RELAXATION IN A SYSTEM SUBJECTED TO SUDDENLY CHANGING PERTURBATIONS IN THE PRESENCE OF CORRELATION BETWEEN SUCCESSIVE VALUES OF THE PERTURBATION

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Submitted to JETP editor 28 January 1966

J. Exptl. Theoret. Phys. (U.S.S.R.) **51**, 1071–1083 (April, 1966)

A method is developed for averaging the characteristics of an arbitrary linear system subjected to the action of a random perturbation of the Markov type. A closed set of integro-differential equations for the averaged density matrix is obtained by means of the method. The behavior of a free atom and an atom in a medium interacting with a monochromatic wave whose phase changes from time to time is considered as an illustration of the possibilities of the method. The method is developed in such a way that the process can be described rigorously even if the phase is not completely changed or even if it remains the same during all the intermediate situations.

RECENTLY one of the authors[1] considered the relaxation produced in a system whose Hamiltonian changes suddenly (accidentally), but remains constant or varies regularly in the intervals between the jumps (Fig. 1). It was possible to describe the average behavior of a system perturbed by a process of this type with perfect rigor by an integral equation of the Volterra type, and several particular problems could be solved on this basis. It was assumed that no correlation existed whatever between the succeeding values of the Hamiltonian, before and after the jump.

In many physical problems this assumption is close to the true situation. It is frequently assumed that a Maxwellian distribution is established in the gas after each collision of the molecules. Then the probability of any particular velocity after the collision does not depend on its magnitude before the collision. This justifies the consideration of the Doppler broadening of the line within the framework of the previously developed formalism.[1] At the same time, a certain correlation between the velocities before and after the collision does exist,[2] and no matter how insignificant it may be, it raises in principle the question of how to further develop the method so as to permit allowance for this correlation. Such an improvement of the method is all the more necessary if each succeeding value of the Hamiltonian depends on the preceding one.

In particular, it is very important to get rid of the assumption that there is no phase correlation in succeeding monochromatic trains—Lorentz radiation, used as the perturbation for the analysis of the interaction between light and an atom in [1,3]. This assumption, while approximately valid for the radiation in the optical band, turns out to be unrealistic in low-frequency magnetic spectroscopy. The point is that the electric interactions are so strong that, in practice, each collision causes a complete collapse of the phase, whereas the magnetic interactions, to the contrary, are too weak to change the phase in one impact. The phase correlation arising in the latter case between the neighboring wave trains complicates the radiation absorption process, which can consequently be described only if the formalism is suitably developed.

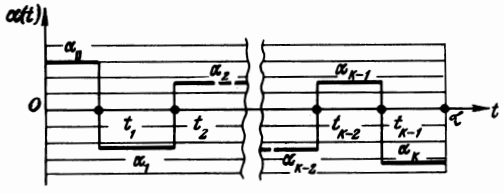

FIG. 1. Variation of random variable in one of the realizations in the absence of correlation.

The referred-to generalization of the method entails only a certain complication of the statistics. Therefore in Sec. 1 of the present article we obtain, by the same method as earlier,[1] an integro-differential equation describing the behavior of the density-matrix components. This equation takes into account the conservation of the phase memory in the perturbation acting on the system. If there values of the Hamiltonian, this equation to that obtained earlier.[1] In the opposite case, when the

correlation is completely conserved, i.e., the is no correlation whatever between the succeeding values of the Hamiltonian, this equation reduces to that obtained earlier.[1] In the opposite case, when the correlation is completely conserved, i.e., the change in the Hamiltonian has a strictly determined character, this equation turns into the usual Schrödinger equation.

By way of an illustration of the method, we consider in Sec. 2 the already mentioned problem of absorption of radiation emitted from a glowing gas by an atomic system. In this case it is possible to obtain the same linear differential equations with constant coefficients as in [1], except that they contain now a phase-correlation parameter Γ. By varying this parameter between unity and zero, it is possible to modify the composition of the acting light, from monochromatic to very highly spread-out with width $1/\tau_0$.

By the same token, the conclusions previously drawn regarding the kinetics of the process are extended in this case also to the case when the collisions in the radiating gas are weak (i.e., the phase is only slightly changed).

In Sec. 3 we show that the region of applicability of the method can be greatly broadened if we include into consideration systems not described by a Schrödinger equation, but specified by relaxation (kinetic) equations. We prove that if these systems are linear we can apply to them the formalism in practically the same form as to the narrower class of dynamic systems. Because of this, the description of relaxation under the influence of a wave with random phase could be extended in the last section to a system experiencing spontaneous decay and interacting with the medium.

1. GENERALIZATION OF THE METHOD

Assume that, as before, the Hamiltonian of the system is of the form $\hat{H} = \hat{H}_0 + \hat{V}(\alpha)$, where α is a jumplike time-varying random parameter (Fig. 1). Accordingly, the perturbation $\hat{V}$ as a function of α remains constant (or varies regularly) over times of the order of τ (distributed in accordance with the law $dW(\tau) = \tau_0^{-1} \times \exp(-\tau/\tau_0)d\tau$), experiencing jumplike changes at the end of each of them, in accordance with the new value of α. As before, the probability of finding a certain value of α at each instant of the process is specified by the same distribution, so that the perturbing random process $\alpha(t)$ is stationary:

$$dW(\alpha) = \varphi(\alpha)\,d\alpha. \tag{1.1}$$

The new factor here is that if a value α_i has already been realized in a given interval, then the probability of appearance of α_{i+1} in the next interval is determined not by (1.1), but by the conditional probability

$$dW_{\alpha_i}(\alpha_{i+1}) = f(\alpha_i, \alpha_{i+1})\,d\alpha_{i+1}.$$

It is obvious that

$$\int \varphi(\alpha_i)\,d\alpha_i = \int f(\alpha_{i-1}, \alpha_i)\,d\alpha_i = 1.$$

At the same time we must bear in mind that the choice of the functions $\varphi(\alpha_i)$ and $f(\alpha_{i-1}, \alpha_i)$ is not arbitrary. The condition for the stationarity of the process presupposes the existence between them of the integral relation

$$\varphi(\alpha_i) = \int f(\alpha_{i-1}, \alpha_i)\,\varphi(\alpha_{i-1})\,d\alpha_{i-1};$$

only in this case does the probability (1.1) remain unchanged when α changes.

The function $f(\alpha_{i-1}, \alpha_i)$ is the only new element which appears in the present variant of the theory, compared with the preceding one. The method developed earlier should now be obtained in the limiting case when

$$f(\alpha_{i-1}, \alpha_i) = \varphi(\alpha_i),$$

i.e., there is no correlation at all with the occurrences in the preceding intervals. To the contrary, if no change in α takes place between intervals, i.e., $f(\alpha_{i-1}, \alpha_i) = \delta(\alpha - \alpha_{i-1})$, then we should expect the process to develop in accordance with the Schrödinger equation (strictly dynamically), and the matter reduces to averaging its solution over α.

The probability that k changes of α actually took place in the interval (0, t) at succeeding instants $t_1, t_2, \ldots, t_k$, and that a certain sequence $\alpha_0, \alpha_1, \ldots, \alpha_k$ was realized between them, is equal to

$$dW(\alpha_0, \alpha_1, \ldots, \alpha_k; t_1, t_2, \ldots, t_k; t) = e^{-t/\tau_0} \prod_{i=1}^{k} \frac{dt_i}{\tau_0^k}\,\varphi(\alpha_0) f(\alpha_0, \alpha_1) \ldots f(\alpha_{k-1}, \alpha_k) \prod_{i=0}^{k} d\alpha_i. \tag{1.2}$$

Using the unitary transformation

$$\rho(t) = \hat{S}(\alpha, t, t')\,\rho(t')\,\hat{S}^{-1}(\alpha, t, t'),$$

which satisfies the Schrödinger equation

$$\frac{d\hat{S}}{dt} = -\frac{i}{\hbar}\hat{H}\hat{S}, \tag{1.3}$$

we can readily find the density matrix, which represents the system at the end of the interval (0, t) if k changes of α took place in it:

$$\rho(\alpha_0, \ldots, \alpha_k;\ t_1, \ldots, t_k;\ t) = S_k \ldots S_1 \rho(0) S_1^{-1} \ldots S_k^{-1},$$

where

$$S_i = S(\alpha_i, t_{i+1}, t_i), \quad t_{k+1} = t, \quad t_0 = 0.$$

Multiplying this expression by its probability (1.2), integrating over all possible positions p_i and values of the parameter α_i, and then summing over the number of jumps, we obtain the sought quantity—the density matrix averaged over all the random variables:

$$\bar{\rho} = \sum_{k=0}^{\infty} \frac{1}{\tau_0^k} \int_{\alpha_0} \ldots \int_{\alpha_k} \int_0^t \ldots \int_0^{t_2} \rho(\alpha_0, \alpha_1, \ldots, \alpha_k; t_1, t_2, \ldots, t_k; t) \times dW(\alpha_0, \alpha_1, \ldots, \alpha_k; t_1, \ldots, t_k; t). \tag{1.4}$$

The correlation existing between the α_i does not make it possible to apply to this series the transformations that previously[1] led directly to an integral equation for α. If we introduce the partial matrix $\rho(t, \beta)$, defined by

$$\bar{\rho} = \int \rho(t, \beta) \varphi(\beta) d\beta, \tag{1.5}$$

i.e., averaged only over those realizations which have identical values of $\alpha_k = \beta$ at the instant t, then we obtain from (1.4) a series for

$$\rho(t, \beta)\varphi(\beta)\exp(t/\tau_0) = \sum_{k=0}^{\infty} \frac{1}{\tau_0^k} \text{ons} \times \int_{\alpha_0} \ldots \int_{\alpha_{k-1}} \int_0^t \ldots \int_0^{t_2} S_k \ldots S_1 \rho(0) S_1^{-1} \ldots S_k^{-1} \times \prod_{i=1}^{k} dt_i\, \varphi(\alpha_0) f(\alpha_0, \alpha_1) \ldots f(\alpha_{k-2}, \alpha_{k-1}) f(\alpha_{k-1}; \beta) \prod_{i=0}^{k-1} d\alpha_i, \tag{1.6}$$

satisfying the equation

$$\rho(\tau, \alpha)\varphi(\alpha)\exp(\tau/\tau_0) = S(\alpha, \tau, 0)\rho(0)S^{-1}(\alpha, \tau, 0) + \frac{1}{\tau_0} \int_0^{\tau} dt \exp\left(\frac{t}{\tau_0}\right) \times \int d\beta\, S(\alpha, \tau, t) \rho(t, \beta) S^{-1}(\alpha, \tau, t) f(\beta, \alpha) \varphi(\beta). \tag{1.7}$$

Indeed, after multiplying (1.6) from the left and from the right by $S(\alpha, \tau, t)$ and $S^{-1}(\alpha, \tau, t)$ respectively, and averaging with weight $\tau_0^{-1} dt$ $f(\beta, \alpha)d\beta$ in the interval $(0, \tau)$ on the right, we obtain the same series as before (with the exception of the first term), but with new arguments (i.e., $\rho(\tau, \alpha)\varphi(\alpha)$), whereas on the left we obtain the integral part of Eq. (1.7). Formally, (1.7) is an integral equation of the mixed type, of the Volterra type in τ and of the Fredholm type in α, and in the latter case the limit can be infinite. Another fact which is new in comparison with the earlier paper[1] is also that this equation defines a function of two variables τ and α, and that its solution is not the final result of the calculation but must be substituted in (1.5) to calculate the complete average $\bar{\rho}(\tau)$.

For a better understanding of the gist of the matter, it is useful to represent (1.7) in the form of a system of integral equations for two equivalent functions $\rho(\tau, \alpha)$ and $\tilde{\rho}(\tau, \alpha)$:

$$\rho(\tau, \alpha) e^{\tau/\tau_0} = S(\alpha, \tau, 0)\rho(0)S^{-1}(\alpha, \tau, 0) + \frac{1}{\tau_0} \int_0 e^{t/\tau_0} S(\alpha, \tau, t) \tilde{\rho}(t, \alpha) S^{-1}(\alpha, \tau, t)\, dt, \tag{1.8a}$$

$$\tilde{\rho}(\tau, \alpha) = \int_{\beta} \frac{f(\beta, \alpha)}{\varphi(\alpha)} \rho(t, \beta)\varphi(\beta) d\beta. \tag{1.8b}$$

From the meaning of these expressions it is seen that $\tilde{\rho}(\tau, \alpha)$ is a subensemble with given α directly after the jump of the perturbation at the instant t. Thus, $\tilde{\rho}(t, \alpha)$ is the initial condition in the subensemble, which then develops regularly from t to τ, so that at the instant τ we get $S(\alpha, \tau, t) \times \tilde{\rho}(t, \alpha)S^{-1}(\alpha, \tau, t)$ from $\tilde{\rho}(t, \alpha)$. A contribution to $\rho(\tau, \alpha)$ is made by all the sub-ensembles with identical α, which differ only in the time t—the instant of the last jump of α prior to τ. Therefore $\rho(\tau, \alpha)$ is the time average (1.8a) of $\tilde{\rho}(t, \alpha)$. To the contrary, $\tilde{\rho}(t, \alpha)$ is obtained by averaging $\rho(t, \beta)$ over the variable β, which directly preceded the given value of α. Inasmuch as nothing happens to the system (to the density matrix) at the instant of a jump of the perturbation, the average (1.8b) reflects simply the reshuffling of the systems among the sub-ensembles: it takes into account the contribution made to $\tilde{\rho}(t, \alpha)$ of each $\rho(t, \beta)$ in proportion to the probability of the replacement of β by α (Fig. 2).

The sought mean value $\bar{\rho}(t)$ can be determined with equal success both in terms of $\rho(t, \beta)$ and in terms of $\tilde{\rho}(t, \alpha)$, for by virtue of relations (1.8b) and (1.2) we have

$$\bar{\rho} = \int_{\alpha} \tilde{\rho}(t, \alpha)\varphi(\alpha) d\alpha = \int_{\beta} d\beta\, \varphi(\beta) \int f(\beta, \alpha) \rho(t, \beta) d\alpha = \int \rho(t, \beta)\varphi(\beta) d\beta.$$

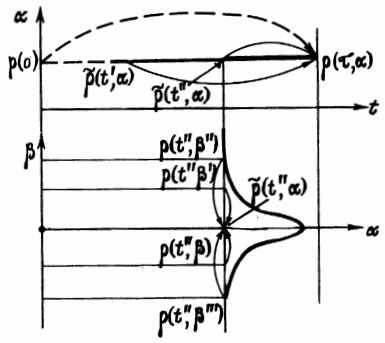

FIG. 2

However, the main difficulty consists not in this concluding averaging procedure, but in the need for solving the integral equations with respect to ρ and $\tilde{\rho}$.

We can see, however, that the integral character of Eq. (1.8a) is immaterial. By directly differentiating it with respect to τ, taking (1.3) into account, we find the differential equation equivalent to it:

$$\frac{d\rho(\tau,\alpha)}{d\tau} = -\frac{i}{\hbar}[\hat{H}(\alpha)\rho(\tau,\alpha)] - \frac{\rho(\tau,\alpha)-\tilde{\rho}(\tau,\alpha)}{\tau_0}. \tag{1.9}$$

If we represent (1.8b) in operator form as $\tilde{\rho} = \hat{T}\rho$, then the formal similarity of (1.9) to the equation describing the relaxation of the system under the influence of the instantaneous perturbation bursts (collisions) becomes obvious.[4] The specific feature and the complexity of the present problem is manifest only in the fact that in this case $\hat{T}$ is much more complicated than the collision operator, and its integral character cannot be eliminated. Nonetheless, in both cases the relaxation process develops in such a way, as if it were attempting to eliminate the difference between $\rho(\tau, \alpha)$ and $\tilde{\rho}(\tau, \alpha) = \hat{T}\rho(\tau, \alpha)$.

In the absence of correlation between successive values of α_i, i.e., when

$$f(\beta,\alpha) = \varphi(\alpha), \tag{1.10}$$

we obtain from (1.8b)

$$\tilde{\rho}(t,\alpha) = \int \rho(t,\beta)\varphi(\beta)\,d\beta = \bar{\rho}(t),$$

and taking this circumstance into account, direct integration of (1.8a) with respect to α yields

$$\bar{\rho}(\tau)e^{\tau/\tau_0} = \int_\alpha S(\alpha,\tau,0)\rho(0)S^{-1}(\alpha,\tau,0)\varphi(\alpha)\,d\alpha + \frac{1}{\tau_0}\int_0^\tau e^{t/\tau_0}\,dt\int_\alpha S(\alpha,\tau,t)\bar{\rho}(t)S^{-1}(\alpha,\tau,t)\varphi(\alpha)\,d\alpha\ldots \tag{1.11}$$

which is the result obtained in [1].

On the other hand, if

$$f(\beta,\alpha) = \delta(\alpha-\beta), \tag{1.12}$$

i.e., no random change in the perturbation takes place, then $\tilde{\rho}(\tau, \alpha) = \rho(\tau, \alpha)$ the relaxation term in (1.9) vanishes, and the equation turns into the usual Schrödinger equation.

In addition to these extreme cases, there are a number of cases in which the correlation at the instant of the jump is violated only partially, and we obtain something intermediate between the purely dynamic and the extremely uncorrelated process. The generality of the method developed here will be brought out most completely in the example considered below.

2. ACTION OF AN IMPACT-BROADENED LINE ON AN ATOM

Let us consider the emission from a radiating gas under conditions when thermal broadening of the line (due to collisions) prevails over the natural and Doppler broadening. Assuming the collisions to be adiabatic, we can visualize this radiation as consisting of a sequence of monochromatic trains of equal amplitude, interrupted in phase at the instants of collision (Fig. 3).

When such radiation acts on an atomic (two-level) system, its Hamiltonian can be represented in the form

$$\hat{H} = \hat{H}_0 + \hat{F}\exp[i\omega t + i\alpha(t)], \quad F_{ii} = 0, \quad (H_0)_{ik} = E_i\delta_{ik}, \tag{2.1}$$

where α is the random phase of the acting wave. If this phase collapses completely during the collision, then the radiation is the Lorentz wave considered earlier.[3] With the aid of the formalism developed here we can now extend the analysis to include also the case when the phase is knocked down only partially.

Let us write Eq. (1.9) for the components of the density matrix ρ_{12} and $\rho_{11} - \rho_{22} = n$:

$$\dot{\rho}_{12} = -i\omega_0\rho_{12} + i\frac{\omega_1}{2}e^{i\omega t+i\alpha}n - \frac{\rho_{12}-\tilde{\rho}_{12}}{\tau_0}, \tag{2.2a}$$

$$\dot{n} = i\omega_1[\rho_{12}e^{-i\omega t-i\alpha} - \rho_{12}^*e^{i\omega t+i\alpha}] - \frac{n-\tilde{n}}{\tau_0}, \tag{2.2b}$$

where

$$\omega_1 = 2F_{12}/\hbar, \quad \omega_0 = (E_2-E_1)/\hbar.$$

The main difficulty in the solution of equations of this type is that $\rho(\tau, \alpha)$ and $\tilde{\rho}(\tau, \alpha)$ are connected by the integral relation (1.8b). However,

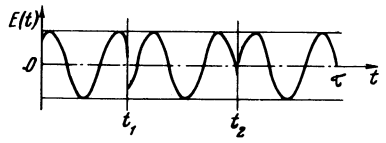

FIG. 3. Lorentz wave.

owing to the special type of the Hamiltonian (2.1) and to the obvious fact that $f(\beta, \alpha) = f(\beta - \alpha)$, this difficulty can be circumvented by establishing with the aid of (1.8b) the following identical relations between the Fourier transforms of the corresponding density matrices:

$$\tilde{\rho}(\tau, k) = \int_{-\infty}^{+\infty} e^{-ik\alpha} \tilde{\rho}(\tau, \alpha)\varphi(\alpha)d\alpha$$
$$= \Gamma_k \int_{-\infty}^{+\infty} e^{-ik\beta} \rho(\tau, \beta)\varphi(\beta)d\beta = \Gamma_k \rho(\tau, k), \qquad (2.3)$$

where, obviously

$$\Gamma_k = \int_{-\infty}^{+\infty} e^{-ik\Delta\alpha} f(\Delta\alpha)d(\Delta\alpha), \qquad (2.4)$$

and $\Delta\alpha = \beta - \alpha$.

If we now average (2.2) with a weight density $\varphi(\alpha)$, after first multiplying the first equation by $\exp(-i\alpha)$, then we can express with the aid of (2.3) all the $\tilde{\rho}(\tau, 1)$ in terms of $\rho(\tau, 1)$ and thus obtain the following system of equations for $\bar{n}(\tau)$ and $\sigma_{12}(\tau, 1) = \rho_{12}(\tau, 1)\exp(-i\omega\tau)$:

$$\dot{\bar{n}} = i\omega_1[\sigma_{12}(\tau, 1) - \sigma_{12}^*(\tau, 1)],$$
$$\dot{\sigma}_{12}(\tau, 1) = -i\Delta\omega\sigma_{12}(\tau, 1) + \frac{i\omega_1}{2}\bar{n}(\tau) - \frac{\sigma_{12}(\tau, 1)[1 - \Gamma_1]}{\tau_0}.$$

Eliminating from it $\sigma_{12}(\tau, 1)$, we obtain the final equation for the relaxation of the populations:

$$\dddot{\bar{n}} + \frac{2(1-\Gamma_1)}{\tau_0}\ddot{\bar{n}} + \left[\frac{(1-\Gamma_1)^2}{\tau_0^2} + \omega_1^2 + \Delta\omega^2\right]\dot{\bar{n}}$$
$$+ \frac{\omega_1^2(1-\Gamma_1)}{\tau_0}\bar{n} = 0. \qquad (2.5)$$

We can similarly obtain an equation for $\bar{\rho}_{12}(\tau)$. To this end it is necessary to average the equations of (2.2) after first multiplying the second of them by $e^{i\alpha}$. But since the system obtained in this case is not closed, it is necessary to obtain one more expression from (2.2a), by multiplying it by $e^{2i\alpha}$ prior to the averaging and taking its complex conjugate. We thus obtain a system of three equations:

$$\dot{\bar{\sigma}}_{12} = -i\Delta\omega\bar{\sigma}_{12} + \frac{i\omega_1}{2}n(\tau, 1),$$
$$\dot{n}(\tau, 1) = i\omega_1[\bar{\sigma}_{12} - \sigma_{21}(\tau, 2)] - \frac{n(\tau, 1)[1-\Gamma_1]}{\tau_0},$$
$$\dot{\sigma}_{21}(\tau, 2) = i\Delta\omega\sigma_{21}(\tau, 2) - \frac{i\omega_1}{2}n(\tau, 1) - \frac{\sigma_{21}(\tau, 2)[1-\Gamma_2]}{\tau_0},$$

where

$$\bar{\sigma}_{12} = \bar{\rho}_{12}(\tau)e^{-i\omega\tau}, \quad n(\tau, 1) = \int_{-\infty}^{+\infty} e^{i\alpha}n(\tau, \alpha)\varphi(\alpha)d\alpha,$$
$$\sigma_{21}(\tau, 2) = \int_{-\infty}^{+\infty} e^{2i\alpha}\sigma_{21}(\tau, \alpha)\varphi(\alpha)d\alpha.$$

Eliminating $n(\tau, 1)$ and $\sigma_{21}(\tau, 2)$, we obtain

$$\dddot{\bar{\sigma}}_{12} + \frac{2-\Gamma_1-\Gamma_2}{\tau_0}\ddot{\bar{\sigma}}_{12} + \left[\frac{(1-\Gamma_1)(1-\Gamma_2)}{\tau_0^2}\right.$$
$$\left. - \frac{i\Delta\omega(1-\Gamma_2)}{\tau_0} + \omega_1^2 + \Delta\omega^2\right]\dot{\bar{\sigma}}_{12}$$
$$+ \left[\frac{\omega_1^2(1-\Gamma_2)}{2\tau_0} + \frac{\Delta\omega^2(1-\Gamma_1)}{\tau_0}\right.$$
$$\left. - \frac{i\Delta\omega(1-\Gamma_1)(1-\Gamma_2)}{\tau_0^2}\right]\bar{\sigma}_{12} = 0. \qquad (2.6)$$

Introducing the notation

$$\frac{1}{\tau_1} = \frac{1-\Gamma_1}{\tau_0}, \quad \frac{1}{\tau_2} = \frac{1-\Gamma_2}{\tau_0}, \qquad (2.7)$$

we can reduce (2.5) and (2.6) to a more compact form

$$\dddot{\bar{n}} + \frac{2}{\tau_1}\ddot{\bar{n}} + \left[\frac{1}{\tau_1^2} + \omega_1^2 + \Delta\omega^2\right]\dot{\bar{n}} + \frac{\omega_1^2}{\tau_1}\bar{n} = 0, \qquad (2.8)$$

$$\dddot{\bar{\sigma}}_{12} + \left[\frac{1}{\tau_1} + \frac{1}{\tau_2}\right]\ddot{\bar{\sigma}}_{12} + \left[\frac{1}{\tau_1\tau_2} - \frac{i\Delta\omega}{\tau_2} + \omega_1^2 + \Delta\omega^2\right]\dot{\bar{\sigma}}_{12}$$
$$+ \left[\frac{\omega_1^2}{2\tau_2} + \frac{\Delta\omega^2}{\tau_1} - \frac{i\Delta\omega}{\tau_1\tau_2}\right]\bar{\sigma}_{12} = 0. \qquad (2.9)$$

These equations are the end result of all the averagings and contain all the information concerning the behavior of the system under the influence of a random process (radiation) of the type under consideration. Inasmuch as they do not differ from the equations obtained earlier,[1] all the information on the relaxation kinetics can be drawn from that paper.

It is now appropriate to turn to the analysis of the limiting situation represented by the distribu-

tions (1.10) and (1.12). After substituting them in (2.4) we obtain respectively

$$\Gamma_1 = \Gamma_2 = 0, \qquad \Gamma_1 = \Gamma_2 = 1,$$

for it has been assumed, as before, that $\varphi(\alpha)$ = const, i.e., all the phases at any instant of the process are equally probable. We recall that in the first case ($\Gamma_1 = \Gamma_2 = 0$) there is no correlation between the neighboring values of the phase, i.e., the phase jump can have any value with equal probability. To the contrary, when $\Gamma_1 = \Gamma_2 = 1$, no jumps occur, the phase is conserved throughout, and the averaging is meaningful only because each term of the ensemble is subject to the action of its unique monochromatic wave, which differs from the others by just this phase.

Thus, the quantities Γ_1 and Γ_2 are measures of the intertrain phase memory in the acting radiation. When there is no memory at all, the coefficients (2.7) are equal to $1/\tau_1 = 1/\tau_2 = 1/\tau_0$, and Eqs. (2.8) and (2.9) themselves reduce as a result to the corresponding equations obtained earlier,[1] as should indeed be the case. To the contrary, for a determined process $1/\tau_1 = 1/\tau_2 = 0$, all the relaxation terms in these equations vanish, leaving the purely dynamic part:

$$\dddot{\bar{n}} + \Omega^2\dot{\bar{n}} = 0, \quad \dddot{\bar{\sigma}}_{12} + \Omega^2\dot{\bar{\sigma}}_{12} = 0, \tag{2.10}$$

where $\Omega^2 = \omega_1^2 + \Delta\omega_1^2 + \Delta\omega^2$. The solution of Eqs. (2.10) under ordinary conditions[1] yields

$$\bar{n}(\tau) = 1 - 2\frac{\omega_1^2}{\Omega^2}\sin^2\frac{\Omega\tau}{2},$$

$$\bar{\sigma}_{12}(\tau) = \frac{\omega_1^2}{\Omega^2}\sin\frac{\Omega\tau}{2} + \cos\Omega\tau + \frac{i\Delta\omega}{\Omega}\sin\Omega\tau.$$

We can easily discern in the behavior of these averages the usual dynamic process—nutation—which is not distorted by averaging over the phase.

For an analysis of the intermediate situations, lying between the considered extreme cases, we can use certain model representations of the function of the phase jumps $f(\Delta\alpha)$. If, in particular, the distribution is of the Lorentz type, $\pi^{-1}a/[a^2 + (\Delta\alpha)^2]$, then we obtain from (2.4)

$$\Gamma_1 = e^{-a}, \qquad \Gamma_2 = e^{-2a} = \Gamma_1^2.$$

If the distribution is Gaussian

$$f(\Delta\alpha) = \frac{1}{a\sqrt{2\pi}}\exp\left[-\frac{(\Delta\alpha)^2}{2a^2}\right],$$

then

$$\Gamma_1 = e^{-a^2/2}, \qquad \Gamma_2 = e^{-2a^2} = \Gamma_1^4.$$

From this we see that a unique correspondence exists between Γ_1 and Γ_2, and both these quantities are exponentially small in a—the value of the phase scatter during the jump, if $a \gg 1$.

For real radiation from a gas, the form of $f(\Delta\alpha)$ is unknown, but even without going into the form of this function we can present a perfectly correct definition for the quantity Γ_k. From the meaning of the averaging (2.4) itself, we see that

$$\Gamma_k = \int e^{ik\Delta\alpha} f(\Delta\alpha)\, d(\Delta\alpha) = \overline{e^{ik\Delta\alpha}}$$

$$= 2\pi\int_0^\infty dW(v)\int_{r_0}^{\sqrt{s/\pi}} \frac{r\,dr}{s}\exp[ik\Delta\alpha(r, v)], \tag{2.11}$$

where r is the impact distance and v is the relative velocity of the molecules during the collision. The normalization constant s, which enters also in the definition of the collision frequency ($1/\tau_0 = ns\bar{v}$, where n is the gas density), is eliminated when the quantity

$$\frac{1}{\tau_k} = \frac{1-\Gamma_k}{\tau_0} = 2\pi n\bar{v}\int_0^\infty dW(v)\int_{r_0}^\infty r\,dr$$

$$\times\,[1 - \exp ik\Delta\alpha(r, v)]$$

is calculated, and therefore it becomes possible to take the limit as $s \to \infty$ in this formula.

The quantity $1/\tau_1$ has the obvious physical meaning of the width of the line of the effective radiation. This can be seen from the fact that it coincides with its usual definition and, in addition, simple calculation of the quantities

$$K(\tau) = \overline{V(\tau)V(0)} = E_0^2 e^{i\omega\tau}\,\overline{\exp[i\alpha(\tau) - i\alpha(0)]}$$

$$= E_0^2\exp(i\omega\tau - \tau/\tau_1),$$

$$\rho(\omega') = \mathrm{Re}\frac{1}{\pi}\int_0^\infty K(\tau)e^{-i\omega'\tau}\,d\tau$$

$$= \frac{E_0^2}{\pi}\frac{\tau_1}{1 + (\omega' - \omega)^2\tau_1^2}$$

confirms that $1/\tau_1$ is identical with the width of the effective spectrum. The quantity $1/\tau_2$ does not have such an analog. It is therefore remarkable that the relaxation of the populations depends only on the composition of the acting light and on nothing else, whereas the relaxation of the phases is determined in the general case not only by the correlation function (τ_1), but also by the more

subtle characteristics of the perturbing process (τ_2).

3. DEVELOPMENT OF A METHOD FOR AN ARBITRARY LINEAR SYSTEM

In the preceding sections and earlier[1,3] we confined ourselves to dynamic systems that obey the Schrödinger equation, so that we could describe their evolution in time by means of a corresponding unitary transformation. However, the existence of a unitary transformation is only a sufficient but not at all necessary condition for the application of the described method. On the other hand, it is easy to indicate systems that are described by kinetic (and not dynamic) equations of the type

$$\frac{\partial \rho_{ik}}{\partial \tau} = -\frac{i}{\hbar}[\hat{H}\rho]_{ik} - Q_{lm}{}^{ik}[\rho_{lm} - \rho_{lm}{}^{0}], \qquad (3.1)$$

and contain as before a Markov variable in the Hamiltonian. These are, for example, a spin interacting weakly with a medium,[5] or a gas atom experiencing a collision with other atoms[4] whose relaxation properties are represented in (3.1) by the operator $\hat{Q}^{ik}$, and the equilibrium state is represented by the matrix ρ^0. In some cases[6] $\rho^0(t)$ varies regularly with time, following the variation of the external field. Thus in expanding the limits of applicability of the method we should deal with a linear equation of the general type:

$$\frac{\partial \rho_{ik}}{\partial \tau} = L_{lm}{}^{ik}(\alpha, \tau)\rho_{lm} + d^{ik}(\tau), \qquad (3.1a)$$

whose solution

$$\rho_{ik}(\tau) = G_{lm}{}^{ik}(\alpha, \tau, t)\rho_{lm}(t) + D^{ik}(\alpha, \tau, t) \qquad (3.2)$$

is the analog of the unitary transformation in the preceding case, if G_{lm}^{ik} satisfies the equation

$$\frac{\partial}{\partial \tau} G_{lm}{}^{ik} = L_{rs}{}^{ik} G_{lm}{}^{rs}, \qquad (3.3)$$

and

$$D^{ik}(\alpha, \tau, t) = \int_t^\tau dt'\, d^{rs}(t') G_{rs}{}^{ik}(\alpha, \tau, t'). \qquad (3.4)$$

Using (3.2)–(3.4), we can easily construct the solution that is obtained at the instant t after k changes of α occurring in the interval (0, t):

$$\rho_{ik}(\alpha_0, \ldots, \alpha_k; t_1, \ldots, t_k; t) = X_k + Y_k + \sum_{r=1}^{k} A_r{}^k$$

$$= G_{lm}{}^{ik}(k) G_{np}{}^{lm}(k-1) \ldots G_{tq}{}^{hs}(0)\rho_{tq}(0)$$

$$+ G_{lm}{}^{ik}(k) G_{np}{}^{lm}(k-1) \ldots G_{tq}{}^{hs}(1) D^{tq}(0)$$

$$+ \sum_{r=1}^{k} G_{lm}{}^{ik}(k) G_{np}{}^{lm}(k-1) \ldots G_{tq}{}^{hs}(r+1) D^{tq}(r), \qquad (3.5)$$

where

$$G(i) = G(\alpha_i, t_{i+1}, t_i); \quad \alpha_k = \beta; \quad t_{k+1} = t; \quad t_0 = 0$$

(and the same for D(i)).

Accordingly, in the averaging of (3.5) over (1.2) (where we left out $d\alpha_k = d\beta$), it turns out that the partial matrix $\rho(t, \beta)$ is a sum of three terms

$$\rho(t, \beta)\varphi(\beta) = X(t, \beta) + Y(t, \beta) + Z(t, \beta),$$

two of which

$$X = \sum_{k=0}^{\infty} X_k \text{ и } Y = \sum_{k=0}^{\infty} Y_k$$

are precisely the same multiplications as in (1.6), and are subsequently transformed in perfect analogy, and only the last term

$$Z(t, \beta) = \sum_{k=0}^{\infty} \overline{\left[\sum_{r=1}^{k} A_r{}^k\right]} = D(\beta, t, 0)\varphi(\beta)e^{-t/\tau_0} + \sum_{k=1}^{\infty}\sum_{r=1}^{k} \overline{A_r{}^k} \qquad (3.6)$$

calls for prior simplification. Carrying out the integration in $\overline{A_r^k}$ over r – 1 time values and r – 1 first parameters, on which the averaged quantity does not depend, we can replace the index r of all the internal variables in the integrals by unity, after which we obtain

$$A_r{}^k = \frac{e^{-t/\tau_0}}{\tau_0^{k-r+1}} \int_{\alpha_{k-r}} \cdots \int_{\alpha_1} \int_0^t \cdots \int_0^{t_2} G_{lm}{}^{ik}(k-r) \ldots$$

$$\ldots G_{tq}{}^{hs}(2) D^{tq}(1) \frac{t_1^{r-1}}{\tau_0^{r-1}(r-1)!}$$

$$\times \varphi(a_1) f(\alpha_1, \alpha_2) \ldots f(\alpha_{k-r}, \beta) \prod_{i=1}^{k-1} dt_i\, d\alpha_i. \qquad (3.7)$$

Substituting this result in (3.6) and changing the order of summation, we can readily note that the power-law series contract into exponentials, so that we ultimately obtain

$$Z(t, \beta) = D(\beta, t, 0)\varphi(\beta)\exp\left(-\frac{t}{\tau_0}\right) + \sum_{i=1}^{\infty}\sum_{r=1}^{\infty} \overline{A_r^{r+i-1}}$$

$$= D^{ik}(0)\varphi \exp\left(-\frac{t}{\tau_0}\right) + \sum_{k=1}^{\infty} G_{lm}{}^{ik}(k) \ldots$$

$$\cdots G_{tq}{}^{hs}(2) D^{tq}(1) \exp\left(\frac{t_1}{\tau_0}\right) = \sum_{k=0}^{\infty} \bar{Z}_{k_2} \qquad (3.8)$$

and the latter expression includes the free term in the common series under the index zero, as was done by us earlier.

It is now easy to see that $Z(t, \beta)$ is perfectly identical in structure with $X(t, \beta)$ and $Y(t, \beta)$, and the series (3.6) therefore satisfies the following integral equation:

$$\rho_{ik}(\tau, \alpha) e^{\tau/\tau_0} = G_{lm}{}^{ik}(\alpha, \tau, 0)\rho_{lm}(0) + \frac{1}{\tau_0}\int_0^{\tau} e^{t/\tau_0} G_{lm}{}^{ik}(\alpha, \tau, t)\,\tilde{\rho}_{lm}(\tau, \alpha)\,dt + D^{ik}(\alpha, \tau, 0) + \frac{1}{\tau_0}\int_0^{\tau} e^{t/\tau_0} D^{ik}(\alpha, \tau, t)\,dt, \quad (3.9)$$

which is a direct generalization of (1.7) for an arbitrary linear system. Moreover, since it is known that $G_{l\,m}^{ik}$ satisfies Eq. (3.3) and D^{ik} satisfies (3.4), we can verify by simple differentiation, just as before, that (3.9) reduces to the differential equation

$$\frac{\partial \rho_{ik}(\tau, \alpha)}{\partial \tau} = L_{lm}{}^{ik}(\tau, \alpha)\rho_{lm}(\tau, \alpha) + d^{ik}(\tau) - \frac{1}{\tau_0}[\rho_{ik}(\tau, \alpha) - \tilde{\rho}_{ik}(\tau, \alpha)] \quad (3.10)$$

with the same definition of $\tilde{\rho}(\tau, \alpha)$ as before.

4. ACTION OF IMPACT-BROADENED RADIATION ON A SUBSTANCE

The broadening attained in this manner by a class of systems in which the method is applicable allows us to describe the action of a wave with variable phase on an atom interacting with a medium, i.e., the absorption of radiation in a substance. The kinetic equations describing such a system differ from the dynamic equations by terms that describe the relaxation of the components of the density matrix under the influence of the medium. By introducing similar terms we can take into account the spontaneous deactivation of the excited states. For a two-level system this means the following modernization of the dynamic equations

$$\frac{\partial \rho_{22}}{\partial \tau} = -\frac{i}{\hbar}[\hat{H}\rho]_{22} - \frac{\rho_{22}}{T_0} - \frac{\rho_{22}}{T_1''} + \frac{\rho_{11}}{T_1'},$$

$$\frac{\partial \rho_{11}}{\partial \tau} = -\frac{i}{\hbar}[\hat{H}\rho]_{11} + \frac{\rho_{22}}{T_1''} - \frac{\rho_{11}}{T_1'},$$

$$\frac{\partial \rho_{12}}{\partial \tau} = -\frac{i}{\hbar}[\hat{H}\rho]_{12} - \frac{\rho_{12}}{T_2}. \quad (4.1)$$

Here T_0 is the spontaneous-emission time, T_1'' and T_1' are the times of adiabatic transitions under the influence of the substance from the upper level to the lower one and in the opposite direction, and T_2 is the phase relaxation time due to all these mechanisms (including adiabatic broadening).

Using (4.1) we get from (3.10)

$$\frac{\partial n}{\partial \tau} = i\omega_1[\sigma_{12}\exp(-i\alpha) - \sigma_{12}^{*}\exp(i\alpha)] - n\left[\frac{1}{T_1''} + \frac{1}{T_1'} + \frac{1}{2T_0}\right] - \left(\frac{1}{T_1''} - \frac{1}{T_1'} + \frac{1}{2T_0}\right) - \frac{n - \tilde{n}}{\tau_0}, \quad (4.2a)$$

$$\frac{\partial \sigma_{12}}{\partial \tau} = -i\Delta\omega\sigma_{12} + \frac{i\omega_1 n \exp(i\alpha)}{2} - \frac{\sigma_{12}}{T_2} - \frac{\sigma_{12} - \tilde{\sigma}_{12}}{\tau_0}. \quad (4.2b)$$

These equations, which differ from (2.2) in allowance for the spontaneous degradation and the interaction of the atom with the medium, do not complicate the calculation in any manner, and the calculation is carried in exact correspondence with the procedure described above. As a result, for example, for the relaxation of the populations we obtain an equation identical to that obtained in [7]:

$$\dddot{\bar{n}} + \left[\frac{2}{\bar{T}_2} + \frac{1}{\bar{T}_1}\right]\ddot{\bar{n}} + \left[\frac{1}{\bar{T}_2{}^2} + \frac{2}{\bar{T}_1\bar{T}_2} + \Delta\omega^2 + \omega_1{}^2\right]\dot{\bar{n}} + \left[\frac{\omega_1{}^2}{\bar{T}_2} + \frac{\Delta\omega^2}{\bar{T}_1} + \frac{1}{\bar{T}_1\bar{T}_2{}^2}\right]\bar{n} = \left(\frac{1}{\bar{T}_1\bar{T}_2{}^2} + \frac{\Delta\omega^2}{\bar{T}_1}\right) n_0, \quad (4.3)$$

in which, however, the phase relaxation times T_2

$$\frac{1}{\bar{T}_2} = \frac{1}{T_2} + \frac{1}{\tau_0} \quad (4.4)$$

and the population relaxation time T_1

$$\frac{1}{\bar{T}_1} = \frac{1}{T_1''} + \frac{1}{T_1'} + \frac{1}{2T_0} \quad (4.5)$$

are determined with allowance for the simultaneous action of the medium, the vacuum, and the broad spectral line, while n_0, defined by the relation

$$n_0\left(\frac{1}{T''} + \frac{1}{T_1'} + \frac{1}{2T_0}\right) = \frac{1}{T_1''} - \frac{1}{T_1'} + \frac{1}{2T_0}, \quad (4.6)$$

represents the stationary population of the levels in the absence of action by the light. When $T_1' = T_1'' = T_2 = T_0 = \infty$, Eq. (4.3) reduces to (2.8), and when $\tau_1 = \tau_2 = \tau_0 = \infty$ it reduces to that obtained in [7]. This equation has thus a general character and only under the conditions spelled out in [7]

does it reduce to the fundamental equation for the transition model describing the kinetics of absorption of only sufficiently weak radiation.

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Translated by J. G. Adashko
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