## OSCILLATION OF PHOTON DENSITY IN A RESONANCE MEDIUM

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Photon density oscillations in a resonant medium of finite size are treated by quantum electrodynamics for the case when the photon losses can be neglected. The oscillation period and the maximum photon density are determined, and the narrowing of the spectral lines with increasing photon density is explained. A comparison with the experimental data is carried out.

As is well known, to develop the theory of the quantum generator and amplifier it is necessary to evaluate the interaction between the radiation field and a system of N identical quantum objects which have two energy levels and are contained within a resonator, i.e., in a limited volume having two parallel reflecting mirrors. For concreteness, we shall call these identical quantum objects molecules, although the deductions derived below pertain to both gaseous and solid substances filling the resonator cavity. We shall consequently take below the words "isolated molecule" to mean also an impurity atom with the two energy levels under consideration, which is situated in the field of the crystal lattice.

An aggregate of identical two-level molecules forms a resonant medium, in which under certain conditions the emission and absorption processes occur in a special manner. We investigate below the time evolution of these processes for the case when there are no quanta at the initial instant and the distribution of the molecules with respect to the levels is fixed. We find that at the start of the process the molecules radiate independently, in accordance with perturbation theory [1]. Subsequently, however, the mechanism of stimulated collective emission comes into play, and the development of the process differs from that given by perturbation theory or the balance equation. Starting with an instant of time which is characteristic for the given medium, the molecules cease to radiate independently, and act like a collective unit. The time after which the radiation process comes into play is several orders of magnitude shorter than the lifetime of the isolated molecule. In this connection, the problem of the oscillation of the photon density in a resonant medium is solved not by using ordinary balance relations, but on the

basis of the general laws of quantum electrodynamics.

Let us investigate first the model problem of the interaction between the radiation field and two-level molecules inside a bounded volume with perfectly reflecting walls. Our aim is to study the emission and absorption of photons in such a resonant medium without using perturbation theory in canonical form<sup>[1]</sup>. The Hamiltonian H of a quantum-mechanical system consisting of N molecules and a radiation field contained in a volume V is written in the form

$$\begin{split} H &= \sum_{j} \frac{\hbar \omega_{0}}{2} \, \sigma_{z} + \sum_{\mathbf{k}\lambda} \hbar \, \left( c_{\mathbf{k}\lambda} b_{\mathbf{k}\lambda}^{+} + c_{\mathbf{k}\lambda}^{+} b_{\mathbf{k}\lambda} \right) + \sum_{\mathbf{k}\lambda} \hbar \omega_{\mathbf{k}} c_{\mathbf{k}\lambda}^{+} c_{\mathbf{k}\lambda}, \\ b_{\mathbf{k}\lambda}^{+} &= \sum_{j} \sigma_{+}^{j} \, \sqrt{\frac{2\pi}{\hbar \omega_{\mathbf{k}} V}} \, \left( \mathbf{M}^{j} \mathbf{l}_{\mathbf{k}}^{\lambda} \right) e^{i\mathbf{k}\mathbf{x}_{j}}, \\ b_{\mathbf{k}\lambda} &= \sum_{j} \sigma_{-}^{j} \, \sqrt{\frac{2\pi}{\hbar \omega_{\mathbf{k}} V}} \, \left( \mathbf{M}^{*j} \mathbf{l}_{\mathbf{k}}^{\lambda} \right) e^{-i\mathbf{k}\mathbf{x}_{j}}, \\ \sigma_{z}^{j} &= \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \qquad \sigma_{+}^{j} &= \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \qquad \sigma_{-}^{j} &= \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}, \end{split}$$

where the first term in H is the operator of the internal energy of the isolated molecules (J = 1, 2, ..., N), and  $\hbar\omega_0$  is the energy difference between the upper and lower levels of the molecule), the second term is the operator of interaction between the molecules and the radiation field, and the last term is the operator of the free quantized radiation field.  $c_{k\lambda}$  and  $c_{k\lambda}^{\dagger}$  are operators of absorption and emission of a photon with momentum  $\hbar k$  and polarization  $l_k^{\lambda}$ , respectively, with

$$c_{\mathbf{k}\lambda}c_{\mathbf{k}'\lambda'}^{\dagger} - c_{\mathbf{k}'\lambda'}^{\dagger}c_{\mathbf{k}\lambda} = \delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'}, \quad \lambda = 1, 2, \quad \omega_{\mathbf{k}}^2 = c^2\mathbf{k}^2;$$

 $\mathbf{x}_j$  is the coordinate of the center of gravity of the j-th molecule,  $M^j$  the matrix element for the transition of the j-th isolated molecule from the lower

level to the upper level with simultaneous absorption of a photon, and  $M^{*j}$  is the matrix element of the inverse transition with emission of a photon. The matrix element  $M^j$  is connected with the probability W of spontaneous dipole emission of a photon per unit time for an isolated molecule by the relation

$$W = \frac{4\omega_0}{3\hbar c^3} |\mathbf{M}^j|^2.$$

In writing down the Hamiltonian H we took into consideration the vanishing of the average dipole moment of the isolated molecule which is in one of the two states in question. In addition, we have discarded the interaction-operator terms

$$\sum_{\mathbf{k}\lambda}\hbar\left(c_{\mathbf{k}\lambda}b_{-\mathbf{k}(3-\lambda)}+c_{\mathbf{k}\lambda}^{+}b_{-\mathbf{k}(3-\lambda)}^{+}\right),$$

which describe effects in which the number of particles (molecules + photons) is not conserved in an individual interaction between the molecule and the radiation field. It can be shown that in the present problem this part of the operator results in a negligible correction.<sup>1)</sup> Finally, the interaction-operator term proportional to the square of the electromagnetic potential has also been omitted, since it makes a negligible contribution.

For the sake of simplicity we used an expansion of the free electromagnetic field in plane waves, corresponding to a volume V in the form of a parallelepiped. In many cases the resonator has a different form, and the expansion must be carried out in the proper solutions for the resonator of the given type. It is easy to show that the replacement of plane waves by other eigenfunctions does not change the final results.

Using the rule for the differentiation for operators we find that, in a volume V, the average number of photons  $n_{\mathbf{k}\lambda}$  with specified momentum  $\hbar \mathbf{k}$  and polarization  $l_{\mathbf{k}}^{\lambda}$ 

$$n_{\mathbf{k}\lambda} = \langle c_{\mathbf{k}\lambda}^{\dagger} c_{\mathbf{k}\lambda} \rangle$$

is determined by the system of coupled equations

$$\begin{aligned} \frac{d}{dt} n_{\mathbf{k}\lambda} &= i \langle c_{\mathbf{k}\lambda} b_{\mathbf{k}\lambda}^{+} \rangle - i \langle c_{\mathbf{k}\lambda}^{+} b_{\mathbf{k}\lambda} \rangle, \\ \frac{d}{dt} \langle c_{\mathbf{k}\lambda} b_{\mathbf{k}'\lambda'}^{+} \rangle &= -i \left( \omega_{\mathbf{k}} - \omega_{0} \right) \langle c_{\mathbf{k}\lambda} b_{\mathbf{k}'\lambda'}^{+} \rangle - i \left\langle b_{\mathbf{k}'\lambda'}^{+} b_{\mathbf{k}\lambda} \right\rangle \\ &- i \sum_{\mathbf{k}''\lambda''} \langle c_{\mathbf{k}\lambda}^{+} \sigma_{\mathbf{k}\lambda} B_{\mathbf{k}'\lambda',\mathbf{k}''\lambda''} \rangle, \\ \frac{d}{dt} \langle c_{\mathbf{k}\lambda}^{+} b_{\mathbf{k}'\lambda'} \rangle &= i \left( \omega_{\mathbf{k}} - \omega_{0} \right) \left\langle c_{\mathbf{k}\lambda}^{+} b_{\mathbf{k}'\lambda'} \right\rangle + i \left\langle b_{\mathbf{k}\lambda}^{+} b_{\mathbf{k}'\lambda'} \right\rangle \\ &+ i \sum_{\mathbf{k}''\lambda''} \left\langle c_{\mathbf{k}\lambda}^{+} c_{\mathbf{k}''\lambda''} B_{\mathbf{k}''\lambda'',\mathbf{k}''\lambda'} \right\rangle, \end{aligned}$$

$$\frac{d}{dt}\langle b_{\mathbf{k}\lambda}^{+}b_{\mathbf{k}'\lambda'}\rangle = i\sum_{\mathbf{k}''\lambda''}(\langle c_{\mathbf{k}''\lambda''}b_{\mathbf{k}\lambda}^{+}B_{\mathbf{k}''\lambda'',\mathbf{k}'\lambda'}\rangle - \langle c_{\mathbf{k}''\lambda''}^{+}B_{\mathbf{k}\lambda,\mathbf{k}''\lambda''}b_{\mathbf{k}'\lambda'}\rangle),$$
(1)

where

$$B_{\mathbf{k}\lambda,\mathbf{k}'\lambda'} \equiv b_{\mathbf{k}\lambda}^{\dagger}b_{\mathbf{k}'\lambda'} - b_{\mathbf{k}'\lambda'}b_{\mathbf{k}\lambda}^{\dagger}$$
$$= \frac{2\pi}{\hbar\sqrt{\omega_{\mathbf{k}}\omega_{\mathbf{k}'}}V} \sum_{\mathbf{j}} \sigma_{z}^{\mathbf{j}} \left(\mathbf{M}^{\mathbf{j}}\mathbf{l}_{\mathbf{k}}^{\lambda}\right) \left(\mathbf{M}^{\star \mathbf{j}}\mathbf{l}_{\mathbf{k}'}^{\lambda'}\right) e^{\mathbf{i}(\mathbf{k}-\mathbf{k}')\mathbf{x}_{\mathbf{j}}}, \qquad (2)$$

and the angle brackets denote quantum-mechanical averaging.

For concreteness we shall assume below that at the initial moment of time t = 0 the number of photons in the volume V is zero, and the number of molecules in the upper and lower levels are respectively  $N_2^0$  and  $N_1^0$ . Then the particle-number conservation law takes the form

$$N_{-} = N_{-}^{0} - 2n, \qquad (3)$$

where  $N_{-} = N_2(t) - N_1(t)$  is the number of active molecules,  $N_{-}^0 = N_2(0) - N_1(0) \equiv N_2^0 - N_1^0$ ,  $N_1(t) + N_2(t) = N$ , and n is the number of photons in the volume V at an arbitrary instant of time t:

$$n=\sum_{\mathbf{k}\lambda}n_{\mathbf{k}\lambda}.$$

The nondiagonal elements of the operator (2) are connected with the spontaneous emission of the individual molecules (radiation damping) and, as shown by the foregoing estimate, they can be left out if the following inequality is satisfied

$$W\tau \ll 1$$
,  $\tau^{-2} = 2\pi c^3 W | N^0_- | /\omega_0^2 V$ 

which as a rule is the case for quantum generators. The smallness of the nondiagonal terms  $B_{\boldsymbol{k}\lambda,\boldsymbol{k}'\lambda'}$  is also confirmed by the following simple reasoning. Since the number of molecules in the volume (V  $\sim 1\ cm^3$ ) is quite large (N  $\sim 10^{19}$ ), the sum over j in (2) can be replaced by an integral with respect to the coordinate of the center of gravity of the molecules. In such an integration it is necessary to take outside the integral sign the average value of the quantity  $(M^j l_k^\lambda)(M^* l_{k'}^\lambda)$ , averaged over all the directions of the vector  $M^j$ , with  $|\ M^j\ |^2 \equiv |\ M\ |^2$ . As a result the nondiagonal terms of the commutator  $B_{\boldsymbol{k}\lambda,\boldsymbol{k}'\lambda'}$  vanish. Consequently, the diagonal matrix elements of the operator (2) are principal terms, which are conveniently written in the form

$$B_{\mathbf{k}\lambda,\mathbf{k}'\lambda'} = \frac{2\pi |\mathbf{M}|^2}{3\hbar\omega_{\mathbf{k}}V} \,\delta_{\mathbf{k}\mathbf{k}'} \delta_{\lambda\lambda'} \sum_{\mathbf{j}} \sigma_z^{\mathbf{j}}.$$
 (4)

If we confine ourselves to a linear approximation in the number of photons n, then relation (4) assumes the form

<sup>&</sup>lt;sup>1)</sup>The correction introduced is of the order of  $(\omega_0 \tau)^{-2}$ , where  $\tau$  is the characteristic time of the problem, which is defined below.

$$b_{\mathbf{k}\lambda}^{+}b_{\mathbf{k}'\lambda'} - b_{\mathbf{k}'\lambda'}b_{\mathbf{k}\lambda}^{+} = \pm \tau^{-2}\delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'}/4.$$

The upper sign preceding  $\tau^{-2}$  is taken everywhere when  $N_{-}^{0} > 0$ , and the lower one when  $N_{-}^{0} < 0$ . Thus,  $b_{k\lambda}$  and  $b_{k\lambda}^{\dagger}$  behave in this case like Bose operators. An analogous situation occurs also with spin waves in a ferromagnet (see, for example, <sup>[2]</sup>).

Let us consider first the initial stage of the process, when  $n \ll |N_{-}^{0}|$ . Retaining in (1) the terms that are linear in the number of photons, i.e., making the substitution

$$\langle \dots \sum_{j} \sigma_{z}^{j} \rangle = \langle \dots \rangle N_{-}^{0},$$

we obtain with allowance for (3) and (4) the follow-ing equation for  $n_{k\lambda}$ :

$$\ddot{n}_{k\lambda} - (\pm \tau^{-2} - (\omega_k - \omega_0)^2) n_{k\lambda} = N_2^0 / 2\tau^2 |N_-^0|.$$
 (5)

In (5) we have discarded terms of order  $(\omega_{\mathbf{k}} - \omega_0)/\omega_0$ , which are beyond the limits of the chosen approximation.

The solution of (5) satisfying the initial conditions  $n_{k\lambda}(0) = \dot{n}_{k\lambda}(0) = 0$ , is of the form

$$n_{\mathbf{k}\lambda} = N_2^0 (\operatorname{ch} \sqrt{\pm \tau^{-2} - (\omega_{\mathbf{k}} - \omega_0)^2} t - 1)/2\tau^2 | N_-^0 | (\pm \tau^{-2} - (\omega_{\mathbf{k}} - \omega_0)^2).$$
(6)\*

If in (6) we go formally to the limit as  $V \rightarrow \infty$ then we obtain from (6) the usual formula<sup>[1]</sup> for the radiation of  $N_2^0$  excited molecules, which radiate independently of each other:

$$n = \sum_{k\lambda} n_{k\lambda} = \int \frac{2N_2^0 W \omega^2 \sin^2 \left[ (\omega - \omega_0) t/2 \right]}{\pi \mathbf{a}_0^2 (\omega - \omega_0)^2} \, d\omega = N_2^0 W t.$$
(7)

For a finite volume V and a sufficiently short time  $t \ll \tau$  expression (6) also leads to the perturbation-theory formula (7). To prove this fact it is sufficient to verify that if we neglect terms of order  $t\tau^{-1}$  then we have the following equality

$$(2\sin^2\frac{1}{2}\sqrt{\mp\tau^{-2}+(\omega-\omega_0)^2}t)/\pi \ (\mp\tau^{-2}+(\omega-\omega_0)^2)t \approx (2\sin^2\frac{1}{2}(\omega-\omega_0)t)/\pi \ (\omega-\omega_0)^2t.$$
 (7')

Indeed, regarding the left and right halves of (7') as functions of  $\omega$ , we see that the height of the main maximum of both functions is the same, and the half-widths are  $2\pi/t \pm t^{-2}/4\pi$  and  $2\pi/t$ , respectively. The behavior of the functions outside the main maximum  $|\omega - \omega_0| > 2\pi/t$  also coincides if we discard small terms of order  $|\omega - \omega_0|^{-1}\tau^{-1} < t\tau^{-1} \ll 1$ . Finally, the infinite integral with respect to  $\omega$  of both halves of (7'), is the same apart from terms of order  $t\tau^{-1}$ . Inasmuch as the right half of (7') is equal to  $\delta(\omega - \omega_0)$  with sufficient accuracy when  $\omega_0^{-1} \ll t$ , we find that (7) is valid for the time interval  $\omega_0^{-1} \ll t \ll \tau$  independently

\*ch = cosh.

of whether the volume V is finite or infinite.

Thus, at instants close to the initial time  $t \ll \tau$ , the molecules inside the volume V radiate spontaneously as independent objects in full accordance with perturbation theory. However, in subsequent instants of time  $\tau \leq t$ , with increasing accumulation of the photons in the volume V, the mechanism of induced collective emission comes into play and the number of photons increases in accordance with laws that differ from those of perturbation theory. For example, for  $N_{-}^{0} > 0$  the number of photons whose frequencies satisfy the inequality  $(\omega_{\mathbf{k}} - \omega_{0})^{2}$  $\tau^{2} < 1$ , increases exponentially. For photons with frequency  $(\omega_{\mathbf{k}} - \omega_{0})^{2}\tau^{2} \ll 1$  at the instants of time  $\tau < t$  we have from (6)

$$n_{\mathbf{k}\lambda} = \frac{N_2^0}{4N_-^0} \exp\left\{ \left( 1 - \frac{(\omega_{\mathbf{k}} - \omega_0)^2 \tau^2}{2} \right) \frac{t}{\tau} \right\}.$$
 (8)

Formula (6), obtained under the assumption that  $W\tau \ll 1$ , cannot be derived for  $\tau \gtrsim t$  by means of the usual perturbation theory<sup>[1]</sup>. This means that when  $W\tau \ll 1$  and  $\tau \gtrsim t$  perturbation theory is not applicable to problems involving the radiation of resonant molecules in a limited volume. In this case the molecules cease to radiate as independent objects in an external field, but radiate collectively as a unit. The strong bonds within this collective unit are due to the strong interaction between the molecules and the resonant quanta of the radiation field, which at the instant  $t \sim \tau$  becomes common to all the molecules.

In the inverse limiting case  $W\tau \gg 1$ , the term  $\tau^{-2}$  in the parentheses  $(\pm \tau^{-2} - (\omega_{\mathbf{k}} - \omega_0)^2)$  in (5) and (6) should be left out, for in writing out (5) we discarded those terms proportional to W which were connected with the radiation damping of the individual molecules (as if they were isolated). Then formula (6) for  $\omega_0^{-1} \ll t$  also leads to the perturbation-theory result (7).

When  $\omega_{\mathbf{k}} = \omega_0$  and  $N_2^0 = N$  formula (6) coincides with the analogous formula obtained by Faĭn<sup>[3]</sup> in a different manner for the case when the molecules interact with only one type of electromagnetic oscillation (one mode) with a wavelength considerably larger than the linear dimensions of the volume V. The presence of an additional factor  $\frac{1}{3}$ in front of  $|\mathbf{M}|^2$  in (6) is due to the already indicated averaging over all directions of the vector  $\mathbf{M}^j$ .

As can be seen from (8), when the number of photons in the volume V increases exponentially, the width  $\Delta \omega$  of the spectral line narrows down in accordance with

$$\Delta \omega = \sqrt{(8/\tau t) \ln 2}.$$
 (9)

Formula (8) enables us also to estimate the time of the "cascade" radiation, i.e., the time T after the lapse of which the number of photons n in the volume V is close to maximal:  $T \sim \tau$ . The width  $\Delta \omega^0$  of the spectral line has then, in accordance with (8) and (9), an order of magnitude  $\Delta \omega^0 \sim \tau^{-1}$ .

The period T of the oscillation of the number of photons n, and also the width  $\Delta\omega^0$  of the spectral line at the instant when the number of photons in the volume V is a maximum, can be determined with sufficient accuracy by solving the system of equations (1) with account of (3) and (4). To this end we note that the terms of (1) that are not linear in n make a noticeable contribution only when the number of photons in the volume V is quite large,  $n \sim N_{-}^0$ . However, when n is large we also have a large  $n_{k\lambda} \gg 1$ , and accurate to terms of order  $n_{k\lambda}^{-1}$  we have

$$\langle c_{\mathbf{k}\lambda}^{+}c_{\mathbf{k}\lambda}\sum_{j}\sigma_{z}^{j}\rangle = \langle c_{\mathbf{k}\lambda}^{+}c_{\mathbf{k}\lambda}\rangle\langle \sum_{j}\sigma_{z}^{j}\rangle = n_{\mathbf{k}\lambda}N_{-},$$
 (10)

Relations (3), (4), and (10) greatly simplify the system (1), reducing it to only two equations

$$\ddot{n}_{\mathbf{k}\lambda} = 2 \langle b_{\mathbf{k}\lambda}^{+} b_{\mathbf{k}\lambda} \rangle + \left( \pm \frac{1}{2\tau^{2}} - (\omega_{\mathbf{k}} - \omega_{0})^{2} \right) n_{\mathbf{k}\lambda} \mp \frac{1}{\tau^{2} N_{-}^{0}} n n_{\mathbf{k}\lambda},$$
$$\frac{d}{dt} \langle b_{\mathbf{k}\lambda}^{+} b_{\mathbf{k}\lambda} \rangle = \pm \frac{1}{4\tau^{2}} \left( 1 - \frac{2n}{N_{-}^{0}} \right) \dot{n}_{\mathbf{k}\lambda}.$$
(11)

Let us investigate the variation of the total number of photons n with time. Using (11) and the initial conditions, we have

$$\ddot{n} + \frac{3}{2\tau^2 |N_-^0|} n^2 - (\pm \tau^{-2} - \overline{\Delta \omega_{\mathbf{k}}^2}) n = \frac{\gamma N_2^0}{2\tau^2 |N_-^0|}, \qquad (12)$$

where

$$\overline{\Delta \omega_{\mathbf{k}}^2} n = \sum_{\mathbf{k}\lambda} (\omega_{\mathbf{k}} - \omega_0)^2 n_{\mathbf{k}\lambda}, \qquad (13)$$

and  $\gamma$  is the number of modes inside the resonator. It will become clear from what follows that exact knowledge of the number of modes  $\gamma$  is not essential, since it enters under the logarithm sign in the expressions for the period of electromagnetic oscillations and the line width.

The quantity  $\overline{\Delta \omega_{\mathbf{k}}^2}$  is connected with the width of the spectral line and is a slow function of the time. According to (6) we have  $\Delta \omega_{\mathbf{k}}^2 \tau^2 \ll 1$  in the region  $\tau < t$  when  $\mathbb{N}^0 > 0$ . Therefore, in order to obtain a solution in this region we can neglect in (12) the quantity  $\overline{\Delta \omega_{\mathbf{k}}^2}$  compared with  $\tau^{-2}$ . The solution obtained must, generally speaking, be a continuation of the function (7), which is the rigorous solution of the problem when  $t \leq \tau$ . Since, however, we are interested in the period T of the variation of the function n(t) and its behavior near the maximum (t ~ T/2,  $\tau \ll$  T), it is sufficient to choose as the additional conditions the zero values of the function n(t) and its derivative when t = 0. Indeed, the continuity conditions  $n(\tau) = N_2^0 W \tau$ ,  $n(\tau) = N_2^0 W$  introduce a small parameter  $W \tau$ , and when n(t) is expanded in powers of this parameter we arrive at a solution with zero initial conditions at t = 0.

The solution of (12) is a periodic function n(t), defined in the region  $\tau \lesssim t \lesssim T - \tau$  by the relation

$$2\tau \bigvee \frac{|N_{-}^{0}|}{\alpha_{1} - \alpha_{2}} F\left( \arcsin \sqrt{\frac{(\alpha_{1} - \alpha_{2})n}{\alpha_{1}(n - \alpha_{2})}}, \sqrt{\frac{\alpha_{1}}{\alpha_{1} - \alpha_{2}}} \right)$$
$$= \begin{cases} t & \text{when } \tau \leq t \leq T/2 \\ T - t & \text{when } T/2 \leq t \leq T - \tau \end{cases},$$
(14)

where the function F is the incomplete elliptic integral of the first kind, and  $\alpha_1$  and  $\alpha_2$  are respectively the positive and negative roots of the quadratic equation

$$x^2 - N_{-}^0 x - \gamma N_2^0 = 0.$$

The period T of the variation of the function n(t) is equal to

$$T = 4\tau \sqrt{\frac{|N_{-}^{0}|}{\alpha_{1} - \alpha_{2}}} F\left(\frac{\pi}{2}, \sqrt{\frac{\alpha_{1}}{\alpha_{1} - \alpha_{2}}}\right), \quad (15)$$

where  $F(\pi/2, \sqrt{\alpha_1/\alpha_1 - \alpha_2})$  is the complete elliptic integral of the first kind. The maximum number of photons  $n_{max}$  in the volume V is equal to

$$n_{max} = \alpha_1. \tag{16}$$

Let us consider formulas (15) and (16) in the following particular case:  $N_{-}^{0} > 0$ ,  $\gamma N_{2}^{0} \ll N_{-}^{02}$ . Then

$$n_{max} = N_{-}^{0}, \ T = 2\tau \ln \left( \frac{16N_{-}^{02}}{\gamma N_{2}^{0}} \right),$$
 (17)

where the term in  $n_{max}$ , proportional to the small parameter  $N_2^0/N_-^{02}$ , has been left out, for in some cases (for example, when  $N_2^0 = N$ ) it makes a correction that lies beyond the limits of the chosen approximation (10).

If 
$$|\mathbb{N}^{0}_{-}| \ll \sqrt{\gamma \mathbb{N}^{0}_{2}}$$
,  $\gamma < \mathbb{N}^{0}_{2}$ , and  $\overline{\Delta \omega_{\mathbf{k}}^{2} \tau^{2}} \ll 1$ , then  
 $n_{max} = \sqrt{\gamma \mathbb{N}^{0}_{2}}, \quad T = \pi \tau \left(4N^{02}_{-}/\gamma N^{0}_{2}\right)^{1/4}.$  (18)

When  $N_{-}^{0} < 0$  and  $\gamma N_{2}^{0} \ll N_{-}^{02}$  the nonlinear term in (12) can be neglected, and the solution of the linearized equation (12) for any t is of the form

$$n(t) = \sum_{\mathbf{k}\lambda} n_{\mathbf{k}\lambda}(t),$$

where  $n_{k\lambda}(t)$  is given by formula (6).

It is interesting to trace the time variation of <u>n</u> in the most important case  $N_{-}^{0} > 0$ ,  $\gamma N_{2} \ll N_{-}^{02}$ ,  $\Delta \omega_{k}^{2} \tau^{2} \ll 1$ , starting from the instant when an avalanche-like increase in the number of photons begins. From (12), in the approximation that is linear in the number of photons n, we have

$$n = (\gamma N_2^0 / 4 N_-^0) e^{t/\tau}, \quad n \ll N_-^0.$$
(19)

Let us determine, further, the oscillation of the number of photons  $n_{k\lambda}$  at an individual mode. As follows from (6) and (8), the greatest contribution to n is made for  $N_{-}^{0} > 0$  only by those modes, whose frequencies lie in the interval  $(\omega_{k} - \omega_{0})^{2} \tau^{2} \leq 1$ . We denote the indicated effective number of modes by  $\gamma$ :

$$\gamma = \sum_{\mathbf{k}\lambda} \mathbf{1} = \frac{8\pi V}{\omega_0 \tau \lambda_0^3},\tag{20}$$

where  $\lambda_0 = 2\pi c/\omega_0$  is the length of the electromagnetic wave. Since exact knowledge of the number of modes is not essential, we can put when solving equation (11) with  $N_-^0 > 0$ , in accordance with (6) and (8),

$$n = \sum_{\mathbf{k}\lambda} n_{\mathbf{k}\lambda} = \gamma n_{\mathbf{k}\lambda}, \qquad (21)$$

where it is sufficient to use Eq. (20) for  $\gamma$ . For small t <  $\tau$ , relations (20) and (21) become invalid, but in this region the terms quadratic in n in (11) are negligibly small. In the region where the terms quadratic in n become significant relations (20) and (21) are satisfied with sufficient accuracy.

Bearing (11) and (21) in mind, we arrive in the case of  $N_{-}^{0} > 0$  at the nonlinear equation

$$\ddot{n}_{\mathbf{k}\lambda} + \frac{3\gamma}{2\tau^2 N_{-}^0} n_{\mathbf{k}\lambda}^2 - \left(\frac{1}{\tau^2} - (\omega_{\mathbf{k}} - \omega_0)^2\right) n_{\mathbf{k}\lambda} = \frac{N_2^0}{2\tau^2 N_{-}^0}$$

the solution of which is of the form

$$2\tau \sqrt{\frac{N_{-}^{0}}{\gamma(\beta_{1}-\beta_{2})}} F\left(\operatorname{arc\,sin} \sqrt{\frac{(\beta_{1}-\beta_{2}) n_{k\lambda}}{\beta_{1}(n_{k\lambda}-\beta_{2})}}, \sqrt{\frac{\beta_{1}}{\beta_{1}-\beta_{2}}}\right)$$
$$= \begin{cases} t & \text{when } 0 \leqslant t \leqslant T/2\\ T-t & \text{when } T/2 \leqslant t \leqslant T \end{cases},$$
(22)

where  $\beta_1$  and  $\beta_2$  are respectively the positive and negative roots of the quadratic equation

$$\gamma x^2 - N_-^0 (1 - (\omega_k - \omega_0)^2 \tau^2) x - N_2^0 = 0.$$

The maximum value of the function  $n_{k\lambda}(t)$  is equal to  $\beta_1$ , and the period of its variation is equal to T.

The use of (21) to solve Eq. (11) represents a certain approximation, which weakens the coupling between the individual modes. However, the closer n is to its maximum value, the more accurate this approximation, since the contribution made to n on the part of the effective modes from the interval (20) is in this case maximal. In this connection, the function (22) defines perfectly satisfactorily the behavior of  $n_{k\lambda}(t)$  not only for small t, when the nonlinearity in the equation can be neglected, but

also for  $t \sim T/2$ . Therefore it is convenient to use the solution (22), for example, when determining the width of the spectral line, for in this case use will be made of the form  $n_{\mathbf{k}\lambda}(T/2)$  (as a function of  $\omega_{\mathbf{k}} = \mathbf{ck}$ ), namely  $n_{\mathbf{k}\lambda}(T/2) \equiv n_{\omega_{\mathbf{k}}}$ , only in the interval of its values from  $n_{\omega_0}$  to  $n_{\omega_0}/2$ . The function  $n_{k\lambda}(t)$  with a frequency  $\omega_k = ck$  that lies far beyond the limits of the width of the spectral line, is described by relation (22) to a much lesser approximation. In particular, the period of variation of  $n_{k\lambda}(t)$  becomes in this case strongly dependent on  $(\omega_{\mathbf{k}} - \omega_0)^2$ , whereas according to (11) the periods n(t) and  $n_{\mathbf{k}\lambda}(t)$  should coincide. This is connected with the fact that the use of (20) and (21) greatly reduces the term  $nn_{k\lambda}$  when the substitution (21) is made when solving Eqs. (11) for  $n_{k\lambda}(t)$  with frequency  $\omega_k = ck$  far beyond the limits of the width of the spectral line.

With the aid of (22) we obtain the width  $\Delta\omega^0$  of the spectral line at the instant when the number of photons n in the volume V is maximal. In the most interesting case  $N_-^0 > 0$ ,  $\gamma N_2^0 \ll N_-^{02}$ ,  $(\tau \Delta \omega^0)^2 \ll 1$  the line width is

$$\Delta\omega^{0} = \frac{4}{\tau} \left( \ln \left( 1 + \sqrt{2} \right) / \ln \frac{16 N_{-}^{02}}{\gamma N_{2}^{0}} \right)^{1/2}$$
 (23)

We have made above, for the sake of simplicity, some definite physical approximations. Thus, for example, we have neglected the motion of the molecules, their collision in the case of gaseous substances, and the level shifts due to the Stark effect in the alternating field of the vibrating lattice. We assume that allowance for these effects does not change the basic principal results of the work.

So far we have considered the idealized problem of oscillations in a resonator with ideally reflecting walls in the presence of some initial conditions. In the real case accumulation of particles in the upper energy level in the resonator occurs gradually, with continuing pump action in the quantum generator. During this period the electromagnetic processes in the resonator depend strongly on the pumping rate and on the emergence of the photons to the outside. However, once the population shift reaches a threshold value  $N_{-}^{0} = N_{-thr}^{0}$ , a rapid avalanche-like coherent radiation begins. During this coherent radiation, the pumping and the emergence of the photons to the outside are slow processes, which influence weakly the development of the photon cascade. Thus, starting with the instant of the threshold  $N_{-}^{0} = N_{-thr}^{0}$  and continuing to the end of the cascade emission (during one spike), conditions are apparently realized in the quantum generator under which the pumping and

the emergence of the photons from the resonator can be neglected, and not only the growth of the number of photons, but also the narrowing down of the spectral line are described with sufficient accuracy by the formulas of the present paper. Such a situation will be repeated for each succeeding spike, although the value of  $N_{-thr}^0$  and the distance between spikes is determined by the pumping rate and by the losses. It is possible that this occurs in each generator in which a spiking mode is clearly observed. It must be noted that for a gasfilled quantum generator, the inequality  $W\tau \gg 1$ is usually satisfied, i.e., the condition for the applicability of the formulas of the present paper is violated.

In experiments with quantum generators there are always losses of the total energy of the photons that are situated inside the cavity, due to relaxation processes and also due to the absorption of photons in the cavity walls and to the departure of the photons from the generator. If these losses, together with the pumping, are introduced into the present problem, then we obtain for the initial change in population  $N^0_-$  a certain threshold value  $N_{-thr}^0 > 0$ , above which  $(N_{-}^0 > N_{-thr}^0)$  there will be observed an exponential increase in the number of photons. Therefore, from the experimental point of view, the most interesting case is when  $N_{-}^{0} > 0$ and  $\gamma N_2^0 \ll N_{-}^{02}$ , and the corresponding formulas are (9), (17), and (23). However, we should now consider among the indicated modes only those in which the vector  $\mathbf{k}$  is approximately perpendicular to the parallel mirrors. In other words, the vector **k** must lie inside a small solid angle  $\Delta \Omega$ , the magnitude of which can be determined experimentally from the light beam emerging from the generator or can be estimated by the characteristic parameters of the problem. It is easy to show that during a time T the number of photons leaving the volume V and having a momentum that does not lie in the solid angle  $\Delta \Omega$  is negligibly small. Thus, as applied to real devices, we can take the number of modes  $\gamma$  in all the formulas to be represented by

$$\gamma = \sum_{\mathbf{k}\lambda} = \frac{2\Delta\omega V}{\omega_0\lambda_0^3}\Delta\Omega,\tag{24}$$

where the width  $\Delta \omega$  of the spectral interval is approximately equal to  $\tau^{-1}$  if  $N_{-}^{0} > 0$ , and equal to  $\Delta \omega^{0}$  from (23) if  $N_{-}^{0} > 0$ , and  $\gamma N_{2}^{0} \ll N_{-}^{02}$ . In other cases  $\Delta \omega$  must be separately calculated.

By virtue of the periodicity of the process, the number of photons n in a real resonator, after going through its maximum, will start decreasing and the excess population  $N_{-}$  will increase and

tend to a maximum value  $N_{-max}$ . However,  $N_{-max}$  remains lower than  $N_{-thr}^{0}$ , owing to the losses due to the increased absorption and the emergence of the photons at the instant when n has a maximum value. Consequently, the device must "wait" until the pumping causes the change in population to reach its threshold value  $N_{-thr}^{0}$  again, after which the process is repeated. The "waiting" time, i.e., the distance between spikes, can also be estimated on the basis of the results obtained, by specifying the rate of pumping and the losses.

We note that in some crystal lattices or in the presence of an external magnetic field, a case can occur when the vectors  $M^j$  of all the active molecules are parallel.  $M^j \equiv M$ . Then there will be no averaging over the directions of  $M^j$  in (4). This will cause the probability of dipole emission W of the isolated molecule to be replaced in the final formulas (17) and (23) by

$$3W (MM^* - (Mn) (M^*n)/MM^*),$$
 (25)

where **n** is a unit vector perpendicular to the two reflecting mirrors of the resonator. In the case of quadrupole radiation, the dependence of the angles between **n** and the axis of the quadrupole-moment tensor of the molecules in (25) is of a more complicated form, but the basic formulas (17) and (23) with averaged direction of  $M^j$  remain valid for any type of radiation.

Let us apply the formulas obtained to a quantum generator using pink ruby  $\lfloor 4-6 \rfloor$ . Starting with the power of the generated light, we can readily calculate the average energy per spike. Then, taking (17) into account, we obtain the threshold value of the excess population  $N_{-}^{0} = N_{-thr}^{0} \approx 10^{-3} N$ , where N  $\approx 3.2 \times 10^{19}$ . The volume of the cavity is  $V \approx 2 \text{ cm}^3$ . The energy difference  $\hbar \omega_0$  between levels of the active molecule corresponds to a wavelength of the photon  $\lambda_0\approx 7\times 10^{-5}~\text{cm}$  . The spontaneous emission probability W of an individual active molecule is  $W \approx 0.3 \times 10^3 \text{ sec}^{-1}$ . In accordance with the angle aperture of the outgoing beam, we put  $\Delta\Omega \approx 10^{-3}$ , and we then obtain for the number of modes<sup>2)</sup>  $\gamma = 10^4$ . Consequently, the duration of the individual spike in the case of a pink-ruby laser is, according to (17),  $T \approx 10^{-8}$  sec, and the width of the spectral line in accordance

<sup>&</sup>lt;sup>2)</sup>If we assume that the process of generation in each individual waveguide of the ruby crystal proceeds independently, then to calculate  $\gamma$  by means of (24) we must take V to mean the volume of the individual waveguide. We then obtain for  $\gamma$  a value of the order of unity. Since  $\gamma$  is under the logarithm sign, such a variation of the number  $\gamma$  will not change noticeably the numerical values of T and  $\Delta \omega^{0}/\omega_{0}$ .

with (23) is  $\Delta \omega^0 / \omega_0 \approx 10^{-6}$ , which is in satisfactory agreement with experiments <sup>[4-6]</sup> in which  $T \approx 10^{-7}$  and  $\Delta \omega^0 / \omega_0$  ranges from  $10^{-5}$  to  $10^{-7}$ .

A better condition for the applicability of (9), (17), and (23) occurs for a laser operating with initial excess population  $N^0_-$  which exceeds greatly  $N^{0}_{-thr}$ , when the losses per spike decrease noticeably. Apparently such a condition is attained, for example, by modulating the Q of the cavity [7-8], thereby ensuring considerable accumulation of active particles during the time of action of the pump. When the Q of the cavity is rapidly increased, the threshold value of the excess population  $N^{0}_{-thr}$  decreases rapidly. As a result, a condition is produced whereby the "initial" excess population  $N^0_-$  greatly exceeds  $N^0_{-thr}$ . During the succeeding "de-excitation" of the molecules, the energy losses are in this case relatively small, and the applicability of formulas (9), (17), and (23) is sufficiently well founded. However, in this case after a lapse of time T the maximum value of the excess population may turn out to exceed the threshold value,  $N_{-max} > N_{-thr}^{0}$ . Therefore a few oscillations will occur during a certain time t<sub>f</sub>, until  $N_{-max}$  drops below the threshold value  $N_{-thr}^0$ .

This circumstance causes the flash time  $t_f$  to exceed the period of the oscillations,  $t_f > T$ , but the width of the spectral line will as before be given by (23).

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