RELAXATION OF QUANTUM SYSTEMS WITH EQUIDISTANT SPECTRA

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The structure of the kinetic equation (3), describing the relaxation of quantum systems with equidistant spectra, is discussed. Examples of such systems are an harmonic oscillator (with linear on nonlinear damping), a spin in a magnetic field, and a collection of two-level atoms interacting with a radiation field (Dicke's problem[1]). It is shown that equations of the type (3) preserve the positive definiteness of the density matrix. A solution of the kinetic equation is obtained for Dicke's problem, and an exact quantum expression is found for the shape of the radiation line. Interference narrowing of a line, associated with the equidistant nature of the spectrum, is discussed.

1. INTRODUCTION

KINETIC equations describing the relaxation of quantum systems were first considered by Landau[2] and Pauli;[3] a number of articles[4-8] are devoted to their general theory. If the possibility of equal or almost equal spacings between the levels is excluded, then the equation for the density matrix ρ has the form (in the interaction representation)

$$\dot{\rho}_{mm} = \sum_n \gamma_{mn}\rho_{nn} - \Gamma_m\rho_{mm}, \tag{1a}$$

$$\dot{\rho}_{mn} = -{}^1/_2(\Gamma_m + \Gamma_n + \Gamma_{mn}')\rho_{mn}, \quad m \neq n, \tag{1b}$$

where γ_{mn} is the probability for a transition from level n to level m, $\Gamma_n = \sum_m \gamma_{mn}$ is the total width of level n, Γ'_{mn} is an additional broadening associated with transverse relaxation. Equation (1a) is the equation of balance for the populations $w_n(t) = \rho_{nn}(t)$. According to Eq. (1b) the nondiagonal matrix elements $\rho_{mn}(t)$ relax independently of each other.

Relaxation in systems with equidistant spectra (an harmonic oscillator, a spin in a magnetic field, a collection of two-level atoms, etc.) possesses certain specific characteristics. Whereas the equation of balance (1a) retains its own form, the time evolution of the nondiagonal elements ρ_{mn} becomes appreciably more complicated: Intermixing of the elements ρ_{mn} having a fixed value for the energy differences $E_m - E_n$ develops.

The purpose of the present article is an investigation of the kinetic equation (3) for these systems. Particular attention is given to the so-called problem of Dicke,[1] i.e., relaxations in a system of identical two-level atoms interacting with a radiation field (see Secs. 4 and 5).

2. THE KINETIC EQUATION FOR THE DENSITY MATRIX

Let us consider a dynamical system whose Hamiltonian describing the interaction with the radiation field has the form

$$\mathcal{H}_{int} = \sum_j \{f_j A^+ b_j e^{i(\omega_0-\omega_j)t} + f_j^* A b_j^+ e^{-i(\omega_0-\omega_j)t}\} \tag{2}$$

(in the interaction representation). The operators A and A^+ refer to the system under consideration, b_j and b_j^+ are the electromagnetic field operators (j is an index which labels the mode), the f_j are the interaction constants. We note that the very possibility of writing $A(t) = Ae^{-i\omega_0 t}$ is related to the equidistant nature of the spectrum ($\hbar\omega_0$ is the energy difference between the levels); in this connection only the resonance terms $\sim\exp\{\pm i(\omega_0 - \omega_j)t\}$ which do not vanish upon averaging over a period are left in (2). The electromagnetic field plays the role of a thermostat. It is assumed that it is found in a state of thermodynamic equilibrium[1)] at a temperature T.

Using the standard techniques of the quantum theory of relaxation,[5-9] one can obtain a kinetic equation for the density matrix $\rho(t)$ of the dynamical system

$$\dot{\rho} = -i[V, \rho] + \frac{\gamma}{2}\{(\nu+1)(2A\rho A^+ - A^+A\rho - \rho A^+A) + \nu(2A^+\rho A - AA^+\rho - \rho AA^+)\}, \tag{3}$$

where

$$\gamma = 2\pi|f_j|^2\rho(\omega_j)|_{\omega_j=\omega_0}, \quad \nu = \left[\exp\left(\frac{\hbar\omega_0}{kT}\right) - 1\right]^{-1}; \tag{4}$$

$$V = \frac{\delta'}{2}(AA^+ + A^+A) + \frac{\delta''}{2}[A, A^+], \tag{5}$$

$$\delta' = P\int d\omega \frac{|f(\omega)|^2\rho(\omega)}{\omega_0 - \omega}, \quad \delta'' = P\int d\omega \frac{|f(\omega)|^2\rho(\omega)}{\omega_0 - \omega}\operatorname{cth}\frac{\hbar\omega}{2kT}.$$

Here $f_j \equiv f(\omega_j)$ and $\rho(\omega)$ is the density of states for the radiation field. The individual terms in Eq. (3) have the following physical interpretation:

1) V denotes a renormalization of the bare Hamiltonian $\mathcal{H}_0$ for the dynamical system, leading to a Lamb shift of the levels. This term is not of any interest as far as relaxation is concerned, and in what follows it is usually omitted.

2) The term $({}^1/_2)\gamma(2A\rho A^+ - A^+A\rho - \rho A^+A)$ describes the spontaneous emission of radiation.

3) The remaining terms in Eq. (3), which are proportional to ν, describe forced absorption and emission of quanta by the dynamical system.

We shall indicate several particular cases of Eq. (3).

1) The harmonic oscillator in the approximation of one-quantum absorption. In this case A = a, $\omega_0 = \omega$, where ω is the frequency of the oscillator, and Eq. (3)

[1)] The specific form of the thermostat is unessential for what follows (the form of the kinetic equation (3) does not depend on this).

turns into the kinetic equation for an oscillator with linear damping, which is considered in detail in[10].

2) For an oscillator with N-quantum absorption we have

$$A = a^N, \quad \omega_0 = N\omega,$$
$$\dot\rho = \frac{\beta}{2}\{(\nu+1)(2a^N\rho a^{+N} - a^{+N}a^N\rho - \rho a^{+N}a^N) + \nu(2a^{+N}\rho a^N - a^N a^{+N}\rho - \rho a^N a^{+N})\}. \quad (6)$$

This equation is of interest in connection with nonlinear optics.[9]

3) A particle of spin s and magnetic moment $\mu = g\mathbf{s}$ situated in a homogeneous magnetic field H possesses a system of (2s + 1) equidistant levels, $E_m = \hbar\omega_e m$ ($\omega_0 = -gH$). In order to be definite, let the gyromagnetic ratio $g < 0$; then the lowest level has $m = -s$, the highest level has $m = s$. Spin relaxation associated with magnetic dipole radiation is described by the equation

$$\dot\rho = -\frac{\gamma}{2}\{(\nu+1)(s_+s_-\rho - 2s_-\rho s_+ + \rho s_+s_-) + \nu(s_-s_+\rho - 2s_+\rho s_- + \rho s_-s_+)\}, \quad (7)$$

where $s_\pm = s_x \pm is_y$ (the z axis is chosen parallel to the magnetic field H). This equation describes "longitudinal" relaxation of the spin. Under real conditions, as is well known,[7,8] a "transverse" relaxation also exists which is not related to actual transitions between the levels. In this connection additional terms appear on the right-hand side of the kinetic equation (7) [in the simplest case, $s = 1/2$, these terms have the form $\gamma_\perp(\sigma_z^2\rho + \rho\sigma_z^2 - 2\sigma_z\rho\sigma_z)$, where $\gamma_\perp$ is the transverse relaxation constant].

4) Depolarization of μ^+ mesons in condensed media has been considered in many articles.[11-13] Due to the smallness of the μ^+ meson's magnetic moment in comparison with the electron's magnetic moment, one can neglect the direct interaction of the μ^+ spin with the medium. The hyperfine splitting of the muon, $\hbar\omega_0 \sim 0.1\,^\circ K$; therefore, at ordinary temperatures $\nu \gg 1$ and $\nu + 1 \approx \nu$. Taking this into consideration, from Eq. (3) we obtain

$$\dot\rho = -i[V, \rho] + W(\rho - s_-\rho s_+ - s_+\rho s_-), \quad (8)$$

where $V = (1/4)\hbar\omega_0 \mathbf{s}\cdot\mathbf{s}_\mu - g_e\mathbf{s}\cdot\mathbf{H}$, s is the electron spin, s_μ is the meson spin, and $W = \nu\gamma$. The kinetic equation (8) corresponds to an infinite temperature for the medium. Comparison with[11-13] indicates that the equations for the muon's density matrix used in these articles actually coincide with Eq. (8) (but the equations are written in a different form). In this connection the constant W was interpreted as the probability for spin-flip of the μ^+ meson.

3. GENERAL PROPERTIES OF THE KINETIC EQUATION

As is well known, the density matrix in quantum mechanics must satisfy three conditions:
(A) Sp ρ = 1 (normalization),
(B) $\rho = \rho^+$ (Hermitian property),
(C) $\langle\psi|\rho|\psi\rangle \geq 0$ for any arbitrary vector $|\psi\rangle$ (positive definiteness).

Since the kinetic equation (3) has a form which differs from the usual Liouville's equation,[2)] $\dot\rho = -i[H, \rho]$, the following question arises: Is Eq. (3) compatible with the requirement that conditions (A)–(C) must be satisfied at any moment of time? Verification of conditions (A) and (B) for Eq. (3) does not involve any difficulty. Therefore we shall only dwell on condition (C), which is important from a fundamental point of view (it guarantees that meaningless quantities of the negative-probability type will not arise during the time evolution of ρ). Let us introduce the notation

$$R_\psi(t) = \langle\psi|\rho(t)|\psi\rangle, \quad (9)$$
$$R(t) = \min_\psi R_\psi(t) \quad \text{for} \quad \langle\psi|\psi\rangle = 1. \quad (9a)$$

Condition (C) is equivalent to the inequality $R(t) \geq 0$.

Let us assume that conditions (A)–(C) are fulfilled for the initial density matrix $\rho(0)$. Then there exist states $|\psi_i\rangle$ such that

$$\rho(0) = \sum_i p_i|\psi_i\rangle\langle\psi_i|, \quad p_i > 0, \quad \langle\psi_i|\psi_j\rangle = \delta_{ij},$$
$$R_\psi(0) = \sum_i p_i|\langle\psi_i|\psi\rangle|^2 \geq 0, \qquad R(0) \geqslant 0. \quad (10)$$

The value of $R_\psi(\Delta t)$ for sufficiently small Δt may become negative only if $R_\psi(0) = 0$, that is, if $\langle\psi_i|\psi\rangle = 0$ for all i. In this connection, however,

$$\rho(0)|\psi\rangle = \sum_i p_i|\psi_i\rangle\langle\psi_i|\psi\rangle = 0, \quad \langle\psi|\rho(0) = 0. \quad (11)$$

Taking (11) into consideration, from Eq. (3) we find

$$\dot R_\psi(0) = \langle\psi|\dot\rho(0)|\psi\rangle = \gamma\sum_i p_i\{(\nu+1)|\langle\psi_i|A^+|\psi\rangle|^2 + \nu|\langle\psi_i|A|\psi\rangle|^2\} \geqslant 0, \quad (12a)$$

i.e., $R_\psi(\Delta t) = \dot R_\psi(0)\Delta t + \ldots \geq 0$. Here an indeterminacy occurs only in the case when $R_\psi(0) = \dot R_\psi(0) = 0$, i.e., $\langle\psi_i|\psi\rangle = \langle\psi_i|A^+|\psi\rangle = \langle\psi_i|A|\psi\rangle = 0$ (for all i). Now, however, in addition to (11) the relations

$$\rho(0)A|\psi\rangle = \rho(0)A^+|\psi\rangle = 0, \quad (13)$$

are satisfied, with the aid of which we obtain

$$\ddot R_\psi(0) = \gamma^2\{(\nu+1)^2\langle\psi_{++}|\hat\rho(0)|\psi_{++}\rangle + \nu(\nu+1)[\langle\psi_{+-}|\hat\rho(0)|\psi_{+-}\rangle + \langle\psi_{-+}|\rho(0)|\psi_{-+}\rangle] + \nu^2\langle\psi_{--}|\hat\rho(0)|\psi_{--}\rangle\} \geqslant 0, \quad (12b)$$

where

$$|\psi_{++}\rangle = A^{+2}|\psi\rangle, \quad |\psi_{+-}\rangle = A^+A|\psi\rangle,$$

and so forth. Thus, $R_\psi(\Delta t) \geq 0$ for every $|\psi\rangle$ and consequently $R(\Delta t) \geq 0$. Therefore $\rho(\Delta t)$ is a nonnegative matrix which one can again bring to diagonal form and repeat the entire argument for the following interval Δt. The positive definiteness of the coefficients in (3) is essential for the proof: $\gamma(\nu + 1) \geq 0$, $\gamma\nu \geq 0$.

It is not difficult to generalize the proof given here to the case of a kinetic equation of somewhat more general form that (3):

$$\dot\rho = \sum_{i,j=1}^{N}\gamma_{ij}\left\{A_i\rho A_j^+ - \frac{1}{2}(A_j^+A_i\rho + \rho A_j^+A_i)\right\}. \quad (14)$$

[2)] In particular, the transformation $\rho(0) \to \rho(t)$ is not unitary, and the eigenvalues of the matrix $\rho(t)$ change in the course of time.

Condition (A) is automatically fulfilled for Eq. (14), and conditions (B) and (C) give $\gamma_{ij}^* = \gamma_{ji}$, $\sum_{i,j} \gamma_{ij} z_i z_j^* \geq 0$ for arbitrary complex numbers z_i (the latter condition is equivalent to positive definiteness of the eigenvalues of the matrix γ_{ij}).

4. SPIN RELAXATION AND DICKE'S PROBLEM

Now let us consider the kinetic equation (7) in more detail. First of all we note that in addition to spin relaxation in the presence of a magnetic field, this equation also describes the relaxation in a system of N identical two-level atoms located in a region whose dimensions are much smaller than the wavelength λ (the problem of Dicke[1]). In such an interpretation $s = \frac{1}{2}\sum_{i=1}^{N} \sigma_i$, where σ_i is the "energy spin" operators for the i-th atom.

Due to the interaction of atoms via the radiation field, their spontaneous emission does not occur independently. One can show[1] that for $|r_i - r_j| \ll \lambda$ and neglecting transverse relaxation, the value of the total "energy spin" S is conserved during the relaxation process, i.e., states with different values of S decay independently, each with its own characteristic lifetime. According to this interpretation, $m = (\frac{1}{2})(n_+ - n_-)$ where $|m| \leq S \leq \frac{1}{2} N$ [here $n_+)n_-$) denotes the number of atoms in the upper (lower) level].

In the occupation number representation, Eq. (7) takes the form ($\tau = \gamma t$)

$$\frac{d\rho_{mn}}{d\tau} = (\nu+1)\sqrt{\lambda_{m+1}\lambda_{n+1}}\,\rho_{m+1,\,n+1} + \nu\sqrt{\lambda_m\lambda_n}\,\rho_{m-1,\,n-1} - \left[(\nu+1)\frac{\lambda_m+\lambda_n}{2} + \nu\frac{\lambda_{m+1}+\lambda_{n+1}}{2}\right]\rho_{mn}, \tag{15}$$

where $\lambda_m = (s - m + 1)(s + m)$. From here it is seen that relaxation of the matrix elements ρ_{mn} takes place along diagonals (we call the set of elements ρ_{mn} having a fixed value for the differences $m - n = k$ the k-th diagonal of the density matrix ρ). For $k = 0$ we obtain the following kinetic equation for the populations $w_n = \rho_{nn}$:

$$\frac{dw_n}{d\tau} = (\nu+1)\lambda_{n+1}w_{n+1} - [(\nu+1)\lambda_n + \nu\lambda_{n+1}]w_n + \nu\lambda_n w_{n-1}, \tag{16}$$

coinciding with Eq. (1a). At the same time a comparison of Eq. (15) with Eq. (1b) shows that the equidistant nature of the level spectrum significantly influences the relaxations of the nondiagonal elements ρ_{mn}: The elements of every diagonal are intermixed with one another. Under these conditions the preservation of the positive definiteness of the density matrix ρ (proved in Sec. 3) appears to be a nontrivial fact.

A stationary solution of Eqs. (15) is given by the Boltzmann distribution

$$\rho_{mn} = \frac{1-\xi}{1-\xi^{2s+1}}\xi^r\delta_{mn}, \quad r = s + m = 0, 1, \ldots, 2s,$$
$$\xi = \exp(-\hbar\omega_0/kT), \quad 0 \leqslant \xi < 1. \tag{17}$$

One can solve Eqs. (15) and (16) with the aid of the Laplace transformation. In this connection a system of $(2s + 1 - k)$ linear equations is obtained for the Laplace transforms of the elements $\rho_{n,n+k}(\tau)$ of the k-th diagonal. We give the answer for the case $\nu = 0$:

$$\rho_{m,\,n}(\tau) = \sum_{l\geqslant 0} G(m, n; m+l, n+l|\tau)\,\rho_{m+l,\,n+l}(0). \tag{18}$$

Introducing the notation

$$g(m, n; l|p) = \int_0^\infty G(m, n; m+l, n+l|\tau)\,e^{-p\tau}d\tau, \tag{19}$$

from Eqs. (15) we obtain

$$g(m, n; l|p) = \frac{\prod_{r=1}^{l}\sqrt{\lambda_{m+r}\cdot\lambda_{n+r}}}{\prod_{r=0}^{l}\left[p + \frac{1}{2}(\lambda_{m+r} + \lambda_{n+r})\right]}. \tag{20}$$

The quantities $G(m, n; m + l, n + l|\tau)$ are determined from here according to standard formulas of operational calculus. As an example we give explicit expressions for the populations $w_n(\tau)$ in the case when the initial state has the projection $m = s$:

a) for spin $s = \frac{1}{2}$

$$w_{\frac{1}{2}}(\tau) = e^{-\tau}, \quad w_{-\frac{1}{2}}(\tau) = 1 - e^{-\tau}, \tag{21a}$$

b) for spin $s = 1$

$$w_1 = e^{-2\tau}, \quad w_0 = 2\tau e^{-2\tau}, \quad w_{-1} = 1 - (1+2\tau)e^{-2\tau}, \tag{21b}$$

c) for spin $s = \frac{3}{2}$

$$w_{\frac{3}{2}} = e^{-3\tau}, \quad w_{\frac{1}{2}} = 3(e^{-3\tau} - e^{-4\tau}), \quad w_{-\frac{1}{2}} = 12[e^{-4\tau} - (1-\tau)e^{-3\tau}]$$
$$w_{-\frac{3}{2}} = 1 + (8 - 12\tau)e^{-3\tau} - 9e^{-4\tau}. \tag{21c}$$

For large values of s the expressions for $w_n(\tau)$ have a complicated form. In order to obtain a qualitative picture, let us consider the limiting case $s \gg 1$, when one can pass from Eq. (16) to the Fokker-Planck equation:

$$\frac{\partial w}{\partial\tau} = \left(\nu + \frac{1}{2}\right)\frac{\partial}{\partial x}\left[(1-x^2)\frac{\partial w}{\partial x}\right] + s\frac{\partial}{\partial x}[(1-x^2)w] \tag{22}$$

($\tau = \gamma t$, $x = m/s$, $-1 < x < 1$). Assuming $s \gg (\nu + \frac{1}{2})$, we first neglect the diffusion term in Eq. (22). The Green's function for the first-order equation which arises in this connection has the form

$$G(x, x_0; \tau) = \delta(x - \xi(\tau)), \quad \xi(\tau) = \frac{x_0 - \operatorname{th} s\tau}{1 - x_0 \operatorname{th} s\tau}. \tag{23}$$

It is easy to see that $\xi(\tau)$ represents the trajectory of a magnetic moment which satisfies the classical equation of Landau and Lifshitz.[14] The diffusion term in Eq. (22) leads to a spreading of the distribution (23), and thus corresponds to taking the quantum corrections into account. Certain approximate formulas for the Green's function of Eq. (22) are given in[15].

It is of interest to determine the spectrum of the photons emitted during the relaxation process (at zero temperature, $\nu = 0$). Let us denote by $g_m^s(\nu)$ the normalized spectral density of the radiation line for the case when, at $t = 0$, the z-component of the spin is equal to m. Then

$$g_m{}^s(\nu) = \frac{s}{\pi(s+m)}\sum_{\sigma=-s}^{m-1}\sum_{\sigma'=-s}^{\sigma}\frac{(s+\sigma)!(s-\sigma')!}{(s-\sigma)!(s+\sigma')!}\cdot$$
$$\cdot\operatorname{Re}\left\{\prod_{n=\sigma'}^{\sigma}\frac{1}{s(s+1) - n^2 + is\nu}\right\}, \tag{24}$$

where

$$v=\frac{\omega-\omega_0}{s\gamma},\quad \int_{-\infty}^{+\infty} g_m^s(v)\,dv=1 \tag{25}$$

(a derivation of formula (24) is given in the Appendix).

For $m=-s+1$ one quantum in all is emitted, and the shape of the line remains Lorentzian: $g^s_{-s+1}(v)=1/\pi(1+v^2)$; the width of this line exceeds the width of the γ line emitted by an individual atom by a factor s. For $m\geq -s+2$ the interference of quanta emitted during successive transitions between equidistant levels occurs, as a consequence of which the shape of the line is modified (see Fig. 1). We note that although

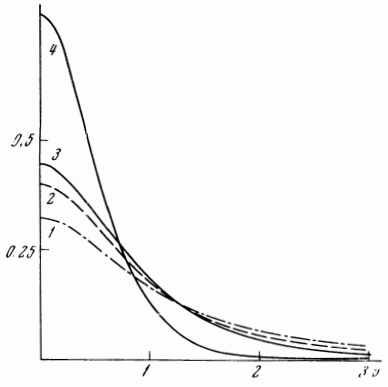

FIG. 1. Shape of the spectral line $g_m^s(v)$ associated with relaxation from the upper level m = s for the following values of the spin: s = 1/2, 1, 3/2, and ∞, which correspond to curves 1, 2, 3, and 4, respectively. Curve 1 has the Lorentzian shape.

the widths of the individual levels $\gamma_m=\lambda_m\gamma\sim s^2\gamma$, the spectral line width $\sim s\gamma$. In this connection, there appears an interference narrowing of the spectral lines radiated by systems possessing equidistant levels (compare with the analogous situation for an harmonic oscillator[10]).

From Eq. (24) we obtain the following result as $v\to\infty$:

$$g_{m^s}(v)=\frac{1}{2\pi v^2}\left(1-\frac{m-1}{s}\right). \tag{26}$$

The shape of the wings of the line is Lorentzian, which is due to the abrupt switching-on of the interaction (preparation of the initial state $\rho_{nn'}(0)=\delta_{mn}\delta_{nn'}$). In the limiting case $s\gg 1$, expression (24) for the spectrum $g^s_m(v)$ can be reduced to the following form:

$$g_{cl}(v,x_0)=\frac{1-x_0}{2\pi}\left|\int_0^\infty \frac{e^{iv\tau}}{\operatorname{ch}\tau-x_0\operatorname{sh}\tau}\,d\tau\right|^2,\quad x_0=\frac{m}{s}, \tag{27}$$

which agrees[3)] with the calculation according to classical theory.[1,16] With the aid of Eq. (27) it is not difficult to find out how the line width depends on the initial value of m. As a measure of the line width it is natural to take the quantity Δ:

$$\Delta(x_0)\equiv\frac{1}{\pi g_{cl}(0,x_0)}=\frac{1+x_0}{2(\arcsin\sqrt{(1+x_0)/2})^2}. \tag{28}$$

This function is shown in Fig. 2. From this figure it is clear that the line width decreases (also see[17]) as $x_0=m/s$ increases, which differs from the analyzed oscillator system.

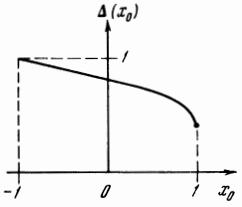

FIG. 2. Dependence of line width $\Delta(x_0)$ on the initial excitation $m=sx_0$ (for the problem of Dickie).

The reason for this difference consists in the following. In the quasiclassical case we have the following expression for the width Δ:

$$\Delta(n_0)\sim n_0\left\{\int_0^{n_0}\frac{dn}{|M(n)|}\right\}^{-2} \tag{29}$$

(for an arbitrary system with an equidistant spectrum). Here $n=0,1,2,\ldots$ labels the successive, equidistant levels, starting from the lowest level; $n_0\gg 1$ is the initial level; $M(n)=\sqrt{\lambda_n}$ is the matrix element for the transition $n\to(n-1)$.

From expression (29) it is clear that the nature of the change of $\Delta(n_0)$ as n_0 increases is determined by the form of $M(n)$. Thus, for an harmonic oscillator $M(n)\propto\sqrt{n}$, and in general Δ does not depend on n_0; for the spin problem $M(n)\propto\sqrt{n(2s-n)}$, which leads to formula (28).

5. DICKE'S PROBLEM FOR ARBITRARY DISTANCES BETWEEN THE ATOMS

Now let us consider Dicke's problem without assuming that $|\mathbf{r}_i-\mathbf{r}_j|\ll\lambda$, assuming however the position of the atoms to be fixed (neglect of the Doppler effect). In this connection the kinetic equation has the form ($\nu=0$):[4)]

$$\rho=-\sum_{i,k=1}^{N}\gamma_{ik}(\sigma_i^{(+)}\sigma_k^{(-)}\rho+\rho\sigma_i^{(+)}\sigma_k^{(-)}-2\sigma_k^{(-)}\rho\sigma_i^{(+)}). \tag{30}$$

One can obtain an expression for γ_{ik} in the same way as formula (4):

$$\gamma_{ik}=\gamma_0\varphi(kr_{ij}),\quad \varphi(x)=\frac{3}{2}\left[\left(1-\frac{1}{x^2}\right)\frac{\sin x}{x}+\frac{\cos x}{x^2}\right] \tag{31}$$

(see, for example,[7]). We note that $\varphi(0)=1$; therefore upon fulfillment of the condition $kr_{ij}\ll 1$ Eq. (30) turns into Eq. (7). The reduction in $\varphi(x)$ for $x\gtrsim 1$ corresponds to a weakening of the correlations in the spontaneous emission of the atoms. If all $r_{ij}\gg\lambda$, then $\varphi(kr_{ij})=\delta_{ij}$, i.e., a transition takes place to a system of independent atoms.

If the conditions $kr_{ij}\ll 1$ (i, j = 1, ..., N) are not satisfied, then the total "energy spin" of the system,

$$\mathbf{s}=\frac{1}{2}\sum_{i=1}^{N}\boldsymbol{\sigma}_i$$

is no longer conserved during the relaxation process, and the results of Section 4 are not applicable. We

[3)] An error was made in the calculation of the spectrum $g_{cl}(v, x_0)$ in article [1], and was corrected in [16,17].

[4)] In Eq. (30) we do not take into consideration terms of the type

$$-\gamma_\perp\sum_i(\sigma_i^{(z)2}\rho+\rho\sigma_i^{(z)2}-2\sigma_i^{(z)}\rho\sigma_i^{(z)}),$$

which correspond to transverse relaxation (i.e., a broadening due to collisions), and also terms of the type $[v,\rho]$ in (3) (renormalization of the frequencies of different modes) are not taken into consideration.

shall limit the investigation to weakly-excited states, when the spin system is equivalent to a set of oscillators. Here the spin operators $\sigma_i^{(-)}$ and $\sigma_i^{(+)}$ play the role of a_i and a_i^+. Equation (30) implies the linearized equations

$$\langle \dot{a}_i \rangle = -\frac{1}{2}\sum_k \gamma_{ik}\langle a_k\rangle, \tag{32}$$

which can be diagonalized by a unitary transformation:

$$a_i' = \sum_k U_{ik}a_k, \quad U^+U = 1. \tag{33}$$

Here $\langle \dot{a}_i' \rangle = -(1/2)\gamma^{(i)}\langle a_i'\rangle$, where the $\gamma^{(i)}$ are the eigenvalues of the matrix γ_{ik}. Choosing the initial state $\langle a_i^+ a_k\rangle_0 = n_0\delta_{ik}$ (equally-probable excitation of all atoms), from Eq. (33) we have $\langle a_i'^+ a_k'\rangle_0 = n_0\delta_{ik}$. Hence for the radiation spectrum averaged over the directions of flight of the photons, we obtain

$$g(\omega) = \frac{1}{2\pi}\int_0^\infty \frac{P(\gamma)\gamma}{(\omega-\omega_0)^2+\gamma^2/4}\,d\gamma, \tag{34}$$

where $P(\gamma)$ is the density of the distribution of eigenvalues $\gamma^{(i)}$, normalized to unity according to

$$\int_0^\infty P(\gamma)\,d\gamma = 1, \quad \int_0^\infty P(\gamma)\gamma\,d\gamma = \gamma_0 \tag{35}$$

(the second condition follows from invariance of the trace, $Sp(\gamma_{ik}) = N\gamma_0$, with respect to the transformations (33)).

Thus, calculation of the shape of the radiation line reduces (under the assumptions made above) to the purely mathematical problem of the distribution $P(\gamma)$ of eigenvalues of the matrices γ_{ij} appearing in the initial equation (30). We have not been able to find the solution of this problem in the literature. We mention only one result which pertains to an integral spectrum. Let us consider the function $G(\Delta\omega)$:

$$G(\Delta\omega) = \int_{-\infty}^{\omega_0-\Delta\omega} g(\omega)\,d\omega + \int_{\omega_0+\Delta\omega}^{\infty} g(\omega)\,d\omega. \tag{36}$$

From Eq. (34) we find

$$G(\Delta\omega) = \frac{2}{\pi}\int_0^\infty P(\gamma)\arctg\frac{\gamma}{2\Delta\omega}\,d\gamma. \tag{37}$$

Since $\tan^{-1} x \le x$ and $P(\gamma) \ge 0$, then from here (with relations (35) taken into consideration) follows the inequality

$$G(\Delta\omega) \leqslant \gamma_0/\pi\Delta\omega. \tag{38}$$

As is well known, for individual modes a_i' the line may be appreciably broadened in comparison with the line for an isolated atom.[1] From inequality (38) it is seen that the total spectrum does not undergo such a broadening; therefore the broadening of the spectrum for some modes must be compensated by a narrowing of other lines. We emphasize, however, that in this derivation substantial use has been made of the assumption concerning the absence of transverse relaxation and Doppler broadening.

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APPENDIX

SHAPE OF THE RADIATION LINE FOR SYSTEMS WITH EQUIDISTANT ENERGY LEVELS

The Heisenberg equations of motion for the field operators follow from the Hamiltonian (2)

$$\dot{b}_i(t) = -i\omega_i b_i(t) - if_i A(t), \tag{A1}$$

from which it follows that

$$b_i(T) = e^{-i\omega_i T}b_i(0) - if_i\int_0^T A(t')e^{-\omega_i(T-t')}dt'. \tag{A.2}$$

We shall calculate the spectrum under the assumption that at $t = 0$ the electromagnetic field is found in the vacuum state $|0\rangle$. Then

$$\langle n_i(T)\rangle = \langle b_i^+(T)b_i(T)\rangle = |f_i|^2\int_0^T\int_0^T dt'\,dt''e^{-i\omega_i(t'-t'')}\langle A^+(t')A(t'')\rangle. \tag{A.3}$$

From here as $T \to \infty$ one obtains the following formula for the spectral density $g(\omega)$:

$$g(\omega) \sim \mathrm{Re}\int_0^\infty dt\int_0^\infty d\tau e^{-i\omega\tau}\langle A^+(t+\tau)A(t)\rangle. \tag{A4}$$

The correlation function $\langle A^+(t+\tau)A(t)\rangle$ is determined by the kinetic equation (3). Using standard techniques (see Section 10 in[7]), we obtain

$$\langle A^+(t+\tau)A(t)\rangle = e^{i\omega_0\tau}\sum_{k\geqslant 0}\rho_{kk}(0)\sum_m \lambda_m^{1/2}G(m,m;k,k|t)\cdot \sum_n \lambda_n^{1/2}G(n-1,n;m-1,m|\tau), \tag{A.5}$$

where $\lambda_m = |(A)_{m-1,m}|^2$ (compare with formula (75) of[10]). Upon substitution of (A5) into (A4) the spectral density $g(\omega)$ is expressed in terms of the Laplace transform of the Green's function G for Eq. (3), for whose determination it is necessary to solve a system of linear equations. In this connection we note that Eqs. (15) and (16) for the above indicated definition of λ_m are valid not only for spin relaxation, but also for an arbitrary system with an equidistant spectrum. Using Eq. (20), from here we obtain the following result for $\nu = 0$:

$$\langle n_i\rangle = \sum_{k\geqslant 0}\rho_{kk}(0)kg_k(\omega_i), \tag{A6}$$

where $g_k(\omega)$ is the shape of the spectral line associated with relaxation from the k-th level:

$$g_k(\omega) = \frac{1}{\pi k}\mathrm{Re}\left\{\sum_{m=0}^{k-1}\sum_{l=0}^{m}\frac{\prod_{r=l+1}^{m}\gamma_r}{\prod_{r=l}^{m}[i(\omega-\omega_0)+1/2\Gamma_{r,r+1}]}\right\}; \tag{A.7}$$

$\Gamma_{r,r+1}$ is the Wigner-Weisskopf expression for the natural width of the line associated with a transition from level $r+1$ to level r:

$$\Gamma_{r,r+1} = \gamma_r + \gamma_{r+1}, \quad \gamma_r = \lambda_r\gamma. \tag{A.8}$$

The numbering of the states in Eqs. (A6) and (A7) is chosen so that the k-th level lies a distance $\hbar\omega_0 k$ from the lowest level; $g_k(\omega)$ is normalized to unity. Expression (24) is obtained from here by substitution of the specific expression $\lambda_m = (s+m)(s-m+1)$.

We note that in Eq. (A7) it is not difficult to take into account a possible violation of the equidistant nature of the levels, and also additional (the so-called "transverse") relaxation which is not associated with the real emission of a photon. For this purpose, in the resonance denominators of formula (A7) instead of ω_0 one should substitute $\omega_{r+1,r} = E_{r+1} - E_r$, and to the $\Gamma_{r+1,r}$ one should add a contribution $\Gamma'_{r+1,r}$ coming from transverse relaxation ($\Gamma'_{r+1,r} \geq 0$); in this connection the numerators are not changed. In the limiting case when the frequencies of the various transitions differ by quantities $\Delta\omega \gg \Gamma$, Eq. (A7) takes the usual form

$$g_k(\omega) \approx \frac{1}{k} \sum_{i=1}^{k} \frac{\Gamma_i}{2\pi[(\omega - \omega_i)^2 + \Gamma_i^2/4]}. \qquad (A.9)$$

However, in the case of systems with equidistant levels $\Delta\omega \lesssim \Gamma$ and in Eq. (A7) it is impossible to neglect the interference terms, which lead to a narrowing of the spectral line and a modification of its shape. Interference narrowing of the spectral line is also discussed in[10,18].

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