

ELECTROMAGNETIC OSCILLATIONS IN A RESONANCE MEDIUM

A. I. ALEKSEEV

Moscow Engineering-physics Institute

Submitted to JETP editor September 9, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) 48, 879-889 (March, 1965)

Electromagnetic oscillations are investigated for a resonant medium consisting of identical two-level molecules; pumping and relaxation losses are not taken into account. The spectral composition of the radiation is determined as well as the width of the spectral line in the extreme cases of small and large overpopulation of the levels.

IN a previous paper^[1], we obtained the oscillation of the density of photons in a resonant medium consisting of two-level molecules, as well as the distribution of photons in the variable $\omega_{\mathbf{k}} = |\mathbf{k}|c$, (here \mathbf{k} is the wave vector). However, when the radiation emerges from the resonant medium the conserved quantity is not $\omega_{\mathbf{k}}$ but the energy of the photons or the frequency ω of oscillation of the electric (magnetic) field intensity vector. Therefore, it is of interest to obtain the spectral decomposition of the radiation in the variable ω . A model problem is considered below on the radiation of identical two-level molecules (without relaxation losses and pumping losses) under the conditions that at the initial instant there are no photons and the distribution of molecules over the levels is fixed. The method proposed can be shown to be useful for the investigation of the general case with relaxation and pumping losses. Inasmuch as only general laws of quantum electrodynamics are used in the solution of the given problem, the results obtained are also important from the viewpoint of principle, since they enable us to estimate to what extent the application of other methods, for example, balance equations, is justified.

The deviation of the results found from the solution of ordinary balance equations^[2] is essentially brought about by the fact that the latter describe the radiation of an individual molecule (or system of independent molecules) in an external electromagnetic field. In addition, in the present problem it is necessary to take into account that a large number of resonant molecules can be found in a volume of the order of the cube of the radiated wavelength; these molecules radiate collectively. Both the law of radiation of resonant molecules obtained below and the spectral line widths which have been obtained, and which have been shown to be proportional to the molecular

density, demonstrate this fact. For the given problem, the ordinary balance equations must be transformed somewhat and supplemented by new terms which take into account the resonance character of the radiation of the system of molecules.

We shall write the Hamiltonian H of the set of identical two-level molecules and the radiation field in the volume V in the form

$$H = \frac{\hbar\omega_0}{2} N_- + \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}} n_{\mathbf{k}} - \frac{1}{c} \sum_{s\mathbf{k}} \mathbf{A}_s(\mathbf{k}) \mathbf{j}_s(\mathbf{k}),$$

where

$$n_{\mathbf{k}} = \sum_{s\lambda} c_{s\mathbf{k}\lambda}^+ c_{s\mathbf{k}\lambda}, \tag{1}$$

$$\mathbf{A}_s(\mathbf{k}) = (2\pi\hbar c^2/\omega_{\mathbf{k}})^{1/2} \sum_{\lambda} (c_{s\mathbf{k}\lambda} \mathbf{l}_{\mathbf{k}}^{\lambda} + c_{s\mathbf{k}\lambda}^+ \mathbf{l}_{\mathbf{k}}^{\lambda}); \tag{2}$$

$$j_{\alpha s}(\mathbf{k}) = \sqrt{2/V} \times \sum_j (k_{\alpha} k_{\beta} / k^2 - \delta_{\alpha\beta}) (\sigma_{+j} M_{\beta}^j + \sigma_{-j} M_{\beta}^{*j}) \Phi_s(\mathbf{k}\mathbf{x}_j), \tag{3}$$

$$\Phi_s(\mathbf{k}\mathbf{x}_j) = \begin{cases} \cos \mathbf{k}\mathbf{x}_j & \text{for } s = 1; \\ \sin \mathbf{k}\mathbf{x}_j & \text{for } s = 2; \end{cases} \quad N_- = \sum_j \sigma_z^j, \tag{4}$$

$$\sigma_z^j = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad \sigma_{+}^j = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \quad \sigma_{-}^j = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}. \tag{5}$$

The first and second components in H are the operators of the energy corresponding to the isolated molecules ($j = 1, 2, \dots, N$) and the free radiation field, while the last component is the operator of their interaction energy; $\hbar\omega_0$ is the difference in energy of the upper and lower levels of the isolated molecule. Furthermore, $c_{s\mathbf{k}\lambda}$ and $c_{s\mathbf{k}\lambda}^+$ are the operators corresponding to absorption and creation of the \mathbf{k} -th photon with polarization $\mathbf{l}_{\mathbf{k}}^{\lambda}$ and the coordinate dependence of the vector potential (4), while

$$c_{s\mathbf{k}\lambda} c_{s'\mathbf{k}'\lambda'}^+ - c_{s'\mathbf{k}'\lambda'}^+ c_{s\mathbf{k}\lambda} = \delta_{ss'} \delta_{\mathbf{k}\mathbf{k}'} \delta_{\lambda\lambda'},$$

$$s = 1, 2, \quad \lambda = 1, 2, \quad \omega_{\mathbf{k}}^2 = \mathbf{k}^2 c^2.$$

Finally, \mathbf{x}_j is the coordinate of the center of mass of the j -th particle while M_{α}^j is the matrix element of transition of the j -th molecule from the lower level to the upper with simultaneous absorption of the k -th photon:

$$M_{\alpha}^j = -i\omega_0 d_{\alpha}^j + \frac{c}{2} e_{\alpha\beta\gamma} \left(\mu_{\gamma}^j \frac{\partial}{\partial x_{\beta j}} - \mu_{\beta}^j \frac{\partial}{\partial x_{\gamma j}} \right) - \frac{i\omega_0}{6} Q_{\alpha\beta}^j \frac{\partial}{\partial x_{\beta j}}$$

where d_{α}^j , μ_{α}^j and $Q_{\alpha\beta}^j$ are the matrix elements of the dipole, magnetic dipole, and quadrupole moments of the j -th isolated molecule, respectively; $e_{\alpha\beta\gamma}$ is a completely antisymmetric unit pseudo-tensor of third rank,^[3] while the derivatives $\partial/\partial x_{\alpha j}$ act on the functions $\Phi_s(\mathbf{k} \cdot \mathbf{x}_j)$, appearing in (3) at the right. The quantities μ_{α}^j and $Q_{\alpha\beta}^j$ are preserved in M_{α}^j , since the long lifetime of the excited isolated molecule gives evidence on the forbiddenness of dipole radiation in many practically important cases. Moreover, the mean quantum mechanical value of the magnetic dipole moment of an isolated molecule found in one of the states considered is assumed to be equal to zero. Repetition of the vector indices is everywhere taken to mean summation from 1 to 3.

In the quantization of the free radiation field, the operator of the vector potential $\mathbf{A}(\mathbf{x})$ is expanded in harmonic functions (4), which are the eigenfunctions of a resonator with volume V and the shape of a parallelepiped:

$$\mathbf{A}(\mathbf{x}) = \sqrt{\frac{2}{V}} \sum_{\mathbf{k}} (\mathbf{A}_1(\mathbf{k}) \cos \mathbf{kx} + \mathbf{A}_2(\mathbf{k}) \sin \mathbf{kx}), \quad (6)$$

where $\mathbf{A}_1(\mathbf{k})$ and $\mathbf{A}_2(\mathbf{k})$ are given in (2) and $\text{div } \mathbf{A}(\mathbf{x}) = 0$. Summation over \mathbf{k} is carried out everywhere over the half-space $k_z \geq 0$. In a number of cases, the volume of the resonator differs from the parallelepiped, and the expansion of the free electromagnetic field must be carried out over the eigenfunctions of the resonator of given shape. However, replacement of the harmonic functions (4) by other eigenfunctions does not change any of the subsequent conclusions, inasmuch as the final results contain only those parameters which are not associated with the choice of the specific eigenfunctions.

For the determination of the width of the spectral lines of the radiation of resonant molecules, it is necessary to investigate the equation for the vector potential. Using the rule of differentiation of operators in quantum mechanics, we easily obtain a closed set of equations for the Fourier components of the operators of the vector potential (2)

and of the polarization current (3), and also the operator N_- of the overpopulation of the levels:

$$\dot{\mathbf{A}}_s(\mathbf{k}) + \omega_k^2 \mathbf{A}_s(\mathbf{k}) = 4\pi c \mathbf{j}_s(\mathbf{k}), \quad (7)$$

$$\dot{\mathbf{j}}_s(\mathbf{k}) + \omega_0^2 \mathbf{j}_s(\mathbf{k}) = -\frac{\omega_0^2}{4\pi c \tau_0^2} N_- \mathbf{A}_s(\mathbf{k}), \quad (8)$$

$$\dot{N}_- = \frac{2}{\hbar \omega_0 c} \sum_{s\mathbf{k}} \mathbf{A}_s(\mathbf{k}) \mathbf{j}_s(\mathbf{k}), \quad (9)$$

which are valid for operators in both the Schrödinger and the Heisenberg representations.

In writing down (8), it was assumed that the orientation of the dipole axis d^j , μ^j and the tensor of the quadrupole moment $Q_{\alpha\beta}^j$ of the individual molecules is random. Therefore, the quantity $M_{\alpha}^j M_{\beta}^{*j}$, averaged over all possible orientations of the axes of the dipoles and of the tensor of the quadrupole moment was taken out from under the summation sign in j :

$$2 \sum_j (M_{\alpha}^j \Phi_s(\mathbf{kx}_j)) (M_{\beta}^{*j} \Phi_s(\mathbf{kx}_j)) \sigma_z^j A_{\beta s}(\mathbf{k}) = \overline{M_{\alpha}^j M_{\beta}^{*j}} N_- A_{\beta s}(\mathbf{k}), \quad (10)$$

$$\overline{M_{\alpha}^j M_{\beta}^{*j}} A_{\beta s}(\mathbf{k}) = \frac{\hbar c^3}{4\omega_0} W A_{\alpha s}(\mathbf{k}), \quad W = W_d + W_{\mu} + W_Q, \quad (11)$$

where W_d , W_{μ} , and W_Q are respectively the probabilities of spontaneous dipole, magnetic dipole, and quadrupole radiation of a photon per unit time for an individual isolated molecule.

The rapidly oscillating functions $\sin^2 \mathbf{k} \cdot \mathbf{x}_j$ and $\cos^2 \mathbf{k} \cdot \mathbf{x}_j$ under the summation over j in (10) are replaced by the mean value $1/2$, since a large number of molecules are distributed randomly in the volume V . Here the derivative operators $\partial/\partial x_{\alpha j}$ entering into M^j on the right hand side of Eq. (10) are replaced by the corresponding components of the vector \mathbf{k} . In (11), the components of order $(\omega_0 - \omega_{\mathbf{k}})/\omega_0$ are omitted as lying beyond the limits of accuracy of calculation. Moreover, the terms associated with radiative decay of the excited state of the individual isolated molecule are neglected in Eq. (8). These terms contribute a trivially small amount to the problem considered below if $W\tau \ll 1$ (see, for example,^[4]), where

$$1/\tau^2 = |N_-^0|/\tau_0^2, \quad 1/\tau_0^2 = 2\pi c^3 W/\omega_0^2 V.$$

As is well known,^[1,4] the overpopulation $N_-(t)$ and the number of photons $n_{\mathbf{k}}(t)$ change little during a time of the order of ω_0^{-1} , if $(\omega_0 \tau)^{-1} \ll 1$. Then the solution of (7)–(9) for the Heisenberg operators can be conveniently sought in the form

$$\dot{\mathbf{A}}_s(\mathbf{k}, t) - i\omega_0 \mathbf{A}_s(\mathbf{k}, t) = \mathbf{X}_{s\mathbf{k}}(t) e^{-i\omega_0 t}, \quad (12)$$

$$\dot{\mathbf{j}}_s(\mathbf{k}, t) - i\omega_0 \mathbf{j}_s(\mathbf{k}, t) = \mathbf{Y}_{s\mathbf{k}}(t) e^{-i\omega_0 t}, \quad (13)$$

where $X_{sk}(t)$ and $Y_{sk}(t)$, as well as $N_-(t)$ and $n_k(t)$, are slowly varying functions of t . Omitting the rapidly oscillating terms with the factor $\exp(\pm 2i\omega_0 t)$, which is equivalent to averaging Eqs. (7)–(9) over an interval of time Δt out of the interval $\omega_0^{-1} \ll \Delta t \ll \tau$, we obtain

$$2i\omega_0 \dot{X}_{sk} = (\omega_k^2 - \omega_0^2)X_{sk} - 4\pi c Y_{sk}, \quad (14)$$

$$2i\omega_0 \dot{Y}_{sk} = \frac{\omega_0^2}{4\pi c \tau_0^2} N_- X_{sk}, \quad (15)$$

$$2i\omega_0^2 \hbar c \dot{N} = \sum_{sk} (X_{sk}^+ Y_{sk} - Y_{sk}^+ X_{sk}), \quad (16)$$

where components which are smaller than the remaining terms by a factor of $(\omega_0 \tau)^{-1}$ are also discarded in Eq. (16). If we keep the discarded components, then we obtain a correction of the order of $(\omega_0 \tau)^{-1}$ to the function n_k , and in the calculation of the frequency of vibration of the vector potential, we obtain a correction of the order $(\omega_0 \tau)^{-2}$.

Combining (14) with (16), and then (15) with (16), and finally, using (14) and (15), we find the integrals of motion:

$$\sum_{sk} X_{sk}^+ X_{sk} + 4\pi c^2 \hbar \omega_0 (N_- - I_1) = 0, \quad (17)$$

$$8\pi \tau_0^2 \sum_{sk} Y_{sk}^+ Y_{sk} + \hbar \omega_0^3 (N_-^2 - 2N_- - I_2) = 0, \quad (18)$$

$$4\pi c (X_{sk}^+ + Y_{sk} + Y_{sk}^+ + X_{sk}) + (\omega_0^2 - \omega_k^2) (X_{sk} + X_{sk} - 4\pi c^2 \hbar \omega_0 I_k) = 0. \quad (19)$$

Here I_1 , I_2 , and I_k are constants determined from the initial conditions.

According to the definition of the operator X_{sk} (12), we have

$$\sum_s X_{sk}^+ X_{sk} = 8\pi c^2 \hbar \omega_0 n_k. \quad (20)$$

Therefore, omitting terms of order $(\omega_k - \omega_0)/\omega_0 \sim (\omega_0 \tau)^{-1} \ll 1$, the relation (17) can also be rewritten in the form of a law of conservation of the number of particles (photons and excited molecules):

$$2n + N_- = I_1, \quad n = \sum_k n_k. \quad (21)$$

By keeping (14)–(21) in mind, it is not difficult to find equations for the operators of overpopulation N_- and the total number of photons n . For example, we obtain the following set of equations for n_k :

$$2\tau_0^2 \ddot{n}_k + (2n - I_1)n_k + 2\tau_0^2 (\omega_k - \omega_0)^2 (n_k - I_k) - 2\tau_0^2 \varphi_k = 0, \quad (22)$$

$$2\tau_0^2 \ddot{\varphi}_k + n \dot{n}_k + \dot{n}_k n + (1 - I_1) \dot{n}_k = 0, \quad (23)$$

where the notation

$$\varphi_k(t) \equiv -\frac{\pi}{\hbar \omega_0^3} \sum_s Y_{sk}^+ Y_{sk}$$

is used.

The equations (22) and (23) are written down for the Heisenberg operators. However, we are interested in the mean quantum mechanical value of the number of photons with given ω_k^2 :

$$\langle n_k \rangle \equiv n_k(t).$$

Therefore, we take the quantum-mechanical mean of the operator equations (22) and (23), using the approximation

$$\langle nn_k \rangle = \langle n \rangle \langle n_k \rangle \equiv nn_k. \quad (24)$$

Equation (24) becomes worse for small n , but the quadratic terms in n are as a rule not important in this region. For large n , Eq. (24) is satisfied with great accuracy.

We shall consider below the more interesting case in which there are no quanta at the initial time $t_0 = 0$, while the number of molecules in the upper and lower levels are equal to $N_2(0) = N_2^0$ and $N_1(0) = N_1^0$, respectively ($N_2^0 - N_1^0 = N_-^0$). If at the initial instant the subsystems (molecules and radiation field) have not interacted and were described by the eigen wave functions, then

$$I_1 = N_-^0, \quad I_k = 0, \quad \varphi_k(0) = 2N_2^0 / \tau_0^2. \quad (25)$$

We shall study first the linear region in n : $n \ll |N_-^0|$ and $|N_-^0| \gg 1$, which is also interesting by reason of the fact that the problem possesses an exact solution. From (22), (23) and (25) we obtain the following equation

$$\ddot{n}_k + \Omega_k^2 n_k = 2N_2^0 / \tau_0^2, \quad (26)$$

the solution of which has the following form for the given initial condition $n_k(0) = \dot{n}_k(0) = 0$:

$$n_k(t) = \frac{4N_2^0}{\tau_0^2 \Omega_k^2} \sin^2 \frac{\Omega_k t}{2},$$

$$\Omega_k^2 = (\omega_k - \omega_0)^2 - \frac{N_-^0}{\tau_0^2}. \quad (27)$$

Summing n_k in (27) over all k , we find the law for the change of the total number of photons with time:

$$n(t) = \sum_k n_k(t) = N_2^0 W \tau \int_0^{t/\tau} J_0(x) dx, \quad N_-^0 < 0. \quad (28)$$

If $N_-^0 > 0$, then the argument x of the Bessel function of zero order $J_0(x)$ is replaced in (28) by the purely imaginary quantity ix .

Close to the initial instant $\omega_0^{-1} \ll t \ll \tau$, Eq. (28) coincides with the ordinary perturbation theory.^[5] This means that at the beginning of the process, the molecules radiate spontaneously as independent objects. For $\tau \ll t$ and a negative overpopulation that is large in modulus $|N_-^0| \gg N_2^0 W \tau$, the total number of photons (28) tends to the equilibrium value:

$$n_{\text{eq}} = N_2^0 W \tau, \quad (29)$$

while the oscillation of the number of photons in the individual mode n_k has always a harmonic character (27).

The asymptotic behavior $n(t)$ for $\tau \ll t$ and a large negative overpopulation $N_-^0 \gg N_2^0 W \tau$ has the form

$$n(t) = N_2^0 W \tau (\tau / 2\pi t)^{1/2} e^{t/\tau}, \quad n(t) \ll N_-^0. \quad (30)$$

At the same time, the asymptote $n_k(t)$ in the same region has another functional dependence on t . This result is brought about by the collective character of the radiation of resonant molecules when $\tau \lesssim t$, and cannot be explained on the basis of the canonical perturbation theory.^[5]

Thus the correct account of all modes reflects the power of radiation of the resonant molecules. In the nonlinear region, and also upon the introduction of pumping and relaxation losses, the coupling of the individual modes n_k is not weakened, and their simultaneous account is quite necessary. This circumstance requires a critical consideration of the theoretical results obtained for the electromagnetic oscillations in a quantum generator with account of only a single mode, if the conditions of the experiment admit the existence of a large number of modes.

The solution of the usual balance equations^[2]

$$\dot{n} = W N_2 (1 + n) - W N_1 n, \quad N_2 = N_2^0 - n,$$

$$N_1 = n + N - N_2^0,$$

written down for our model problem with the initial condition $n(0) = 0$ and $N_2(0) = N_2^0$, contradicts (28). Consequently, the balance equation, which is essentially based on canonical perturbation theory, applicable to an individual molecule in an external electromagnetic field, cannot describe the collective character of the radiation processes of a system of resonant molecules.

If the overpopulation at the initial instant was negative and $N_2^0 W \tau \ll |N_-^0|$, $W \tau \ll 1$, then only an insignificant number of molecules, given by Eq. (29), subsequently luminesce, and the overwhelming majority continue to remain in the excited state. Naturally, the resulting equilibrium is

dynamic. If we now increase N_2^0 adiabatically, then the equilibrium density of photons in the volume is increased in correspondence with the change in N_2^0 . However, as only the negative overpopulation $N_-^0 = N_2^0 - N_1^0$ in such an adiabatic increase reaches the region $|N_-^0| \lesssim N_2^0 W \tau$, the problem becomes nonlinear from a certain time and Eq. (29) loses force.

We now determine the spectral composition of the radiation (28), (29). Inasmuch as the function $n(t)$ is aperiodic, the spectral decomposition of the radiation energy in the variable ω_k can easily be determined from (28) by averaging $n_k(t)$ over the time: $\bar{n}_k(t) \equiv n(\omega_k)$. The width of the spectral line in the variable ω_k is equal to $2/\tau$. However, upon emission of radiation from the volume, it is not ω_k that is conserved but the frequency ω of oscillation of the vector potential (or of the electric field intensity vector $\mathbf{E} = -\dot{\mathbf{A}}/c$). In order to determine the spectral composition of the radiation relation to the variable ω , it is necessary to establish the connection between ω_k and ω . For this purpose, we solve Eqs. (14) and (15) with the initial condition $n_k(0) = 0$ and $N_2(0) = N_2^0 \ll N$, which, upon account of (12), gives the following expression for the mean field:

$$A_s(\mathbf{k}, t) = (4\pi c^2 \hbar N_2^0 / \omega_0 \tau_0^2 \Omega_h^2)^{1/2} (\cos \omega_1 t - \cos \omega_2 t), \quad (31)$$

$$\omega_{1,2} = \omega_0 + (\omega_k - \omega_0 \pm [(\omega_k - \omega_0)^2 + \tau^{-2}]^{1/2}) / 2. \quad (32)$$

The vector $\mathbf{A}_s(\mathbf{k}, t)$ lies in a plane perpendicular to the vector \mathbf{k} , while the radiation (31) is unpolarized. The coordinate dependence of the vector potential is determined by the expansion (6).

Thus, to each oscillation in the number of photons n_k with a fixed wavelength (or ω_k), there correspond two oscillations of the electric field intensity vector with frequencies ω_1 and ω_2 , equal amplitudes but with phases differing by π . Superposition of these two oscillations of the vector field intensity lead to beats, which appear in the form of oscillations of the mean density of photons n_k/V , where n_k is given in (27). In the functional relation of ω_k with ω according to (32) there are two branches; therefore, in the spectral decomposition of the function $n(\omega_k) \equiv n_\omega$ in the variable ω , two symmetric maxima appear at the points $\omega = \omega_0 - 1/2\tau$ and $\omega = \omega_0 + 1/2\tau$. In order to determine the distribution of radiation energy over the variable ω , it is necessary to multiply n_ω by the density of energy states of a photon with given ω . As a result, the radiation energy $d\mathcal{E}_\omega$ in the volume V , occurring in the frequency range $d\omega$, is written in the form

$$d\mathcal{E}_\omega = \frac{N_2^0 W}{2\pi} \frac{\hbar \omega_0}{(\omega - \omega_0)^2 + (2\tau)^{-2}} d\omega.$$

Consequently, in the adiabatic emission of radiation from the volume a Lorentz shape will be observed for the spectral line with width $1/\tau$. Here, the intensity of radiation is a constant according to Eq. (29).

In the nonlinear region, the problem of the width of the spectral line is greatly complicated, since the electromagnetic oscillations are no more harmonic and each type of oscillations n_k , which is characterized by the parameter ω_k^2 , depends essentially on all the other oscillations with all possible $\omega_k^2 \neq \omega_k^2$. However, the nonlinear problem yields an approximate solution in the special case of a large positive initial overpopulation $N_-^0 \gg N_2^0 W \tau$, when the intense (cascade) radiation and the subsequent absorption of photons take place over a time interval

$$\tau \ll |t - mT| \leq T/2, \quad m = 0, 1, 2, \dots,$$

where T is the period of oscillation of $n(t)$.

The analysis of the start of the radiation (30) leads to the conclusion that only a limited number of modes n_k take part in the process of cascade radiation and absorption of photons. These can easily be determined from the asymptotic expansion of $n(t)$ (30) and $n_k(t)$ (27), which yield

$$n(t) = \gamma_{\text{eff}}(t) n_k(t), \quad (33)$$

$$\gamma_{\text{eff}}(t) = \begin{cases} N_-^0 W \tau (\tau/2\pi t)^{1/2} & \text{for } \tau \ll t \lesssim T/2 \\ N_-^0 W \tau (\tau/2\pi(T-t))^{1/2} & \text{for } \tau \ll T-t \lesssim T/2 \end{cases}, \quad (33')$$

where T is the period of the functions $n(t)$ and $n_k(t)$ from (33), and $j_{\text{eff}}(t)$ is a slowly changing function of time in comparison with $n_k(t)$. The function $j_{\text{eff}}(t)$ determines the effective number of modes n_k in the volume at each instant of time t :

$$\gamma_{\text{eff}}(t) = \sum_k 1 = \frac{V \omega_0^2 \Delta \omega_k}{(2\pi)^2 c^3}.$$

Here $\Delta \omega_k \equiv 2 |\omega_k^{\text{max}}(t) - \omega_0|$ is the effective width of the spectral interval in the variable ω_k . It is easy to prove (see [1]) that $(\omega_k^{\text{max}}(t) - \omega_0)^2 \tau^2 \ll 1$; therefore, one can set $\omega_k = \omega_0$ in the function for $n_k(t)$ (33) if necessary.

At a moment close to $t = T/2$, the number of modes $\gamma_{\text{eff}}(t)$ can differ insignificantly from (33'); however the exact knowledge of the value of $\gamma_{\text{eff}}(t)$ is not obligatory, since it enters under the logarithm in the determination of the width of the spectral line, and the maximum of $n(t)$ does not depend on γ_{eff} at all. Therefore, in what follows, we shall set t in the function for $\gamma_{\text{eff}}(t)$ (33) equal to $T/2$ and define

$$\gamma_{\text{eff}}(T/2) = \gamma_0. \quad (34)$$

After this, the set of equations (22) and (23), with account of (33), (34), reduces to a single equation:¹⁾

$$n_k + \frac{3\gamma_0}{2\tau_0^2} n_k^2 + \Omega_k^2 n_k = \frac{2N_2^0}{\tau_0^2} \quad (35)$$

with the initial condition $n_k(0) = \dot{n}_k(0) = 0$. If $N_-^{02} \gg \gamma_0 N_2^0$ and $N_-^0 > 0$, then one can use Eq. (35) over the entire time interval, because at the moment of time $|t - mT| \lesssim \tau$, $m = 0, 1, 2, \dots$, the number of photons is small and quadrature over n_k of the terms in (35) is insignificant, while for $\tau \ll |t - mT| \leq T/2$, $m = 0, 1, 2, \dots$, the values (33) and (34) are correct.

We shall write the solution of Eq. (35) in the form

$$n_k(t) = \frac{\beta_2 \kappa^2 \text{sn}^2(u, \kappa)}{\kappa^2 \text{sn}^2(u, \kappa) - 1} = \beta_1 \text{cn}^2(u + K, \kappa), \quad (36)$$

where

$$u = \frac{2Kt}{T}, \quad T = \frac{4\tau_0 K}{(\gamma_0(\beta_1 - \beta_2))^{1/2}}$$

$$K = F\left(\frac{\pi}{2}, \kappa\right), \quad \kappa^2 = \frac{\beta_1}{\beta_1 - \beta_2}.$$

Here $\text{sn}(u, \kappa)$ and $\text{cn}(u, \kappa)$ are the elliptic sine and cosine,^[6] T is the period of the function $n_k(t)$, K is the complete elliptic integral of the first kind and $\beta_1 > 0$ and $\beta_2 < 0$ are equal to

$$\beta_{1,2} = (-\tau_0^2 \Omega_k^2 \pm [\tau_0^4 \Omega_k^4 + 16\gamma_0 N_2^0]^{1/2}) / 2\gamma_0.$$

The maximum of the function $n_k(t)$ is equal to $n_k(T/2) = \beta_1$.

If $N_-^{02} \gg \gamma_0 N_2^0$ and $N_-^0 < 0$, then the solution of (36) is identical with (27) for any t (nonlinear effects are negligibly small).

If $N_-^{02} \gg \gamma_0 N_2^0$ and $N_-^0 > 0$, then in the region that is linear in n_k , at the beginning of the process, the solution (36) is also identical with (27), and the total number of photons is given by Eq. (28).

In the nonlinear region, $\tau \ll t < T$, the total number of photons, according to (33)–(36), is equal to $n(t) = \gamma_0 n_k(t)$, while the maximum value is achieved at $t = T/2$ and is equal to N_-^0 . Here T is period of the function $n(t)$, which is identical with the period of $n_k(t)$, and is equal to

$$T = 2\tau \ln(4N_-^{02} / \gamma_0 N_2^0).$$

In the linear region $0 < T - t \lesssim \tau$, close to the instant $t = T$, the solution (36) goes over into Eqs. (27) and (28) with replacement of t by $T - t$ in them. Thereafter, the process is again repeated. Consequently, there is an integral contri-

¹⁾ Equation (35) is identical with the similar equation from [1] if it is taken into account that $n_k = \sum_{\lambda} (n_{k\lambda} + n_{-k\lambda})$, where $n_{k\lambda}$ is the number of photons with a wave vector k and polarization $\pm 1_{k\lambda}$, and $4\gamma_0 = \gamma$.

bution of these regions, where (33) is inapplicable the mean value over a period of the number of photons \bar{n} in the volume V is negligibly small, so that one can set

$$\bar{n} = \frac{1}{T} \int_0^T n(t) dt = \frac{\gamma_0}{T} \int_0^T n_h(t) dt = \frac{2N_-^0}{\ln(4N_-^0/\gamma_0 N_2^0)}. \quad (37)$$

If we denote the quality factor of the resonator by Q , then in the adiabatic emission of radiation from the volume, the mean energy of radiation averaged over a single period dE/dt emanating from the volume per unit time is equal to

$$-\frac{d\mathcal{E}}{dt} = \frac{\omega_0}{Q} \mathcal{E} = \frac{2\hbar\omega_0^2 N_-^0}{Q \ln(4N_-^0/\gamma_0 N_2^0)}. \quad (38)$$

In the determination of the spectral composition of the radiation, (37) and (38), we note that in accord with (12) the frequency ω_m on which the Fourier component of the slowly changing function $X_{Sk}(t)$ depends, is connected with the frequency ω of the Fourier component of the vector potential $A_S(\mathbf{k}, t)$ by the relation $\omega - \omega_0 = \omega_m$. Therefore the Fourier decomposition of the slowly changing functions $X_{Sk}(t)$ and $n_k(t)$ determines the scatter of the frequencies of the vector potential near the fundamental frequency ω_0 . Expanding $\text{cn}(u + K, \kappa)$ in a Fourier series

$$\text{cn}(u + K, \kappa) = -\frac{i\pi}{kK} \sum_{m=-\infty}^{\infty} \frac{(-1)^m q^{m+1/2}}{1 + q^{2m+1}} e^{-i(2m+1)\pi t/T}$$

and averaging (36) over the time we get in place of (37)

$$\bar{n} = \gamma_0 \bar{n}_h = \frac{\pi^2 \gamma_0 \beta_1}{\kappa^2 K^2} \sum_{m=-\infty}^{\infty} \frac{q^{2m+1}}{(1 + q^{2m+1})^2}, \quad (39)$$

where

$$q = \exp\left(-\frac{\pi K'}{K}\right), \quad K' = F\left(\frac{\pi}{2}, \kappa'\right), \quad \kappa'^2 = 1 - \kappa^2.$$

The component of the series (39) with index m is proportional to the average over the period of the energy of radiation frequency $\omega = \omega_0 + (2m + 1)\pi/T$, where T is the period of the function $n_k(t)$. It is not difficult to determine in (39) the number m of the component which is half the value of the fundamental term with $m = 0$. This gives the following expression for the width $\Delta\omega = 2|\omega - \omega_0|$ of the spectral line:

$$\Delta\omega = \ln 6 / \pi\tau, \quad (40)$$

which is valid for $\ln(4N_-^0/\gamma_0 N_2^0) \gg 1$.

Strictly speaking, the frequency region $\Delta\omega$ (40) contains a large number of discrete lines with frequencies $\omega = \omega_0 + (2m + 1)\pi/T$, where $m = 0, \pm 1, \pm 2, \dots$, and only apparatus with insufficient

resolving power will show a single spectral line of width given by Eq. (40).

As has been pointed out earlier,^[1] the solution of the present model problem can under certain conditions be compared with experimental data on the radiation of a quantum generator in the peak regime. Here the width of the spectral line (40) is in excellent agreement with the experimental results.^[7]

In the general case, pumping and relaxation processes can be taken into account by introducing the corresponding phenomenological terms in Eqs. (7)–(9) and assuming here that A_S , j_S , and N_- are classical (or mean quantum-mechanical) quantities. Here we note that account of the radiation damping of the excited state of the individual molecules in the present model problem leads to the appearance of an additional component Wj_S on the left hand side of Eq. (8) (see^[4]). Considering the radiation damping as one of the possible relaxation processes which leads to a broadening of the excited level of the molecule, we make the generalization that the other relaxation processes, which broaden the excited level of the molecule, can also be taken into account by replacing the component Wj_S by the quantity $(1/T_2)j_S$ where T_2 is the characteristic time of the relaxation processes leading to the broadening of the excited level of the molecule by an amount \hbar/T_2 . Then in place of (8), we get

$$\begin{aligned} \ddot{j}_s(\mathbf{k}, t) + \frac{1}{T_2} \dot{j}_s(\mathbf{k}, t) + \omega_0^2 j_s(\mathbf{k}, t) \\ = -\frac{\omega_0^2}{4\pi c\tau_0^2} N_- A_s(\mathbf{k}, t), \end{aligned} \quad (41)$$

where $A_S(\mathbf{k}, t)$, $s = 1, 2$ are the Fourier components of the vector potential

$$A_s(\mathbf{x}, t) = \sqrt{\frac{2}{V}} \sum_{\mathbf{k}} (A_1(\mathbf{k}, t) \cos \mathbf{kx} + A_2(\mathbf{k}, t) \sin \mathbf{kx}),$$

and $j_s(\mathbf{k}, t)$ is the Fourier component of the polarization current.

The second component of the left side of (41) characterizes the relaxation losses which are due both to spontaneous emission and to nonradiative transitions of molecules from an excited level. By the introduction of T_2 , as is evident, one can in the same way take account of small deviations of the energy levels of the two-level molecules from exact resonance. In the latter case, the value of \hbar/T_2 has the physical meaning of the width of the scattering of levels of the molecules near the resonance value. In both cases, the parameter $1/T_2$ coincides numerically with the value of the spectral line of the fluorescent molecules.

The losses of electromagnetic energy are easily

taken into account phenomenologically in (7) by introducing the "effective conductivity" of the medium, which leads (according to the Maxwell equations) to the following expression for the vector potential:

$$\ddot{\mathbf{A}}_s(\mathbf{k}, t) + \frac{1}{T_1} \dot{\mathbf{A}}_s(\mathbf{k}, t) + \omega_k^2 \mathbf{A}_s(\mathbf{k}, t) = 4\pi c \mathbf{j}_s(\mathbf{k}, t), \quad (42)$$

where T_1 is a phenomenological constant determined by the experiment. Usually it is expressed in terms of the quality factor of the generator $T_1 = Q/\omega_k$.

We take the pumping into account by the usual method of balance relations, introducing another experimental constant W_p , which characterizes the power of the pumping, namely:

$$\begin{aligned} \dot{N}_- = & W_p(N - N_-) - W_{nr}(N + N_-) \\ & + \frac{2}{\hbar\omega_0 c} \sum_{\mathbf{sk}} \mathbf{A}_s(\mathbf{k}, t) \cdot \mathbf{j}(\mathbf{k}, t), \end{aligned} \quad (43)$$

where W_{nr} is the probability of nonradiative transition of the molecule from the second (upper) to the first (lower) energy level, which, in the absence of a spread of levels, is equal to $W_{nr} = 1/T_2 - W$. The first component of the right hand side of (43) describes the transition as a result of pumping, the second, due to relaxation losses, which are brought about by the nonradiative transitions of the molecules. The relaxation losses by spontaneous radiation are retained in the last component on the right hand side (43). The same component takes into account the change in the overpopulation as a result of coherent radiation (absorption) of quanta, and also contains the losses of coherent quanta as the result of absorption in the walls and escape to the outside. Actually, transition to the slowly changing functions by the method of (12)–(21) transforms equation (43), for a double mirror cavity with closed lateral walls, to the following form:

$$\begin{aligned} \frac{d}{dt} (N_-/2 + n) = & W_p N_1 - (W_{nr} + W) N_2 - n/T_1; \\ \omega_0 T_1 \gg 1, \quad \omega_0 T_2 \gg 1, \quad \omega_0 \tau_0 \gg \sqrt{N}. \end{aligned} \quad (44)$$

The relation (44), being multiplied by $\hbar\omega_0$, represents the equation of energy balance, in which $\hbar\omega (N_-/2 + n)$ is the total energy of the system. Both (43) and (44) are valid for the case in which the probability W_p of transition per unit time of a molecule from the first energy level to the third (auxiliary) as a result of the irradiation is much less than the probability of nonradiative transition from the third level to the second. In the opposite case, it is necessary to consider the system with three-level quantum objects.

If the resonant molecules are located in a medium whose dielectric constant ϵ (without resonant molecules) differs significantly from

unity, then Eqs. (41) and (42) are somewhat changed. Inasmuch as in the given problem there exists a narrow range of frequencies close to the fundamental frequency ω_0 , then the dielectric constant ϵ of the medium (without resonant molecules) can be considered constant in this band of frequencies. In such a case, the right side of Eq. (41) is multiplied by ϵ and the right side of (42) is divided by ϵ . Here, $\omega_k^2 = k^2 c^2 / \epsilon$ and the parameter τ_0^{-2} will have the form

$$\tau_0^{-2} = 2\pi c^3 W / \omega_0^2 V \epsilon^{3/2},$$

where W is the probability per unit time of spontaneous emission of a quantum of frequency ω_0 for a solitary molecule in a medium with dielectric permittivity ϵ . The physical meaning of T_1 , T_2 , W_p , and W_{nr} remains the same as before.

The sets of equations (41)–(43) are similar to analogous equations suggested in a number of researches,^[8–11] but the latter do not coincide with the present set completely. Equations (41)–(43) describe electromagnetic oscillations in a resonant medium with account of pumping and losses. These equations bear a postulative character. The basis of the introduction of the phenomenological terms lies in the comparison of the results of the solution with experimental data.

The author is grateful to V. M. Galitskiĭ for repeated discussions on a number of questions related to the present research.

¹ Alekseev, Vdovin and Galitskiĭ, JETP 46, 320 (1964) Soviet Phys. JETP 19, 220 (1964).

² N. Bloembergen, Phys. Rev. 104, 324 (1956).

³ L. D. Landau and E. M. Lifshitz, Teoriya polya (Theory of Fields) Fizmatgiz, 1962, p. 29.

⁴ A. I. Alekseev and V. M. Galitskiĭ, JETP 47, 1893 (1964), Soviet Phys. JETP 20.

⁵ W. Heitler, Quantum Theory of Radiation, (Russian Translation), IIL 1956, p. 162. [Oxford, 1954].

⁶ E. T. Whittaker and G. N. Watson, A Course of Modern Analysis, (Russian Translation), Fizmatgiz, 1963, p. 375 [Cambridge, 1940].

⁷ Maiman, Hoskins, D'Haensens, Asawa, and Evtuhov, Phys. Rev. 123, 1151 (1961).

⁸ V. M. Faĭn, JETP 33, 945 (1957), Soviet Phys. JETP 6, 726 (1958).

⁹ A. N. Oraevskiĭ, Radiotekhnika i elektronika 4, 718 (1959): Trudy, (Physics Institute, Acad. Sci. USSR) 21, 3 (1963).

¹⁰ L. W. Davis, Proc. IEEE 51, 112 (1963).

¹¹ V. V. Korobkin and A. V. Uspenskiĭ, JETP 45, 1003 (1963), Soviet Phys. JETP 18, 693 (1963).