PROPAGATION OF PHOTONS IN A MEDIUM OF RESONANT MOLECULES

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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. 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

$$H = \frac{\omega_0}{2} \sum_i \hat{a}_i^\dagger \hat{a}_i + \sum_{k,k'} (c_{k,k'} b_{k'}^\dagger b_k + c_{k,k'} b_k^\dagger b_{k'}^\dagger) + \sum_{k,k'} \omega_{k,k'} \hat{c}_{k,k'}^\dagger \hat{c}_{k,k'},$$ (1)
\[ b_{k\lambda} = \sum_j \sigma_j \left( \frac{2\pi}{\omega_0 V} \right) (M^j k) e^{-ikx_j} = \sum_j \sigma_j Q_{k\lambda} e^{-ikx_j}. \]

We use a system of units in which \( \hbar = 1 \). Here \( x_0 \) 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, \( \sigma_{\pm} \) are the operators of the absorption and emission of a photon with momentum \( k \) and polarization \( \Lambda_j^\pm \), respectively (\( \Lambda = 1, 2 \)), \( \omega_0^2 = c^2 \lambda^2 \), \( \sigma_x \) and \( \sigma_\pm \) are Pauli matrices, and \( M^j \) 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 \( M^j \) and the probability \( W \) of spontaneous dipole radiation of a photon per unit time by an isolated molecule are connected by the relation

\[ W = \frac{4}{3} \frac{\omega_0}{c^2} |M|^2 = \frac{2}{3} |Q_{k\lambda}|^2 \frac{\omega_0^2 V}{\pi c^3}. \]

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

\[ \frac{1}{4\tau^2} = \frac{|Q_{k\lambda}|^2}{3} N = \frac{2}{3} |M|^2 N \left( \frac{\omega_0}{V} \right)^2 \frac{\pi}{2} \rho \lambda W_0. \]

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

In the operators \( b_{k\lambda} \) and \( b_{k\lambda}^\dagger \) only the commutator

\[ [b_{k\lambda}, b_{k'\lambda'}^\dagger] = -\sum_j \sigma_j Q_{k\lambda} Q_{k'\lambda'} e^{-i(k-k')x_j} \]

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 \( \sigma_j^1 \rightarrow -1 \), so that the commutator of the operators \( b_{k\lambda} \) and \( b_{k'\lambda'}^\dagger \) will be a number. By virtue of the phase relations, this number has a maximum when \( k', \lambda' = k, \lambda \).

The difference between the commutator and zero when \( k', \lambda' = k, \lambda \) is connected with the scattering of the quanta and will be taken into account below.

In this approximation the operators \( b_{k\lambda} \) and \( b_{k\lambda}^\dagger \) can be regarded as Bose operators with commutation relations (see\(^2\))

\[ \frac{[b_{k\lambda} b_{k\lambda}^\dagger]}{[b_{k\lambda}^\dagger b_{k\lambda}]} = \frac{4}{3} \tau \lambda \delta_{k\lambda}. \]

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 \( a_{k\lambda}^{(1)} \) and \( a_{k\lambda}^{(2)} \)

\[ a_{k\lambda}^{(r)} = u_k^{(r)} b_{k\lambda} + v_k^{(r)} c_{k\lambda}, \quad r = 1, 2. \]

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

\[ H = -\frac{\omega_0}{2} N + \sum_{k\lambda} \omega_{k\lambda} (a_{k\lambda}^{(1)})^2 + \sum_{k\lambda} \omega_{k\lambda} (a_{k\lambda}^{(2)})^2. \]

Here

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

\[ u_k^{(2)} = 2\tau v_k^{(1)} = 2\tau (1 + \Delta / \Omega)^{1/2}. \]

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 \(-\omega_0 N / 2\). The state \( \alpha_{k\lambda}^{(1)+} |0 \rangle \) 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 \( \omega^2 = \omega_0^2 \).

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...
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 $k_0$ and polarization $\lambda_0$, that is, the state is described by a wave function $c_{k_0\lambda_0}|0\rangle$, or else one quantum of the medium, that is, the state $\alpha(t) = c_{k_0\lambda_0}|0\rangle$. 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

$$
\psi(t) = \sum_{\lambda} f_{\lambda}(k, t)c_{k\lambda}^*|0\rangle e^{i\omega_{\lambda}t/2} + \sum_j q_j(t)\sigma_j^+|0\rangle e^{i\Omega_{\lambda_j}t/2}.
$$

(11)

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

$$
if_{\lambda}(k, t) = \omega_0 f_{\lambda}(k, t) + \sum_j Q_{\lambda\lambda_j} e^{-i\omega_{\lambda_j}t} q_j(t),
$$

$$
i\varphi_j(t) = \omega_0 \varphi_j(t) + \sum_k Q_{\lambda\lambda_k} e^{i\omega_{\lambda_k}t} f_{\lambda}(k', t).
$$

(12)

It is necessary to solve these equations with initial conditions

$$
f_{\lambda}(k, 0) = \delta_{k\lambda}\delta_{\lambda\lambda_0}, \quad \varphi_j(0) = 0.
$$

(13)

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

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

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

$$
(\omega - \omega_0) f_{\lambda}(k, \omega) - \frac{1}{(\omega - \omega_0)} \sum_{\lambda_0} f_{\lambda_0}(k, \lambda') f_{\lambda'}(k', \omega) = i\delta_{k\lambda_0}\delta_{\lambda\lambda_0},
$$

(14)

$$
J_{\lambda\lambda'}(k, k') = \sum_j Q_{\lambda\lambda_j} Q_{\lambda_j\lambda'}^* e^{i(k - k_0)j}.
$$

(15)

We see that the amplitude $f_{\lambda}(k, \omega)$ 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

$$
f_{\lambda}(0, \omega) = iG_0(\omega_0, \omega) \delta_{k_0, \lambda}, \quad \delta_{k_0, \lambda} = i\delta_{k_0, \delta_{\lambda\lambda_0}/(\omega - \omega_0)}.
$$

(16)

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

$$
\langle f_{\lambda}(k, \omega) \rangle = iG_0(k, \omega).
$$

(17)

Carrying out the iterations, we obtain the following series

$$
G_{\lambda}(k, \omega) = G_0 + G_0 \frac{J}{\omega - \omega_0} G_0 + G_0 \frac{J}{\omega - \omega_0} G_0 + \ldots
$$

(18)

When averaging over the coordinates $x_j$ 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. 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 $J$ to a point. In each term of the series (18) we average the product of the operators $J$. Let us examine the averaging method, using as an example a third-order term containing three operators $J$. 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 $J$. 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 $J$ with subsequent averaging will be denoted on the diagrams by dashed lines joining the corresponding nodes. The averaging of exponential functions contained in $J$ 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

$$
Q_{\lambda\lambda_0} Q_{\lambda_0\lambda}^*/(\omega - \omega_0).
$$

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

$$
\frac{1}{(\omega - \omega_0)^2(\omega - \omega_{k_0})^3} \sum_j |Q_{\lambda\lambda_j}|^2 \sum_j |Q_{\lambda_j\lambda}|^2 \sum_j |Q_{\lambda_j\lambda_0}|^2.
$$

After integrating over the vector $k_i$ and subsequently averaging over the directions of the vectors

![FIG. 1](attachment:image.png)
We have left out the real part of the integral with respect to $\omega_k$, 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

$$I = \sum_{n=1}^{\infty} \frac{1}{(\omega - \omega_0)^n} \left( \sum_{k\ell} Q_{k\ell}(\omega - \omega_k) \right)^n$$

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

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:

$$\Sigma^{(1)} = \frac{1}{\omega - \omega_0} \sum_j |Q_{k\ell}(\omega - \omega_k)|^2 = \frac{1}{4\tau^2} \frac{t}{\omega - \omega_0}.$$  

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 $(\omega_0\tau)^{-1}$

$$\Sigma^{(0)} = \frac{1}{\omega - \omega_0} \sum_j |Q_{k\ell}(\omega - \omega_k)|^2 = \frac{1}{4\tau^2} \frac{t}{\omega - \omega_0}. \tag{20}$$

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

$$G_{\omega}(\mathbf{k}, \omega) = \frac{\delta_{\mathbf{k}\mathbf{k'}}}{\omega - \omega_0 - \frac{iW}{2}}.$$  

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

$$\omega = \omega_0 + \frac{i\omega W}{2} \pm \frac{1}{2} (\Delta^2 + \tau^2 + i\omega W - \frac{i}{4} W^2)^{1/2}. \tag{23}$$

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\cite{1}.

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

$$\omega_1 = \omega_0 - \frac{i\omega W}{2} \pm \frac{1}{2} (\Delta^2 + \tau^2 + i\omega W - \frac{i}{4} W^2)^{1/2}; \quad \omega_2 = \omega_0 + \frac{i\omega W}{2} \pm \frac{1}{2} (\Delta^2 + \tau^2 + i\omega W - \frac{i}{4} W^2)^{1/2}. \tag{24}$$

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

$$G(\omega_0, \omega) = \frac{1}{\omega - \omega_0 + i\Gamma/2}. \tag{26}$$

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 $\Gamma = \sigma_0 \sigma_1 \epsilon_0 c$ (see \cite{1}). The mean free path $l$ is in this case of the order of $c/\Gamma \sim (\omega_0^2 \tau)^{-1}$. 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 $l \ll \lambda$.

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

$$G(\omega_0, \omega) = \frac{\omega - \omega_0 + i\omega W/2}{(\omega - \omega_1 + i\omega W/2) \omega - \omega_2 + iW/2}. \tag{27}$$
where $\omega_{1,2}$ are given by expressions (9) and

$$W_1 = \frac{1}{2} W (1 - \Delta / \Omega), \quad W_2 = \frac{1}{2} W (1 + \Delta / \Omega). \tag{28}$$

In this case the Green's function describes

the quanta of the electromagnetic field in the medium

(quanta of the medium), that is, unlike the pre­

ceeding 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

(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

(1 $\tau^{-1}$) of the medium. Indeed, using (7) and

we can find

transitions between different sorts of quanta of the

medium. The latter circumstance is connected with

the fact that the energy difference of these

states is 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 condi­

$$\eta^{(r)}(k, t = 0) = \delta_{kk'0}, \quad \eta^{(t)}(k, t = 0) = \delta_{kk',0}. \tag{32}$$

Let us find the amplitudes $\delta^{(r)}_\lambda(k, \omega)$, averaged

over the coordinates of the centers of gravity of

the molecules of the medium. In analogy with (17),

we put

$$\eta^{(r)}(k, \omega) = \frac{i}{W} G_{\lambda}^{(r)}(k, \omega). \tag{33}$$

Using the diagram technique we developed, we

readily obtain ($r = 1, 2$)

$$G_{\lambda}^{(r)}(k, \omega) = G_r(\omega_0, \omega) \delta_{kk',0} \frac{\delta_{kk',0}}{\omega - \omega_0 + iW/2}. \tag{34}$$

It is easy to see that the Green's function (27) can be

expressed in terms of a superposition of the

functions (34)

$$G(\omega_0, \omega) = \sum_{r=1}^{2} |v_{r}(\omega)|^2 G_r(\omega_0, \omega). \tag{35}$$

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

$$\lambda_0 \gg k.$$
null
Relation (46) determines $K(\omega_k, \omega, \omega')$ as a function of the Laplace variables $-\text{i}\omega$ and $-\text{i}\omega'$. 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 $n(\omega_k, t)\,\text{d}\omega_k$ the average number of the photons in the interval $\text{d}\omega_k$. 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 $\frac{\text{d}\omega}{2\pi}$ [see (37)]:

$$n(\omega_k, \omega') = \frac{i}{\Gamma} \delta(\omega_k - \omega_k)$$

Changing over to the time domain

$$n(\omega_k, t)\,\text{d}\omega_k = \left[ \int_{-\infty}^{\infty} n(\omega_k, \omega) e^{-\text{i}\omega t} \frac{\text{d}\omega}{2\pi} \right] \,\text{d}\omega_k$$

$$= e^{-r_1} \delta(\omega_k - \omega_k) \,\text{d}\omega_k$$

$$+ \frac{\Gamma}{\pi(\omega^2 + \Gamma^2)} \left[ 1 - e^{-r_1} \left( \cos \frac{\omega}{\Gamma} t + \frac{\Gamma}{\omega} \sin \frac{\omega}{\Gamma} t \right) \right] \,\text{d}\omega_k. \quad (48)$$

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

$$n(\omega_k)\,\text{d}\omega_k = \Gamma \,\text{d}\omega_k / \pi(\omega_k^2 + \Gamma^2). \quad (49)$$

We see from this relation that the quantum energy relative to the initial energy $\omega_k$ 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 $W_r \ll 1$. 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 $K^0_{r,r'}$ and $K^1_{r,r'}$:

$$K^0_{r,r'}(\omega_k, \omega, \omega') = G_r(\omega_k, \omega) G^+_r(\omega_k, \omega' - \omega), \quad (50)$$

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

$$G_r(\omega_k, \tilde{\omega}) = \delta(\omega + \omega_r + \text{i}W_r/2)^{-1}, \quad r = 1, 2. \quad (51)$$

The function $K^1_{r,r'}$ averaged over the directions of the vector $k$ is defined by the relation

$$K^1_{r,r'}(\omega_k, \omega, \omega') = \frac{\text{i}W}{4\pi^2} \left\langle \eta_k^{(r)}(k, \omega) \eta_k^{(r')}(k, \omega'-\omega) \right\rangle. \quad (52)$$

It can be shown, on the basis of (31), that the function $K^0_{r,r'}$, which represents the density matrix of the photons of the medium with respect to the indices $r_1$ and $r_2$, satisfies the equation

$$K^0_{r,r'}(\omega_k, \omega, \omega') = K^0_{r,r'}(\omega_k, \omega, \omega') \rho_k \delta(\omega_k - \omega_k)$$

$$+ \frac{8}{9} N \left| Q_k \right|^2 D_0(\omega_k) K^0_{r,r'}(\omega_k, \omega, \omega')$$

$$\times \int \frac{\text{d}\omega_k \rho_k}{\left| Q_k \right|^2} \left[ 2 Q_k^* Q_k^2 \right] \eta_k^{(r)}(k, \omega_k) \eta_k^{(r')}(k, \omega_k) K^0_{r,r'}(\omega_k', \omega, \omega'), \quad (53)$$

where $r_1' = 3 - r_1$ and $r_2' = 3 - r_2$. The functions $K$ and $K^0_{r,r'}$ like the functions $K^1$ and $K^0_{r,r'}$, satisfy the following relation, which is derivable from (35):

$$K = \sum_{r_1, r_2} \left| \eta_k^{(r)} \right|^2 \left| \eta_k^{(r')} \right|^2 K^0_{r,r'}. \quad (54)$$

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 $\text{d}\omega/2\pi$, take the form

$$n_{r,r'}(\omega_k, \omega') = \frac{i}{\omega' - \text{i}W_r} \delta(\omega_k - \omega_k)$$

$$+ \frac{iW}{4\pi^2 \left( \omega_k^2 + (W_r - \text{i} \omega')^2 \right)^2} \eta_k^{(r)}(k, \omega_k) \eta_k^{(r')}(k, \omega_k). \quad (55)$$

$$\eta_1 = \omega_k - \omega_k, \quad \eta_2 = \omega_k - \omega_k.$$  

$$\eta_1 \approx \frac{1}{2} (1 + \Delta / \Omega) \eta_2 \approx \frac{1}{2} \eta_2 (1 - \Delta / \Omega). \quad (56)$$

In the time domain

$$n_{r,r'}(\omega_k, t)\,\text{d}\omega_k = e^{-\text{i}W_r} \delta(\omega_k - \omega_k)\,\text{d}\omega_k$$

$$+ \frac{W}{4\pi^2 \left( \omega_k^2 + W_r^2 \right)^2} \left[ \cos \frac{\omega_k}{\omega_k} t + \frac{W_r}{\omega_k} \sin \frac{\omega_k}{\omega_k} t \right] \,\text{d}\omega_k. \quad (57)$$

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

The function $n(\omega_k, t)\,\text{d}\omega_k$ is defined in the upper half plane of the complex variable $\omega_k$. In the lower half plane it is necessary to use analytic continuation of the obtained expressions.
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:

\[
\int n_{rr}(\omega_h, t) d\omega_h = \int n_{rr}(\omega_r, t) d\omega_r = 1. \tag{57'}
\]

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

\[
n_{rr}(\omega_h, t) d\omega_h = W_r \frac{d\omega_r}{\pi \left( (\omega_h - \omega_h)^2 + W_r^2 \right)}. \tag{58}
\]

Analogously, we obtain from \( (53) \) the functions \( n_{12}(\omega_h, t) \) and \( n_{21}(\omega_h, t) = n_{21}^*(\omega_h, t) \).

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

\[
n_{12}(\omega_h, t) = e^{-i\omega_h rt} \delta(\omega_h - \omega_h) 
+ \frac{W \exp[-i(\Omega + x\Delta/2)t - W^t/2]}{\pi(x^2 + W^2)} \times \left\{ \cos x t - \frac{W}{x} \sin x t - \exp\left[-\frac{W^t}{2} \left( 1 - \frac{\Omega}{\Omega} \right) \right] \right\} \times \left\{ \cos x t - \frac{W}{x} \sin x t \right\}, \tag{59}
\]

Unlike the functions \( n_{11} \) and \( n_2 \), 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, [4]).

If at the initial instant of time there was a vacuum photon \( \phi(0,0) \), then we have for the average number of photons at the instant time \( t \), in accord with \( (54), (57) \), and \( (59) \),

\[
n(n, t) d\omega_h = \sum_{rr} \left\{ |\langle \gamma_r | \gamma_0 \rangle|^2 |\langle \gamma_0 | \gamma_r \rangle|^2 n_{rr}(\omega_h, t) d\omega_h \right\}
= \frac{1}{4} \left\{ \left( 1 + \frac{\Omega}{\Omega} \right)^2 e^{-W t} + \left( 1 - \frac{\Omega}{\Omega} \right)^2 e^{-W t} \right\}
+ \frac{2}{\Omega^2} e^{-W t} \cos \Omega t \delta(\omega_h - \omega_h) d\omega_h
+ \frac{W d\omega_h}{4\pi\Omega^2 t} \left\{ \left[ x^2 + W^2 \left( \frac{\Omega - \Delta}{\Omega + \Delta} \right)^2 \right]^{-1} \times \left[ 1 - e^{-W t} \left( \cos x t + \frac{W}{x} \sin x t \right) \right] \right\}
+ \left[ x^2 + W^2 \left( \frac{\Omega + \Delta}{\Omega - \Delta} \right)^2 \right]^{-1} \times \left[ 1 - e^{-W t} \left( \cos x t + \frac{W}{x} \sin x t \right) \right] \right\}
+ \frac{2e^{-W t} \cos (\Omega + x\Delta/2)t}{x^2 + W^2} \left[ \cos \frac{x}{2} t - \frac{W}{x} \sin \frac{x}{2} t \right]
- \exp\left[-\frac{W}{2} \left( 1 - \frac{\Delta}{\Omega} \right) t \right]
\times \left\{ \cos \frac{x}{2} \Omega t - \frac{W}{x} \sin \frac{x}{2} \Omega t \right\}, \tag{60}
\]

where \( \Omega \) and \( \Delta \) depend on \( \omega_{k0} \). 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 \( (27), (39), (41) \)]

\[
K^0(\omega_h, \omega_e) = \frac{\left( \omega_h - \omega_e + iW/2 \right) \left( \omega_h - \omega_e - \omega_e' - iW/2 \right)}{\left( \omega_h - \omega_e + iW/2 \right) \left( \omega_h - \omega_e - iW/2 \right)}.
\]

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

\[
n(\omega_h) d\omega_h = \frac{W d\omega_h}{4\pi\Omega^2 t} \left\{ \left( \omega_h - \omega_h \right)^2 + W^2 \left( \Omega - \Delta \right)^2 \right\}^{-\frac{3}{2}}
+ \frac{W d\omega_h}{4\pi\Omega^2 \left( \omega_h - \omega_h \right)^2 + W^2}.
\]

For the total number of quanta we have

\[
\int n(\omega_h, t) d\omega_h = \frac{1}{2} \left( 1 + \frac{\Delta}{\Omega} \right)^2 + \frac{1}{t^2} F(t) \tag{63}
\]

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

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 \( e^{-W t} \Omega^3/7 t^3 \) we have

\[
n = \frac{1}{4} \left[ 1 + \frac{\Delta}{\Omega} + \frac{1}{t^2} \exp\left(-W t + \frac{W|\Delta|}{2\Omega} t \right) \cos \Omega t \right]. \tag{65}
\]

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 \( \left( 1 + \Delta^2/\Omega^2 \right)/2 \) 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)]:

\[
c_{k_0}\alpha\beta|0\rangle = \sum_{\alpha,\beta} c_{k_\alpha\beta}|\alpha\beta\rangle \tag{66}
\]

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 \( \alpha_{\lambda\alpha}^{(1)}|0\rangle \) the photon is represented with probability \( \alpha_{\lambda\alpha}^{(1)}|0\rangle \), we obtain for the number of photons \( n \)

\[
n = |v_{k}^{(1)}|^4 + |v_{k}^{(2)}|^4 = \frac{1}{2} \left( 1 + \frac{\Delta^2}{\Omega^2} \right)
\]

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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