# KINETICS OF INDUCED RELAXATION

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An exact description is obtained for the kinetics of quantum transitions between two levels, induced by Lorentz radiation or by a wave whose amplitude is modulated by a normal process. The conditions under which the kinetics may be satisfactorily described by a statistical transition model (perturbation theory) are considered, and the anomalies which occur under conditions for which the model is apparently not valid are also studied.

## 1. RELAXATION OF AN ATOM UNDER THE ACTION OF LORENTZ RADIATION

IN the Lorentz approximation, radiation emitted by a luminous gas (broadened as a result of collisions) can be regarded as a monochromatic wave randomly interrupted in phase at the instants of the collisions (Fig. 1). The correlation function of such radiation and its spectral composition are determined by the formulas

$$K(\tau)=\overline{E(t)E(t+\tau)}=E_0^2\exp\left[i\omega\tau-\frac{|\tau|}{\tau_0}\right],$$
$$\rho(x)=\frac{1}{2\pi}\int_{-\infty}^{+\infty}K(\tau)e^{ix\tau}d\tau=\frac{1}{\pi}\frac{E_0^2\tau_0}{1+(x-\omega)^2\tau_0^2}, \quad (1.1)$$

where $E(t)=E_0e^{i\omega t+i\alpha(t)}$ [$\alpha(t)$ is the randomly changing phase, $E_0$ the amplitude of the wave], and $\tau_0$ is the mean duration of the interval of monochromaticity. If radiation of such a type, regarded as an external perturbation, acts on an atom—a two-level system—in such a fashion that only the element

$$\hat{V}_{12}=-\langle\hat{\mu}\mathbf{E}\rangle_{12}=\tfrac{1}{2}\omega_1\hbar e^{i\omega t+i\alpha(t)},\ \omega_1=2\mu_{12}E_0/\hbar \quad (1.2)$$

in the perturbation operator $V(t)$ ($H=H_0+V$) differs from zero, then, by choice of the phases of representation (of the appropriate system of coordinates) one can always arrange it that the matrix element of the dipole moment would be a real, positive definite quantity. The time change of this perturbation has a random character because of the drift of the phase $\alpha$, which changes jumpwise over an interval $\Delta t$, the length of which is given by the time distribution of the path length in the radiating gas:

$$dW(\Delta t)=\exp\left[-\frac{\Delta t}{\tau_0}\right]\frac{d(\Delta t)}{\tau_0}.$$

The change in the density matrix of the atom under the action of the perturbation just described is given by the equations

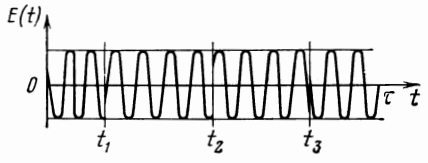


FIG. 1. Lorentz wave.

$$i\frac{d(\sigma_{11}-\sigma_{22})}{dt}=\omega_1\{\sigma_{12}e^{-i\Delta\omega t-i\alpha(t)}-\sigma_{12}^*e^{i\Delta\omega t+i\alpha(t)}\}, \quad (1.3a)$$

$$i\frac{d\sigma_{12}}{dt}=\frac{1}{2}\omega_1(\sigma_{11}-\sigma_{22})e^{i\Delta\omega t+i\alpha(t)}, \quad (1.3b)$$

where $\Delta\omega=\omega-\omega_0$, $\omega_0$ is the frequency of the atom. We shall consider the relaxation taking place under these conditions in the populations $\sigma_{11}(t)$ and $\sigma_{22}(t)$ in the spirit of the usual problem on transitions: for $t=0$, the atom is in the lowest energy state, $\sigma_{11}=1$, $\sigma_{22}=\sigma_{12}=0$, while subsequently the populations of both terms are equalized. The simplicity of the acting noise allows us to consider the kinetics of this relaxation exactly, without application of perturbation theory.

The basic difficulty of problems of this type lies in the correct averaging of the density matrix of the atom $\sigma_{ik}(t)$ over all realizations of the actual noise [random perturbations $E(t)$]. Inasmuch as we do not know in what way one must perform this operation directly in Eq. (1.3), the necessary preliminary stage is the solution of these equations for the purpose of obtaining the functional dependence $\sigma[E(t)]$ in explicit form, a dependence by means of which one can then carry out the averaging over the different realizations of the argument. In the general case, it is not possible to integrate (1.3) in quadratures, but in the given problem this situation is actually made easier by the fact that in the limits of each interval of mono-

chromaticity ($\alpha$ = const) such a solution is found without difficulty.[1] Moreover, if we had a diagonal matrix in the initial interval, then subsequently the difference of its diagonal elements

$$n(t) = \sigma_{11}(t) - \sigma_{22}(t) = n(0)X(t) \tag{1.4}$$

depends only on the value of these elements at the initial instant of time: $n(0) = \sigma_{11}(0) - \sigma_{22}(0)$, but does not depend on the phase of the real noise $\alpha$. The universal function $X(t) = 1 - 2P_{12}$, where $P_{12}(t)$ is the probability of finding the atom in the second state if at $t = 0$, it was entirely located in the first state. This probability necessarily oscillates ("nutates") in time according to the well known dynamic law,[1] so that

$$X(t) = 1 - 2\frac{\omega_1^2}{\Omega^2}\sin^2\frac{\Omega t}{2}, \tag{1.5}$$

where $\Omega = (\Delta\omega^2 + \omega_1^2)^{1/2}$ is the nutation frequency. This information is sufficient to carry out averaging with complete rigor.

We separate the set of realizations $E(t)$ in which k changes in phase take place in the interval $(0\tau)$ in the successive moments of time $t_1, t_2, \ldots, t_k$ (Fig. 2). In the interval $(0t_1)$ the change in $n(t)$ in any of these realizations chosen, which differ from one another only in the different sequence of phases, will be determined by Eq. (1.4) with $n(0) = 1$, in correspondence with the initial condition of the problem. The different realizations $\sigma_{ik}(t)$ at the time $t_1$ will therefore differ only by the phase of the nondiagonal element $\sigma_{12}$, remaining identical in all other respects. The value of the phase factor in $\sigma_{12}$ is as a whole determined by the phase of the perturbation $\alpha$ which was realized in the interval $(0t_1)$. The further behavior of the system in the interval $(t_1, t_2)$ will now depend on this phase, which controls the accumulation of $\sigma_{12}$ in the interval $(0t_1)$, and on the new phase of the perturbation $\alpha'$ which replaces it. Since at the beginning of the new interval $\sigma_{ik}(t_1)$ is no longer diagonal in an arbitrary realization of the process, it is impossible to expect such a simple change in $n(t)$ as in (1.4).

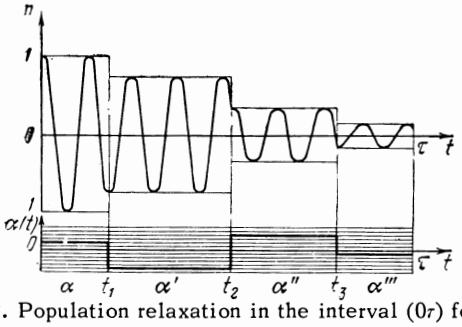


FIG. 2. Population relaxation in the interval $(0\tau)$ for k = 3. In the lower part of the figure the shading indicates the range of possible phases; the bold line indicates its sequential changes in one realization of perturbation consistent with the indicated distribution of the times $t_1$, $t_2$, and $t_3$.

However, inasmuch as any $\alpha'$ can be preceded by any given value of $\alpha$ (there is no phase correlation whatever between the trains), it is much simpler to obtain immediately an idea of the average change $\overline{n}(t)$ by first averaging over all $\alpha$. Because of this averaging, which does not affect $\sigma_{11}$ and $\sigma_{22}$, and consequently does not affect n, $\overline{\sigma}_{12}(t, \alpha)$ vanishes (since $dW(\alpha) = d\alpha/2\pi$), and it is again necessary to take the diagonal matrix as an average initial condition in $(t_1, t_2)$. This makes possible the repeated use of (1.4), but with $n(0) = n(t_1)$. The entire reasoning can be repeated:

$$n(t_2) = n(t_1)X(t_2 - t_1),$$

$$n(t_3) = n(t_2)X(t_3 - t_2), \ldots, n(\tau) = n(t_k)X(\tau - t_k).$$

Following this logic, one can easily trace the behavior of $n(\tau)$ for any number of phase shifts: $n(\tau) = X(t_1)X(t_2 - t_1)\ldots X(\tau - t_k)$. Although the average sign is omitted everywhere, it is necessary to keep it in mind that this value has already been averaged over the k + 1 phase of the perturbation $\alpha, \alpha', \alpha'', \ldots$ for a given arrangement $t_1, t_2 \ldots, t_i, \ldots, t_k$.

Inasmuch as all the phase shifts (collisions in the radiating gas) are statistically independent events, we can determine the probability of the given arrangement $t_i$ in the interval $(0\tau)$ as

$$dW(t_1, t_2, \ldots, t_k; \tau) = \exp\left(\frac{t_k - \tau}{\tau_0}\right)\prod_{i=1}^{k}\exp\left(\frac{t_{i-1} - t_i}{\tau_0}\right)\frac{dt_i}{\tau_0}$$

$$= \exp\left(-\frac{\tau}{\tau_0}\right)\prod_{i=1}^{k}\left(\frac{dt_i}{\tau_0}\right). \tag{1.6}$$

Taking it into account that any of these k − 1 times can change in the interval $0 \le t_i \le t_{i+1}$, and the last $t_k$ from 0 to $\tau$, and assuming that in the interval $(0\tau)$ there can take place 0, 1, 2, ..., k, ... up to $\infty$ phase shifts, we get for the values of $n(\tau)$ averaged over all of these possibilities

$$n(\tau) = e^{-\tau/\tau_0}\Big\{X(\tau) + {\sum_{k=1}^{\infty}}' \frac{1}{\tau_0^k}\int_0^{\tau} dt_k X(\tau - t_k)$$

$$\times \int_0^{t_k} dt_{k-1}X(t_k - t_{k-1})\int_0^{t_{k-1}}\ldots\int_0^{t_2} dt_1 X(t_1)\Big\}. \tag{1.7}$$

Multiplying both sides by $X(T - \tau)e^{\tau/\tau_0}d\tau/\tau_0$, and integrating over $\tau$ from 0 to T, we get the integral kinetic equation

$$n(T)e^{T/\tau_0} = X(T) + \frac{1}{\tau_0}\int_0^T X(T-\tau)n(\tau)e^{\tau/\tau_0}d\tau. \quad (1.8)$$

The simplicity of the kernel makes it possible to eliminate (by three successive differentiations) the integral part of the equation and reduce it to a differential equation:

$$n''' + \frac{2}{\tau_0}n'' + \left(\frac{1}{\tau_0^2} + \Delta\omega^2 + \omega_1^2\right)n' + \frac{\omega_1^2}{\tau_0}n = 0. \quad (1.9)$$

The initial conditions determined in passing have the form

$$n(0) = 1, \quad n'(0) = 0, \quad n''(0) = -\omega_1^2. \quad (1.10)$$

The general solution of (1.9) with account of these conditions can be represented in the form

$$n(t) = \frac{z_2z_3 - \omega_1^2}{(z_2 - z_1)(z_3 - z_1)}e^{z_1t} + \frac{z_3z_1 - \omega_1^2}{(z_3 - z_2)(z_1 - z_2)}e^{z_2t} + \frac{z_1z_2 - \omega_1^2}{(z_1 - z_3)(z_2 - z_3)}e^{z_3t}, \quad (1.11)$$

where $z_1$, $z_2$, $z_3$ are the roots of the characteristic equation of third degree:

$$z\left(z + \frac{1}{\tau_0}\right)^2 + \omega_1^2\left(z + \frac{1}{\tau_0}\right) + \Delta\omega^2 z = 0. \quad (1.12)$$

Thus the exact description of the relaxation under the action of a Lorentz wave reduces to the solution of an algebraic problem.

The solution of (1.11) does not contain spontaneous emission of light, since the electromagnetic field has been considered quasiclassically from the very beginning. For this reason, the kinetics of the process is correctly described by them only at times which are small in comparison with the time of spontaneous decay, and then only in the case in which the damping decrement of (1.11) is much shorter than it. This limitation, which is unimportant in the microwave region, must have a significant effect in the optical range.

## 2. CORRESPONDENCE WITH THE MODEL OF THE TRANSITIONS

In the framework of the simplest statistical scheme of transitions induced by light, this same relaxation process is described by the equation

$$dn/dt = -2Wn, \quad (2.1)$$

which gives a simple aperiodic solution $n = e^{-2Wt}$ as an alternative to (1.11). The value of the transition probability

$$W = \frac{2\pi}{\hbar^2}\mu_{12}^2\rho(\omega_0) = 2\frac{\mu_{12}^2E_0^2}{\hbar^2}\frac{\tau_0}{1 + \Delta\omega^2\tau_0^2} = \frac{1}{2}\frac{\omega_1^2\tau_0}{1 + \Delta\omega^2\tau_0^2} \quad (2.2)$$

is obtained by time-dependent perturbation theory applied to this problem which, in combination with certain statistical assumptions,[2] frequently implicit, makes it possible to establish the fundamental kinetic equation partially. The presence of an exact solution however makes it possible to go somewhat further—to make clear by direct comparison the degree of imperfection and the region of applicability of the results obtained even approximately according to an effective perturbation method.

First let us turn attention to the fact that the solution (1.11) generally contains oscillatory components—a consequence of the dynamic process which completely disappears in the crude statistical scheme (2.11). The oscillating relaxation regime does not exist everywhere; in a narrow region near resonance the process has an aperiodic character, but even in this case, (1.11) differs appreciably from (2.1), since it contains three exponents with different decay parameters. A simple analysis of Eq. (1.12), carried out at the suggestion of Ya. M. Buzhdan to the author, shows that the equation of the curve separating the periodic and aperiodic solutions and representing those points on the plane $\Delta\omega/\omega_1$, $1/\omega_1\tau_0$ (Fig. 3) at which there exists a double degeneracy of the roots of the characteristic equation, has the form

$$1/\omega_1\tau_0 = [(3 \mp \lambda)^3/8(1 \mp \lambda)]^{1/2},$$
$$\lambda = [1 - 8(\Delta\omega/\omega_1)^2]^{1/2}. \quad (2.3)$$

The upper sign in this formula refers to the rising branches of the curve, and the lower to its horizontal branch. It is significant that the regime of damped oscillations is realized not only under conditions of relatively strong perturbation $\omega_1\tau_0 \gg 1$, but also for the very weak case; the only exception is resonance relaxation ($\Delta\omega = 0$), which preserves its aperiodic character even as $\omega_1 \to 0$.

Although one can speak only of an approximate

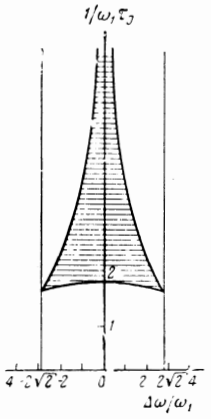


FIG. 3. Regions of periodic and aperiodic solutions (the aperiodic region is shaded).

correspondence of (1.9) and (2.1) in the arbitrary case, it is natural to expect that, at small $\omega_1$, the solutions of these equations must differ only trivially from one another. The roots of the characteristic equation corresponding to this limiting situation are equal to 0 and $-1/\tau_0$ in the zeroth approximation, whereas, in the next approximation,

$$z_1 = -2W, \quad z_{2,3} = -\frac{1}{\tau_0} - \frac{z_1}{2} \pm i\left(\Omega^2 + \frac{z_1}{\tau_0} + \frac{3}{4}z_1^2\right)^{1/2}$$

$$\approx -\frac{1}{\tau_0} + W \pm i(\Delta\omega^2 - W^2)^{1/2}, \qquad (2.4)$$

where W is determined in the same fashion as in (2.2). This approximation can be completely satisfied if $|z_1| \ll 1/\tau_0$, that is, if the inequality

$$2W\tau_0 \ll 1, \qquad (2.5)$$

is satisfied, which coincides with the ordinary condition of applicability of perturbation theory.

However, this result can also be improved. Thus, in the second approximation for $z_1$, which is necessary for the estimate of $z_{2,3}$ in (2.4), we have

$$z_1 = -2W\left[1 + 2W\tau_0 \frac{1 - \Delta\omega^2\tau_0^2}{1 + \Delta\omega^2\tau_0^2}\right]. \qquad (2.6)$$

It is not without interest to note that the correction of this approximation appearing in (2.6) cannot be obtained in successive orders of perturbation theory, leading to (2.2). This is connected with the fact that both the approximate solution of the dynamic problem and the rough averaging considered as a statistically independent random perturbation of the system and its reaction on itself underlie the perturbation method.[2]

Depending on the sign of the integrand in (2.4), we have either three real roots or one real and two complex roots. Accordingly, substitution of (2.4) in (1.11) gives either

$$n(t) = [1 + 2W\tau_0]\, e^{-2Wt}$$

$$- 2W\tau_0\, e^{-t/\tau_0}\left\{1 + \frac{\Delta\omega^2}{\omega_1^2}[1 - e^{-2Wt}]\right\} \qquad (2.7)$$

for the aperiodic region, or

$$n(t) = \left[1 - 2W\tau_0 \frac{\delta^2 - 1}{\delta^2 + 1}\right] e^{-2Wt} + 2W\tau_0\, e^{-t/\tau_0}$$

$$\times \left[\frac{\delta^2 - 1}{\delta^2 + 1}\cos\Delta\omega t + \frac{2\delta}{\delta^2 + 1}\sin\Delta\omega t\right] \qquad (2.8)$$

for the regime of damped oscillations ($\delta = \Delta\omega\tau_0$). In either case of departure from ideal kinetics, $n(t) = e^{-2Wt}$ holds only at the beginning of the process, in a small interval of the order of $\tau_0$; subsequently, in a scale of time commensurate with the relaxation time $(2W)^{-1}$, only the first component which represents the asymptote of $n(t)$ is preserved; this is identical with the ideal case upon neglect of corrections of the order of $W\tau_0$ and less.

Solution of (2.7) and (2.8) satisfactorily describes the physical situation which corresponds to points lying in the upper part of the plane (Fig. 3) but becomes inapplicable for a sufficiently close approach to the abscissa ($\omega_1 \to \infty$). In order to obtain a representation of the character of the deviations from ideal kinetics which take place here, let us consider another approximate solution, valid for

$$\omega_1\tau_0 \gg 1. \qquad (2.9)$$

The roots of (1.12) corresponding to this limiting situation are equal to

$$z_1 = -\frac{\omega_1^2}{\Omega^2\tau_0}, \quad z_{2,3} = -\frac{1}{2\tau_0}\left[1 + \frac{\Delta\omega^2}{\Omega^2}\right] \pm i\Omega, \qquad (2.10)$$

while the relaxation process is described by the oscillating function

$$n(t) = \frac{\Delta\omega^2}{\Omega^2}\exp\left(-\frac{\omega_1^2 t}{\Omega^2\tau_0}\right) + \frac{\omega_1^2}{\Omega^2}\exp\left[-\frac{t}{2\tau_0}\left(1 + \frac{\Delta\omega^2}{\Omega^2}\right)\right]$$

$$\times\left\{\cos\Omega t + \frac{\Omega^2 + 3\Delta\omega^2}{2\Omega^3\tau_0}\sin\Omega t\right\}, \qquad (2.11)$$

inasmuch as the aperiodic regime under these conditions is not realized everywhere, as is evident from (2.9) and Fig. 3. The solution (2.11) propagates in two essentially different regions. For $\Delta\omega \gg \omega_1$, the conditions (2.5) and (2.9) are compatible and therefore it is natural that, setting $\Omega \approx \Delta\omega$ in (2.11), and taking it into account in (2.8) that $\Delta\omega\tau_0 \gg \omega_1\tau_0 \gg 1$, one can reduce these formulas to identical correspondence.

An entirely different situation arises for

$$\omega_1 \gg \Delta\omega. \qquad (2.12)$$

The condition (2.5) is violated in this case and we get

$$n(t) = \frac{\Delta\omega^2}{\omega_1^2}e^{-t/\tau_0} + \left(1 - \frac{\Delta\omega^2}{\omega_1^2}\right)e^{-t/2\tau_0}\left[\cos\Omega t + \frac{1}{2\omega_1\tau_0}\sin\Omega t\right]$$

$$\approx e^{-t/2\tau_0}\cos\Omega t, \qquad (2.13)$$

that is, the regime of damped oscillations which can in no way be put in correspondence with the model of the transitions. On the other hand, it recalls very much the purely dynamic process (1.5) induced by monochromatic radiation. In view of (2.9) and (2.12), $\Omega\tau_0 \gg 1$ and, consequently, the atom can undergo many times the transition from one state to the other before the range of

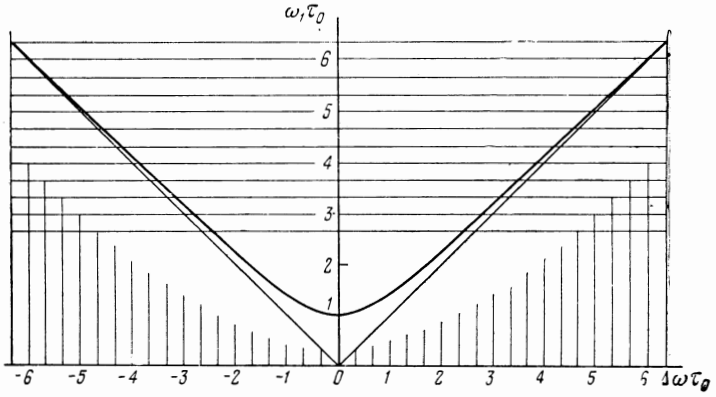


FIG. 4. Limit of applicability of perturbation theory (bold line). Solutions (2.7) and (2.8) are real in the region of vertical shading, (2.11) – in the region of horizontal shading.

monochromaticity is completed. Under these conditions, the shift in the phase of the perturbation bringing about the reduction (which gives the meaning of population to the diagonal elements of the density matrix) forces the atom into one of the possible states with a probability $\frac{1}{2}$. As a result, the damping decrement in (2.13) is twice as large as $\tau_0$.

Thus the sufficient condition for the reducibility of (1.9) to (2.1)

$$\omega_1^2\tau_0^2 \ll 1 + \Delta\omega^2\tau_0^2, \tag{2.14}$$

which follows from (2.5) is at the same time the necessary condition. The upper limit of the light intensity for which the relaxation process can still be regarded within the framework of the transitions model is established by these conditions. As is seen from Fig. 4, the region in which this condition is satisfied is by no means universal. Beyond its limits are physical situations which are of undoubted interest in spectroscopy, which uses powerful radiation sources with a narrow spectrum.

## 3. EXACT RESONANCE

As is seen from (2.13) and Fig. 4, under the conditions of resonance perturbation, with which one most frequently deals in a real experiment, the deviations from ideal kinetics brought about by the increase in power set in earlier than in any other case, and are most clearly expressed. The resonance relaxation is described by the approximate formulas (2.7) and (2.13) only in limiting situations: $\omega_1\tau_0 \ll 1$, and $\omega_1\tau_0 \gg 1$, respectively, whereas in the intermediate region, $\Delta\omega = 0$, neither the one nor the other is propagated. Fortunately, at $\Delta\omega = 0$, one can obtain an entirely rigorous solution, since (1.2) can be solved exactly:

$$z_1 = -\frac{1}{\tau_0}, \quad z_{2,3} = -\frac{1}{2\tau_0}[1 \mp (1 - 4\omega_1^2\tau_0^2)^{1/2}]. \tag{3.1}$$

Upon substitution of (3.1) in the general solution (1.11), the coefficient for the first component vanishes, and the two remaining yield

$$n(t) = \frac{1 - 2\gamma + (1-4\gamma)^{1/2}}{1 - 4\gamma + (1-4\gamma)^{1/2}} \exp\left[-\frac{t}{2\tau_0}(1 - (1-4\gamma)^{1/2})\right] + \frac{1 - 2\gamma - (1-4\gamma)^{1/2}}{1 - 4\gamma - (1-4\gamma)^{1/2}} \exp\left[-\frac{t}{2\tau_0}(1 + (1-4\gamma)^{1/2})\right], \tag{3.2}$$

where $\gamma = \omega_1^2\tau_0^2$. It is easy to establish the fact that for $\gamma \ll 1$, this expression reduces to (2.7), while for $\gamma \gg 1$, to (2.13).

Inasmuch as the oscillatory relaxation regime at $\omega_1\tau_0 \gg 1$ is essentially the new result of the exact equation (1.9), it is necessary in contrast with (2.1), to visualize in what measure this result is connected with the specific noise chosen—the Lorentz radiation. For this case we shall consider a resonance relaxation under the action of a perturbation of very general form $E(t) = E_0(t) \exp[i\omega_0 t + i\alpha(t)]$. It is described by the system of equations

$$i\frac{d[\sigma_{11} - \sigma_{22}]}{dt} = \frac{2\mu_{12}}{\hbar}E_0(t)\{\sigma_{12}e^{-i\alpha(t)} - \sigma_{12}^* e^{i\alpha(t)}\}, \tag{3.3a}$$

$$i\frac{d\sigma_{12}}{dt} = \frac{\mu_{12}}{\hbar}E_0(t)[\sigma_{11} - \sigma_{22}], \tag{3.3b}$$

which, in spite of the significant simplification in comparison with (1.3), achieved at the price of a restriction to the case $\Delta\omega = 0$, is generally speaking not solved. In this respect the exact solution (3.2) is only a partial success, since it is valid only for $E_0(t) = \text{const}$, and for such a special phase change which is an extreme simplification of the natural picture. In reality, the phase change of the radiation in the gas is due to the random frequency shifts of the atom at the moment of collision, generally speaking, these are not at all instantaneous, and do not entirely remove the phase correlation between neighboring trains. Unfortunately, each complication of the time dependence $\alpha(t)$ eliminates the application

of the method, developed in the present article and is evidently associated with insurmountable difficulties. However, one can point out such a type of random perturbation—a wave with unchanging phase, but with randomly changing amplitude (in time)—which in some sense is an alternative to what was considered, and at the same time admits an entirely rigorous averaging under conditions of resonance. Actually, introducing the variables $n = \sigma_{11} - \sigma_{22}$ and $m = -2 \operatorname{Im} \sigma_{12}$, we get from (3.3) (for $\alpha = 0$):

$$\frac{dn}{dt} = -\frac{2\mu_{12}}{\hbar} E_0(t) m = -\omega_1(t) m, \tag{3.4a}$$

$$\frac{dm}{dt} = \frac{2\mu_{12}}{\hbar} E_0(t) n = \omega_1(t) n. \tag{3.4b}$$

Reduction of the problem to such equations, which have the evident solution

$$n = \cos\left[\int_0^t \omega_1(t')\,dt'\right], \quad m = \sin\left[\int_0^t \omega_1(t')\,dt'\right], \tag{3.5}$$

becomes possible only at conditions of resonance, since for any other case the presence of the time factor $e^{i\Delta\omega t}$, as in (1.3), does not allow us to separate it from the real part of $\sigma_{12}$. On the other hand, the solution of (3.5), to which is added the dynamic part of the problem, and which establishes the functional dependence $n(t)$ on $\omega(t)$ in explicit form, is real for any form of $\omega_1(t)$. As has already been pointed out, the existence of such a solution makes it possible to proceed to the standard method of the theory of random processes, while in the given specific case, we are treating with fluctuations of the modulation of a harmonic oscillation with frequency $\omega_1(t)$.

Although we have a certain freedom of choosing the type of random process $\omega_1(t)$, it is physically clear that when one is concerned with fluctuations of the amplitude of radiation $E_0(t)$, it is more reasonable to consider this process to be normal (jumpwise changes would in this case be unnatural). If the random process is centered, by separating the mean value of the amplitude $\bar{E}_0$ from it,

$$E(t) = [\bar{E}_0 + \Delta E_0(t)]\, e^{i\omega t},$$

where $\Delta\bar{E}_0(t) = 0$, then it is seen that

$$n = \cos\left[\bar{\omega}_1 t + \int_0^t \Delta\omega_1\,dt'\right], \quad \bar{\omega}_1 = \frac{2\mu_{12}\bar{E}_0}{\hbar},$$

$$\Delta\omega_1(t) = \frac{2\mu_{12}}{\hbar}\Delta E_0(t). \tag{3.6}$$

If the fundamental characteristics of the normal random perturbation of frequency $\Delta\omega(t)$ are given:

$$\overline{\Delta\omega} = 0, \quad \overline{\Delta\omega^2} = d, \quad K(\tau) = \overline{\Delta\omega_1(t)\,\Delta\omega_1(t+\tau)} = dr(\tau),$$

$$\tau_0 = \int_0^\infty r(\tau)\,d\tau, \tag{3.7}$$

then the solution of the problem in the very general case has the form [3]

$$n(t) = \exp\left[-\frac{d}{2}\int_0^t\int_0^t r(s_1 - s_2)\,ds_1\,ds_2\right]\cos\bar{\omega}_1 t. \tag{3.8}$$

The quantity $\tau_0$, defined in (3.7), represents the correlation time of the effective noise, generalized to the case in which its normal correlation coefficient is not exponential, as in (1.1).

If $t \gg \tau_0$, then the general expression (3.8) is well approximated by the approximate form

$$n(t) = e^{-d\tau_0 t}\cos\bar{\omega}_1 t. \tag{3.9}$$

It is not difficult to identify the decay parameter which appears in this asymptotic expression with the general definition of the transition probability (2.2) in which, however, it is necessary to use the previously centered noise: $\Delta E(t) = \Delta E_0(t)\, e^{i\omega t}$ (earlier, this requirement was satisfied automatically because of the phase disorder). In this case, we have

$$2W = \frac{4\pi\mu_{12}^2}{\hbar^2}\rho_{\Delta E}(\omega_0) = \left(\frac{\overline{2\mu_{12}\Delta E_0}}{\hbar}\right)^2\int_0^\infty r(t)\,dt$$

$$= \overline{\Delta\omega_1^2}\,\tau_0 = d\tau_0. \tag{3.10}$$

It is evident that Eq. (3.9) is a satisfactory description in practice on all time axes if the following inequality is satisfied

$$\overline{\Delta\omega_1^2}\,\tau_0^2 \ll 1, \tag{3.11}$$

This expression is identical in meaning with (2.14) but takes into account the specifics of the given problem. This specific character lies in the fact that we here have three independent parameters: $\tau_0$, d, and $\bar{\omega}_1$, whereas in the Lorentz form there are two, $\tau_0$ and $\omega_1$ ($d = \omega_1^2$). Therefore, in spite of the identical character of the decay parameter in (3.9) with the quantity considered according to perturbation theory (3.10), we still do not have the right to claim that the kinetics of the process are close to ideal (non-oscillating). This happens only if the additional condition $\bar{\omega}_1\tau_0 \ll 1$, which does not follow from (3.11), is satisfied.

For small $t \ll \tau_0$, behavior of $n(t)$ is approximately described by the formula

$$n(t) = e^{-dt^2/2}\cos\bar{\omega}_1 t, \tag{3.12}$$

which can be used in practice everywhere if $\overline{\Delta\omega_1^2}\tau_0^2 \gg 1$, that is, if the process is actually

ended at the time when the asymptotic representation (3.9) goes into effect. It is evident that in this case the specifics of the problem appear more sharply—the decay of the state takes place according to a Gaussian law (not exponentially), and the decay parameter d/2 does not change upon subsequent lengthening of $\tau_0$, in contrast with that which was the case in (2.13). However, in this case, as earlier, there is an oscillating relaxation regime with period $\omega_1^{-1}$.

From a comparison of the two perturbations of different character considered, it is possible to draw a conclusion which has evidently a general character: in the limits of its applicability, the perturbation method gives a universal recipe for calculation of the asymptotic decay parameter in the first non-vanishing approximation, whereas the decrement and the character of the damping beyond these limits depend essentially on the specific character of the actual noise, and their estimate requires a rather rigorous special consideration.

[1] L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics) Gostekhizdat, 1948, p. 171.

[2] A. I. Burshteĭn, FTT **5**, 1244 (1963), Soviet Phys. Solid State **5**, 908 (1963).

[3] L. A. Vainshteĭn and V. D. Zubakov, Vydelenie signalov na fone sluchaĭnykh pomekh (Separation of signals from the noise of random interferences) Soviet Radio, Moscow, 1960, Ch. X.

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