

*RADIATION FROM A SYSTEM OF RESONANT MOLECULES WITH ENERGY-LEVEL  
SCATTER*

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The radiation from a macroscopic system of identical two-level molecules is determined under the condition that the energy levels of the molecules deviate slightly from exact resonance. The limiting cases of narrow and broad level spreads are investigated. For a small spread (narrow resonance) the molecule emission is of collective nature and the usual balance equations no longer hold. For the case of a broad spread (broad resonance) a region is found in which the emission law is the same as that calculated by perturbation theory, and the balance equations are valid. The problem of "burning out" of the initially excited molecules is considered and the limits of applicability of the linear approximation in problems pertaining to the interaction of radiation with resonant molecules are determined.

SEVERAL recent papers (see for example, the reviews<sup>[1-4]</sup>) deal with electromagnetic radiation of a macroscopic system of resonant two-level molecules. Owing to the presence of stimulated emission, electromagnetic processes in such a system differ noticeably from the usual case of independent emission of photons from individual molecules. The question of the radiation from a system consisting of a large number of resonant molecules is macroscopic in nature. It is frequently solved by using balance equations in which the probabilities of the elementary processes are calculated by perturbation theory for the isolated molecule. Yet the validity of such equations is not obvious. Indeed, if the wavelength of the radiated light is large compared with the mean distance between the molecules, and the resonance is sufficiently narrow, then a strong resonant interaction is established between the molecules even before the field radiated in each individual act reaches the wave zone. In this case even the elementary act of emission of a quantum has a collective character. A more consistent solution of the problem is then obtained by using the macroscopic averaged equations of electrodynamics<sup>[5,6]</sup>. As the resonance broadens, the collective character of the elementary emission act is lost, and the emission process is described by the aforementioned balance equations.

The approaches frequently used in the limiting cases of narrow and broad resonance, which are different in principle, make it possible neither

to establish the region of their applicability, nor to obtain a solution in the intermediate case. It is therefore natural to attempt to obtain a general solution of the problem on the basis of exact quantum-mechanical equations. For the case of strict resonance, such a solution was obtained earlier<sup>[7]</sup>. Analogous solutions were obtained also for the diffusion of a quantum in a resonant medium<sup>[8]</sup>, with the obtained solutions going over into the two indicated limiting cases.

In this paper we generalize the previously developed method<sup>[7]</sup> to include the case when like energy levels of the molecules are scattered about some average position, so that the resonance is not exact. The physical reason for the scatter is not ascertained in detail. The scatter of the levels may be due, for example, to the Stark effect for the impurity atoms situated in the alternating field of the vibrating lattice, or to the Doppler effect in the case of a gas. It is also assumed that only two levels of molecules participate in the radiation. The remaining nonresonant levels can in principle be taken into account with the aid of the dielectric constant of the medium. It is assumed in the investigation of the radiation that at the initial instant of time the molecule distribution over the levels is fixed and there are no quanta. The solution of the problem for arbitrary initial conditions is obtained in a similar manner.

1. Inclusion of only two levels allows us to employ a spin description of the molecules, whereby the molecule at the upper and lower level corre-

sponds to "spin" projections  $+1/2$  and  $-1/2$ , respectively. Then the Hamiltonian  $H$  of molecules in a radiation field contained in a volume  $V$  is written in the form

$$\begin{aligned} H = & \sum_r \frac{\hbar\omega_r}{2} N_r + \sum_{k\lambda} \hbar\omega_k c_{k\lambda}^+ c_{k\lambda} \\ & + \sum_{r,k\lambda} \hbar(c_{k\lambda} b_{k\lambda}^{r+} + c_{k\lambda}^+ b_{k\lambda}^r), \\ b_{k\lambda}^{r+} = & \sum_j' \sigma_{+j} \left( \frac{2\pi}{\hbar\omega_k V} \right)^{\eta_k} (\mathbf{M}^j l_{k\lambda}) e^{ikx_j}, \\ b_{k\lambda}^r = & \sum_j' \sigma_{-j} \left( \frac{2\pi}{\hbar\omega_k V} \right)^{\eta_k} (\mathbf{M}^{*j} l_{k\lambda}) e^{-ikx_j}, \\ N_r = & \sum_j' \sigma_z^j, \quad \sigma_z^j = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \\ \sigma_+^j = & \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \quad \sigma_-^j = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}. \end{aligned} \quad (1)$$

The first and second terms in (1) are the energy operators of the molecules and of the free radiation field, respectively, and the last term is the operator of interaction of the molecules with the radiation field;  $\hbar\omega_r$  is the energy difference between the upper and lower levels of the individual molecules;  $c_{k\lambda}$  and  $c_{k\lambda}^+$  are the operators of absorption and production, respectively, of a photon with momentum  $\hbar k$  and polarization vector  $\mathbf{l}_{k\lambda}^\lambda$ , with

$$c_{k\lambda} c_{k'\lambda'}^+ - c_{k'\lambda'}^+ c_{k\lambda} = \delta_{kk'} \delta_{\lambda\lambda'}, \quad \lambda = 1, 2, \quad \omega_k^2 = \mathbf{k}^2 c^2; \quad (2)$$

$x_j$  is the coordinate of the center of gravity of the  $j$ -th molecule, and  $\mathbf{M}^j$  is the matrix element of the transition of the  $j$ -th molecule from the lower level to the upper one with simultaneous absorption of a photon. The matrix element  $\mathbf{M}^j$  is connected with the probability  $W$  of spontaneous dipole emission of a photon per unit time for the isolated  $j$ -th molecule by the relation

$$W = \frac{4\omega_j}{3\hbar c^3} |\mathbf{M}^j|^2$$

where  $\hbar\omega_j$  is the energy difference between the upper and lower levels of the  $j$ -th molecule. The prime on the sign of summation with respect to the index  $j$  denotes throughout summation over all the molecules in the volume  $V$ , the molecule energy being  $\hbar\omega_r$  (it is assumed that the number of such molecules is sufficiently large). The physical quantities pertaining to this group of molecules are tagged by the index  $r$ .

As before<sup>[7]</sup>, we have left out of the Hamiltonian (1) the interaction-operator terms that describe effects in which the number of particles (photons

and excited molecules) are not conserved in the individual act of emission or absorption, and also the terms which are quadratic in the vector potential. These terms make a negligibly small contribution in the present problem<sup>[6,7]</sup>.

We shall need in what follows the commutation relations of the operators contained in (1). In addition to (2) we have

$$\begin{aligned} b_{k\lambda}^r N_{-r} - N_{-r} b_{k\lambda}^r &= 2\delta_{rr'} b_{k\lambda}^r, \\ b_{k\lambda}^{r+} N_{-r'} - N_{-r'} b_{k\lambda}^{r+} &= -2\delta_{rr'} b_{k\lambda}^{r+}, \\ b_{k\lambda}^{r+} b_{k'\lambda'}^{r'} - b_{k'\lambda'}^{r'} b_{k\lambda}^{r+} &= \delta_{rr'} B_{k\lambda, k'\lambda'}^r, \end{aligned} \quad (3)$$

where  $N_r^r = N_2^r - N_1^r$  is the excess population in the group of molecules with an energy-level difference  $\hbar\omega_r$ , and

$$\begin{aligned} B_{k\lambda, k'\lambda'}^r &= \frac{2\pi}{\hbar(\omega_k \omega_{k'})^{1/2} V} \sum_j' \sigma_z^j (\mathbf{M}^j l_{k\lambda}) (\mathbf{M}^{*j} l_{k'\lambda'}^*) \\ &\times \exp(i(\mathbf{k} - \mathbf{k}') \mathbf{x}_j). \end{aligned} \quad (4)$$

The off-diagonal ( $\mathbf{k} \neq \mathbf{k}'$ ) terms of the operator (4) vanish upon averaging. A nonvanishing result is obtained from the quadratic terms, an account of which leads to a natural broadening of the spectral line<sup>[6,8]</sup>. We shall assume that the natural width is much smaller than the assumed scatter of the levels, so that the off-diagonal terms in (4) can be neglected. Then the commutation relation (3) takes the form

$$\begin{aligned} b_{k\lambda}^{r+} b_{k'\lambda'}^{r'} - b_{k'\lambda'}^{r'} b_{k\lambda}^{r+} &= \delta_{rr'} \delta_{kk'} \delta_{\lambda\lambda'} N_r^r / 4\tau_0^2, \\ \tau_0^{-2} &= 2\pi c^3 W / \omega_0^2 V, \end{aligned} \quad (5)$$

where in the right side of (5) the frequencies  $\omega_r$  are set equal to the resonant value  $\omega_0$  about which the scatter takes place.

The quantity  $N_r^r$  can be regarded as the  $z$ -th projection of the total "spin" of the  $r$ -th group of molecules. If the upper and lower level numbers of the molecules are not close to one another,  $N_r^r \gg 1$ , then  $N_r^r$  is much larger in absolute value than its commutators (3). In this case  $N_r^r$  can be regarded as a c-number, just as in the analysis of spin-waves in a ferromagnet<sup>[9]</sup>. This closes the system of commutators for the operators  $c_{k\lambda}^+$ ,  $c_{k\lambda}^r$ ,  $b_{k\lambda}^r$ , and  $b_{k\lambda}^{r+}$ .

We denote the mean quantum-mechanical values of the number of photons  $n_{k\lambda}$  and of the excess population  $N_r^r$  by

$$n_{k\lambda} = \langle c_{k\lambda}^+ c_{k\lambda} \rangle, \quad N_r^r = \left\langle \sum_j' \sigma_z^j \right\rangle.$$

Using the rules for the differentiation of operators, we obtain, taking (2) and (5) into account, the follow-

ing system of equations for the quantum-mechanical mean values:

$$i \frac{d}{dt} n_{k\lambda} = \sum_r (\langle c_{k\lambda} b_{k\lambda}^{r+} \rangle - \langle c_{k\lambda} b_{k\lambda}^{r-} \rangle), \quad (6)$$

$$i \frac{d}{dt} \langle c_{k\lambda} b_{k\lambda}^{r+} \rangle = (\omega_k - \omega_r) \langle c_{k\lambda} b_{k\lambda}^{r+} \rangle + \frac{1}{4\tau_0^2} N_-^{-r} n_{k\lambda} \\ + \sum_{r'} \langle b_{k\lambda}^{r+} b_{k\lambda}^{r'} \rangle, \quad (7)$$

$$i \frac{d}{dt} \langle c_{k\lambda} b_{k\lambda}^{r-} \rangle = (\omega_r - \omega_k) \langle c_{k\lambda} b_{k\lambda}^{r-} \rangle - \frac{1}{4\tau_0^2} N_-^{-r} n_{k\lambda} \\ - \sum_{r'} \langle b_{k\lambda}^{r-} b_{k\lambda}^{r'} \rangle, \quad (8)$$

$$i \frac{d}{dt} \langle b_{k\lambda}^{r+} b_{k\lambda}^{r'} \rangle = (\omega_{r'} - \omega_r) \langle b_{k\lambda}^{r+} b_{k\lambda}^{r'} \rangle \\ + \frac{1}{4\tau_0^2} (N_-^{-r} \langle c_{k\lambda} b_{k\lambda}^{r-} \rangle - \langle c_{k\lambda} b_{k\lambda}^{r+} \rangle N_-^{-r'}), \quad (9)$$

$$i \frac{d}{dt} N_-^{-r} = 2 \sum_{k\lambda} (\langle c_{k\lambda} b_{k\lambda}^{r+} \rangle - \langle c_{k\lambda} b_{k\lambda}^{r-} \rangle). \quad (10)$$

2. Let us consider the initial stage of the process, in which  $N_-^r$  can be regarded as a constant in (6)–(9) equal to the initial excess population:  $N_-^r = N_2^r - N_1^r$ . Then Eq. (10) must be discarded and the remaining system of equations is linear. To solve it, we must specify the initial conditions. Assume that there are no photons at the initial instant  $t = 0$ , meaning that

$$n_{k\lambda}|_{t=0} = \langle c_{k\lambda} b_{k\lambda}^{r+} \rangle|_{t=0} = \langle c_{k\lambda}^+ b_{k\lambda}^r \rangle|_{t=0} = 0.$$

In an approximation analogous to that used earlier for the operator (4), the initial value of the quantity  $\langle b_{k\lambda}^{r+} b_{k\lambda}^r \rangle$  is written in the form

$$\langle b_{k\lambda}^{r+} b_{k\lambda}^r \rangle|_{t=0} = N_2^r \delta_{rr}/4\tau_0^2.$$

It is natural to use further a Laplace transformation with respect to the variable  $t$ . Denoting the transform by the same symbol as the original, we eliminate the quantity  $\langle b_{k\lambda}^{r+} b_{k\lambda}^r \rangle$  from (7) and (8) with the aid of (9). We then get

$$ip n_{k\lambda}(p) = -\frac{1}{4\tau^2} \int f(\omega) [v_{k\lambda}(\omega, p) - \bar{v}_{k\lambda}(\omega, p)] d\omega, \quad (11)$$

$$\left( \omega - \omega_k + ip + \frac{1}{4\tau^2} \frac{1}{\omega - \omega_0 + i(p + \delta/2)} \right) v_{k\lambda}(\omega, p) \\ = n_{k\lambda}(p) + \frac{N_2^0}{p N_-^0} - \frac{1}{4\tau^2} \int \frac{f(\omega') \bar{v}_{k\lambda}(\omega', p) d\omega'}{\omega' - \omega - ip}, \quad (12)$$

$$\left( \omega - \omega_k - ip + \frac{1}{4\tau^2} \frac{1}{\omega - \omega_0 - i(p + \delta/2)} \right) \bar{v}_{k\lambda}(\omega, p) \\ = n_{k\lambda}(p) + \frac{N_2^0}{p N_-^0} - \frac{1}{4\tau^2} \int \frac{f(\omega') v_{k\lambda}(\omega', p) d\omega'}{\omega' - \omega + ip}, \quad (13)$$

where we introduce the notation

$$\sum_{\substack{r \\ \omega \leq \omega_r \leq \omega + d\omega}} \langle c_{k\lambda} b_{k\lambda}^{r+} \rangle = \frac{v_{k\lambda}(\omega, p)}{4\tau^2} f(\omega) d\omega, \\ \sum_{\substack{r \\ \omega \leq \omega_r \leq \omega + d\omega}} \langle c_{k\lambda} b_{k\lambda}^{r-} \rangle = \frac{\bar{v}_{k\lambda}(\omega, p)}{4\tau^2} f(\omega) d\omega, \\ \tau^2 = |N_-^0|/\tau_0^2$$

and we assume for the sake of concreteness that the distribution of the molecules with respect to the groups characterized by the quantity  $\omega_r$ ,

$$\sum_r N^r = \int N(\omega) d\omega \equiv N \int f(\omega) d\omega,$$

and also the distribution over the excitations at the initial instant of time

$$\sum_{\substack{r \\ \omega \leq \omega_r \leq \omega + d\omega}} N_2^r = N_2^0 f(\omega) d\omega, \quad \sum_{\substack{r \\ \omega \leq \omega_r \leq \omega + d\omega}} N_-^{-r} = N_-^0 f(\omega) d\omega,$$

are given by the same distribution function  $f(\omega)$ , which we take in the form

$$f(\omega) = \frac{1}{2\pi} \frac{\delta}{(\omega - \omega_0)^2 + \delta^2/4}, \quad (14)$$

where  $N_2^0$  is the total number of molecules at the upper level, and  $N_-^0$  is the total excess population at the initial instant. The width  $\hbar\delta$  of the scatter of the molecule levels should be sufficiently small ( $\delta/\omega_0 \ll 1$ ), in order for the Hamiltonian of the system to retain the form (1).

From Eqs. (11)–(13) with account of (14) we obtain finally the following expression for the Laplace transform  $n_{k\lambda}(p)$  of the function  $n_{k\lambda}(t)$ :

$$n_{k\lambda}(p) = \frac{N_2^0}{2\tau_0^2} \times \frac{(p + \delta)(p + \delta/2)}{p \{p(p + \delta)[(\omega_k - \omega_0)^2 + (p + \delta/2)^2] - (p + \delta/2)^2 \tau^2\}}. \quad (15)$$

Taking the inverse transform, we write

$$n_{k\lambda}(t) = \frac{N_2^0}{2\tau_0^2} \left\{ \frac{e^{-\delta t/2}}{p_1^2 - p_2^2} [\text{ch}(p_1 t + \theta_1) + \cos(|p_2| t + \theta_2)] - 2\tau^2 \right\}, \quad (16)*$$

$$p_{1,2} = \frac{1}{2} \left\{ \frac{1}{\tau^2} + \frac{\delta^2}{4} - (\omega_k - \omega_0)^2 \right\} \\ \pm \left[ \left( \frac{1}{\tau^2} + \frac{\delta^2}{4} - (\omega_k - \omega_0)^2 \right)^2 + \delta^2 (\omega_k - \omega_0)^2 \right]^{1/2}, \\ \theta_1 = \ln \frac{p_1 + \delta/2}{p_1 - \delta/2}, \quad \theta_2 = 2 \arctg \frac{2|p_2|}{\delta}. \quad (17)$$

Expressions (15)–(17) are valid for a positive initial excess population  $N_-^0$ . For negative  $N_-^0$  it is necessary to reverse the sign of  $\tau^2$ .

\*ch = cosh; arctg = tan<sup>-1</sup>.

3. As expected, for a macroscopic system the number of photons  $n_{k\lambda}(t)$  of each mode is a finite quantity. From (17) we see that  $p_1^2 > 0$  and  $p_2^2 < 0$ . For a positive initial excess population, the following inequality is satisfied

$$p_1^2 > \delta^2/4$$

for arbitrary values of  $\omega_k$ . This means an exponential growth of the number of quanta with arbitrary  $k$ . Such a result is a natural consequence of neglecting the escape of the quanta from the system. For long times, it is sufficient to retain in (16) only the exponentially growing term:

$$n_{k\lambda}(t) = \frac{N_2^0}{4\tau_0^2} \frac{p_1 + \delta/2}{p_1 - \delta/2} \frac{\exp\{(p_1 - \delta/2)t\}}{p_1^2 - p_2^2}. \quad (18)$$

The dependence on the frequency  $\omega_k$  is determined here essentially by the argument of the exponent. The maximum of this argument occurs at a value  $\omega_k = \omega_0$ . Expanding near this maximum, we obtain

$$\begin{aligned} n_{k\lambda}(t) &= \frac{N_2^0}{4N_0^0} \left[ 1 + \left( 1 + \frac{4}{\delta^2\tau^2} \right)^{-1/2} \right]^2 \\ &\times \exp \left\{ \left[ \left( \frac{1}{\tau^2} + \frac{\delta^2}{4} \right)^{1/2} - \frac{\delta}{2} \right] t - \frac{(\omega_k - \omega_0)^2 \tau t}{2(1 + \delta^2\tau^2/4)^{1/2}} \right\}. \end{aligned}$$

Thus, the effective line width  $|\omega_k - \omega_0|_{\text{eff}}$  decreases with time like

$$|\omega_k - \omega_0|_{\text{eff}} \sim \tau^{-1} (1 + \delta^2\tau^2/4)^{1/2} (\tau/t)^{1/2}.$$

In the case of a negative initial excess population we have

$$p_1^2 < \delta^2/4,$$

so that, unlike (18), the function (16) tends to a constant limit if  $t \rightarrow \infty$ .

Summing  $n_{k\lambda}(t)$  in (16) over all values of  $k$  and  $\lambda$ , we obtain the total number of photons  $n(t)$  as a function of the time:

$$n(t) = N_2^0 W \left( F(t) + \delta \int_0^t F(t') dt' \right), \quad (19)$$

$$F(t) = e^{-\delta t/2} \int_0^t I_0 \left( t' \left( \frac{\delta^2}{4} + \frac{1}{\tau^2} \right)^{1/2} \right) I_0 \left( \frac{(t-t')\delta}{2} \right) dt', \quad (20)$$

where the sign of  $\tau^2$  is reversed if the initial excess population  $N_0^0$  is negative. If  $\delta = 0$ , then formula (19) coincides with that obtained earlier.<sup>[10]</sup> Here  $I_0(x) \equiv J_0(ix)$  is a Bessel function of zero order of pure imaginary argument.

As  $V \rightarrow \infty$  and for arbitrary  $\delta$ , the expression (19) for a fixed number of molecules goes over into the perturbation-theory formula

$$n(t) = N_2^0 W t. \quad (21)$$

For a finite volume  $V$  and small  $t$ , expression

(19) also coincides with (21), and as  $t \rightarrow \infty$  the form of the function (19) depends on the sign of  $N_0^0$ . If  $N_0^0 > 0$ , then  $n(t)$  increases in the linear region exponentially with time:

$$\begin{aligned} n(t) &= N_2^0 W \tau \left[ \left( 1 + \frac{\delta^2\tau^2}{4} \right)^{1/2} + \frac{\delta\tau}{2} \right]^2 \left( 1 + \frac{\delta^2\tau^2}{4} \right)^{-1/4} \left( \frac{\tau}{2\pi t} \right)^{1/2} \\ &\times \exp \left\{ \left[ \left( \frac{\delta^2}{4} + \frac{1}{\tau^2} \right)^{1/2} - \frac{\delta}{2} \right] t \right\}, \\ &[(\delta^2/4 + 1/\tau^2)^{1/2} - \delta/2] t \gg 1. \end{aligned}$$

For  $N_0^0 < 0$  and  $t \rightarrow \infty$ , the function  $n(t)$  increases with time in the linear region like the square root

$$n(t) = 2N_2^0 W \tau (\delta t / \pi)^{1/2}. \quad (22)$$

At large values of  $t$ , this quantity becomes larger than  $N_0^0$ , because the assumptions made above are not valid in this time region.

4. Let us consider the limiting cases of broad and narrow resonances,  $\delta^2\tau^2 \gg 1$  and  $\delta^2\tau^2 \ll 1$ , respectively. Let  $\delta^2\tau^2 \gg 1$ , so that  $\tau^{-2}$  can be neglected in (16) and retained only as a common factor. Then formula (16) becomes

$$\begin{aligned} n_{k\lambda}(t) &= \frac{N_2^0}{2\tau_0^2} \left\{ \frac{\delta t/2}{(\omega_k - \omega_0)^2 + \delta^2/4} + \frac{(\omega_k - \omega_0)^2 - \delta^2/4}{[(\omega_k - \omega_0)^2 + \delta^2/4]^2} \right. \\ &\times [1 - e^{-\delta t/2} \cos(\omega_k - \omega_0)t] - \frac{(\omega_k - \omega_0)\delta}{[(\omega_k - \omega_0)^2 + \delta^2/4]^2} \\ &\left. \times e^{-\delta t/2} \sin(\omega_k - \omega_0)t \right\}. \end{aligned} \quad (23)$$

For  $\delta \gg 1$ , the second and third terms of (23) can be neglected. The remaining expression can be readily seen to coincide with that obtained by perturbation theory for independent radiation of  $N_2^0$  molecules. Indeed, multiplying  $n_{k\lambda}$  by the number of states in the interval  $d\omega_k$ , we obtain

$$\begin{aligned} dn &= \frac{\delta}{2\pi} \frac{N_2^0 W t}{(\omega_k - \omega_0)^2 + \delta^2/4} d\omega_k \\ &= N_2^0 W t d\omega_k \int \delta(\omega_k - \omega) f(\omega) d\omega. \end{aligned} \quad (24)$$

The difference between (23) and (24) when  $t$  is small is connected with the following circumstance. For one molecule with an energy-level difference  $\hbar\omega$ , perturbation theory yields directly the following formula for the probability per unit time of emitting a quantum whose energy is in the interval  $d\omega_k$ :

$$dW = W \frac{2 \sin^2[(\omega_k - \omega)t/2]}{\pi(\omega_k - \omega)^2} d\omega_k,$$

and for the emission of  $N_2^0$  molecules we have

$$dn(t) = N_2^0 W t d\omega_k \int \frac{2 \sin^2[(\omega_k - \omega)t/2]}{\pi(\omega_k - \omega)^2 t} f(\omega) d\omega. \quad (25)$$

The first factor in the integrand is a sharp function of width  $t^-$ . However,  $f(\omega)$  is also a sharp function of width  $\delta$ . Naturally the first factor can be regarded as a  $\delta$ -function only if  $t^- \ll \delta$ . It must be noted that expression (23) can be obtained by direct integration in (25) using a function  $f(\omega)$  from (14).

There is no stimulated emission of molecules in formula (23), because the quantity  $\tau^{-2}$  has been neglected. This neglect means that we have set  $N_- = 0$ , so that the number of absorption events is equal to the number of stimulated-emission events. Let us now take into account the next terms of the expansion in the parameter  $(\delta\tau)^{-2} \ll 1$ . Then for large  $t \gg \delta^{-1}$  formula (16), regardless of the sign of  $N_-^0$ , becomes

$$n_{k\lambda}(t) = \frac{N_2^0}{N_-^0} \left\{ \exp \left[ \frac{N_-^0 t}{4\tau_0^2} \frac{\delta}{(\omega_k - \omega_0)^2 + \delta^2/4} \right] - 1 \right\}. \quad (26)$$

Expression (26) is a solution of the balance equation in which the molecule emission probability is calculated by perturbation theory. Indeed, the quantity

$$W_{k\lambda,\omega} = \frac{\pi^2 c^3 W}{\omega_0^2 V} \delta(\omega_k - \omega) = \frac{\pi}{2\tau_0^2} \delta(\omega_k - \omega) \quad (27)$$

is the perturbation-theory probability that an isolated molecule with an energy-level difference  $\hbar\omega$  will emit in a unit of time a quantum with given  $k$  and  $\lambda$ . Consequently, the sought balance equation can be written in the form

$$\frac{d}{dt} n_{k\lambda} = \int d\omega W_{k\lambda,\omega} N_2^0(\omega) (n_{k\lambda} + 1) - \int d\omega W_{k\lambda,\omega} N_1^0(\omega) n_{k\lambda}$$

with solution (26).

For not too large  $t$ , formula (26) admits of an expansion of the exponential, leading to an expression corresponding to (24). For example, for the center of the spectral line ( $\omega_k = \omega_0$ ) this is possible for a time  $t \ll \delta\tau^2$ , which is much larger than  $t \sim \delta^{-1}$ . Thus, in the limiting case of a broad scatter of the levels,  $\delta^2\tau^2 \gg 1$ , there is a time interval

$$\delta^{-1} \ll t \ll \delta\tau^2,$$

in which perturbation theory is valid and stimulated emission has not yet begun. In other words, the stimulated emission proceeds here in the real-quantum stage.

A different picture is encountered in the opposite limit case  $\delta^2\tau^2 \ll 1$  (narrow resonance). Here the distribution of the quanta relative to the variable  $\omega_k$  is very close to the perturbation-theory result only for a small time  $t \ll \tau$ , and the indicated similarity disappears in the region  $\tau \lesssim t \ll W^{-1}$  (when the spectral line is still quite broad). In this case the stimulated emission begins already during the virtual-quantum stage. A more accurate statement is that the emission process has a

selective character, so that the emission of the quantum proceeds with formation of a "spin" wave.

5. So far we have investigated the variation of the number of quanta with time. It is also of interest to determine the time variation of the number of molecules at the upper level:

$$\sum_{\substack{r \\ \omega \leq \omega_r \leq \omega + d\omega}} N_2^r(t) = N_2(\omega, t) d\omega,$$

$$N_2(\omega, t) + N_1(\omega, t) = N(\omega),$$

$$N_2(\omega, t) - N_1(\omega, t) = N_-(\omega, t).$$

The above-noted predominant increase in the number of quanta in the center of the spectral distribution leads to the "burning out" of  $N_2(\omega, t)$  and  $N_-(\omega, t)$  near  $\omega = \omega_0$  as a result of the stimulated emission. For example, for  $\delta^2\tau^2 \gg 1$  and  $t \gg \delta^{-1}$  we obtain the following expression for  $N_-(\omega, t)$

$$N_-(\omega, t) = N_-^0(\omega)$$

$$- \frac{4}{\pi} N_2^0 W \tau^2 \left[ \exp \left( \frac{t}{4\tau^2} \frac{\delta}{(\omega - \omega_0)^2 + \delta^2/4} \right) - 1 \right],$$

which allows us to find the time interval in which the linear approximation is valid. Indeed, by definition, the linear approximation is valid in the region where the relative change in the excess population is small:

$$[N_-^0(\omega) - N_-(\omega, t)] / |N_-^0(\omega)| \ll 1,$$

$$N_-^0(\omega) = N_-(\omega, 0).$$

For example, for the center of the spectral distribution we obtain the following time limitation in the case of a large positive initial excess population:

$$t \ll T = \tau^2 \delta \ln \frac{N_-^0}{2N_2^0 W \tau^2} \sim \tau^2 \delta \ln \frac{N_-^{02}}{N_2^0 \gamma},$$

where  $\gamma$  is the effective number of modes in the volume  $V$ , and the parameter  $T$  coincides in order of magnitude with the period of the oscillation of the photon density in the resonant medium (cf. [7]).

For small  $t$ , the distribution  $N_-(\omega, t)$  has as a function of  $\omega$  a single maximum at  $\omega = \omega_0$ . For a sufficiently large positive initial excess population and for large  $t$ :

$$t > \tau^2 \delta \ln (N_-^0 / 2N_2^0 W t)$$

the extremum at the point  $\omega = \omega_0$  becomes a minimum and the curve  $N_- = N_-(\omega, t)$  assumes a "two-hump" form with two identical maxima, lying at points which are symmetrical with respect to  $\omega_0$ ; these points are given by the equation

$$(\omega - \omega_0)^2 = \frac{\delta^2}{4} \left( \frac{1}{\tau^2 \delta \ln (N_-^0 / 2N_2^0 W t)} - 1 \right).$$

This formula determines the location of the maxima approximately, since the change in the form of the curve  $N_- = N_-(\omega, t)$  occurs near the limit of applicability of the linear approximation.

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