.5a) is equal to  $\sqrt{\pi}/2 \approx 0.887$ .

It can be shown that the range of applicability of the result (8.5) is not limited to the condition  $\nu T_2 \ll 1$ . Moreover, the indicated result is valid independently of the degree of correlation ( $\gamma$ ) of the random process.

<sup>1)</sup>An exception is the previously mentioned real normal process, strictly resonant to the atomic transition.

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## Acceleration of atoms by a strong resonance field

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A consistent investigation is carried out of the acceleration of an atom in the field of a traveling monochromatic wave, taking into account the quantization of the translational coordinates of the atom. It is shown that in the case of metastable working levels the momentum distribution arising in the process of acceleration is exponential and, consequently, its width  $\Delta p$  is of the order of the average momentum  $\langle p \rangle$  transferred to the atom. Thus, the usually adopted description by means of an average force is incorrect in the general case. But if the lower working level is the ground level, then in the case of a large number of photons scattered by the atom  $n \gg 1$  the momentum distribution is Gaussian, with  $\Delta p \ll \langle p \rangle$ . The origin of the uncertainty  $\Delta p$  is determined by two causes: recoil on spontaneous emission of photons and the uncertainty  $\Delta n$  in the number of photons scattered by the atom. It is shown that for  $n \gg 1$  the first cause always leads to a small uncertainty  $\Delta p \ll \langle p \rangle$ , while it is specifically the second cause that leads to a large uncertainty  $\Delta p \sim \langle p \rangle$  in the case of metastable levels.

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### 1. INTRODUCTION

Acceleration of atoms by a resonance field has been investigated in a number of papers<sup>[1–4]</sup> on the assumption that both the field and the translational motion of the atom can be treated as classical. The effect of acceleration was described by means of an average force

$$\mathbf{F}(t) = \nabla(\mathbf{E}(\mathbf{r}, t)\langle \mathbf{d}(t) \rangle), \quad (1)$$

where  $\nabla$  operates only on the intensity  $\mathbf{E}(\mathbf{r}, t)$  of the electric field, and  $\langle \mathbf{d}(t) \rangle$  is the quantum average of the dipole moment of the atom.<sup>[2–4]</sup>

The quantum fluctuations in this force were taken into