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Abstract

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ON THE THEORY OF RELAXATION UNDER THE ACTION OF AN EXTERNAL RANDOM FIELD

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1. In the present work the behavior of a two-level system under the action of a monochromatic wave with a randomly varying phase is studied in a case close to resonance. Such a problem arises in the study of the interaction of molecules with radiation "broadened" as a result of collisions. Under certain conditions the relaxation process of a two-level system can be described by means of a kinetic equation (of the balance-equation type—see, for example, ([1])). Of special interest in the problem considered is the question of describing the relaxation process of the system in the case when the balance equations are not valid (the latter is usually associated with a violation of the random-phase approximation). The problem described above was solved in ([2]) under the assumption that there are no temporal correlations whatever between the phases of the external field*. Below, the problem of relaxation of a two-level system induced by an external field with a randomly shifting phase is solved in a sufficiently general form, with very weak restrictions on the random law governing the behavior of the field phases. The method developed in the work admits generalization to the case of more complicated systems.

2. The equations for the components of the density matrix describing the behavior of a two-level system under the action of an external field specified in the form of a monochromatic wave with a randomly varying phase have the form

$$dn/dt = 2i\left[F^*\rho_{12}e^{i\varepsilon t+i\varphi(t)} - F\rho_{21}e^{-i\varepsilon t-i\varphi(t)}\right],$$
$$d\rho_{12}/dt = iFe^{-i\varepsilon t+i\varphi(t)}n, \qquad d\rho_{21}/dt = -iF^*e^{i\varepsilon t+i\varphi(t)}n, \tag{1}$$
$$n = \rho_{11} - \rho_{22}, \qquad \rho_{11}+\rho_{22} = 1, \qquad \varepsilon = \omega - \omega_0 \quad (\hbar = 1),$$

where ρ_{ik} are the components of the density matrix; ω is the frequency of the external field; ω_0 is the transition frequency of the two-level system; F is the off-diagonal matrix element of the perturbation; $\varphi(t)$ is the phase varying according to a prescribed random law. In writing system (1) it was assumed that

$$\varepsilon \ll \omega_0 \tag{2}$$

in accordance with the condition of nearness to resonance, and the term giving a contribution $\sim \varepsilon/\omega_0$ was discarded. Below a special form of the law of variation of the phase $\varphi(t)$ is chosen, which will make it possible subsequently to carry out the generalization to a more general case easily. Let $\varphi(t)$ have the form

$$\varphi(t) = \alpha \int_0^t \sum_k \delta(t' - t_k)\, dt', \tag{3}$$

i.e., the phase increases discontinuously at the points t_k falling in the interval $(0, t)$, each time by the amount α**. The distribution of the phase-shift points t_k (henceforth, simply kicks) is assumed random and specified in the form of a Poisson distribution. In other words, the probability of the occurrence of a kick in the interval $(t + dt, t)$ is equal to $\lambda\, dt \equiv dt/\tau_0$, or the probability

* A. I. Burshtein has informed us that at present he has obtained results more general than those in work ([2]).

** The value of the phase at $t = 0$ is included in F.

the fact that the time interval between any two successive kicks lies in the interval $(t, t + dt)$, is equal to $e^{-\lambda t} dt$. The time τ_0 has the meaning of the mean time between kicks. Our ultimate goal is to determine the ρ_{ik} averaged over the random process specified above.

3. Let us introduce the notation

$$\rho_1 = \rho_{12} e^{i\varepsilon t + i\varphi(t)}, \qquad \rho_2 = \rho_1^* \tag{4}$$

and rewrite system (1) in the new variables:

$$\dot{n} = 2i\left(F^* \rho_1 - F \rho_2\right),$$

$$\dot{\rho}_1 = i\left(\varepsilon + \alpha \sum_k \delta(t - t_k)\right)\rho_1 + iFn, \tag{5}$$

$$\dot{\rho}_2 = -i\left(\varepsilon + \alpha \sum_k \delta(t - t_k)\right)\rho_1 - iF^* n.$$

Let us now consider the phase space of the random variables (n, ρ_1, ρ_2) and find an equation describing the change with time of the distribution function $f(n, \rho_1, \rho_2, t)$. The form of system (5) makes it possible to use for this purpose a known method, employed, for example, in works ([3],[4]). Denoting, for convenience, the variables (n, ρ_1, ρ_2) by a single letter x, we write the balance equation for $f(x,t)$, describing the change in the number of points in the element of phase-space volume $dx = dn\, d\rho_1\, d\rho_2$ with coordinate x during the time dt:

$$f(x(t+dt), t+dt)\, dx(t+dt) - f(x,t)\, dx = -\lambda dt \cdot f(x,t)\, dx + \lambda dt \cdot f(\bar{x}, t)\, d\bar{x}, \quad (6)$$

where the first term on the right-hand side takes into account the departure of points from dx due to a collision, and the second term the arrival, due to a collision, from the region $d\bar{x}$ with coordinate $\bar{x}$. From equations (5) we have:

$$n(t+dt) = n + 2i(F^* \rho_1 - F\rho_2)\, dt,$$

$$\rho_1(t+dt) = \rho_1 + (i\varepsilon\rho_1 + iFn)\, dt, \qquad \rho_2(t+dt) = \rho_2 - (i\varepsilon\rho_2 + F^* n)\, dt,$$

$$\bar{n} = n, \qquad \bar{\rho}_1 = \rho_1 e^{-i\alpha}, \qquad \bar{\rho}_2 = \rho_2 e^{+i\alpha}, \quad (7)$$

$$dx(t+dt) = dx, \qquad d\bar{x} = dx.$$

Substituting (7) into (6) and passing to the limit $dt \to 0$, we obtain

$$\frac{\partial f}{\partial t} = -2i(F^*\rho_1 - F\rho_2)\frac{\partial f}{\partial n} - i\frac{\partial}{\partial \rho_1}[(\varepsilon\rho_1 + Fn)f] +$$

$$+i\frac{\partial}{\partial \rho_2}[(\varepsilon\rho_2 + F^* n)f] + \lambda f(n, \rho_1 e^{-i\alpha}, \rho_2 e^{i\alpha}, t) - \lambda f, \quad (8)$$

$$f = f(n, \rho_1, \rho_2, t).$$

Solving equation (8) is difficult for arbitrary α. We, however, need to find only the moments of the function $f(n, \rho_1, \rho_2, t)$. Multiplying (8) successively by n, ρ_1, ρ_2 and integrating over the whole phase space, we obtain:

$$\frac{d\langle n\rangle}{dt} = 2iF^*\langle\rho_1\rangle - 2iF\langle\rho_2\rangle,$$

$$\frac{d\langle\rho_1\rangle}{dt} = -iF\langle n\rangle + \left[i\varepsilon + \lambda(e^{i\alpha} - 1)\right]\langle\rho_1\rangle, \tag{9}$$

$$\frac{d\langle\rho_2\rangle}{dt} = -iF^*\langle n\rangle + \left[-i\varepsilon + \lambda(e^{-i\alpha} - 1)\right]\langle\rho_2\rangle.$$

Seeking the solution of system (9) in the form $e^{\Gamma t}$, we find

$$\Gamma^3 + 2\lambda(1 - \cos\alpha)\Gamma^2 + \left[4|F|^2 + (\varepsilon + \lambda\sin\alpha)^2 + \lambda^2(1 - \cos\alpha)^2\right]\cdot\Gamma + 4|F|^2\lambda(1 - \cos\alpha) = 0. \tag{10}$$

For $\lambda = 0$ or $\alpha = 0; 2\pi$ we obtain Γ, corresponding to the solution of system (1) in the absence of the random process (see, for example, ([5])). Each

which of the moments $\langle n\rangle$, $\langle\rho_{1,2}\rangle$ is expressed as a linear combination of solutions of the type $e^{\Gamma t}$, corresponding to the three roots of equation (10) and to the initial conditions. The problem, however, has been solved only for the diagonal elements $\langle n\rangle$, since the variables $\rho_{1,2}$ are of no interest to us. Let us now turn to averaging the off-diagonal element ρ_{12}.

4. Introduce the notation

$$\rho_0 = ne^{-i\varepsilon t - i\varphi(t)}, \qquad \rho_3 = \rho_{21}e^{-2i\varepsilon t - 2i\varphi(t)} \tag{11}$$

and rewrite system (1) in the form:

$$\frac{d\rho_0}{dt} = -i\left[\varepsilon + \alpha\sum_k \delta(t - t_k)\right]\rho_0 + 2i(F^*\rho_{12} - F\rho_3),$$

$$\frac{d\rho_{12}}{dt} = iF\rho_0, \tag{12}$$

$$\frac{d\rho_3}{dt} = -iF^*\rho_0 - 2i\left[\varepsilon - \alpha\sum_k \delta(t - t_k)\right]\rho_3.$$

Now we can consider the phase space of the random variables $(\rho_0, \rho_{12}, \rho_3)$ and obtain an equation for the distribution function $f(\rho_0, \rho_{12}, \rho_3, t)$, which will make it possible to calculate $\langle\rho_{12}\rangle$. Analogously to the derivation of equation (8), we obtain

$$\frac{\partial f}{\partial t} = -\frac{\partial}{\partial\rho_0}\left[(-i\varepsilon\rho_0 + 2iF^*\rho_{12} - 2iF\rho_3)f\right] - iF\rho_0\frac{\partial f}{\partial\rho_{12}} +$$

$$+\frac{\partial}{\partial \rho_3}\left[(2i\varepsilon\rho_3 + iF^*\rho_0)f\right] + \lambda f(\rho_0 e^{i\alpha}, \rho_{12}, \rho_3 e^{2i\alpha})e^{3i\alpha} - \lambda f, \tag{13}$$

$$f = f(\rho_0, \rho_{12}, \rho_3, t).$$

Equation (13) leads to the following equations for the moments:

$$\frac{d\langle\rho_0\rangle}{dt} = \left[-i\varepsilon + \lambda(e^{i\alpha} - 1)\right]\langle\rho_0\rangle + 2iF^*\langle\rho_{12}\rangle - 2iF\langle\rho_3\rangle,$$

$$\frac{d\langle\rho_{12}\rangle}{dt} = iF\langle\rho_0\rangle, \tag{14}$$

$$\frac{d\langle\rho_3\rangle}{dt} = -iF^*\langle\rho_0\rangle + \left[-2i\varepsilon + \lambda(e^{-2i\alpha} - 1)\right]\langle\rho_3\rangle.$$

Seeking a solution of system (14) in the form $e^{\gamma t}$, we obtain, for the determination of the three complex roots γ, the characteristic equation

$$\gamma^3 + \gamma^2\left[3i\varepsilon + \lambda(2 - e^{-i\alpha} - e^{-2i\alpha})\right] +$$

$$+\gamma\left\{4|F|^2 - 2\varepsilon^2 - i\varepsilon\lambda(e^{-2i\alpha} + 2e^{-i\alpha} - 3) + \lambda^2(e^{-i\alpha} - 1)(e^{-2i\alpha} - 1)\right\} +$$

$$+2|F|^2\left[2i\varepsilon - \lambda(e^{-2i\alpha} - 1)\right] = 0. \tag{15}$$

To avoid cumbersome expressions, we shall not write out explicitly the roots $\Gamma_{1,2,3}$, $\gamma_{1,2,3}$ of equations (10), (15). It is clear, however, that, once certain initial conditions have been specified, one can now write down the time behavior of the components $\langle\rho_{ik}\rangle$, averaged over the given random process of phase jumps.

5. Let us consider some limiting cases. First of all, let us show under what conditions a solution arises that corresponds to the random-phase approximation, i.e., to a kinetic equation of the balance-equation type. Put

$$\nu = |F|\tau_0/\sqrt{1 - \cos\alpha} \ll 1, \qquad \varepsilon = 0. \tag{16}$$

From (10), (15) we have:

$$\Gamma_{1,2} = -\frac{1 - \cos\alpha + i\sin\alpha}{\tau_0}\left[1 + O(\nu)\right], \qquad \Gamma_3 = -\frac{4|F|^2\tau_0}{(1 - \cos\alpha)^2 + \sin^2\alpha}\left[1 + O(\nu)\right],$$

$$\gamma_{1,2} \simeq \Gamma_{1,2}, \qquad \gamma_3 \simeq \frac{1}{2}\Gamma_3. \tag{17}$$

If we now choose the initial conditions $\langle n\rangle_{t=0} = 1$, $\langle \rho_{12}\rangle_{t=0} = 0$, then from (9), (14), (16), (17) it follows that

$$\langle n\rangle \approx e^{-t/\tau_R}[1 + O(\nu)], \qquad \tau_R \equiv -\Gamma_3^{-1},$$

$$\langle \rho_{12}\rangle \approx iF\tau_0\{e^{-t/2\tau_R} - e^{-|\Gamma_2|t}\}[1 + O(\nu)]. \tag{18}$$

In writing (18) we have omitted terms in $\langle n\rangle$ that decay much more rapidly with time $\left(\sim \exp\{-|\Gamma_{1,2}|t\}\right)$, and an inessential factor in $\langle \rho_{12}\rangle$ depending on α. It follows from (18) that, although at $t = 0$ the off-diagonal element was absent, subsequently it appears with a small coefficient $\sim \nu$ and relaxes to zero in a time twice as long as the relaxation time of the diagonal elements.

If at the initial time the phase φ_0 $(F = |F|e^{i\varphi_0})$ is random with distribution $W(\varphi_0)d\varphi_0 = d\varphi_0/2\pi$, then averaging over the initial phase leads to the disappearance of $\langle \rho_{12}\rangle$ at all times (see the second formula in (18)). This result corresponds to the well-known approximation of deriving the kinetic equation (6).

In the case when $\nu \gg 1$, the principal characteristic time of the problem becomes not τ_R, as in the preceding case, but τ_0. The relaxation of $\langle \rho_{ik}\rangle$ occurs over times $\sim \tau_0$ with superposed oscillations, and the approach to equilibrium is nonmonotonic. Since these results readily follow from (10), (15), we omit the corresponding formulas.

6. The results obtained are easily generalized to more general types of the random process $\varphi(t)$. Suppose that at each collision the probability that α lies in the interval $(\alpha, \alpha + d\alpha)$ is $w(\alpha)d\alpha$, the same for each collision. Then in (8) the last two terms are replaced by

$$\lambda \int_{-\pi}^{\pi} d\alpha\, w(\alpha)\{f(n, \rho_1 e^{-i\alpha}, \rho_2 e^{i\alpha}, t) - f\}. \qquad \left(\int_{-\pi}^{\pi} w(\alpha)\, d\alpha = 1\right). \tag{19}$$

Equation (10) is replaced by

$$\Gamma^3 + 2\lambda(1 - \overline{\cos\alpha})\Gamma^2 + [4|F|^2 + (\varepsilon + \lambda\,\overline{\sin\alpha})^2 + \lambda^2(1 - \overline{\cos\alpha})^2]\Gamma + 4|F|^2\lambda(1 - \overline{\cos\alpha}) = 0, \tag{20}$$

where

$$\overline{\psi(\alpha)} = \int_{-\pi}^{\pi} \psi(\alpha) w(\alpha)\, d\alpha.$$

Analogous changes are also made in equations (13)–(15). If, for example, the distribution in α (including the initial phase included in F!) is uniform, i.e. $w(\alpha) = 1/2\pi$, then we obtain the results of works (2). If, in addition to everything else, $\lambda = \lambda(\alpha)$, then in expression (19) one must put $\lambda(\alpha)$ under the integral sign, with all the consequent changes.

We note that there is a physically inessential restriction on the random process of change of the phase of the external field, connected with the neglect of terms of order ε/ω_0 in solving system (1). For the case, for example, of a Poisson distribution of collisions it has the form

$$\alpha/\omega_0\tau_0 \ll 1. \tag{21}$$

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Note: Figure translations are in progress. See original paper for figures.