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RADIATION DIFFUSION IN A RESONANT MEDIUM

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Propagation of radiation in a resonant medium of two-level molecules[3,4] is studied by quantum-mechanics methods. A distribution function n(**r**, t) for the excited molecules is found and a kinetic equation is then set up for it.

1. INTRODUCTION

THE propagation of radiation in a resonant medium has been studied many times both theoretically and experimentally. Among the earlier papers we can mention the theoretical work by Compton[1] and the experimental work by Zemansky[2]. Compton assumed that a quantum executes in a resonant medium a motion similar to Brownian motion of a particle, since it experiences a number of absorption and re-radiation acts prior to leaving the volume[2]. Therefore the motion of the excitation in the system can be judged from the density n(**r**, t) of the excited atoms. Compton obtained for the function n(**r**, t) a diffusion equation with a diffusion coefficient

$$D = \gamma / 3k_0^2, \tag{1}$$

where $1/k_0$ is the mean free path of the quantum and $1/\gamma$ is the lifetime of the individual excited atom. Holstein[3] and Biberman[4] have shown that the diffusion approximation was insufficient for the analysis of this question. They obtained phenomenologically an integro-differential equation for the function n(**r**, t). An equation for the function n(**r**, t) *was also derived* by quantum-mechanical means by D'yakonov and Perel'[5] for the case of a Doppler scatter of the atomic levels.

In this paper we determine the distribution function n(**r**, t) of the excited atoms by solving the Schrödinger equation. A general expression is obtained for the function n(**r**, t) when the resultant scatter of the atomic levels is determined by two processes, one of which gives the Doppler line width and the other the dispersion width. All the calculations are carried through to conclusion for the dispersion line shape only. The kinetic equation satisfied by the function n(**r**, t) is derived. Unlike the equation obtained by Holstein[3] or Biberman[4], our equation is characterized by a collision term which is integral in time. In addition, we took retardation into account when calculating the kernel of the equation. The transition to the Biberman-Holstein equation is realized in the same manner as in the paper of D'yakonov and Perel'[5]. In considering this question we used the mathematical formalism developed in the papers of Alekseev, Vdovin, and Galitskiĭ[6-8].

2. GENERAL EXPRESSION FOR THE FUNCTION n(r, t)

We consider a medium consisting of an aggregate of two-level molecules. It is assumed that the transition frequencies ω_j (j is the number of the molecules) are distributed in a certain interval about an average value ω_0. If the line broadening is determined by two processes, one of which gives a dispersion line shape and the other a Doppler line shape, then the resultant distribution function of the frequencies ω_j takes the following form:

$$\chi(\omega_j - \omega_0) = \int_{-\infty}^{\infty} d\omega_j' \chi_1(\omega_j - \omega_j') \chi_2(\omega_j' - \omega_0),$$

$$\chi_1(\omega_j - \omega_j') = \frac{\delta}{2\pi}[(\omega_j - \omega_j')^2 + \delta^2/4]^{-1}, \tag{2}$$

$$\chi_2(\omega_j - \omega_0) = \frac{1}{\beta\sqrt{\pi}} \exp\left[-\frac{(\omega_j - \omega_0)^2}{\beta^2}\right] \tag{3}$$

The medium is assumed sufficiently rarefied, so that the condition $\rho\lambdabar_3 \ll 1$ is satisfied, where $\rho = N/V$ is the density of the molecules of the medium and $\lambdabar = c/\omega_0$ is the wavelength of the resonance radiation. When this condition is satisfied, it is possible to disregard processes in which the excitation is directly transferred from molecule to molecule via the static Coulomb field[10]. The spatial distribution of the molecules of the medium and their orientation are assumed to be random.

In analogy with the procedure first used by Alekseev, Vdovin and Galitskiĭ[6], we write the Hamiltonian of a system of N two-level molecules in a radiation field, contained in a volume V, in the form

$$H = \frac{1}{2}\sum_j \omega_j \sigma_z{}^j + \sum_{j\mathbf{k}\lambda} (Q_{\mathbf{k}\lambda}{}^{j*} e^{i\mathbf{k}\mathbf{x}_j} C_{\mathbf{k}\lambda}\sigma_+{}^j + Q_{\mathbf{k}\lambda}{}^j e^{-i\mathbf{k}\mathbf{x}_j} C_{\mathbf{k}\lambda}{}^+\sigma_-{}^j)$$

$$+ \sum_{\mathbf{k}\lambda} \omega_k C_{\mathbf{k}\lambda}{}^+ C_{\mathbf{k}\lambda}, \tag{4}$$

$$Q_{\mathbf{k}\lambda}{}^j = \left(\frac{2\pi}{\omega_k V}\right)^{1/2} (\mathbf{M}^{j*}\mathbf{l}_{\mathbf{k}}{}^\lambda), \quad \sigma_z{}^j = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}_j,$$

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

We use here a system of units in which $\hbar = 1$; $\mathbf{x}_j$ are the coordinates of the centers of gravity of the molecules; $C_{\mathbf{k}\lambda}$ and $C^+_{\mathbf{k}\lambda}$ are the operators for absorption and creation of a photon with momentum $\mathbf{k}$ and polarization λ ($\lambda = 1, 2$); $\mathbf{M}^j$ is the matrix element of the transition of the j-th isolated molecule from the lower level to the upper one with simultaneous absorption of a quantum; $\mathbf{l}_k$ is the quantum polarization vector; $\omega_k^2 = c^2k^2$. The quantity $\mathbf{M}^j$ is connected with γ (the probability of spontaneous dipole emission of a photon per unit time by an isolated molecule) in the following manner:

$$\gamma = \frac{4\omega_0}{3c^3} |\mathbf{M}^j|^2. \tag{6}$$

In the Hamiltonain (4) we have left out small nonresonant terms $\sim (\omega_0\tau)^{-2}$, where

$$\tau^{-2} = 2\pi\rho\lambdabar^3\gamma\omega_0 \tag{7}$$

The time τ is the characteristic time for the different electromagnetic processes occurring in such a medium[6-8].

The Hamiltonian (4) conserves the number of excitations, and therefore the wave function at any instant of time can be written in the form[7]

$$\psi(t) = \sum_j \varphi_j(t)\sigma_+{}^j \left|0\right\rangle \exp\left(\frac{i}{2}\sum_j \omega_j t\right)$$

$$+ \sum_{\mathbf{k}\lambda} f_\lambda(\mathbf{k}, t) C_{\mathbf{k}\lambda}{}^+ \left|0\right\rangle \exp\left(\frac{i}{2}\sum_j \omega_j t\right) \tag{8}$$

Here $|0\rangle$ denotes the state of the "vacuum": there are no photons and the molecules are not excited.

Assume that at the initial instant of time the molecule numbered j_0 is excited, i.e., the initial condition for the function $\psi(t)$ is of the form:

$$\varphi_j(0) = \delta_{j,j_0}, \qquad f_\lambda(\mathbf{k}, 0) = 0 \tag{9}$$

From Schrödinger's equation we obtain an equation for the amplitudes $\varphi_j(t)$ and $f_\lambda(\mathbf{k}, t)$, from which, by eliminating the amplitude $f_\lambda(\mathbf{k}, t)$, we arrive at an equation for the function $\varphi_j(\omega)$, which is the Laplace transform of the function $\varphi_j(t)$:

$$\varphi_j(\omega) = i\delta_{j,j_0}(\omega - \omega_j + i\gamma/2)^{-1} + (\omega - \omega_j + i\gamma/2)^{-1}$$

$$\times \sum_{j'\neq j} \varphi_{j'}(\omega) \sum_{\mathbf{k}\lambda} \frac{Q_{\mathbf{k}\lambda}{}^{j'} Q_{\mathbf{k}\lambda}{}^{j*}}{\omega - \omega_k} \exp[i\mathbf{k}(\mathbf{x}_j - \mathbf{x}_{j'})], \tag{10}$$

$$\varphi_j(\omega) = \int_0 dt\, e^{-pt}\varphi_j(t), \quad \omega = ip. \tag{11}$$

We are interested in the quantity $|\varphi_j(t)|^2$, which is the probability that at the instant of time t there will be excited a molecule located at a distance $\mathbf{r}_j = \mathbf{x}_j - \mathbf{x}_{j_0}$ from the initially excited molecule. We are considering here the case $j \neq j_0$; the case $j = j_0$ calls for a separate analysis, which was carried out in[11]. Equation (10) is solved by an iteration method. A similar iteration series is obtained for the function $\varphi_j^+(\omega')$, which is the Laplace transform of the function $\varphi_j^*(t)$.

The averaging over the coordinates of the centers of gravity of the molecules, used in the calculation of the product of the series $\varphi_j(\omega)\varphi_j^+(\omega_1 - \omega)$, is denoted by the angle brackets. This averaging makes it possible to use a diagram technique developed in detail as for problems of this type in the papers of Vdovin and Galitskiĭ[7,8]. Since we are using in the present paper the same diagrams as in[8], we shall not discuss this question in detail, and will recall only that the diagrams are selected with respect to the parameter $\rho\lambdabar^3 \ll 1$. The expression obtained for the function $\langle|\varphi_j(t)|^2\rangle$ depends on the angles between the directions of $\mathbf{r}_j$, $\mathbf{M}^j$, and $\mathbf{M}^{j_0}$. Since the medium is assumed isotropic, we shall average over the directions of $\mathbf{M}^j$ and $\mathbf{M}^{j_0}$. This averaging will be denoted by a superior bar. After this, $\langle|\varphi_j(t)|^2\rangle$ depends only on $|\mathbf{x}_j - \mathbf{x}_{j_0}|$, therefore we omit the index j of $\mathbf{r}_j$ and introduce the function

$$n(r, t) = \langle\overline{|\varphi_j(t)|^2}\rangle \tag{12}$$

For the function $n(\mathbf{r}, t)$ we have

$$n(r, t) = \sum_{\varkappa} e^{i\varkappa\mathbf{r}} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} e^{-i\omega_1 t} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} (-1)(\omega - \omega_j + i\gamma/2)^{-1}$$

$$\times (\omega - \omega_j - \omega_1 - i\gamma/2)^{-1} (\omega - \omega_{j_0} + i\gamma/2)^{-1} (\omega - \omega_{j_0}$$

$$-\omega_1 - i\gamma/2)^{-1} \sum_{\mathbf{k}\lambda\lambda'} \frac{1}{3} Q_{\mathbf{k}+\varkappa/2} \cdot Q_{\mathbf{k}-\varkappa/2} (\mathbf{l}^\lambda_{\mathbf{k}+\varkappa/2}\mathbf{l}^{\lambda'}_{\mathbf{k}-\varkappa/2}) K_{\lambda\lambda'}(\mathbf{k}$$

$$+ \varkappa/2, \mathbf{k} - \varkappa/2, \omega, \omega_1), \tag{13}$$

where

$$Q_{\mathbf{k}} = \left(\frac{2\pi}{\omega_k V}\right)^{1/2} |\mathbf{M}|.$$

This expression does not include as yet the averaging over the frequencies ω_j and ω_{j_0}. Such an averaging will be made in the concrete calculation of the function n(**r**, t). The function $K_{\lambda\lambda'}(\mathbf{k}+\boldsymbol{\kappa}/2, \mathbf{k}-\boldsymbol{\kappa}/2, \omega, \omega_1)$ satisfies the integral equation

$$K_{\lambda\lambda'}(\mathbf{k}+\boldsymbol{\varkappa}/2, \mathbf{k}-\boldsymbol{\varkappa}/2, \omega, \omega_1)$$

$$= \frac{1}{3} Q_{\mathbf{k}+\varkappa/2} Q_{\mathbf{k}-\varkappa/2} (\mathbf{l}^{\lambda}_{\mathbf{k}+\varkappa/2} \mathbf{l}^{\lambda'}_{\mathbf{k}-\varkappa/2}) \cdot K^{(0)} (\omega_{\mathbf{k}+\varkappa/2}, \omega_{\mathbf{k}-\varkappa/2}, \omega, \omega_1)$$

$$+ \frac{N}{15} Q_{\mathbf{k}+\varkappa/2} Q_{\mathbf{k}-\varkappa/2} (\omega_1 + i\gamma)^{-1} \cdot K^{(0)} (\omega_{\mathbf{k}+\varkappa/2}, \omega_{\mathbf{k}-\varkappa/2}, \omega, \omega_1)$$

$$\times [D(\beta; \omega - \omega_0) + D(\beta; \omega_1 - \omega + \omega_0)]$$

$$\times \sum_{\mathbf{k}_1\lambda_1\lambda_1'} Q_{\mathbf{k}_1+\varkappa/2} Q_{\mathbf{k}_1-\varkappa/2} [(\mathbf{l}^{\lambda}_{\mathbf{k}+\varkappa/2} \mathbf{l}^{\lambda'}_{\mathbf{k}-\varkappa/2}) (\mathbf{l}^{\lambda_1}_{\mathbf{k}+\varkappa/2} \mathbf{l}^{\lambda_1'}_{\mathbf{k}_1-\varkappa/2})$$

$$+ (\mathbf{l}^{\lambda}_{\mathbf{k}+\varkappa/2} \mathbf{l}^{\lambda_1}_{\mathbf{k}_1+\varkappa/2}) \cdot (\mathbf{l}^{\lambda'}_{\mathbf{k}-\varkappa/2} \mathbf{l}^{\lambda_1'}_{\mathbf{k}_1-\varkappa/2}) + (\mathbf{l}^{\lambda}_{\mathbf{k}+\varkappa/2} \mathbf{l}^{\lambda_1'}_{\mathbf{k}_1-\varkappa/2}) (\mathbf{l}^{\lambda'}_{\mathbf{k}-\varkappa/2} \mathbf{l}^{\lambda_1}_{\mathbf{k}_1+\varkappa/2})]$$

$$\times K_{\lambda_1\lambda_1'} (\mathbf{k}_1 + \varkappa/2, \mathbf{k}_1 - \boldsymbol{\varkappa}/2, \omega, \omega_1) \tag{14}$$

We have introduced here the notation

$$K^{(0)}(\omega_k, \omega_{k'}, \omega, \omega_1) = \left[\omega - \omega_k - \frac{D(\beta; \omega - \omega_0)}{4\tau^2}\right]^{-1}$$

$$\times \left[\omega_1 - \omega + \omega_{k'} - \frac{D(\beta; \omega_1 - \omega + \omega_0)}{4\tau^2}\right]^{-1}; \tag{15}$$

$$D(\beta; \omega) = -\frac{i\sqrt{\pi}}{\beta}\left[1 - \Phi\left(\frac{\xi/2 + \omega/i}{\beta}\right)\right]$$

$$\times \exp\left[\frac{(\xi/2 + \omega/i)^2}{\beta^2}\right], \qquad \xi = \gamma + \delta. \tag{16}$$

The function $\Phi(x)$ is the usual probability integral[12]. The function $K_{\lambda\lambda'}(\mathbf{k}+\boldsymbol{\kappa}/2, \mathbf{k}-\boldsymbol{\kappa}/2, \omega, \omega_1)$ has the meaning of the photon density matrix (apart from a multiplier). This is seen from the fact that when $\kappa = 0$ and $\lambda = \lambda'$ Eq. (14) goes over into the equation for the function determining the probability of finding a photon with momentum **k** and polarization λ in such a system[8,13].

Equation (14) is solved accurately only for $\kappa = 0$. In the general case the solution of (14) is obtained approximately. Use is made here of the fact that the most effective in the sums of (13) are those values of κ and k for which

$$\varkappa/k \ll 1 \quad (k \sim \omega_0/c; \quad \varkappa \sim 1/r; \quad \varkappa/k \sim \bar{\lambda}/r \ll 1).$$

After substituting the obtained solution in (13) and summing over **k**, λ, and λ' we obtain the following expression for the function n(r, t):

$$n(r, t) = \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} e^{-i\omega_1 t} \int_{-\infty}^{\infty} \frac{d\omega_\varkappa}{2\pi} e^{i\omega_\varkappa r/c} n(\omega_\varkappa, \omega_1), \tag{17}$$

$$n(\omega_\varkappa, \omega_1) = \frac{i\gamma^2}{8(\omega_0 r/c)^2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} (\omega - \omega_j + i\gamma/2)^{-1} (\omega - \omega_j$$

$$- \omega_1 - i\gamma/2)^{-1} \cdot (\omega - \omega_{j_0} + i\gamma/2)^{-1} (\omega - \omega_{j_0} - \omega_1$$

$$- i\gamma/2)^{-1} \left\{ (\omega_1 - \omega_\varkappa)(\omega_1 + i\gamma) - \frac{\omega_1}{4\tau^2} [D(\beta; \omega - \omega_0)\right.$$

$$\left. + D(\beta; \omega_1 - \omega - \omega_0)] \right\}^{-1}, \tag{18}$$

where $\omega_\kappa = c\kappa$. The factor r^{-2} has already been separated in the expression for $n(\omega_\kappa, \omega_1)$. This is connected with the fact that only the terms that attenuate least when $r \to \infty$ have been retained in n(r, t). Actually we have again expanded here in the parameter $\bar{\lambda}/r \ll 1$.

3. CASE OF DISPERSION SCATTER OF THE LEVELS

The integration in formulas (17) and (18) cannot be carried out for arbitrary values of δ and β. The calculations can be carried through to conclusion only when the scatter of the molecular levels of the solid molecules is determined by the dispersion formula ($\beta \to 0$). The distribution of the transition frequencies for the excited molecules can be arbitrary, but we assume for simplicity that it is the same as for the unexcited ones. Averaging (18) over the frequencies ω_j and ω_{j_0} with the function $\chi_1(\omega_j - \omega_0)$ (with $\beta \to 0$, and $\chi(\omega_j - \omega_0) \to \chi_1(\omega_j - \omega_0)$, and integrating with respect to ω, we obtain

$$n(\omega_\varkappa, \omega_1) = \frac{\gamma^2\tau^2}{2(\omega_0 r/c)^2} \omega_1^{-1} (\omega_1 + i\gamma)^{-1}$$

$$\times \left\{\left[1 - \frac{\omega_1}{\tau^2(\omega_1 - \omega_\varkappa)(\omega_1 + i\xi)(\omega_1 + i\gamma)}\right]^{-1/2} - 1\right\}. \tag{19}$$

The expression obtained for $n(\omega_\kappa, \omega_1)$ is substituted in (17) and reduced, after integration with respect to ω_κ, to the following expression for the function n(r, t):

$$n(r, t) = \frac{\gamma^2}{4(\omega_0 r/c)^2} \int_{-i\infty+\sigma}^{i\infty+\sigma} \frac{dp}{2\pi i} \frac{e^{p(t-r/c)}}{(p+\xi)(p+\gamma)^2}$$

$$\times \exp\left[-\frac{pr}{2c\tau^2(p+\gamma)(p+\xi)}\right] \left\{ I_0\left(\frac{pr}{2c\tau^2(p+\gamma)(p+\xi)}\right)\right.$$

$$\left. - I_1\left(\frac{pr}{2c\tau^2(p+\gamma)(p+\xi)}\right)\right\}.$$

It is impossible to carry out the integration in (20) for an arbitrary instant of time. This is not important in principle, since the function n(r, t) itself is not measured in the experiments. We note that the function n(r, t) determines the probability that a molecule will be excited at the point r at the instant t. The function of the density of the excited molecules can be obtained by multiplying n(r, t) by ρ. It follows therefore, in particular, that when

$\rho \to 0$ the density of the excited molecules also tends to zero, whereas n(r, t) tends to a finite limit, which can be readily obtained from (20):

$$n(r,t) = \frac{\gamma^2 e^{-\gamma(t-r/c)}}{4\delta(\omega_0 r/c)^2}\left[t - \frac{r}{c} + \frac{1}{\delta}\left(e^{-\delta(t-r/c)} - 1\right)\right],$$

$$t - r/c > 0, \quad n(r,t) = 0, \qquad t - r/c < 0. \tag{21}$$

For the case $t - r/c \ll 1/\gamma$, $1/\delta$ we can obtain (21) directly from general considerations[14].

Let us consider the behavior of the function n(r, t) for small values of the time, when

$$\xi(t - r/c) \ll 1.$$

In this case formula (20) yields

$$n(r,t) = \frac{\gamma^2\tau^2(t - r/c)}{2\omega_0^2 (r/c)^3} J_1^2\left(\sqrt{\frac{r(t - r/c)}{c\tau^2}}\right), \quad t - r/c > 0. \tag{22}$$

We see therefore directly that the density of the excited molecules also experiences oscillations with a characteristic time τ, just as the photon density in such a medium[6-8].

Let us calculate $\langle r^2(t)\rangle$—the mean-square path traversed by the excitation in such a medium by the instant t. It is necessary to note here the following. As seen from[8,18], the probability of observing a photon in the medium differs from zero for any instant of time $t > 0$. This indicates that actually we always have only a partial dragging of the radiation. The parameter which determines the degree of dragging of the radiation is the quantity $\xi\gamma\tau^2$. If $\xi\gamma\tau^2 \gg 1$, i.e., the medium is highly rarefied or the scatter of the levels is very large, the principal part of the excitation is carried by the free photons, as can be readily verified by investigating the results of Vdovin[15] for $\sum_{k\lambda} |f_\lambda(k, t)|^2$. In this case the fraction of the excitation which is carried by the absorption and reradiation processes is of the order of $(\xi\gamma\tau)^{-1} \ll 1$.

This result can also be obtained from the following considerations. The time that the quantum spends in the atom is $\tau_a \sim 1/\gamma$. The time during which it exists as a free quantum is $\tau \sim (\rho c\sigma)^{-1}$, where $\sigma \sim \pi\lambda\!\!\!^{-2}\gamma/\xi$. Then the fraction of the excitation transported by the absorption and re-radiation processes is of the order of

$$\tau_a / (\tau_\gamma + \tau_a) \sim (\xi\gamma\tau^2)^{-1}.$$

The radiation is strongly dragged when the $\xi\gamma\tau^2 \ll 1$. In this case almost all the excitation propagates as a result of absorption and re-radiation acts. The fraction of the excitation, which in this case is carried by the free photons, is of the order of $(\xi\gamma\tau^2)^{1/2} \ll 1$. Therefore the formulas presented above have a real meaning only for the case $\xi\gamma\tau^2 \ll 1$.

In the case of greatest interest $\gamma t \gg 1$, the expression for $\langle r^2(t)\rangle$ is of the form

$$\langle r^2(t)\rangle = \int \rho\, d\mathbf{r}\, n(r,t) r^2 \approx \frac{c^2 t^2}{8}(\xi\gamma\tau^2)^{1/2}. \tag{23}$$

Let us now discuss which quantity should be regarded as the time needed for the excitation to leave the system. In the literal sense of the problem as here formulated, no such quantity can be introduced, since the problem is solved either for an unbounded medium or for a medium contained in a box with ideally reflecting walls. One can speak of a departure time in this case only in the following arbitrary sense, as is done, for example, in the paper by Veklenko[16]. We separate mentally a sphere of radius R around an initially excited molecule, and define as the time of departure of the excitation from the system as the time of departure of the radiation from the given sphere. It is clear that the departure time defined in this manner will always exceed the corresponding departure time for a bounded medium contained in a sphere of the same radius R, owing to the reaction of the molecules situated outside the sphere.

It follows from the experimental papers[17-19] that the experimentally measured quantity is J(R, t) (or its time derivative), which determines the total probability that the radiation will leave the volume by the instant t. Knowledge of n(r, t) enables us to calculate this quantity directly:

$$J(R,t) = 1 - e^{-\gamma t} - 4\pi\rho \int_0^R dr r^2 n(r,t). \tag{24}$$

The time of departure of the radiation from the system is determined from a reduction of the plot of J(R, t) (or $\partial J(R/t)/\partial t$)[17]. At present, however, we know of no experiments in which the dispersion width is much larger than the Doppler width.

4. KINETIC EQUATION FOR THE FUNCTION n(r, t)

We now construct the kinetic equation which is satisfied by the function n(r, t), for the case of the dispersion broadening. We recall that n(r, t) does not describe the behavior of the initially excited molecule, which is therefore an external source with respect to the remaining molecules. Consequently, the equation for the obtained function n(r, t) will be inhomogeneous, unlike the phenomenological equations of Holstein[3] and Biberman[4]. The equation obtained by quantum mechanical means in the paper of D'yakonov and Perel'[5] for the function n(r, t) is in general inhomogeneous, the inhomogeneous term describing just the action of the excitation source.

Let us write down the connection between the function n(r, t) and $n(\omega_\kappa, \omega_1)$:

$$n(\mathbf{r}, t) = \int \frac{d\varkappa}{(2\pi)^3} e^{i\varkappa\mathbf{r}} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} e^{-i\omega_1 t} n(\omega_\varkappa, \omega_1), \tag{25}$$

$$n(\omega_\varkappa, \omega_1) = \int d\mathbf{r}\, e^{-i\varkappa\mathbf{r}} \int_0^\infty dt\, e^{-pt} n(\mathbf{r}, t) \qquad (p = -i\omega_1) \tag{26}$$

Comparing expressions (25) and (17), we can easily obtain from (19) the following expression for $n(\omega_\kappa, \kappa_1)$:

$$n(\omega_\varkappa, \omega_1) = \frac{\gamma}{\rho} \frac{1}{\omega_1} \frac{1}{\omega_1 + i\gamma}$$
$$\times \left\{ \left[1 - \frac{\omega_1}{\tau^2 (\omega_1 - \omega_\varkappa)(\omega_1 + i\xi)(\omega_1 + i\gamma)} \right]^{-1/2} - 1 \right\}. \tag{27}$$

We now write down the algebraic identity

$$-i\omega_1 n(\omega_\varkappa, \omega_1) + \gamma n(\omega_\varkappa, \omega_1)$$
$$= -\gamma \left\{ \left[1 - \frac{1}{\tau^2 (\omega_1 - \omega_\varkappa)(\omega_1 + i\xi)} \right]^{-1/2} - 1 \right\}$$
$$\times\, n(\omega_\varkappa, \omega_1) + \frac{\gamma}{i\rho\omega_1(\omega_1 + i\gamma)}$$
$$\times \left\{ \left[1 - \frac{\omega_1}{\tau^2 (\omega_1 - \omega_\varkappa)(\omega_1 + i\xi)(\omega_1 + i\gamma)} \right]^{-1/2} - 1 \right\}$$
$$\times \left\{ \omega_1 + i\gamma \left[1 - \frac{1}{\tau^2 (\omega_1 - \omega_\varkappa)(\omega_1 + i\xi)} \right]^{-1/2} \right\}. \tag{28}$$

The transition in formula (28) from ω_κ and ω_1 to the variables r and t by means of formula (25), and replacing $n(\omega_\kappa, \omega_1)$ in the right side of (28) by expression (26), we arrive at the following equation:

$$\frac{\partial n(r, t)}{\partial t} + \gamma n(r, t) = \gamma \int_0^\infty dt' \int d\mathbf{r}'\, G(|\mathbf{r} - \mathbf{r}'|, t - t') n(r', t)$$
$$+ F(r, t), \tag{29}$$

where

$$G(r, t) = \frac{e^{-\xi(t - r/c)}}{8\pi c \tau^2 r^2} \left[J_0^2 \left(\sqrt{\frac{r(t - r/c)}{c\tau^2}} \right) \right.$$
$$\left. - J_1^2 \left(\sqrt{\frac{r(t - r/c)}{c\tau^2}} \right) \right], \qquad t - r/c > 0;$$

$$G(r, t) = 0, \qquad t - r/c < 0. \tag{30}$$

Since the concrete form of F(r, t) is connected with the given choice of the initial conditions, it is meaningless to present here the explicit expression for F(r, t).

We note that the kernel of the obtained equation depends strongly on the time. The transition to the Holstein equation is carried out in the same manner as in the paper of D'yakonov and Perel'[5], by replacing G(r, t) by $\tilde{G}(r)\delta(t - r/c)$, where

$$\tilde{G}(r) = \int_0^\infty dt\, G(r, t).$$

Using (30), we obtain

$$\tilde{G}(r) = \frac{k_d e^{-k_d r/2}}{8\pi r^2} \{ I_0(k_d r/2) - I_1(k_d r/2) \},$$
$$k_d = (c\xi\tau^2)^{-1}. \tag{31}$$

The kernel $\tilde{G}(r)$ coincides fully with the corresponding kernel of Holstein's paper[3]. For complete agreement with Holstein's equation[3] it is necessary also to neglect the delay and, naturally, leave out F(r, t). The neglect of the retardation is valid if $\gamma R/c \ll 1$, since the characteristic time of variation of the function n(r, t) is of the order of $1/\gamma$ (R is the linear dimension of the system). This criterion was first indicated in[3].

Integration with respect to time in (29) takes into account the contribution made to the change in the density of the excited atoms, at the point r at the instant t, not only by the quanta received from other volume elements at the given instant of time, i.e., for which $\Delta t = t - t' - |\mathbf{r} - \mathbf{r}'|/c = 0$, but also of quanta arriving earlier, for which $\Delta t > 0$. It is clear that the contribution of such quanta to the excitation decreases by the instant t, this being reflected in the factor $\exp(-\xi\Delta t)$ in the kernel. From (30) we see that the kernel experiences oscillations with a characteristic time τ. This is apparently due to exchange of excitations with the free quanta, whose density experiences similar oscillations.

The function n(r, t) has two characteristic times, within which it changes appreciably. One is the time of the rapid oscillations and is of the order of τ, while the other is the characteristic damping time of the function and is of the order of $1/\gamma$. Since $\gamma\tau \ll 1$, the transition to the kernel (31) denotes, probably, an averaging over a time interval Δt, with $\tau \ll \Delta t \ll 1/\gamma$. Thus, the transition to the equation given by Holstein[3] and Biberman[4] is equivalent in a certain sense to the approximation of slowly varying amplitudes, which is usually used in investigations of processes involving the oscillation of the photon density in the resonant medium[20].

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