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# *PROPAGATION OF PHOTONS IN A MEDIUM OF RESONANT MOLECULES*

Yu. A. VDOVIN and V. M. GALITSKIĬ

Moscow Engineering Physics Institute

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We investigate the lower excited states of a system consisting of strictly resonant two-level molecules and an electromagnetic field. We introduce the operators of elementary excitations —quanta in the medium. We investigate the damping of the excitations, and develop a suitable diagram technique. We determine the Green functions of the photon in such a medium. We investigate the propagation of photons in such a system of molecules, under the condition that either a specified photon or a quantum of the medium is present at the initial instant of time. We take into account both the ''departure'' and the ''arrival'' of photons in a given state. The energy distribution function of the photons is determined.

## 1. INTRODUCTION

WE consider in this paper the interaction between an electromagnetic field and an aggregate of identical resonant molecules. This problem is of considerable interest in connection with the extensive research being carried out presently on resonant processes in interactions between photons and a medium. It is assumed that the resonance is sufficiently narrow and only the molecular levels are of importance for the interaction in the given frequency region. The energy difference of these levels (the transition frequency $\omega_0$) is assumed to be same for all molecules. We start from the assumption that the molecules have a random distribution in space, something which is true of a gaseous medium or of impurities in a solid. We disregard the broadening of the lines due to the motion of molecules, the Stark effect, etc.

We investigate in this paper weakly excited states of the system, so that the interaction between the quanta themselves can be neglected. In the case of a medium of low density these excitations are divided into individual excitations of the molecules and photons. The damping of the former is determined by the radiation of the isolated molecules, and that of the latter by the processes of resonant scattering of the quanta from the molecules[1]. With increasing density of the medium, such a separation becomes meaningless. This is seen, for example, from the fact that the mean free path of the quantum becomes smaller than its wavelength. However, it becomes necessary even sooner to take into account the coherent interaction of the photons with the medium, corresponding to the presence of a dielectric constant. In this case there are no individual molecular excitations or photons, and their place is taken by the quanta of the electromagnetic field of a medium having a resonant dielectric constant. We find the damping of these states.

We also investigate the kinetics of the process. We take into account both ''departure'' of the photons from a given state and their ''arrival.'' The energy distributions of the photons and of the quanta of the medium are found.

The oscillation of the photon density in such a resonant system was investigated by Alekseev and the authors[2]. A somewhat different approach was developed by Kazantsev and Smirnov[3] and by James and Cummings[4]. However, no account of relaxation processes was taken in these papers, and the structure of the excited states was not investigated. In an earlier paper[5], the influence of the resonant nature was taken into account within the framework of the dielectric constant of the medium.

## 2. ELEMENTARY EXCITATIONS WITHOUT ACCOUNT OF DAMPING

When considering the behavior of quanta in a medium, it is useful to introduce the concept of ''quanta of a medium'' which take into account the dielectric constant. Without taking damping into consideration, these quanta can be introduced by means of a canonical transformation. The Hamiltonian H of a system of N molecules and a radiation field, contained in a volume V, is of the form

$$H = \frac{\omega_0}{2}\sum_j \sigma_z{}^j + \sum_{\mathbf{k}\lambda}(c_{\mathbf{k}\lambda}b^+_{\mathbf{k}\lambda} + c^+_{\mathbf{k}\lambda}b_{\mathbf{k}\lambda}) + \sum_{\mathbf{k}\lambda}\omega_k c^+_{\mathbf{k}\lambda}c_{\mathbf{k}\lambda}, \quad (1)$$

$$b_{\mathbf{k}\lambda}=\sum_j \sigma_-{}^j\left(\frac{2\pi}{\omega_k V}\right)^{1/2}(\mathbf{M}^{j*}\mathbf{l}_{\mathbf{k}}{}^{\lambda})\,e^{-i\mathbf{k}\mathbf{x}_j}=\sum_j \sigma_-{}^j Q_{k\lambda}{}^j e^{-i\mathbf{k}\mathbf{x}_j}. \qquad (2)$$

We use a system of units in which $\hbar = 1$. Here $\mathbf{x}_j$ is a coordinate of the center of gravity of the j-th molecule, $\omega_0$ the energy differs between the upper and lower levels of the molecules, $c_{k\lambda}$ and $\overset{+}{c}_{k\lambda}$ are the operators of the absorption and emission of a photon with momentum **k** and polarization $\mathbf{l}_k^{\lambda}$ respectively ($\lambda = 1, 2$), $\omega_k^2 = c^2k^2$, $\sigma_z$ and $\sigma_\pm$ are Pauli matrices, and $\mathbf{M}^j$ is the matrix element of the transition of the j-th isolated molecule from the lower level to the upper level with simultaneous absorption of a photon. The matrix element $\mathbf{M}^j$ and the probability W of spontaneous dipole radiation of a photon per unit time by an isolated molecule are connected by the relation

$$W=\frac{4}{3}\frac{\omega_0}{c^3}|\mathbf{M}^j|^2=\frac{2}{3}|Q_k|^2\frac{\omega_0{}^2V}{\pi c^3}. \qquad (3)$$

As before[2], we have left out from the Hamiltonian the terms proportional to the square of the electromagnetic potential and the terms which give a small contribution of the order of $(\omega_0\tau)^{-2}$, where

$$\frac{1}{4\tau^2}=\frac{|Q_k|^2N}{3}=\frac{2\pi|\mathbf{M}^j|^2N}{3V\omega_0}=\frac{\pi}{2}\rho\bar{\lambda}^3W\omega_0 \qquad (4)$$

(the factor 1/3 is due to the averaging over the directions of the vector $\mathbf{M}^j$), $\rho$ is the density of the molecules, and $\bar{\lambda} = \omega_0^{-1}c$. Terms of the order $(\omega_0\tau)^{-1}$ relative to the principal terms will henceforth be disregarded.

In the operators $b_{k\lambda}$ and $\overset{+}{b}_{k\lambda}$ only the commutator

$$[b_{\mathbf{k}\lambda},\, b^{+}_{\mathbf{k}'\lambda'}]=-\sum_j \sigma_z{}^j Q^j_{\mathbf{k}\lambda}Q^{j*}_{\mathbf{k}'\lambda'}e^{i(\mathbf{k}'-\mathbf{k})\mathbf{x}_j} \qquad (5)$$

differs from 0. If almost all the molecules are at the lower energy level, then we can make on the right side of the sum the substitution $\sigma_z^j \to -1$, so that the commutator of the operators $b_{k\lambda}$ and $\overset{+}{b}_{k'\lambda'}$ will be a number. By virtue of the phase relations, this number has a maximum when $\mathbf{k}', \lambda' = \mathbf{k}, \lambda$. The difference between the commutator and zero when $\mathbf{k}', \lambda' \neq \mathbf{k}, \lambda$ is connected with the scattering of the quanta and will be taken into account below.

In this approximation the operators $b_{k\lambda}$ and $\overset{+}{b}_{\lambda k}$ can be regarded as Bose operators with commutation relations (see [2]) [1)]

$$[b_{\mathbf{k}\lambda},\, \overset{+}{b}_{\mathbf{k}'\lambda'}]=\frac{1}{4}\tau^{-2}\delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'}. \qquad (6)$$

The corresponding Hamiltonian H can be readily diagonalized with the aid of a canonical transformation. We introduce in lieu of the operators $b_{k\lambda}$ and $c_{k\lambda}$ new Bose operators $\alpha_{k\lambda}^{(1)}$ and $\alpha_{k\lambda}^{(2)}$

$$\alpha_{\mathbf{k}\lambda}{}^{(r)}=u_k{}^{(r)}b_{\mathbf{k}\lambda}+v_k{}^{(r)}c_{\mathbf{k}\lambda},\quad r=1,2. \qquad (7)$$

Choosing the functions $u_k^{(r)}$ and $v_k^{(r)}$ in suitable fashion, we obtain

$$H=-\frac{\omega_0}{2}N+\sum_{\mathbf{k}\lambda}\omega_1\alpha_{\mathbf{k}\lambda}^{(1)+}\alpha_{\mathbf{k}\lambda}^{(1)}+\sum_{\mathbf{k}\lambda}\omega_2\alpha_{\mathbf{k}\lambda}^{(2)+}\alpha_{\mathbf{k}\lambda}^{(2)}; \qquad (8)$$

$$\omega_1=\omega_0+{}^1/_2\Delta+{}^1/_2\Omega,\quad \omega_2=\omega_0+{}^1/_2\Delta-{}^1/_2\Omega,$$

$$\Delta=\omega_k-\omega_0,\quad \Omega=(\Delta^2+\tau^{-2})^{1/2}. \qquad (9)$$

Here

$$u_k{}^{(1)}=-2\tau v_k{}^{(2)}=2^{1/2}\tau(1-\Delta/\Omega)^{1/2},$$

$$u_k{}^{(2)}=2\tau v_k{}^{(1)}=2^{1/2}\tau(1+\Delta/\Omega)^{1/2}. \qquad (10)$$

Relations (7)—(10) determine the weakly-excited states in the system of resonant molecules. The ground state $|0\rangle$ of the system is characterized by the fact that all the molecules are in the lower energy state and there are no photons. We note that this state is also the exact ground state of the Hamiltonian (1). The energy of this state is equal to $-\omega_0N/2$. The state $\alpha_{k\lambda}^{(r)+}|0\rangle$ is characterized by the presence in the system of one elementary excitation with energy $\omega_r$. These excitations constitute quanta of the electromagnetic field in a medium consisting of resonant molecules. The frequencies $\omega_1$ and $\omega_2$ are found by solving the dispersion equation $\omega^2\epsilon = \omega_k^2$ for this case. Far from resonance, when $\omega_k \gg \omega_0$, the energy $\omega_1$ goes over into the energy $\omega_k$ of the free photon, while the energy $\omega_2$ goes over into the energy $\omega_0$ of the isolated excited molecule. When $\omega_k \ll \omega_0$, to the contrary, $\omega_1$ goes over into $\omega_0$ and $\omega_2$ into $\omega_k$.

We shall show later that the concept of quanta in a medium is useful only in the case of sufficiently high density. In the case of low density it is more advantageous to use the concept of a quantum in vacuum (the state $\overset{+}{c}_{k\lambda}|0\rangle$).

## 3. DAMPING OF EXCITATIONS. PHOTON PROPAGATION FUNCTION

Let us examine the behavior of one quantum in a medium of resonant molecules. The solution will actually be valid also in the case of a large

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[1)] A similar situation occurs in the investigation of weakly excited states – spin waves in ferromagnets (cf., for example, [6]).

number of quanta, provided, however, that their density is sufficiently low to neglect their interaction with one another.

We assume that in the initial state the medium contains either one quantum with momentum $\mathbf{k}_0$ and polarization $\lambda_0$, that is, the state is described by a wave function $\overset{+}{c}_{\mathbf{k}_0\lambda_0}|0\rangle$, or else one quantum of the medium, that is, the state $\alpha^{(r)}_{\mathbf{k}_0\lambda_0}|0\rangle$. The Hamiltonian (1) conserves the number of excitations, and therefore the state will remain single-particle at any instant of time. The wave function of an arbitrary single-particle state can be written in the form

$$\psi(t)=\sum_{\mathbf{k}\lambda} f_\lambda(\mathbf{k},t)\,c_{\mathbf{k}\lambda}{}^{+}|0\rangle\, e^{iN\omega_0 t/2}+\sum_j \varphi_j(t)\,\sigma_+{}^{j}|0\rangle\, e^{iN\omega_0 t/2}. \tag{11}$$

From the Schrödinger equation we obtain the following expressions for the amplitudes $f_\lambda(\mathbf{k}, t)$ and $\varphi_j(t)$:

$$i\dot{f}_\lambda(\mathbf{k},t)=\omega_k f_\lambda(\mathbf{k},t)+\sum_j Q^j_{\mathbf{k}\lambda}e^{-i\mathbf{k}\mathbf{x}_j}\varphi_j(t),$$

$$i\dot{\varphi}_j(t)=\omega_0\varphi_j(t)+\sum_{\mathbf{k}'\lambda'} Q^{j*}_{\mathbf{k}'\lambda'}e^{i\mathbf{k}\mathbf{x}_j}f_{\lambda'}(\mathbf{k}',t). \tag{12}$$

It is necessary to solve these equations with initial conditions

$$f_\lambda(\mathbf{k},0)=\delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0},\qquad \varphi_j(0)=0. \tag{13}$$

It is convenient to go over from the time-dependent amplitudes $f_\lambda(\mathbf{k}, t)$ and $\varphi_j(t)$ to their Laplace transforms $f_\lambda(\mathbf{k}, \omega)$ and $\varphi_j(\omega)$, for example

$$f_\lambda(\mathbf{k},\omega)=\int_0^\infty e^{-pt}f_\lambda(\mathbf{k},t)\,dt,$$

where we put $ip = \omega$. Eliminating from the resultant system of equations the function $\varphi_j(\omega)$, we have equations for the amplitudes $f_\lambda(\mathbf{k}, \omega)$

$$(\omega-\omega_k)f_\lambda(\mathbf{k},\omega)-\frac{1}{(\omega-\omega_0)}\sum J_{\lambda\lambda'}(\mathbf{k},\mathbf{k}')f_{\lambda'}(\mathbf{k}',\omega)$$

$$=i\delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0}, \tag{14}$$

$$J_{\lambda\lambda'}(\mathbf{k},\mathbf{k}')=\sum_j Q_{\mathbf{k}\lambda}{}^{j}Q^{j*}_{\mathbf{k}'\lambda'}\,e^{i(\mathbf{k}'-\mathbf{k})x_j}. \tag{15}$$

We see that the amplitude $f_\lambda(\mathbf{k}, \omega)$ is proportional to the Green's function of (14) or to the retarded propagation function of the photon. The interaction of the molecules with the electromagnetic field is described by the operator J. In the zeroth approximation in the interaction we have

$$f_\lambda{}^{(0)}(\mathbf{k},\omega)=iG_0(\omega_k,\omega)\,\delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0}=i\delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0}/(\omega-\omega_k). \tag{16}$$

We are interested in the amplitude $f_\lambda(\mathbf{k}, \omega)$ averaged over the coordinates of the centers of gravity of all the molecules. Denoting this averaging by angle brackets, we write

$$\langle f_\lambda(\mathbf{k},\omega)\rangle=iG_\lambda(\mathbf{k},\omega). \tag{17}$$

Carrying out the iterations, we obtain the following series

$$G_\lambda(\mathbf{k},\omega)=G_0+G_0\left\langle\frac{\hat{J}}{\omega_0-\omega_0}\right\rangle G_0$$

$$+\,G_0\left\langle\frac{\hat{J}}{\omega-\omega_0}G_0\frac{\hat{J}}{\omega-\omega_0}\right\rangle G_0+\ldots \tag{18}$$

When averaging over the coordinates $\mathbf{x}_j$ it becomes possible to use the well developed methods of the diagram technique. (In the simplest cases this technique was initially developed in the papers of Abrikosov and Gor'kov or of Edwards[7].) We set in correspondence with each term of the series (19) a diagram in which the Green's function G corresponds to a solid line and the operator $\hat{J}$ to a point. In each term of the series (18) we average the product of the operators $\hat{J}$. Let us examine the averaging method, using as an example a third-order term containing three operators $\hat{J}$. This expression contains terms in which all three indices j are different. The average of such terms breaks up into a product of averages. There are in addition terms with a pair of coinciding indices j. Their average breaks up into a product of averages of one and of two operators $\hat{J}$. Finally, there are terms in which all three indices j coincide. Thus, the average breaks up into a sum of different averages. The equating of the indices j of different operators $\hat{J}$ with subsequent averaging will be denoted on the diagrams by dashed lines joining the corresponding nodes. The averaging of exponential functions contained in $\hat{J}$ gives rise to a $\delta$-symbol, which denotes the momentum conservation law: the sum of all the momenta of the lines entering the nodes joined by a dashed line is equal to the sum of the momenta of the lines leaving them. To each node there corresponds a factor

$$Q_{\mathbf{k}_1\lambda_1}Q^{j*}_{\mathbf{k}_2\lambda_2}/(\omega-\omega_0).$$

Thus, for example, the term corresponding to the diagram of Fig. 1 is

$$\frac{1}{(\omega-\omega_0)^3(\omega-\omega_{k_0})^3}\sum_{j_1}|Q^{j_1}_{\mathbf{k}_0\lambda_0}|^2\sum_j|Q^j_{\mathbf{k}_0\lambda_0}|^2\sum_{\mathbf{k}_1\lambda_1}\frac{|Q^j_{\mathbf{k}_1\lambda_1}|^2}{(\omega-\omega_{k_1})}.$$

After integrating over the vector $\mathbf{k}_1$ and subsequently averaging over the directions of the vectors

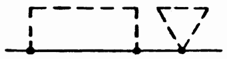

FIG. 1

$\mathbf{M}^{j}$ and $\mathbf{M}^{j_1}$ we obtain

$$\frac{1}{(\omega-\omega_0)^3(\omega-\omega_{k_0})^3}\left(\frac{1}{4\tau^2}\right)^2\left(\frac{-iW}{2}\right).$$

We have left out the real part of the integral with respect to $d\omega_k$, since its role reduces only to a small radiative frequency shift $\omega_0$. In addition, we took into account the fact that averaging over the directions of the vector $\mathbf{M}^{j}$ gives rise to a $\delta$-symbol expressing the conservation of the polarization of the quantum.

We introduced the concept of a generalized node, defining it by means of the sum of the diagrams of Fig. 2. Denoting the generalized node by I, we get

$$I = Q^{j}_{\mathbf{k}_1\lambda_1}Q^{j*}_{\mathbf{k}_2\lambda_2}e^{-i(\mathbf{k}_1-\mathbf{k}_2)\mathbf{x}_j}\sum_{n=1}^{\infty}\frac{1}{(\omega-\omega_0)^n}\left(\sum_{\mathbf{k}\lambda}\frac{|Q^{j}_{\mathbf{k}\lambda}|^2}{\omega-\omega_k}\right)^n$$
$$= \frac{Q^{j}_{\mathbf{k}_1\lambda_1}Q^{j*}_{\mathbf{k}_2\lambda_2}}{(\omega-\omega_0+iW/2)}e^{-i(\mathbf{k}_1-\mathbf{k}_2)\mathbf{x}_j}. \tag{19}$$

We note that the generalized node (19) differs from $\hat{J}/(\omega-\omega_0)$ in that the denominator contains an additional $iW/2$, that is, account is taken of the finite lifetime in the excited state of the molecule.

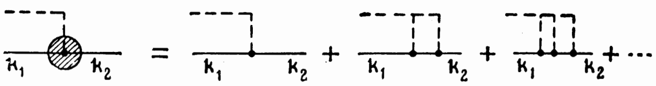

FIG. 2

Diagrams with generalized nodes are constructed in accordance with the same principle as non-generalized nodes, with the exception of the case of the simplest self-energy loop, in which only one of the nodes should be replaced by a generalized one.

The Green's function G can be expressed in the usual manner in terms of the self-energy part $\Sigma$, where $\Sigma$ is the sum of all the irreducible diagrams. In first order, $\Sigma$ is determined by the diagram of Fig. 3:

$$\Sigma^{(1)} = \frac{1}{\omega-\omega_0}\sum_{j}|Q^{j}_{\mathbf{k}_0\lambda_0}|^2 = \frac{1}{4\tau^2}\frac{1}{\omega-\omega_0}. \tag{20}$$

A definite sequence of diagrams of the next-higher orders can be expressed in terms of the generalized node, so that $\Sigma^{(2)}$ is represented by the diagram of Fig. 4. The contribution from the remaining diagrams can be neglected, accurate to terms of order $(\omega_0\tau)^{-1}$

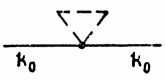

FIG. 3

$$\Sigma^{(2)} = \frac{1}{4\tau^2}\left(\frac{-iW}{2}\right)\frac{1}{(\omega-\omega_0+iW/2)(\omega-\omega_0)}. \tag{21}$$

The Green's function (18) has accordingly the form

$$G_\lambda(\mathbf{k},\omega) = G(\omega_k,\omega)\delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0}$$
$$= \frac{\delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0}}{\omega-\omega_k-[4\tau^2(\omega-\omega_0+iW/2)]^{-1}}. \tag{22}$$

The Green's function (22) has two poles in $\omega$. The pole values are respectively equal to [see Eq. (9) for the notation]

$$\omega = \omega_0 + {}^1\!/_2\Delta - {}^1\!/_4 iW \pm {}^1\!/_2(\Delta^2+\tau^{-2}+iW\Delta-{}^1\!/_4W^2)^{1/2}. \tag{23}$$

The expressions for the pole values of $\omega$ and for the respective Green's function (22) simplify greatly if the inequalities $W\tau \gg 1$ and $W\tau \ll 1$ are satisfied. Actually, in the optical region, the inequality $W\tau \ll 1$ is satisfied as a rule. However, the opposite limiting case (very highly rarefied medium) is also of certain interest. The results obtained then go over into the well known formulas of resonant fluorescence for a single molecule[1].

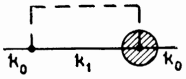

FIG. 4

We consider first the case $W\tau \gg 1$. The corresponding pole values of the frequencies are

$$\omega_1 = \omega_k - {}^1\!/_2 i\Gamma, \qquad \omega_2 = \omega_0 - {}^1\!/_2 iW + {}^1\!/_2 i\Gamma; \tag{24}$$

$$\Gamma = \frac{1}{4\tau^2}\frac{W}{\Delta^2+W^2/4}. \tag{25}$$

We see that the poles corresponding to the photon and the molecule have separated, and the admixture of the molecular state is negligible, so that the Green's function is of the form

$$G(\omega_k,\omega) = 1/(\omega-\omega_k+i\Gamma/2). \tag{26}$$

This case corresponds to a small coherent interaction between the electromagnetic field and the molecules, giving rise to the dielectric constant of the medium. The main process is resonant scattering, so that $\Gamma = \rho\sigma_{\mathrm{res}}c$ (see [1]). The mean free path $l$ is in this case of the order of $c/\Gamma \sim (\rho ƛ^2)^{-1}$. Extension of this formula to include a system with arbitrary density, that is, into the region $W\tau \ll 1$, leads to the absurd result $l \ll ƛ$.

We now consider the case $W\tau \ll 1$. The Green's function is of the form

$$G(\omega_k,\omega) = \frac{\omega-\omega_0+iW/2}{(\omega-\omega_1+iW_1/2)(\omega-\omega_2+iW_2/2)}, \tag{27}$$

where $\omega_{1,2}$ are given by expressions (9) and

$$W_1 = {}^1/_2 W(1 - \Delta / \Omega), \qquad W_2 = {}^1/_2 W(1 + \Delta / \Omega). \quad (28)$$

In this case the Green's function describes quanta of the electromagnetic field in the medium (quanta of the medium), that is, unlike the preceeding case, the coherent part of the interaction, which is of the order of $1/\tau$ (the real part of $\Sigma$) is appreciably larger than the incoherent part ($\mathrm{Im}\ \Sigma$), which is of the order of W. The quanta of the medium contain an admixture of the state of the molecule at the upper level, leading to damping with probabilities $W_1$ and $W_2$. The factors $(1 \mp \Delta/\Omega)/2$ correspond to the relative time that the quantum stays in the molecular state.

The Green's function (27) can be represented by a superposition of Green's functions corresponding to the quanta of the first and second kind in the medium. Indeed, using (7) and (10), we can find directly the dampings of the elementary excitations in the medium, that is, find the retarded propagation function of a quantum of the medium. For single-particle states, the commutator of the operators $b_{\mathbf{k}\lambda}$ and $b^{+}_{\mathbf{k}'\lambda'}$ is strictly a number. Therefore the initial state of the type $\alpha^{(r)+}_{\mathbf{k}\lambda} |0\rangle$ at any instant of time can be rigorously expressed in the form

$$\psi(t) = \sum_{r=1}^{2} \sum_{\mathbf{k}\lambda} \eta_\lambda{}^{(r)}(\mathbf{k}, t)\, \alpha_{\mathbf{k}\lambda}{}^{(r)+} |0\rangle\, e^{iN\omega_0 t/2}. \quad (29)$$

According to (7) and (11), the amplitudes $\eta^{(r)}_{\lambda}(\mathbf{k}, t)$ and $f_\lambda(\mathbf{k}, t)$ are connected by the relation

$$f_\lambda(\mathbf{k}, t) = \sum_{r=1}^{2} \eta_\lambda{}^{(r)}(\mathbf{k}, t)\, v_k{}^{(r)}. \quad (30)$$

From Schrödinger's equation, using expression (1) for the Hamiltonian H, we obtain equations for the amplitudes $\eta^{(r)}_{\lambda}(\mathbf{k}, t)$. For the Laplace transforms $\eta^{(r)}_{\lambda}(\mathbf{k}, \omega)$, these equations take the form

$$(\omega - \omega_r)\, \eta_\lambda{}^{(r)}(\mathbf{k}, \omega) = i\eta_\lambda{}^{(r)}(\mathbf{k}, t = 0)$$

$$+ 2\,|v_k{}^{(r)}| \sum_{\substack{\mathbf{k}\neq\mathbf{k}' \\ \lambda\neq\lambda'}} J_{\lambda\lambda'}(\mathbf{k}, \mathbf{k}')\, \frac{\tau\Omega'}{\omega - \omega'_{(3-r)}}\, |v_{k'}{}^{(3-r)}|\, \eta_{\lambda'}{}^{(r)}(\mathbf{k}', \omega), \quad (31)$$

r = 1, 2; the primes denote quantities which depend on $\mathbf{k}'$. This is an approximate form of the system of equations. We have left out small correction terms of the order of $W\tau \ll 1$. Accurate to such small terms, we have independent equations for the amplitudes $\eta^{(1)}_{\lambda}(\mathbf{k}, \omega)$ and $\eta^{(2)}_{\lambda}(\mathbf{k}, \omega)$. Consequently, in this approximation there are no transitions between different sorts of quanta of the medium. The latter circumstance is connected with the fact that the energy difference of these states are equal to $\omega_1 - \omega_2 \sim 1/\tau$, whereas the relaxation widths are of the order of W.

We solve Eqs. (31) subject to the initial conditions

$$\eta_\lambda{}^{(1)}(\mathbf{k}, t = 0) = \delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0}, \quad \eta_\lambda{}^{(2)}(\mathbf{k}, t = 0) = \delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0}. \quad (32)$$

Let us find the amplitudes $\eta^{(r)}_{\lambda}(\mathbf{k}, \omega)$, averaged over the coordinates of the centers of gravity of the molecules of the medium. In analogy with (17), we put

$$\langle \eta_\lambda{}^{(r)}(\mathbf{k}, \omega) \rangle = iG_\lambda{}^{(r)}(\mathbf{k}, \omega). \quad (33)$$

Using the diagram technique we developed, we readily obtain (r = 1, 2)

$$G_\lambda{}^{(r)}(\mathbf{k}, \omega) = G_r(\omega_k, \omega)\, \delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0} = \frac{\delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0}}{\omega - \omega_r + iW_r/2}. \quad (34)$$

It is easy to see that the Green's function (27) can be expressed in terms of a superposition of the functions (34)

$$G(\omega_k, \omega) = \sum_{r=1}^{2} |v_k{}^{(r)}|^2\, G_r(\omega_k, \omega). \quad (35)$$

This equation corresponds exactly to relation (30), if initial conditions (13) are taken into account.

We note that when $W\tau \ll 1$, the relaxation times turn out to be of the order of 1/W and the mean free path is

$$l \sim \lambda\!\!\!^{-}\omega_0 / W \gg \lambda\!\!\!^{-}.$$

## 4. ENERGY DISTRIBUTION OF THE PHOTONS

Let us find the energy distribution of the photons at an arbitrary instant of time t, under the condition that one photon $\mathbf{k}_0\lambda_0$ existed in the system at the initial instant of time t = 0, that is, the wave function was of the form $c^{+}_{\mathbf{k}_0\lambda_0}|0\rangle$. The average number of the photons in the state $\mathbf{k}, \lambda$ is determined, in accordance with (11), by the formula

$$n_{\mathbf{k}\lambda}(t) = \langle |f_\lambda(\mathbf{k}, t)|^2 \rangle. \quad (36)$$

The amplitude $f_\lambda(\mathbf{k}, t)$, or more exactly its Laplace transform $f_\lambda(\mathbf{k}, \omega)$, satisfies Eq. (14). Averaging the quantity $|f_\lambda(\mathbf{k}, \omega)|^2$ gives rise to averages of the operators $\hat{J}$ which enter into one of the functions f or $f^{+}$, and to averages of the operators $\hat{J}$ belonging to the functions f and $f^{+}$. An account of the former averages leads to the appearance of the function G. In the time-dependent representation, this function is attenuated because of the departure of the quanta from the initial state. This process is offset by arrival from other states, resulting from the averaging of the operators $\hat{J}$ entering in different Green's function.

In the Laplace representation, formula (36) takes the form

$$n_{\mathbf{k}\lambda}(t) = \iint \frac{d\omega\, d\widetilde{\omega}}{(2\pi)^2} \langle f_\lambda(\mathbf{k}, \omega) f_\lambda^+(\mathbf{k}, \widetilde{\omega})\rangle e^{-i(\omega+\widetilde{\omega})t}$$
$$= \int\limits_{-\infty+i\delta}^{\infty+i\delta} \frac{d\omega'}{2\pi} n_{\mathbf{k}\lambda}(\omega') e^{-i\omega' t}, \qquad (37)$$

where $\omega' = \omega + \widetilde{\omega}$ and the function $\overset{+}{f}_\lambda(\mathbf{k}, \widetilde{\omega})$ is the Laplace transform of the function $f^*(\mathbf{k}, t)$.

Denoting, in analogy with (17),

$$\langle f_\lambda^+(\mathbf{k}, \widetilde{\omega})\rangle = -iG_\lambda^+(\mathbf{k}, \widetilde{\omega}), \qquad (38)$$

we obtain for the function $\overset{+}{G}_\lambda(\mathbf{k}, \omega)$, in analogy with (22),

$$G_\lambda^+(\mathbf{k}, \widetilde{\omega}) = G^+(\omega_k, \widetilde{\omega})\,\delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0}$$
$$= -\frac{\delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0}}{\widetilde{\omega} + \omega_k - [4\tau^2(\widetilde{\omega} + \omega_0 + iW/2)]^{-1}}. \qquad (39)$$

We introduce the function $K_{\lambda\lambda_0}(\mathbf{k}, \omega, \omega'; \mathbf{k}_0)$, defined by

$$K_{\lambda\lambda_0}(\mathbf{k}, \omega, \omega'; \mathbf{k}_0) = \langle f_\lambda(\mathbf{k}, \omega) f_\lambda^+(\mathbf{k}, \omega' - \omega)\rangle, \qquad (40)$$

where the angle brackets denote averaging over the coordinates of the molecules. Using again the diagram technique, we find that the function $K_{\lambda\lambda_0}$ is the sum of all possible diagrams, in which it is necessary to take into account the connections both between the nodes taken on one line and between the nodes taken on different lines. We represent graphically the function $K_{\lambda\lambda_0}$ in the form shown in Fig. 5. In addition, we introduce the function $K_0$ shown in Fig. 6:

$$K^0(\omega_k, \omega, \omega') = G(\omega_k, \omega)G^+(\omega_k, \omega' - \omega). \qquad (41)$$

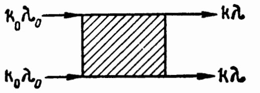

FIG. 5

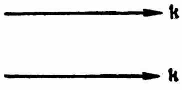

FIG. 6

Using the diagram technique, we can set up for the function K the equation shown graphically in Fig. 7. Its analytic form is

$$K_{\lambda\lambda_0}(\mathbf{k}, \omega, \omega'; \mathbf{k}_0) = K^0(\omega_k, \omega, \omega')\,\delta_{\mathbf{k}\mathbf{k}_0}\delta_{\lambda\lambda_0}$$
$$+ K^0(\omega_k, \omega, \omega') \sum_{\mathbf{k}'\lambda'} U_{\lambda\lambda'}(\mathbf{k}, \mathbf{k}') K_{\lambda'\lambda_0}(\mathbf{k}', \omega, \omega'; \mathbf{k}_0). \qquad (42)$$

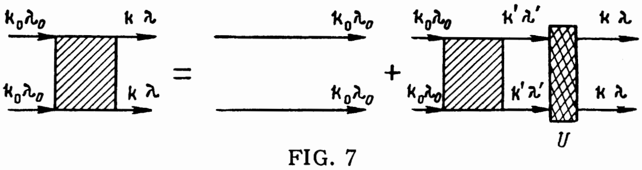

FIG. 7

The kernel $U_{\lambda\lambda'}(\mathbf{k}, \mathbf{k}')$ is expressed in terms of the sum of all possible diagrams which cannot be divided into parts joined by two solid lines only. In the approximation used by us to calculate the self-energy part Σ, the kernel U is determined by the diagram of Fig. 8 (we note that the difference of momenta in the upper and lower lines is conserved in the direction of the arrows)

$$U_{\lambda\lambda'}(\mathbf{k}, \mathbf{k}') = \frac{\sum |Q_{\mathbf{k}\lambda}{}^j|^2 |Q^j_{\mathbf{k}'\lambda'}|^2}{(\omega - \omega_0 + iW/2)(\omega - \omega_0 - \omega' - iW/2)}. \qquad (43)$$

We are interested in the function K averaged over the initial polarizations of the photon and summed over its final polarizations. Apart from this, we are not interested in the angular distributions of the photons and we shall therefore average the function K over the directions of the vector **k**. Then (42) takes the form

$$K(\omega_k, \omega, \omega') = K^0(\omega_k, \omega, \omega') \frac{1}{\rho_k} \delta(\omega_k - \omega_{k_0})$$
$$+ \frac{2}{9} |Q_k|^2 N K^0(\omega_k, \omega, \omega')$$
$$\times \frac{\int d\omega_{k'} \rho_{k'} |Q_{k'}|^2 K(\omega_{k'}, \omega, \omega')}{(\omega - \omega_0 + iW/2)(\omega - \omega_0 - \omega' - iW/2)}, \qquad (44)$$

where $\rho_\mathrm{k}$ is the state density in **k**-space.

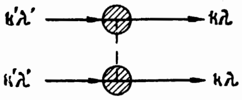

FIG. 8

Let us first find the solution of (44) in the region $W\tau \gg 1$. In this case, according to (41) and (26),

$$K^0(\omega_k, \omega, \omega')$$
$$= (\omega - \omega_k + i\Gamma/2)^{-1}(\omega - \omega' - \omega_k - i\Gamma/2)^{-1}. \qquad (45)$$

We see from this expression that $\omega$ differs from $\omega_\mathrm{k}$ by an amount $\sim\Gamma$. The quantity $\omega'$ is of the same order. Recognizing that $\Gamma \ll W$, the factor in the denominator of the integral term in (44) can be represented in the form

$$(\omega - \omega_0 + iW/2)(\omega - \omega' - \omega_0 - iW/2) \approx \Delta^2 + W^2/4.$$

Multiplying (44) by $|Q_\mathrm{k}|^2\rho_\mathrm{k}$ and integrating over $d\omega_\mathrm{k}$, we readily obtain

$$K(\omega_k, \omega, \omega') = K^0(\omega_k, \omega, \omega') \frac{1}{\rho_k} \delta(\omega_k - \omega_{k_0})$$
$$+ \frac{2}{9} \frac{N|Q_k|^2 |Q_{k_0}|^2}{(\Delta^2 + W^2/4)} K^0(\omega_k, \omega, \omega') K^0(\omega_{k_0}, \omega, \omega') \frac{\omega' + i\Gamma}{\omega'}. \qquad (46)$$

Relation (46) determines $K(\omega_k, \omega, \omega')$ as a function of the Laplace variables $-i\omega$ and $-i\omega'$. The Laplace transforms are defined in the upper half planes of the complex variables $\omega$ and $\omega'$. In the lower half plane it is necessary to use analytic continuation of the obtained expressions.

We denote by $n(\omega_k, t)\,d\omega_k$ the average number of the photons in the interval $d\omega_k$. The Laplace transform $n(\omega_k, \omega')$ of the function $n(\omega_k, t)$ is obtained by multiplying (46) by $\rho_k$ and integrating this expression with respect to $d\omega/2\pi$ [see (37)]:

$$n(\omega_k, \omega') = \frac{i}{\omega' + i\Gamma}\delta(\omega_k - \omega_{k_0}) + \frac{i\Gamma}{\pi[\varkappa^2 + (\Gamma - i\omega')^2]\,\omega'}, \tag{47}$$

where $\kappa = \omega_k - \omega_{k_0}$.

Changing over to the time domain

$$n(\omega_k, t)\,d\omega_k = \left[\int_{-\infty+i\delta}^{\infty+i\delta} n(\omega_k, \omega')\,e^{-i\omega' t}\frac{d\omega'}{2\pi}\right]d\omega_k$$

$$= e^{-\Gamma t}\delta(\omega_k - \omega_{k_0})\,d\omega_k + \frac{\Gamma}{\pi(\varkappa^2 + \Gamma^2)}\left[1 - e^{-\Gamma t}\left(\cos\varkappa t + \frac{\Gamma}{\varkappa}\sin\varkappa t\right)\right]d\omega_k. \tag{48}$$

We see that the quantum relaxation time is given by the quantity $1/\Gamma$. For large times, the distribution becomes stationary and is of the form

$$n(\omega_k)\,d\omega_k = \Gamma d\omega_k / \pi[(\omega_k - \omega_{k_0})^2 + \Gamma^2]. \tag{49}$$

We see from this relation that the quantum energy relative to the initial energy $\omega_{k_0}$ is equal to $\Gamma$, that is, to the reciprocal of the mean free time, although the width of the resonance is equal to W—the lifetime of the isolated molecule. The distribution (48) is normalized to unity.

We now consider the region $W\tau \ll 1$. In this region, the natural physical objects are the quanta of the electromagnetic field in the medium. The field quanta in vacuum are superpositions of these quanta. Since there are two types of quanta in the medium, we determine the functions $K^0_{r_1 r_2}$ and $K_{r_1 r_2}$:

$$K^0_{r_1 r_2}(\omega_k, \omega, \omega') = G_{r_1}(\omega_k, \omega)\,G_{r_2}^{+}(\omega_k, \omega' - \omega), \tag{50}$$

where [see also (34)], in analogy with (39),

$$G_r^{+}(\omega_k, \tilde{\omega}) = -(\tilde{\omega} + \omega_r + iW_r/2)^{-1}, \qquad r = 1,2. \tag{51}$$

The function $K_{r_1 r_2}$, averaged over the directions of the vector **k**, is defined by the relation

$$K_{r_1 r_2}(\omega_k, \omega, \omega') = \sum_\lambda \int \frac{d\Omega_{\mathbf{k}}}{4\pi}\langle \eta_\lambda^{(r_1)}(\mathbf{k}, \omega)\,\eta_\lambda^{(r_2)+}(\mathbf{k}, \omega' - \omega)\rangle. \tag{52}$$

It can be shown, on the basis of (31), that the function $K_{r_1 r_2}$, which represents the density matrix of the photons of the medium with respect to the indices $r_1$ and $r_2$, satisfies the equation

$$\begin{aligned} K_{r_1 r_2}(\omega_k, \omega, \omega') &= \overset{0}{K}_{r_1 r_2}(\omega_k, \omega, \omega')\,\rho_k^{-1}\delta(\omega_k - \omega_{k_0}) \\ &+ \frac{8}{9}N|Q_k|^2 v_k^{(r_1)} v_k^{(r_2)} \overset{0}{K}_{r_1 r_2}(\omega_k, \omega, \omega') \\ &\times \int d\omega_{k'}\rho_{k'}|Q_{k'}|^2\tau^2 Q'^2 v_{k'}^{(r_1')} v_{k'}^{(r_2')} \overset{0}{K}_{r_1' r_2'}(\omega'_k, \omega, \omega') \\ &\times K_{r_1 r_2}(\omega'_k, \omega, \omega'), \end{aligned} \tag{53}$$

where $r_1' = 3 - r_1$ and $r_2' = 3 - r_2$. The functions K and $K_{r_1 r_2}$, like the functions $K^0$ and $K^0_{r_1 r_2}$, satisfy the following relation, which is derivable from (35):

$$K = \sum_{r_1 r_2} |v_k^{(r_1)}|^2 |v_k^{(r_2)}|^2 K_{r_1 r_2}. \tag{54}$$

It follows from the form of (53) that the type of the quantum of the medium (first or second type) does not change during the course of propagation. This result is natural from the physical point of view. The quantum energy scatter connected with the relaxation process has a width of the order of W, whereas the energy difference between the quanta of the first and second type is of the order of $1/\tau \gg W$.

A solution of (53) is obtained in analogy with (46). The functions $K_{11}$ and $K_{22}$ are of interest in themselves, since they give the distribution of the medium quanta of the first and second type, if a single quantum of the first or the second type, respectively, existed during the initial instant of time. These functions, when integrated with respect to $d\omega/2\pi$, take the form

$$n_{rr}(\omega_k, \omega') = \frac{i}{\omega' + iW_r}\delta(\omega_k - \omega_{k_0}) + \frac{iW}{4\pi\Omega^2\tau^2[\varkappa_r^2 + (W_r - i\omega')^2]\,\omega'}; \tag{55}$$

$$\varkappa_1 = \omega_{1k} - \omega_{1k_0} \approx {}^1/_2(1 + \Delta/\Omega)\varkappa, \quad \varkappa_2 = \omega_{2k} - \omega_{2k_0} \approx {}^1/_2\varkappa(1 - \Delta/\Omega). \tag{56}$$

In the time domain

$$n_{rr}(\omega_k, t)\,d\omega_k = e^{-W_r t}\,\delta(\omega_k - \omega_{k_0})\,d\omega_k + \frac{W}{4\pi\Omega^2\tau^2(\varkappa_r^2 + W_r^2)} \times\left[1 - e^{-W_r t}\left(\cos\varkappa_r t + \frac{W_r}{\varkappa_r}\sin\varkappa_r t\right)\right]d\omega_k. \tag{57}$$

For the number of photons in the frequency interval $d\omega_r$ and not in the interval $d\omega_k$ we obtain a distribution analogous to (48), with the substitutions $\Gamma \rightarrow W_r$ and $\kappa \rightarrow \kappa_r$.

We see that the energy distribution widths are determined by the values of $W_r$, that is, by the reciprocal relaxation times of the quanta. We note that the distribution (57) is normal, that is, the total number of quanta of either type is conserved in time:

$$\int n_{rr}(\omega_k, t)\,d\omega_k = \int n_{rr}(\omega_r, t)\,d\omega_r = 1. \tag{57'}$$

At times that are large compared with the relaxation time, the distribution is of the form

$$n_{rr}(\omega_k, t)\,d\omega_r = W_r d\omega_r / \pi[(\omega_{rk} - \omega_{rk_0})^2 + W_r^2]. \tag{58}$$

Analogously, we obtain from (53) the functions

$$n_{12}(\omega_k, t) \text{ and } n_{21}(\omega_k, t) = n_{12}^*(\omega_k, t).$$

For example, for the function $n_{12}$ we have for $t \gg \tau$ and $t \ll \tau$,

$$\begin{aligned} n_{12}(\omega_k, t) &= e^{-i\Omega t - Wt/2}\,\delta(\omega_k - \omega_{k_0}) \\ &+ \frac{W \exp[-i(\Omega + \varkappa\Delta/2\Omega)t - Wt/2]}{\pi(\varkappa^2 + W^2)} \\ &\times \left\{\cos\frac{\varkappa}{2}t - \frac{W}{\varkappa}\sin\frac{\varkappa}{2}t - \exp\left[-\frac{Wt}{2}\left(1 - \frac{|\Delta|}{\Omega}\right)\right]\right. \\ &\times \left.\left(\cos\frac{\varkappa\Delta}{2\Omega}t - \frac{W}{\varkappa}\sin\frac{\varkappa|\Delta|}{2\Omega}t\right)\right\}. \end{aligned} \tag{59}$$

Unlike the functions $n_{11}$ and $_2$, these quantities tend to 0 when $t \gg 1/W$. This tendency of the nondiagonal elements of the density matrix to 0 is a characteristic of relaxation processes (see, for example, [8]).

If at the initial instant of time there was a vacuum photon $c^+_{k_0\lambda_0}|0\rangle$, then we have for the average number of photons at the instant time t, in accord with (54), (57), and (59),

$$\begin{aligned} n(\omega_k, t)\,d\omega_k &= \sum_{r_1 r_2} |v_k^{(r_1)}|^2 |v_k^{(r_2)}|^2 n_{r_1 r_2}(\omega_k, t)\,d\omega_k \\ &= \frac{1}{4}\left\{\left(1 + \frac{\Delta}{\Omega}\right)^2 e^{-W_1 t} + \left(1 - \frac{\Delta}{\Omega}\right)^2 e^{-W_2 t}\right. \\ &+ \left.\frac{2}{\Omega^2\tau^2} e^{-Wt/2}\cos\Omega t\right\}\delta(\omega_k - \omega_{k_0})\,d\omega_k \\ &+ \frac{W\,d\omega_k}{4\pi\Omega^2\tau^2}\left\{\left[\varkappa^2 + W^2\left(\frac{\Omega - \Delta}{\Omega + \Delta}\right)^2\right]^{-1}\right. \\ &\times \left[1 - e^{-W_1 t}\left(\cos\varkappa_1 t + \frac{W}{\varkappa_1}\sin\varkappa_1 t\right)\right] \\ &+ \left[\varkappa^2 + W^2\left(\frac{\Omega + \Delta}{\Omega - \Delta}\right)^2\right]^{-1} \\ &\times \left[1 - e^{-W_2 t}\left(\cos\varkappa_2 t + \frac{W_2}{\varkappa_2}\sin\varkappa t\right)\right] \\ &+ \frac{2e^{-Wt/2}\cos(\Omega + \varkappa\Delta/2\Omega)t}{\varkappa^2 + W^2}\left[\cos\frac{\varkappa}{2}t - \frac{W}{\varkappa}\sin\frac{\varkappa}{2}t\right. \\ &- \left.\exp\left[-\frac{W}{2}\left(1 - \frac{|\Delta|}{\Omega}\right)t\right] \right. \\ &\times \left.\left.\left(\cos\frac{\varkappa}{2}\frac{\Delta}{\Omega}t - \frac{W}{\varkappa}\sin\frac{\varkappa}{2}\frac{|\Delta|}{\Omega}t\right)\right]\right\}, \end{aligned} \tag{60}$$

where $\Omega$ and $\Delta$ depend on $\omega_{k_0}$. The last term holds for the times $t \gg \tau$ and $t \ll \tau$.

We arrive at the same result if we use (54) directly and take for the function $K^0$ its value at $W\tau \ll 1$ [see (27), (39), (41)]

$$\begin{aligned} K^0(\omega_k, \omega, \omega') &= \frac{(\omega - \omega_0 + iW/2)(\omega - \omega_0 - \omega' - iW/2)}{(\omega - \omega_1 + iW_1/2)(\omega - \omega_2 + iW_2/2)} \\ &\times [(\omega - \omega_1 - \omega' - iW_1/2)(\omega - \omega_2 - \omega' - iW_2/2)]^{-1}. \end{aligned} \tag{61}$$

We see that in this case all the relaxation times are of the order of $1/W$.

When $t \gg W^{-1}$ the photon distribution takes the form

$$\begin{aligned} n(\omega_k)\,d\omega_k &= \frac{W\,d\omega_k}{4\pi\Omega^2\tau^2}\left\{\left[(\omega_k - \omega_{k_0})^2 + W^2\left(\frac{\Omega - \Delta}{\Omega + \Delta}\right)^2\right]^{-1}\right. \\ &+ \left.\left[(\omega_k - \omega_{k_0})^2 + W^2\left(\frac{\Omega + \Delta}{\Omega - \Delta}\right)^2\right]^{-1}\right\} \\ &= \frac{W_1(\Omega + \Delta)^2\,d\omega_1}{4\pi\Omega^2[(\omega_{1k} - \omega_{1k_0})^2 + W_1^2]} \\ &+ \frac{W_2(\Omega - \Delta)^2\,d\omega_2}{4\pi\Omega^2[(\omega_{2k} - \omega_{2k_0})^2 + W_2^2]}. \end{aligned} \tag{62}$$

For the total number of quanta we have

$$n = \int n(\omega_k, t)\,d\omega_k = \frac{1}{2}\left(1 + \frac{\Delta^2}{\Omega^2} + \frac{1}{\tau^2\Omega^2}F(t)\right); \tag{63}$$

$$\begin{aligned} F(t) &= e^{-Wt}\left\{\cosh\left(\frac{W\Delta}{2\Omega}t\right)\cos\Omega t\right. \\ &+ \tau\int_0^{t/\tau}\left[\frac{W}{2}\cosh\left(\frac{W\Delta}{2\Omega}\tau y\right)\cos\Omega\tau y\right. \\ &- \left.\Delta\sinh\left(\frac{W\Delta}{2\Omega}\tau y\right)\sin(\Omega\tau y)\right]J_0\left(\frac{t}{\tau} - y\right)dy. \end{aligned} \tag{64}$$

Formula (63) has been presented for the entire time interval, and the terms of order $W\tau$ should be omitted. When $t \gg \tau$ it simplifies appreciably. Accurate to terms of the order of $e^{-Wt}O(\sqrt{\tau/t})$ we have

$$n = \frac{1}{2}\left[1 + \frac{\Delta^2}{\Omega^2} + \frac{1}{\tau^2\Omega^2}\exp\left(-Wt + \frac{W|\Delta|}{2\Omega}t\right)\cos\Omega t\right]. \tag{65}$$

This result follows directly from (60). From relations (63) and (65) it follows that the quantity n decreases from unity at $t = 0$ to a value $(1 + \Delta^2/\Omega^2)/2$ when $t \gg 1/W$.

The decrease in the number of photons with time is connected with the following physical circumstance. At the initial instant of time the photon is a superposition of quanta of the medium with amplitudes $v_{k_0}^{(1)}$ and $v_{k_0}^{(2)}$ [see (7) and (9)]:

$$c_{k_0\lambda_0}|0\rangle = \sum_{r=1}^{2} v_{k_0}^{(r)} \alpha_{k_0\lambda_0}^{(r)+}|0\rangle, \qquad (66)$$

so that the number of quanta of the first type is $|v_{k_0}^{(1)}|^2$ and that of the second type is $|v_{k_0}^{(2)}|^2$. This number is conserved in the course of time [see (57′)], but as a result of the relaxation processes the quanta become noncoherent and the superposition (66) turns into a mixture. Bearing in mind that in a state with one quantum of the medium of the first (second) type $\alpha_{k\lambda}^{(r)+}|0\rangle$ the photon is represented with probability $|v_k^{(r)}|^2$, we obtain for the number of photons n

$$n = |v_{k_0}^{(1)}|^4 + |v_{k_0}^{(2)}|^4 = \frac{1}{2}\left(1 + \frac{\Delta^2}{\Omega^2}\right)$$

when $t \gg W^{-1}$.

On moving away from resonance $|\Delta| \gg 1/\tau$, all the results obtained under the condition $W\tau \gg 1$ and $W\tau \ll 1$ coincide.

We note in conclusion that in this paper we assumed all the molecules to be strictly resonant, that is, we disregarded the scatter in the molecular levels. We assume that an allowance for this scatter, if it is large, leads to some effective replacement of the natural width W by the width of this scatter.

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