

## RELAXATION OF PHOTON DENSITY IN A RESONANT MEDIUM

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Submitted to JETP editor June 29, 1965

J. Exptl. Theoret. Phys. (U.S.S.R.) 50, 395-403 (February, 1966)

We investigate relaxation electromagnetic processes in a resonant medium. The distribution of the medium molecules is assumed to be random. Account is taken of the nonresonant nature of the molecules, i.e., the scatter of the molecular levels of the molecules is introduced. The time variation of the photon density distribution over the states with given wave vector is obtained. The relaxation times and distribution widths are determined. The corresponding kinetic equations are derived. Allowance for the relaxation process leads to a collision term in a form that is integral with respect to time.

## 1. INTRODUCTION

WE investigate in this article electromagnetic relaxation processes in a resonant medium. The molecules are assumed to be randomly distributed in the medium. In the quantum formulation for a system of strictly resonant molecules, the relaxation problem was considered by Galitskiĭ and the author in an earlier paper,<sup>[1]</sup> where we dealt with the time development of a process for a specified initial state, characterized by the presence of a photon or a quantum of the medium in the system. In this paper we consider the problem with allowance for the scatter of the energy levels of the molecules of the medium. This scatter is due to such processes as the Doppler effect, the Stark effect, etc. The transition frequencies  $\omega_j$  ( $j$  is the number of molecules) lie in this case in certain intervals  $\delta$  about their mean value  $\omega_0$ . As will be shown below, the presence of a spread  $\delta$  of the levels greatly influences the evolution of the process in time. Under certain conditions (not too rarefied a medium and not too large  $\delta$ ) the relaxation time is determined by the quantity  $1/\delta$ . During the course of the relaxation, photons are produced in the system with wave vectors  $\mathbf{k}$  ( $\omega_{\mathbf{k}}^2 = c^2 \mathbf{k}^2$ ) that differ from the wave vector of the initial photon  $\mathbf{k}_0$  ( $\omega_{\mathbf{k}} \neq \omega_{\mathbf{k}_0}$ ). The width of the photon distribution with respect to  $\omega_{\mathbf{k}}$  changes with time. The scatter of the molecular levels greatly influences the width of this distribution.

We formulate also the corresponding kinetic equations. It will be shown that they are characterized by a collision term which is integral with respect to the time. This form of the collision

term is caused by the photon-energy uncertainty resulting from the relaxation process.

The medium is assumed to be sufficiently rarefied, so that the condition  $\rho \bar{\lambda}^3 \ll 1$  is satisfied, where  $\rho$  is the density of the medium molecules,  $\rho = N/V$ , and  $\bar{\lambda} = c/\omega_0$ . Under these conditions we can disregard direct transfer of excitation from one molecule to the other via the static Coulomb field. In other words, the condition  $\rho \bar{\lambda}^3 \ll 1$  denotes that excitation transfer takes place in the wave zone and consequently only the resonance radiation field is of importance for the transfer of the excitation. We can also state that the frequency of the collisions that lead to exchange of excitations is of the order of  $\rho \bar{\lambda}^3 w$ , as first shown in a paper by Fursov and Vlasov<sup>[2]</sup> ( $w$  is the natural width of the excited level of the molecule). Consequently, these collisions are not significant under the conditions in question.

The influence of the scatter of the levels on the photon-density oscillations in the resonant medium was considered in a paper by Alekseev and Galitskiĭ.<sup>[3]</sup>

## 2. RELAXATION OF PHOTON DENSITY

Our task is to find the distribution function  $K_{\lambda\lambda_0}(\mathbf{k}, t; \mathbf{k}_0)$  of the photons with respect to the states  $\mathbf{k}$  and  $\lambda$  at an arbitrary instant of time  $t > 0$  under the condition that at the initial instant of time  $t = 0$  there was present in the system a photon with specified wave vector  $\mathbf{k}_0$  and polarization  $\lambda_0$ . The molecules of the medium at the initial instant of time are assumed to be unexcited.

For a specified distribution over the initial

states  $\mathbf{k}_0$  and  $\lambda_0$ , the distribution of the photons is obtained by summing the functions  $K$  with respect to  $\mathbf{k}_0\lambda_0$  with a suitable weighting factor.

As in the earlier papers<sup>[1, 3, 4]</sup> we shall start with the Hamiltonian for the quantized electromagnetic field and a system of  $N$  two-level molecules contained in a volume  $V$ . From the Schrödinger equation we obtain an equation for the amplitude of the probability  $f_\lambda(\mathbf{k}, t)$  of observing in the system the photon  $\mathbf{k}, \lambda$  at an arbitrary instant of time  $t$ . The derivation of this equation is similar to that given in the earlier paper by the author and Galitskiĭ.<sup>[1]</sup> In the Fourier representation we have

$$(\omega - \omega_k) f_\lambda(\mathbf{k}, \omega) - \sum_{\mathbf{k}'\lambda'} J_{\lambda\lambda'}(\mathbf{k}, \mathbf{k}') f_{\lambda'}(\mathbf{k}', \omega) = i\delta_{\mathbf{k}\mathbf{k}_0} \delta_{\lambda\lambda_0}, \quad (1)$$

where

$$J_{\lambda\lambda'}(\mathbf{k}, \mathbf{k}') = \sum_j \frac{Q_{\mathbf{k}\lambda^j} Q_{\mathbf{k}'\lambda'^j}^*}{\omega - \omega_j} e^{i(\mathbf{k}' - \mathbf{k})\mathbf{x}_j}.$$

Here

$$Q_{\mathbf{k}\lambda^j} = (2\pi\omega_j^2 / \omega_k V)^{1/2} (d^j \mathbf{1}_k^\lambda),$$

$d^j$  is the matrix element of the dipole transition of the  $j$ -th molecule (located at the point  $\mathbf{x}_j$ ) from the lower level to the upper level with simultaneous absorption of a photon,  $\mathbf{1}_k^\lambda$  is the photon polarization vector ( $\lambda = 1, 2$ ). We use a system of units in which  $\hbar = 1$ . The distribution of the photons over the states with given  $\mathbf{k}, \lambda$  is given by the function  $K_{\lambda\lambda_0}(\mathbf{k}, t; \mathbf{k}_0)$ :

$$\begin{aligned} K_{\lambda\lambda_0}(\mathbf{k}, t; \mathbf{k}_0) &= \langle f_\lambda(\mathbf{k}, t) f_{\lambda_0}^*(\mathbf{k}_0, t) \rangle \\ &= \int_{-\infty+i\sigma}^{\infty+i\sigma} \frac{d\omega'}{2\pi} e^{-i\omega' t} K_{\lambda\lambda_0}(\mathbf{k}, \omega'; \mathbf{k}_0), \end{aligned}$$

where

$$\begin{aligned} K_{\lambda\lambda_0}(\mathbf{k}, \omega'; \mathbf{k}_0) &= \int_{-\infty+i\sigma}^{\infty+i\sigma} \frac{d\omega}{2\pi} K_{\lambda\lambda_0}(\mathbf{k}, \omega, \omega'; \mathbf{k}_0) \\ &= \int_{-\infty+i\sigma}^{\infty+i\sigma} \frac{d\omega}{2\pi} \langle f_\lambda(\mathbf{k}, \omega) f_{\lambda_0}^*(\mathbf{k}_0, \omega - \omega') \rangle. \end{aligned}$$

The averaging over the coordinates of the centers of gravity of the molecules, denoted by the angle brackets, makes it possible to use diagram-technique methods.<sup>[5]</sup> As applied to these problems, this technique was developed by the author with Galitskiĭ earlier.<sup>[1]</sup> Using the developed procedure, we can formulate an equation for the function  $K_{\lambda\lambda_0}$ . We are not interested in this paper in the angular distribution of the photons, and therefore we introduce a function  $K(\omega_k, \omega, \omega')$  such that  $K d\omega_k$  has the meaning of the number of photons in the interval  $d\omega_k$ . For the function  $K$  we

have the equation

$$\begin{aligned} K(\omega_k, \omega, \omega') &= K^0(\omega_k, \omega, \omega') \delta(\omega_k - \omega_{k_0}) \\ &- \frac{w}{8\pi\tau^2} K^0(\omega_k, \omega, \omega') \int \frac{d\omega_j \chi(\omega_j - \omega_0)}{(\omega - \omega_j + i w/2)(\tilde{\omega} + \omega_j + i w/2)} \\ &\times \int d\omega_{k'} K(\omega_{k'}, \omega, \omega'), \end{aligned} \quad (2)$$

where  $\tilde{\omega} = \omega' - \omega$ , the function  $\chi(\omega_j - \omega_0)$  characterizes the distribution of the molecules over the frequency  $\omega_j$ ,  $w$  is the natural width of the excited level of the molecule, and  $1/\tau$  has the meaning of the product of the Langmuir frequency by the corresponding oscillator strength:

$$w = \frac{4}{3} \frac{\omega_0^3}{c^3} |\mathbf{d}|^2, \quad \frac{1}{\tau^2} = \frac{8\pi}{3} \omega_0 |\mathbf{d}|^2 \rho = 2\pi\rho \tilde{\lambda}^3 w \omega_0.$$

The function  $K^0$  is of the form

$$\begin{aligned} K^0(\omega_k, \omega, \omega') &= [\omega] \overline{\varepsilon(\omega) - \omega_k}^{-1} \\ &\times [\omega] \overline{\varepsilon^*(-\tilde{\omega}) - \omega_k - \omega'}^{-1}, \end{aligned}$$

where  $\varepsilon(\omega)$  is the dielectric constant of the medium ( $\omega \approx \omega_0$ )

$$\varepsilon(\omega) = 1 - \frac{1}{2\tau^2 \omega_0} \int d\omega_j \frac{\chi(\omega_j - \omega_0)}{\omega - \omega_j + i w/2}.$$

Let us integrate (2) with respect to  $\omega_k$ . We note that since  $\rho \tilde{\lambda}^3 \ll 1$ , we have  $|1 - \varepsilon(\omega)| \ll 1$ . Further, we take into account the fact that the effective values are  $\omega \sim \omega_0$  and  $\omega' \ll \omega_0$ . As a result we obtain

$$\begin{aligned} K(\omega_k, \omega, \omega') &= K^0(\omega_k, \omega, \omega') \delta(\omega_k - \omega_{k_0}) \\ &+ \frac{w\omega}{4\pi\omega'} [\varepsilon(\omega) - \varepsilon^*(\omega - \omega')] K^0(\omega_k, \omega, \omega') K^0(\omega_{k_0}, \omega, \omega') \\ &\times \frac{\omega' + \omega [\varepsilon(\omega) - \varepsilon^*(\omega - \omega')]/2}{\omega' + i w + \omega [\varepsilon(\omega) - \varepsilon^*(\omega - \omega')]/2}. \end{aligned} \quad (3)$$

To integrate with respect to  $\omega$  and  $\omega'$  it is necessary to know the concrete form of the distribution function  $\chi(\omega_j - \omega_0)$ . We confine ourselves first to the assumption that this distribution has a dispersion character:

$$\chi(\omega_j - \omega_0) = \frac{\delta}{2\pi} \frac{1}{(\omega_j - \omega_0)^2 + \delta^2/4}. \quad (4)$$

Later on we shall generalize the obtained results, dispensing with the choice of the concrete form of the distribution function.

Under condition (4) all the integrals in (3) with respect to  $\omega_j$  can be evaluated directly, and this relation takes the following form:

$$\begin{aligned} K(\omega_k, \omega, \omega') &= K^0(\omega_k, \omega, \omega') \delta(\omega_k - \omega_{k_0}) \\ &+ \frac{w}{8\tau^2\pi} \frac{\omega' + i\xi}{(\omega - \omega_0 + i\xi/2)(\omega - \omega' - \omega_0 - i\xi/2)(\omega' + i w)} \end{aligned}$$

$$\times K^0(\omega_k, \omega, \omega') K^0(\omega_{k_0}, \omega, \omega') \left[ 1 + \frac{iw}{4\tau^2\omega'} \frac{(\omega' + i\xi)}{(\omega' + iw)(\omega - \omega_0 + i\xi/2)(\omega - \omega' - \omega_0 - i\xi/2) + (\omega' + i\xi)/4\tau^2} \right], \quad (5)$$

where  $\xi = w + \delta$  and

$$K^0(\omega_k, \omega, \omega') = \left[ \omega - \omega_k - \frac{1}{4\tau^2} \left( \omega - \omega_0 + \frac{i\xi}{2} \right)^{-1} \right]^{-1} \times \left[ \omega - \omega' - \omega_k - \frac{1}{4\tau^2} \left( \omega - \omega' - \omega_0 - \frac{i\xi}{2} \right)^{-1} \right]^{-1}. \quad (6)$$

The pole values of the frequency  $\omega$  in (6) take the simplest form if the inequalities  $\xi\tau \ll 1$  and  $\xi\tau \gg 1$  are satisfied. Let us consider first the inequality  $\xi\tau \ll 1$ . This case corresponds to not too rarefied a medium ( $w\tau \ll 1$ ) and not too large a scatter of the energy levels of the molecules ( $\delta\tau \ll 1$ ). The condition  $w\tau \ll 1$  signifies that the energy width  $\Gamma$ , due to the resonant scattering of the photons by the molecules (see, for example, [6]),

$$\Gamma_w = \rho\sigma_{\text{res}}c = \frac{1}{4\tau^2} \frac{w}{(\omega - \omega_0)^2 + w^2/4}, \quad (7)$$

is large compared with the width of the excited level  $w$ ,  $\Gamma_w \gg w$ . The condition  $\delta\tau \ll 1$  signifies that the additional broadening connected with the scatter of the levels likewise leaves this condition unchanged. Then the resultant distribution widths (formulas (10) and (11)) are determined by the values of  $w$  and  $\delta$  and finally depend on the initial state (the values of  $\omega_{k_0}$ ).

The case  $w\tau \gg 1$  corresponds to a very highly rarefied medium, when  $\Gamma_w \ll w$ . The distribution widths are determined in this case by the quantity  $\Gamma_\xi$  ( $\Gamma_\xi$  differs from  $\Gamma_w$  in that the width  $w$  is replaced by the width  $\xi$ ). When  $w\tau \ll 1$ , but  $\delta\tau \gg 1$ , the medium is in effect highly rarefied with respect to the given photon, and the distribution width is likewise determined by the value of  $\Gamma_\xi$ . When  $\xi\tau \ll 1$  the function (6) is of the four-pole type, and is conveniently represented in the form

$$K^0(\omega_k, \omega, \omega') = \sum_{r_1, r_2} (v_k^{(r_1)})^2 (v_k^{(r_2)})^2 K_{r_1 r_2}^0(\omega_k, \omega, \omega'), \quad (8)$$

where  $r_1, r_2 = 1, 2$ ,

$$v_k^{(1)} = \sqrt{\frac{1}{2} \left( 1 + \frac{\Delta}{\Omega} \right)}, \quad v_k^{(2)} = -\sqrt{\frac{1}{2} \left( 1 - \frac{\Delta}{\Omega} \right)},$$

$$\Omega = \sqrt{\Delta^2 + \frac{1}{\tau^2}}, \quad \Delta = \omega_k - \omega_0;$$

$$K_{r_1 r_2}^0(\omega_k, \omega, \omega')$$

$$= [(\omega - \omega_{r_1} + i\xi_{r_1}/2)(\omega - \omega_{r_2} + i\xi_{r_2}/2)]^{-1};$$

$$\omega_{1,2} = \omega_0 + \frac{1}{2}\Delta \pm \frac{1}{2}\Omega,$$

$$\xi_{1,2} = \frac{1}{2}\xi(1 \mp \Delta/\Omega), \quad w_{1,2} = \frac{1}{2}w(1 \mp \Delta/\Omega).$$

A decomposition similar to (8) holds also for the function  $K$ . Carrying out these decompositions in (5), we obtain a relation for the function  $K_{r_1 r_2}$ . The function  $K^0(\omega_{k_0}, \omega, \omega')$  which enters in this relation is likewise conveniently represented in the form (8). Then the main contribution in the expression for the function  $K_{r_1 r_2}$  is made by that component of the function  $K^0(\omega_{k_0}, \omega, \omega')$  which depends on the same indices  $r_1$  and  $r_2$ . This circumstance is connected with the fact that the interval between the pole values of the frequencies is of the order of  $1/\tau$ , in a broad range of values of  $\omega_{k_0}$ , and the width of the scatter with respect to  $\omega_k$ , as will be shown, is of the order of  $\delta$  ( $\delta \ll 1/\tau$ ). The physical meaning of the function  $K_{r_1 r_2}$  was discussed earlier.<sup>[1]</sup> Namely, the functions  $K_{11}$  and  $K_{22}$  give the distribution of the quanta of the first and second types in the medium (with energies  $\omega_1$  and  $\omega_2$ , respectively) for a specified quantum in the medium at the initial instant of time.

Integrating the obtained relations with respect to  $\omega$ , we get

$$K_{11}(\omega_1, \omega') = \frac{i}{\omega' + i\xi_1} \delta(\omega_1 - \omega_{10}) - \frac{i\omega_1}{\pi} \frac{\omega' + i\xi}{\omega'(\omega' + iw + i\delta_1)} \frac{1}{(\omega' + i\xi_1)^2 - \kappa_1^2}, \quad (9)$$

where

$$\kappa_{1,2} = \omega_{1,2}(k) - \omega_{1,2}(k_0) \cong \frac{1}{2}\kappa(1 \pm \Delta/\Omega),$$

$$\omega_{r0} = \omega_r(k_0), \quad \kappa = \omega_k - \omega_{k_0};$$

$$K_{11}(\omega_1, \omega') = K_{11}(\omega_k, \omega') d\omega_k / d\omega_1.$$

Going over to the time domain, we have

$$K_{11}(\omega_1, t) = e^{-\xi_1 t} \delta(\omega_1 - \omega_{10}) + \frac{w_1 \xi_1}{\pi(w + \delta_1)} \frac{1}{\kappa_1^2 + \xi_1^2} \times \left[ 1 - e^{-\xi_1 t} \left( \cos \kappa_1 t + \frac{\xi_1}{\kappa_1} \sin \kappa_1 t \right) \right] - \frac{w_1}{\pi} \frac{\delta_2}{w + \delta_1} \frac{1}{\kappa_1^2 + w_2^2} \times \left[ e^{-w_1 t - \delta_1 t} - e^{-\xi_1 t} \left( \cos \kappa_1 t - \frac{w_2}{\kappa_1} \sin \kappa_1 t \right) \right]. \quad (10)$$

The expression for the function  $K_{22}$  is obtained by making the substitutions  $\chi_1 \rightarrow \chi_2$ ,  $w_{1,2} \rightarrow w_{2,1}$ , and  $\delta_{1,2} \rightarrow \delta_{2,1}$ .

The distribution for the functions  $K_{12}$  and  $K_{21} = K_{12}^*$  has a more complicated character. Unlike the functions  $K_{11}$  and  $K_{22}$ , these functions tend to zero as  $t \rightarrow \infty$ . In the time domain, in the region that is significant with respect to  $\omega_k$  ( $\chi \ll 1/\tau$ ),

we have

$$\begin{aligned}
 K_{12}(\omega_k, t) + K_{21}(\omega_k, t) &= 2 \cos \Omega t e^{-\xi t/2} \delta(\omega_k - \omega_{k_0}) \\
 &+ \frac{2w}{\pi} (\kappa^2 + w^2)^{-1} \cos \left( \Omega_0 + \frac{\kappa \Delta}{2\Omega} \right) \\
 &\times t e^{-\xi t/2} \left[ \left( \cos \frac{\kappa}{2} t - \frac{w}{\kappa} \sin \frac{\kappa}{2} t \right) \right. \\
 &- \frac{1}{2} e^{-w_1 t} \left( \cos \frac{\kappa \Delta}{2\Omega} t - \frac{w}{\kappa} \sin \frac{\kappa \Delta}{2\Omega} t \right) \\
 &- \left. \frac{1}{2} e^{-w_2 t} \left( \cos \frac{\kappa \Delta}{2\Omega} t + \frac{w}{\kappa} \sin \frac{\kappa \Delta}{2\Omega} t \right) \right] - \frac{2w \Delta e^{-wt}}{\pi (\kappa^2 + w^2)} \\
 &\times \int_0^t F(t-t') e^{-\delta t'/2} \sin \left( \Omega_0 + \frac{\kappa \Delta}{2\Omega} \right) t' \\
 &\times \left( \operatorname{sh} \frac{w \Delta}{2\Omega} t' \cos \frac{\kappa \Delta}{2\Omega} t' - \frac{w}{\kappa} \operatorname{ch} \frac{w \Delta}{2\Omega} t' \sin \frac{\kappa \Delta}{2\Omega} t' \right) dt', \quad (11)^*
 \end{aligned}$$

where  $\Omega_0$  depends on  $\omega_{k_0}$  (where possible, we neglect the difference between  $\omega_k$  and  $\omega_{k_0}$ );

$$F(t) = \frac{d}{dt} e^{-\delta t/2} \int_0^t I_0 \left[ \frac{\delta}{2} (t-t') \right] J_0 \left( \frac{t'}{\tau} \right) dt', \quad (12)$$

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$$K(\omega, \omega') = \int K(\omega_k, \omega, \omega') d\omega_k = K^0(\omega_{k_0}, \omega, \omega') \left[ 1 + \frac{iw}{4\tau^2 \omega'} \frac{\omega' + i\xi}{(\omega' + iw)(\omega - \omega_0 + i\xi/2)(\omega - \omega' - \omega_0 - i\xi/2) + (\omega' + i\xi)/4\tau^2} \right]. \quad (15)$$

Integrating with respect to  $\omega$  and going over to the time domain, we get

$$\begin{aligned}
 K_{11}(t) &= \frac{1}{w + \delta_1} (w + \delta_1 e^{-wt - \delta_1 t}), \\
 K_{12}(t) + K_{21}(t) &= 2 \cos \Omega t e^{-wt - \delta t/2} \operatorname{ch} \frac{w \Delta}{2\Omega} t \quad (16)
 \end{aligned}$$

$$+ 2\Delta e^{-wt} \int_0^t F(t-t') e^{-\delta t'/2} \sin \Omega t' \operatorname{sh} \frac{w \Delta}{2\Omega} t' dt'. \quad (17)$$

Here the function  $F$  is defined by relation (12). The quantities  $\Omega$ ,  $\Delta$ , and  $\delta_1$  in (16) and (17) depend on the value of  $\omega_{k_0}$ .

We now consider the limiting case  $\xi \tau \gg 1$ , which signifies either a very highly rarefied medium or a large scatter of the energy levels. The width  $\Gamma_\xi$  due to the process of resonant scattering of photons by the molecules is then small compared with the width  $w$ . The Green's function has in this case, in fact, a single pole with respect to  $\omega$ , and all the pertinent results are obtained from (10) and (16) by means of the substitution

$$\kappa_1 \rightarrow \kappa, \quad \omega_1 \rightarrow \omega_k, \quad \xi_2, w_2 \rightarrow \xi, w, \quad \xi_1, \delta_1 \rightarrow \Gamma_\xi, \quad w_1 \rightarrow \Gamma_\xi w / \xi,$$

where

$$\Gamma_\xi = \xi [4\tau^2(\Delta^2 + \xi^2/4)]^{-1}. \quad (7')$$

\*sh = sinh, ch = cosh.

$I_0$  and  $J_0$  are Bessel functions. In the region  $\tau \ll t < 1/\delta$  we have

$$F(t) = e^{-\delta t/2} J_0(t/\tau). \quad (13)$$

From (10) and (11) it follows that if a photon with  $k = k_0$  was present at the initial instant of time, then photons with  $\omega_k \neq \omega_{k_0}$  are produced during the relaxation process, and the width of the distribution with respect to  $\omega_k$  is determined by the values of  $w$  and  $\delta$ , that is, by the natural width of the level and by the width of the scatter. The same quantities determine also the time variation of the relaxation. After a time  $t \gg 1/\delta$  the distribution becomes stationary and takes the form

$$\begin{aligned}
 K(\omega_k) &= \frac{w \xi}{\pi} \{ [(w + \delta_1)(\kappa_1^2 + \xi_1^2)]^{-1} \\
 &+ [(w + \delta_2)(\kappa_2^2 + \xi_2^2)]^{-1} \}. \quad (14)
 \end{aligned}$$

We now consider not the differential but the integral characteristics. Integrating (5) with respect to  $\omega_k$ , we obtain

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We now generalize the obtained results, disregarding the concrete form of the distribution function (4). We assume that the function  $\chi(\omega_j - \omega_0)$  characterizes a certain distribution with effective width  $\delta$  which decreases sufficiently rapidly when  $|\omega_j - \omega_0| > \delta$ . This may be, for example, a Doppler distribution. We assume that the effective width of the distribution is large compared with the natural width of the level, that is,  $\delta \gg w$ .

We assume first that the distribution is not very broad, that is,  $\delta \tau \ll 1$ . In this case for effective frequencies  $\omega$  satisfying the inequality  $|\omega - \omega_0| > \delta$ , the distribution with respect to  $\omega_j$  does not affect the results, and the photon distribution is described by the formulas (10) and (11) derived above, in which we must put  $\delta = 0$ . This circumstance is connected with the fact that when  $\delta \tau \ll 1$  the influence of the distribution over the frequencies can affect only the incoherent part of the interaction, that is, the imaginary part of the corresponding pole values of the frequencies. An additional contribution to the absorption (compared with the contribution due to the natural width of the level  $w$ ) can come only from molecules for which  $\omega_j \approx \omega$ . But if the distribution given by the functions  $\chi(\omega_j - \omega_0)$  decreases much more rapidly than (4), and the effective frequencies are  $|\omega - \omega_0| > \delta$ ,

then the corresponding contribution can be neglected. In the resonance region at effective frequencies  $\omega - \omega_0 \sim \delta$ , the influence of the distribution of the frequencies causes the effective width  $w$  to be replaced by the effective width  $\delta$ .

Let us assume now that the effective width of the distribution  $\delta$  is much larger than  $1/\tau$ , namely  $\delta\tau \gg 1$ . In this case the contribution to the absorption is made in the resonance region only by molecules with  $\omega_j \approx \omega$ , that is, the principal role is played in the integral contained in  $\epsilon(\omega)$  by the pole term. The pole value of the frequencies is determined by the condition

$$\omega \left[ 1 - \frac{i\pi}{2\omega\tau^2} \chi(\omega - \omega_0) \right] = \omega_k.$$

When  $\delta\tau \gg 1$  the second term in the square bracket is small (in absolute value) compared with unity. In the resonance region we have  $\Delta < \delta$ ,  $\chi(\omega - \omega_0) = 2C/\delta$ , where  $C$  is a numerical factor of the order of unity, and we have for the pole value of the frequency

$$\omega - i\pi C/\tau^2\delta = \omega_k.$$

Accordingly, the distribution function is obtained from (10) by means of the substitution

$$\kappa_1 \rightarrow \kappa; \quad \omega_1 \rightarrow \omega_k; \quad \xi_2, w_2 \rightarrow \xi, w;$$

$$\xi_1, \delta_1 \rightarrow \frac{\pi C}{\tau^2\delta}; \quad w_1 \rightarrow \frac{w}{\xi} \frac{\pi C}{\tau^2\delta}.$$

In the region far from resonance,  $\Delta \gg \delta$ , the effect of the distribution with respect to  $\omega_j$  is not felt for reasons discussed above, and when  $w\tau \ll 1$  the distribution is described by formulas (10) and (11) (with the condition  $\delta = 0$ ). When  $w\tau \gg 1$  the corresponding distribution is obtained from (10) by making the substitutions  $\kappa_1 \rightarrow \kappa$ ,  $\omega_1 \rightarrow \omega_k$ ,  $\delta \rightarrow 0$ ,  $w_2 \rightarrow w$ , and  $w_1 \rightarrow \Gamma_w$ .

### 3. KINETIC EQUATIONS

The obtained functions  $K_{R_1 R_2}(\omega_k, t)$  for  $\xi\tau \ll 1$  and  $K(\omega_k, t)$  for  $\xi\tau \gg 1$  satisfy definite kinetic equations. The derivation of these equations is a more general problem than the direct derivation of the results (10) and (11), and is of great interest. We confine ourselves here to a derivation of the equations for the functions  $K_{R_1 R_2}(\omega_k, t)$  and  $K(\omega_k, t)$ , which give the distribution of the field quanta in the medium. We use for this purpose a method developed by Galitskiĭ and Yakimets.<sup>[7]</sup>

Let us formulate the kinetic equation satisfied by the function  $K_{11}(\omega_1, \omega')$ . This equation should have the following form:

$$(\xi_1 - i\omega')K_{11}(\omega_1, \omega') = \delta(\omega_1 - \omega_{10}) + \int d\omega_1' S(\omega_1 - \omega_1', \omega') K_{11}(\omega_1', \omega'), \quad (18)$$

with the kernel  $S(\omega_1 - \omega_1', \omega')$  depending only on the frequency difference  $\kappa_1' = \omega_1 - \omega_1'$ . The latter circumstance is due to the fact that the solution of the equation—the function  $K_{11}$  (see (9))—depends only on the frequency difference  $\kappa_1$ . (In view of the narrowness of the distribution, we disregard the variation of the quantities  $w_1$  and  $\xi_1$  in the integration.)

We take the Fourier transform  $K_{11}(y, \omega')$  of (18) with respect to the variable  $\omega_1$ :

$$(\xi_1 - i\omega')K_{11}(y, \omega') = 1/2\pi + 2\pi S(y, \omega')K_{11}(y, \omega'). \quad (19)$$

Using (9), we obtain

$$K_{11}(y, \omega') = \frac{i}{2\pi} \frac{1}{\omega' + i\xi_1} \left( 1 + \frac{iw_1(\omega' + i\xi)}{\omega'(\omega' + iw + i\delta_1)} \times \exp\{i|y|(\omega' + i\xi_1)\} \right). \quad (20)$$

Substituting (20) in (19), we get

$$S(y, \omega') = \frac{1}{2\pi} (\xi_1 - i\omega') \left[ 1 - \frac{i\omega'(\omega' + iw + i\delta_1)}{w_1(\omega' + i\xi)} \times \exp\{|y|(\xi_1 - i\omega')\} \right]^{-1}. \quad (21)$$

Taking the inverse Fourier transform, we obtain the kernel  $S(\kappa_1', \omega')$ :

$$S(\kappa_1', \omega') = \frac{w_1(\omega' + i\xi)}{2\pi(\omega' + iw)} \frac{1}{(\xi_1 - i\omega')^2 + \kappa_1'^2} \times \left[ (\xi_1 - i\omega' - i\kappa_1') F\left(1, 1, 2 + \frac{i\kappa_1'}{\xi_1 - i\omega'}; z\right) + (\xi_1 - i\omega' + i\kappa_1') F\left(1, 1, 2, -\frac{i\kappa_1'}{\xi_1 - i\omega'}; z\right) \right]; \quad (22)$$

$$z = \frac{iw_1(\omega' + i\xi)}{(\omega' + i\xi_1)(\omega' + iw)},$$

where  $F$  is a hypergeometric function.

The kernel (22) is quite complicated. The main features of the process can be described also by an approximate expression for the kernel  $S$  obtained by letting  $w_1 \rightarrow 0$  in the arguments of the hypergeometric functions. Then

$$S(\kappa_1', \omega') = \frac{w_1}{\pi} \frac{\omega' + i\xi}{\omega' + iw} \frac{\xi_1 - i\omega'}{(\xi_1 - i\omega')^2 + \kappa_1'^2}. \quad (23)$$

With such a simplified kernel the kinetic equation in the time domain takes the form

$$K_{11}(\omega_1, t) = -\xi_1 K_{11}(\omega_1, t) + \frac{w_1}{\pi} \int d\omega_1' \int_0^t dt' K_{11}(\omega_1', t - t')$$

$$\times \left\{ e^{-\xi_1 t'} \cos \kappa_1' t' + \frac{\delta}{\kappa_1'^2 + (\xi_1 - w)^2} [(\xi_1 - w) e^{-wt'} + e^{-\xi_1 t'} (\kappa_1' \sin \kappa_1' t' - (\xi_1 - w) \cos \kappa_1' t')] \right\}. \quad (24)$$

When  $\xi\tau \gg 1$  the equation for the function K is obtained from (18), (22), and (24) by means of the substitutions  $\xi_1 \rightarrow \Gamma_\xi$  (see (7')),  $w_1 \rightarrow \Gamma_\xi w/\xi$ , and  $\omega_1 \rightarrow \omega_K$ . If we forego the concrete form of the distribution (4), the generalization for the expression for the kernel S is carried out in the same manner as was done above for the function K.

The derived equations are characterized by the appearance of a collision term which is integral with respect to time. This form of the collision term is due to the photon-energy uncertainty resulting from the relaxation processes. Because of this uncertainty, the  $\delta$ -function which expresses the conservation of energy in each individual scattering act goes over into a function of the form  $\gamma/(\omega_{k_1} - \omega_{k_2})^2 + \gamma^2$ , the width  $\gamma$  of which is time dependent. When  $\gamma$  tends to zero, the equation takes on the usual form. If, however, we are interested in the distribution width, then it is essen-

tial to retain the collision operator in a form that is integral with respect to time.

The author thanks V. M. Galitskiĭ for valuable discussion of this work.

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Translated by J. G. Adashko