# PROPAGATION OF RADIATION IN A RESONANT MEDIUM

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A graph technique is developed for studying kinetic problems connected with the propagation of radiation in a resonant medium with $n\lambda^3 \ll 1$. A kinetic equation is set up for the density matrix of excited atoms, assuming an arbitrary relation between the Doppler and natural line widths. The shape of the spectral line due to radiation diffusion is investigated.

IN the present paper we investigate the propagation of electromagnetic excitations in a gaseous resonant medium. In a previous paper of one of the authors[1] the spatial distribution of the excited atoms was considered, neglecting their thermal motion. In the present paper we consider a gaseous medium consisting of a set of two-level atoms (molecules). The medium is assumed to be sufficiently dilute so that the condition $n\lambda^3 \ll 1$ is fulfilled, where n is the density of atoms, and $\lambda$ is the wavelength of the resonance radiation ($\lambda = c/\omega_0$, where $\omega_0$ is the energy difference of the levels of the atoms). Under these conditions one can neglect the resonant dipole-dipole interaction between the atoms.[2] Then the interaction between the atoms reduces to the absorption and re-emission of quanta, and the propagation of an excitation in the medium can be treated in terms of the density of excited atoms.

The term "radiation diffusion" which is commonly used in the description of the propagation of such a "captive" radiation does not truly reflect the essence of the process, since the diffusion approximation[3] is insufficient. This was first pointed out by Biberman[4] and Holstein.[5] In these papers a phenomenological formulation of the kinetic equation for the density of excited atoms was given. The kinetic equation for the density of excited atoms was obtained by a quantum mechanical approach by D'yakonov and Perel',[6] using a graph technique developed earlier by Konstantinov and Perel'.[7]

In Sec. 1 of the present paper we develop a graph technique particularly adapted to the consideration of kinetic problems connected with the propagation of an excitation in a resonant medium. In Sec. 2, the rules of this technique are used to set up a kinetic equation for the density matrix for the excited atoms $f_{mm'}(\mathbf{r}, \mathbf{p}, t)$, where m and m′ are the quantum numbers for the projection of the total angular momentum of the upper level on an arbitrarily oriented axis. The kinetic equation is obtained for an arbitrary relation between $\gamma$ (the natural line width) and $\beta$ (the Doppler width), taking account of the retardation, in contrast to the case considered in [6] ($\beta \gg \gamma$, retardation neglected). In Sec. 3 a kinetic equation is constructed for the density of quanta in such a medium, and the shape of the line is found which results when the radiation is adiabatically emitted from the volume. The results obtained for the shape and width of the spectral line are compared with the experimental data of Tomiser.[8]

## 1. GRAPH TECHNIQUE FOR PROBLEMS CONNECTED WITH THE PROPAGATION OF AN EXCITATION IN A RESONANT MEDIUM

The Hamiltonian for the medium and the radiation field in the volume V is written in the form

$$H = \sum_{\mathbf{p}\mu} \frac{p^2}{2M} a^+_{\mathbf{p}\mu} a_{\mathbf{p}\mu} + \sum_{\mathbf{p}m} \left(\frac{p^2}{2M} + \omega_0\right) b^+_{\mathbf{p}m} b_{\mathbf{p}m} + \sum_{\mathbf{k}\lambda} \omega_k c^+_{\mathbf{k}\lambda} c_{\mathbf{k}\lambda} + \sum_{\substack{\mathbf{p}m\mu \\ \mathbf{k}\lambda}} [Q^{m\mu}_{\mathbf{k}\lambda} c_{\mathbf{k}\lambda} b^+_{\mathbf{p}m} a_{\mathbf{p}-\mathbf{k}\mu} + Q^{m\mu *}_{\mathbf{k}\lambda} c^+_{\mathbf{k}\lambda} b_{\mathbf{p}m} a^+_{\mathbf{p}-\mathbf{k}\mu}]. \quad (1)$$

Here the indices $\mu$ and m characterize the projections of the total angular momentum of the lower and upper levels (the angular momentum of the lower level is $j_0$, that of the upper level, $j_1$; we assume that a dipole transition is allowed between these levels); the operators $a^+_{\mathbf{p}\mu}$, $a_{\mathbf{p}\mu}$ and $b^+_{\mathbf{p}m}$, $b_{\mathbf{p}m}$ are creation and annihilation operators for the unexcited and excited atoms; the operators $c^+_{\mathbf{k}\lambda}$, $c_{\mathbf{k}\lambda}$ are creation and annihilation operators for quanta with the momentum $\mathbf{k}$ and polarization $\lambda$ ($\lambda = 1, 2$); $\omega^2_{\mathbf{k}} = c^2\mathbf{k}^2$;

$$Q_{\mathbf{k}\lambda}{}^{m\mu} = \left(\frac{2\pi}{\omega_k V}\right)^{1/2} (\mathbf{M}_\mu{}^{m*} \mathbf{l}_{\mathbf{k}}{}^{\lambda}), \quad (2)$$

where $\mathbf{M}^m_\mu$ is the matrix element for a dipole transition in an isolated atom from the lower level characterized by the index $\mu$ to the upper level (index m) accompanied by the absorption of a quantum; $l^\lambda_{\mathbf{k}}$ is the polarization vector of the quantum ($\hbar = 1$).

In the following we consider the case of weakly excited states, where the density of excited atoms is much smaller than the density of unexcited atoms. Then the state of the unexcited atoms remains practically unchanged, and the average number of unexcited atoms with projection of total angular momentum $\mu$ is

$$n_\mu(\mathbf{p}) = \frac{n}{(2j_0 + 1)} \varphi(\mathbf{p}), \quad (3)$$

where $n = N/V$ is the average density of atoms and $\varphi(\mathbf{p})$ is the Maxwell distribution function. The gas is assumed to be nondegenerate, i.e., the average occupation numbers $n(\mathbf{p}) \ll 1$. For definiteness, we assume that the operators $a_{\mathbf{p}\mu}$ and $b_{\mathbf{p}m}$ are boson operators, although this is of no significance in the present circumstances.

We denote a state where no excited atoms or quanta are present by $|0, 0, s\rangle$, where s is the state of unex-

cited atoms. The derivation of the rules for the construction of graphs will be illustrated by a concrete example, where a molecule with momentum $\mathbf{p}_0$ and polarization $m_0$ is excited at the initial moment t = 0. The state vector of the system at t = 0 has the form

$$\Phi(t=0) = b^{+}_{\mathbf{p}_0 m_0}|0,0,s\rangle. \tag{4}$$

Then the formal solution of the Schrödinger equation with the Hamiltonian (1) is

$$\Phi(t) = e^{-iHt} b^{+}_{\mathbf{p}_0 m_0}|0,0,s\rangle.$$

Let us ask for the probability of observing at time t an excited molecule with momentum $\mathbf{p}$ and polarization m. Since the Hamiltonian (1) conserves the total number of excitations, the amplitude for this probability is given by the expression

$$\langle s',0,0|b_{\mathbf{p}m}e^{-iHt}b^{+}_{\mathbf{p}_0 m_0}|0,0,s\rangle.$$

For the probability summed over the final states of the unexcited atoms and averaged over their initial states with the density matrix

$$\rho = \exp\left[\frac{1}{T}(\Omega + \mu N - H_{\text{av}})\right], \qquad H_{\text{av}} = \sum_{\mathbf{p}\mu}\frac{p^2}{2M}a_{\mathbf{p}\mu}{}^{+}a_{\mathbf{p}\mu}, \tag{5}$$

we then obtain

$$f_m(\mathbf{p},\mathbf{p}_0,m_0,t) = \sum_{ss'}\exp\left[\frac{1}{T}(\Omega+\mu N - H_{\text{cp}})\right]\cdot$$

$$\cdot\langle s,0,0|b_{\mathbf{p}_0 m_0}e^{iHt}b^{+}_{\mathbf{p}m}|0,0,s'\rangle\langle s',0,0|b_{\mathbf{p}m}e^{-iHt}b^{+}_{\mathbf{p}_0 m_0}|0,0,s\rangle. \tag{6}$$

Let us go over to Heisenberg operators in (6) and introduce the single-particle Green's functions

$$\hat{G}_{mm'}(\mathbf{p},\mathbf{p}',t-t') = -i\langle 0,0|T\{b_{\mathbf{p}m}(t)b^{+}_{\mathbf{p}'m'}(t')\}|0,0\rangle, \tag{7}$$

which are operators in the variables of the unexcited atoms. Analogous operator Green's functions were introduced by Galitskiĭ and Yakimets.[9] Then we obtain for $f_m(\mathbf{p}, \mathbf{p}_0, m_0, t)$

$$f_m(\mathbf{p},\mathbf{p}_0,m_0,t) = \{\langle s|\hat{G}^{*}_{mm_0}(\mathbf{p},\mathbf{p}_0,t)\hat{G}_{mm_0}(\mathbf{p},\mathbf{p}_0,t)|s\rangle\}_s, \tag{8}$$

where $\{\ldots\}_s$ denotes the average over the states of the unexcited atoms with the density matrix 5). We note that $\hat{G}_{mm'}(\mathbf{p},\mathbf{p}',t) = 0$ for $t < 0$, so that the distribution function (8) has meaning only for positive times. This property of the Green's functions allows one to apply a Laplace transformation in the time. In the following, we denote the Laplace variable by $\omega$ ($\omega = ip$, where p is the usual Laplace variable). The rules of the graph technique for the distribution function are obtained by the usual procedure, i.e., by going over to the interaction picture and expanding the S matrix in a series in $H_{\text{int}}$. The interaction Hamiltonian $H_{\text{int}}$ is the expression inside the square brackets in (1). As a result we obtain the following rules for writing down analytic expressions for the graphs and for the construction of the graphs.

Three lines enter at each vertex: a solid line, depicting the propagation of excited atoms, a wavy line, depicting the propagation of quanta, and a dotted line, depicting the propagation of unexcited atoms. The solid and wavy lines have a definite direction: in the graphs for the Green's function $\hat{G}_{mm'}$ they are directed from left to right in the direction of increasing time; for the function $\hat{G}^{*}_{mm'}$, they are directed from right to left. Some graphs referring to the operator functions $\hat{G}_{mm'}$ and $\hat{G}^{*}_{mm'}$ have the form

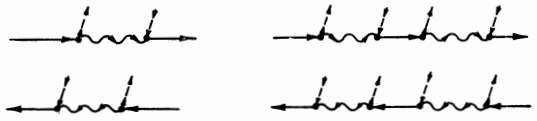
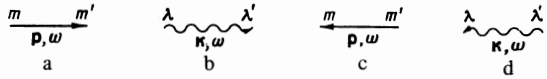


FIG. 1.

In these functions we have not yet averaged over the operators $a_{\mathbf{p}\mu}$, $a^{+}_{\mathbf{p}\mu}$. This averaging is carried out when the distribution function (8) is calculated, and leads to the appearance of dotted lines connecting two vertices at the ends of the same or different lines. An "upper line" is a line going from left to right and consisting of solid and wavy lines, i.e., a line characterizing the function $\hat{G}_{mm'}$. Analogously, a "lower line" is a line characterizing the function $\hat{G}^{*}_{mm'}$.

The solid and wavy lines




a b c d

FIG. 2.

correspond to the following analytic expressions:

a) $G^{(0)}_{mm'}(\mathbf{p},\omega) = \delta_{mm'}(\omega - \omega_0 - \varepsilon_p)^{-1}$;

b) $D^{(0)}_{\lambda\lambda'}(\mathbf{k},\omega) = \delta_{\lambda\lambda'}(\omega - \omega_k)^{-1}$;

c) $G^{(0)+}_{mm'}(\mathbf{p},\omega) = -\delta_{mm'}(\omega + \omega_0 + \varepsilon_p)^{-1}$;

d) $D^{(0)+}_{\lambda\lambda'}(\mathbf{k},\omega) = -\delta_{\lambda\lambda'}(\omega + \omega_k)^{-1}$.

Each dotted line is characterized by a momentum $\mathbf{p}$, an energy $\varepsilon_p = p^2/2M$ and polarizations $\mu$ and $\mu'$. If this line connects vertices belonging either only to an upper or a lower line, and is "regular," i.e., its direction coincides with the direction of the corresponding solid and wavy lines, then it corresponds to the factor $\delta_{\mu\mu'}$. If it is "irregular," i.e., its direction is opposite to the direction of the corresponding solid and wavy lines, then it corresponds to the factor

$$\frac{n}{(2j_0+1)}\varphi(\mathbf{p})\delta_{\mu\mu'}. \tag{9}$$

The dotted lines which run from an upper line to a lower line are regarded as "regular" and those running from a lower to an upper line as "irregular." In the vertex we write $Q^{m\mu *}_{\mathbf{k}\lambda}$ if a wavy line leaves from it, and $Q^{m\mu}_{\mathbf{k}\lambda}$ if a wavy line enters in it. There are in all four types of vertices. At each vertex momentum is conserved: the momentum of the solid line is equal to the sum of the momenta of the wavy and dotted lines. At each vertex the frequency is conserved (for a dotted line, the role of frequency is played by the energy $\varepsilon_p$). In the vertices of an upper line the frequency of the solid line is equal to the sum of the frequencies of the wavy and dotted lines, and in the vertices of the lower lines the

frequency of the solid line is equal to the difference of the frequencies of the wavy and dotted lines. We sum over all intermediate values $\mathbf{p}$, $\mathbf{k}$, $\lambda$, m, and $\mu$ and integrate over the intermediate frequencies $\left(\int_{-\infty}^{\infty} d\omega \ldots\right)$.

## 2. KINETIC EQUATION FOR THE DENSITY OF EXCITED ATOMS

We consider a function describing the space-momentum distribution of the excited atoms for given initial conditions. The graphical form of this distribution function $f_{mm'}(\mathbf{p}, \kappa, \omega, \tilde{\omega})$ is shown in Fig. 3.

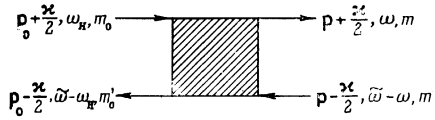



FIG. 3.

For brevity of writing we have here omitted the dependence of the function $f_{mm'}$ on the initial values $\mathbf{p}_0$, $m_0$, $m_0'$, and $\omega_H$. The distribution function in the variables $\mathbf{r}$, t is related to $f_{mm'}(\mathbf{p}, \kappa, \omega, \tilde{\omega})$ by

$$f_{mm'}(\mathbf{p}, \mathbf{r}, t) = \sum_{\kappa} e^{i\kappa r} \int_{-\infty}^{\infty} \frac{d\tilde{\omega}}{2\pi} e^{-i\tilde{\omega}t} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} f_{mm'}(\mathbf{p}, \kappa, \omega, \tilde{\omega}). \quad (10)$$

The diagonal elements of the matrix $f_{mm'}$ describe the population of the sublevels with a given projection of the angular momentum. Therefore the total density of excited atoms, which must be compared with the function introduced by Holstein,[5] is given by the sum of the diagonal elements of this matrix:

$$n(\mathbf{r}, \mathbf{p}, t) = \mathrm{Sp}\, f_{mm'}(\mathbf{p}, \mathbf{r}, t). \quad (11)$$

For definiteness, we assume $j_0 = 0$, $j_1 = 1$ in the further calculations. Analogous computations can be carried out for other values of the angular momenta; for example, in Sec. 3 we consider transitions with $j_0 = 0$, $j_1 = 1/2$, and $j_1 = 3/2$. The last case corresponds to the doublets in alkaline metals.

We now write down an equation for the function $f_{mm'}(\mathbf{p}, \kappa, \omega, \omega)$. Its graphical form is shown in Fig. 4.

Analytically, it can be written in the following form:

$$f_{mm'}(\mathbf{p}, \kappa, \omega, \tilde{\omega}) = G_{mm_0}(\mathbf{p}+\kappa/2, \omega) G^{+}_{m'm_0'}(\mathbf{p}-\kappa/2, \tilde{\omega}-\omega)\delta_{\mathbf{p}, \mathbf{p}_0} + G_{mm_2}(\mathbf{p}+\kappa/2, \omega) G^{+}_{m'm_2'}(\mathbf{p}-\kappa/2, \tilde{\omega}-\omega) \cdot \sum_{\mathbf{p}_1} \int d\omega_1 U^{m_1 m_1'}_{m_2 m_2'}(\mathbf{p}, \mathbf{p}_1, \kappa, \omega, \omega_1, \tilde{\omega}) f_{m_1 m_1'}(\mathbf{p}_1, \kappa, \omega_1, \tilde{\omega}). \quad (12)$$

Here a summation over identical indices m is implied.

All solid lines in this equation are "heavy," i.e., correspond to non-operator Green's functions which appear through connecting the operators of the unexcited atoms at the ends of the same line. The Dyson equation for the function $G_{mm'}(\mathbf{p}, \omega)$ with the simplest self-energy part $\Sigma_{m_1 m_1'}(\mathbf{p}, \omega)$ is shown in Fig. 5.

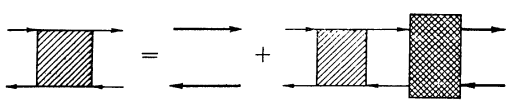


FIG. 4.

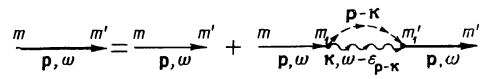



FIG. 5.

All corrections connected with "converting the intermediate photon line into a heavy line" and with the inclusion of more complicated irreducible graphs for the self-energy part $\Sigma_{mm'}$ are proportional to the small parameter $n\lambda^3$. Taking only the imaginary part of $\Sigma_{mm'}$ into account (we neglect the radiation shift), we therefore obtain for the function $G_{mm'}(\mathbf{p}, \omega)$

$$G_{mm'}(\mathbf{p}, \omega) = \delta_{mm'}[\omega - \omega_0 - \varepsilon_p + i\gamma/2]^{-1}, \quad (13)$$

where $\gamma$ is the natural line width of the level, which is related to $M^m_\mu$ by

$$\gamma = \frac{4\omega_0}{3c^3} \sum_{\mu} |M^m_\mu|^2. \quad (14)$$

Analogously, we have for the function $G^{+}_{mm'}(\mathbf{p}, \omega)$

$$G^{+}_{mm'}(\mathbf{p}, \omega) = -\delta_{mm'}[\omega - \omega_0 + \varepsilon_p + i\gamma/2]^{-1}. \quad (15)$$

In lowest approximation in the parameter $n\lambda^3$ we obtain for the kernel $U^{m_1 m_1'}_{m_2 m_2'}$ an expression which corresponds to the graph of Fig. 6.

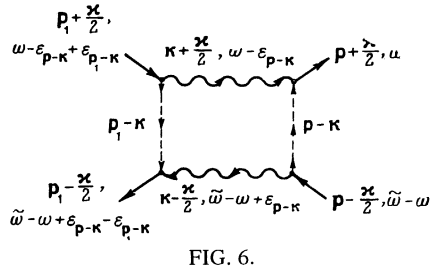



FIG. 6.

The wavy lines in the kernel $U^{m_1 m_1'}_{m_2 m_2'}$ are heavy, i.e., describe the propagation of a quantum in the medium (not in the vacuum). The Dyson equation for the function $D_{\lambda\lambda'}(\mathbf{k}, \omega)$ with a polarization operator with the simplest dependence on the parameter $n\lambda^3$ is shown in Fig. 7.

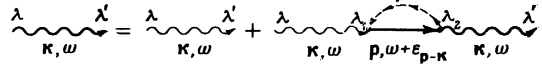



FIG. 7.

Solving this graphical equation, we obtain

$$D_{\lambda\lambda'}(\mathbf{k}, \omega) = \delta_{\lambda\lambda'}[\omega - \omega_k - \Pi(\omega - \omega_0)]^{-1}, \quad (16)$$

where in the present approximation

$$\Pi(\omega) = -\frac{3i\sqrt{\pi}}{4\tau^2\beta}\left\{1 - \Phi\left(\frac{\gamma/2 - i\omega}{\beta}\right)\right\} \exp\left[\frac{(\gamma/2 - i\omega)^2}{\beta^2}\right]. \quad (17)$$

Here $\Phi(x)$ is the usual probability integral,[10] $\beta = v_0\omega_0/c$ is the Doppler width, $v_0 = (2T/M)^{1/2}$, and $\tau$

is a characteristic time for electromagnetic processes in a resonant medium, introduced earlier:[11]

$$\tau^{-2} = 2\pi n \lambda\!\!\!^{-3} \gamma \omega_0. \tag{18}$$

Up to a factor $^3/_4\tau^2$ the function $\Pi(\omega)$ agrees with the function $D(\beta, \omega)$ introduced in [1].

Analogously we calculate the function $D^+_{\lambda\lambda'}(\mathbf{k}, \omega)$ corresponding to a quantum belonging to a lower line:

$$D^+_{\lambda\lambda'}(\mathbf{k}, \omega) = -\delta_{\lambda\lambda'}[\omega + \omega_k - \Pi(\omega + \omega_0)]^{-1}. \tag{19}$$

Let us write down the analytic expression corresponding to Fig. 6. Since $G_{mm'}$ and $G^+_{mm'}$ are proportional to $\delta_{mm'}$, we have

$$\begin{aligned}
&\delta_{mm_1}\delta_{m'm_1'} U^{m_1m_1'}_{m_2m_2'}(\mathbf{p}, \mathbf{p}_1, \varkappa, \omega, \omega_1, \tilde{\omega}) = \\
&= \sum_{\mathbf{k}\lambda\lambda'} Q^{m}_{\mathbf{k}+\varkappa/2,\lambda} Q^{m_1^*}_{\mathbf{k}+\varkappa/2,\lambda} Q^{m'^*}_{\mathbf{k}-\varkappa/2,\lambda'} Q^{m_1'}_{\mathbf{k}-\varkappa/2,\lambda'} n\varphi(\mathbf{p}-\mathbf{k}) \cdot \\
&\cdot [\omega - \omega_{\mathbf{k}+\varkappa/2} - \varepsilon_{\mathbf{p}-\mathbf{k}} - \Pi(\omega - \omega_0 - \varepsilon_{\mathbf{p}-\mathbf{k}})]^{-1} \cdot \\
&\cdot [\tilde{\omega} - \omega + \omega_{\mathbf{k}-\varkappa/2} + \varepsilon_{\mathbf{p}-\mathbf{k}} - \Pi(\tilde{\omega} - \omega + \omega_0 + \varepsilon_{\mathbf{p}-\mathbf{k}})]^{-1}.
\end{aligned} \tag{20}$$

To evaluate the sum $\Sigma_{\mathbf{k}}$ further we make the following simplifications. We use again the effective smallness of the ratio $\kappa/k$,[1] in particular, $\omega_{\mathbf{k}\pm\kappa/2} \approx \omega_{\mathbf{k}} \pm c\kappa q/2$, where $\mathbf{q} = \mathbf{k}/k$. Since, effectively, the momentum of the quantum is small compared to the thermal momentum, $\varphi(\mathbf{p}-\mathbf{k}) \approx \varphi(\mathbf{p})$. The sums over $\mathbf{k}$ and $\mathbf{p}$ are replaced by integrals in the usual way:

$$\sum_{\mathbf{k}} \to \frac{V}{(2\pi)^3} \int d\mathbf{k}.$$

Since the D functions are sharp functions of $\omega_{\mathbf{k}}$, we replace the other functions under the integral by their values for $\omega_{\mathbf{k}} = \omega_0$ in the integration over $\omega_{\mathbf{k}}$, and introduce a function similar to the one introduced in [6]:

$$P^{m_1m_1'}_{mm'}(\omega_0\mathbf{q}) = V^2 \sum_{\lambda\lambda'} Q_{\mathbf{k}\lambda} {}^{m} Q^{m_1^*}_{\mathbf{k}\lambda} Q^{m'^*}_{\mathbf{k}\lambda'} Q^{m_1'}_{\mathbf{k}\lambda'}. \tag{21}$$

In the following, the vector $\mathbf{k}$ will always be interpreted as $\mathbf{k} = \omega_0\mathbf{q}/c$. The product of G functions (without $\delta_{mm'}$) appearing in the inhomogeneous term in (12) and in the integral is written in the following way:

$$\begin{aligned}
&[\omega - \omega_0 - \varepsilon_{p+\varkappa/2} + i\gamma/2]^{-1}[\tilde{\omega} - \omega + \omega_0 + \varepsilon_{p-\varkappa/2} + i\gamma/2]^{-1} \\
&\qquad = (\tilde{\omega} - p\varkappa/M + i\gamma) B(\mathbf{p}, \omega, \tilde{\omega}),
\end{aligned} \tag{22}$$

where

$$\begin{aligned}
B(\mathbf{p}, \omega, \tilde{\omega}) = &\left[\omega - \omega_0 - \varepsilon_p - \frac{\tilde{\omega}}{2} + \frac{\tilde{\omega} + i\gamma}{2}\right]^{-1} \\
&- \left[\omega - \omega_0 - \varepsilon_p - \frac{\tilde{\omega}}{2} - \frac{\tilde{\omega} + i\gamma}{2}\right]^{-1}.
\end{aligned} \tag{23}$$

The function $B(\mathbf{p}, \omega, \tilde{\omega})$ is a sharp function of the variable $\omega$ near the point $\omega = \omega_0 + \varepsilon_p$. In particular, if we disregard the quantum indeterminacy between energy and time, i.e., set $\tilde{\omega} = 0$ in this function, then the width of the distribution in $\omega$ near the point $\omega_0 + \varepsilon_p$ is equal to the natural line width $\gamma$. The widths of the other functions entering in the kernel of the equation are considerably larger than $\gamma$ owing to the integration over $\mathbf{p}$. Therefore the distribution function $f_{mm'}(\mathbf{p}, \kappa, \omega, \tilde{\omega})$ can be written in the form

$$f_{mm'}(\mathbf{p}, \varkappa, \omega, \tilde{\omega}) = B(\mathbf{p}, \omega, \tilde{\omega}) \varphi_{mm'}(\mathbf{p}, \varkappa, \tilde{\omega}), \tag{24}$$

where the function $\varphi_{mm'}$ is independent of $\omega$ (strictly speaking, $\varphi_{mm'}$ is a slowly varying function of $\omega$, so that it can be set equal to its value at the point $\omega = \omega_0 + \varepsilon_p$ in the integration over $\omega$). According to (24) we can interpret the frequency $\omega$ as the energy of the excited atom. Therefore the function $f_{mm'}(\mathbf{p}, \kappa, \omega, \tilde{\omega})$ describes not only the time variation of the space-momentum distribution of the excited atoms but also their energy distribution. The usual distribution function with respect to $\mathbf{r}$ and $\mathbf{p}$,[5,6] which we denote by $f_{mm'}(\mathbf{p}, \kappa, \tilde{\omega})$, is obtained from the function $f_{mm'}(\mathbf{p}, \kappa, \omega, \tilde{\omega})$ by integration over $\omega$. Multiplying both sides of (12) by $-i(\tilde{\omega} - p\kappa/M + i\gamma)$ and going over to the variables $\mathbf{r}$ and $t$ with the help of (10), we obtain the following equation for the distribution function of the excited atoms:

$$\begin{aligned}
&\frac{\partial f_{mm'}(\mathbf{p}, \mathbf{r}, t)}{\partial t} + \mathbf{v}_p \nabla_r f_{mm'}(\mathbf{p}, \mathbf{r}, t) + \gamma f_{mm'}(\mathbf{p}, \mathbf{r}, t) \\
&= \gamma \int_0^\infty dt' \int d\mathbf{p}_1 \int d\mathbf{r}' \sum_{m_1m_1'} K^{m_1m_1'}_{mm'}(\mathbf{r} - \mathbf{r}', \mathbf{p}, \mathbf{p}_1, t - t') f_{m_1m_1'}(\mathbf{p}_1, \mathbf{r}', t').
\end{aligned} \tag{25}$$

The kernel of the equation has the form

$$\begin{aligned}
K^{m_1m_1'}_{mm'}(\mathbf{r}, \mathbf{p}, \mathbf{p}_1, t) = &-\frac{\omega_0^2 n\varphi(\mathbf{p})}{(2\pi)^5 c^4 \gamma r^2} P^{m_1m_1'}_{mm'}(\omega_0\mathbf{q}) \cdot \\
&\cdot \int_{-\infty}^{\infty} \frac{d\tilde{\omega}}{2\pi} \exp\{-i\tilde{\omega}(t - r/c)\} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} B(\mathbf{p}, \omega, \tilde{\omega}) B(\mathbf{p}_1, \omega + \varepsilon_{\mathbf{p}_1-\mathbf{k}} \\
&- \varepsilon_{\mathbf{p}-\mathbf{k}}, \tilde{\omega}) \exp\left\{-\frac{ir}{c}[\Pi(\omega - \omega_0 - \varepsilon_{\mathbf{p}-\mathbf{k}}) + \right. \\
&\left. + \Pi(\tilde{\omega} - \omega + \omega_0 + \varepsilon_{\mathbf{p}-\mathbf{k}})]\right\},
\end{aligned} \tag{26}$$

where

$$\mathbf{k} = \omega_0\mathbf{q}/c, \quad \mathbf{q} = \mathbf{r}/r, \quad \mathbf{v}_p = \mathbf{p}/M.$$

It follows at once from the known properties of the Laplace transformation that $K^{m_1m_1'}_{mm'} = 0$ for $t = r/c < 0$. Let us obtain an expression for the kernel K in the two limiting cases where $\beta \gg \gamma$ and $\beta \ll \gamma$. If $\beta \gg \gamma$ and if we are not interested in the rapid oscillations of the kernel with the characteristic period of order $\tau$,[1] we can simplify (26) considerably. The neglect of the oscillations implies that $\tilde{\omega}$ in the function $\Pi(\omega)$ must be set equal to zero. The same limit must also be taken in the functions $B(\mathbf{p}, \omega, \tilde{\omega})$ (neglect of the quantum indeterminacy between energy and time). Then the integration over $\tilde{\omega}$ in (26) leads to the appearance of the $\delta$ function $\delta(t - r/c)$ which guarantees the usual retardation, and the function B has the form

$$B(\mathbf{p}, \omega, \tilde{\omega} \to 0) = -2\pi i \delta_\gamma(\omega - \omega_0 - \varepsilon_p). \tag{27}$$

The index $\gamma$ of the $\delta$ function indicates that this function was obtained in the limit $\gamma \to 0$. Using further

$$\Pi(\omega - \omega_0) + \Pi(-\omega + \omega_0) = -ick(\omega), \tag{28}$$

where $k(\omega)$ is the usual absorption coefficient,[12] we have

$$\begin{aligned}
K^{m_1m_1'}_{mm'}(\mathbf{r}, \mathbf{p}, \mathbf{p}_1, t) = &\delta(t - r/c)\delta(\mathbf{v}_p\mathbf{q} - \mathbf{v}_{p_1}\mathbf{q}) \frac{\omega_0 n\varphi(\mathbf{p})}{(2\pi)^4 r^2 c^3 \gamma} \cdot \\
&P^{m_1m_1'}_{mm'}(\omega_0\mathbf{q}) \exp\{-rk(\omega_0 + \mathbf{v}_p\mathbf{q}\omega_0/c)\}.
\end{aligned} \tag{29}$$

When the retardation is neglected, expression (29) coincides with the result obtained in [6] if $\gamma = 0$ in the expression for $k(\omega)$.

In the other limiting case, $\gamma \gg \beta$, an expression for

the kernel was obtained in [1], where the spatial distribution of fixed excited atoms was investigated. The results of [1] follow from (26) in the limit of infinitely heavy atoms.

## 3. SHAPE OF THE LINE OF EMISSION FOR ADIABATIC EMISSION OF THE QUANTA FROM THE SYSTEM

The behavior of the quanta in a medium of two-level atoms has been investigated repeatedly in recent times. Our method permits a generalization of the results of a number of papers[11, 13-15] taking account of the heat motion of the atoms and of the degeneracy of the upper as well as lower levels. Let us consider the problem of the shape of the spectral line of the quantum caused by the radiation diffusion in a system in which the radiation is emitted adiabatically, i.e., at $t \to \infty$. We emphasize that the entire discussion rests on the assumption $n\lambda\!\!\!^{-3} \ll 1$, so that the resonant dipole-dipole interaction plays practically no role and the width (which turns to be $\sim 1/\tau \gg \beta, \gamma$) is the collective width due to the numerous absorption and re-emission processes. As is known,[12, 16] the shape of the line of emission depends on the character of the excitation; we therefore investigated the cases where there is an excited molecule or quantum in the system at $t = 0$. The graphical equations for the density function of the quanta $f_{\lambda\lambda'}(\mathbf{k}, \kappa, \omega, \tilde{\omega})$ are shown in Fig. 8.

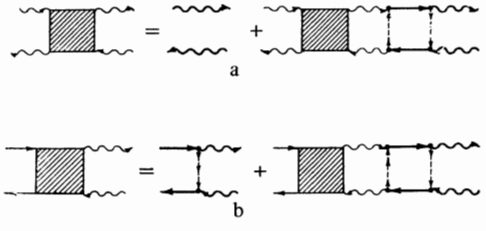


FIG. 8.

The solution of the analytic equations corresponding to Fig. 8 was found with the same simplifications which were used in Sec. 2. Moreover, we have neglected the Doppler broadening. Because of this last simplification, the solution with the quantum in the initial state (Fig. 8, a) is evidently not very interesting, since in this case we obtain two narrow lines displaced from $\omega_0$ by a distance of the order $\pm 1/\tau$, with a width of order $\gamma$ (the shift and the width depend on $\Delta = \omega_{k_0} - \omega_0$). In the case where there is an excited molecule in the initial state or where the irradiation is done with a broad spectral beam, we obtain the following line shape:

$$f(\omega) \sim \left[ (\omega - \omega_0)^2 + \frac{\gamma^2}{4} + \frac{1}{4\tau^2} \frac{(2j_1+1)}{(2j_0+1)} \right]^{-1}. \qquad (30)$$

An analogous result without account of the degeneracy of the levels was obtained by Alekseev[14] and one of the authors.[15]

Thus the spectral line due to the diffusion radiation has a Lorentzian shape with the width

$$\Gamma = \frac{1}{\tau} \left( \frac{2j_1+1}{2j_0+1} \right)^{1/2}. \qquad (31)$$

It follows from (18) that $\Gamma \sim \sqrt{n}$ ($\Gamma \gg \gamma$). It is possible that the large widths obtained in the experiments of Tomiser[8] and Moser and Schulz[17] are due precisely to the effect of the diffusion stage of the process. The widths observed in these works are about $10^3$ times larger than the values which follow from the shock and statistical theories. We note that in earlier works[18-20] such anomalously large widths have not been observed. In [8, 17] the resonance lines of the doublets of alkaline metals have been investigated, for which $j_0 = 1/2$, $j_1 = 1/2$ and $3/2$; therefore

$$\Gamma_{3/2} / \Gamma_{1/2} = \sqrt{2}\, \tau_{1/2} / \tau_{3/2},$$

where $\Gamma_{3/2}, \tau_{3/2}$ refer to the line with $j_1 = 3/2$, and $\Gamma_{1/2}, \tau_{1/2}$ refer to the line with $j_1 = 1/2$. Since for both lines of the doublet the lifetimes and the frequencies of the transitions do not differ strongly,[12] we have $\Gamma_{3/2}/\Gamma_{1/2} \approx \sqrt{2}$.

The data quoted in the review paper of Chen and Takeo[21] show that this ratio is indeed close to $\sqrt{2}$. There it is also noted that for small pressures the broadening is symmetric and is well described by a Lorentzian formula. An analogous dependence of the width on the statistical weights of the levels in the form $[(2j_1 + 1)/(2j_0 + 1)]^{1/2}$, but with a linear dependence on the density, was obtained by Foley.[22] We note that the result of Foley is based on the assumption that the broadening is due to the resonant dipole-dipole interaction. This assumption seems questionable under conditions where $n\lambda\!\!\!^{-3} \ll 1$. The experiments[8, 17] were performed over a wide range of densities from $n\lambda\!\!\!^{-3} \ll 1$ to $n\lambda\!\!\!^{-3} > 1$. The shape of the absorption line was observed, so that one should not expect exact agreement with (31). For a comparison, we quote the widths taken from [8] and calculated according to (31). These data refer to the resonance line of the sodium doublet with the wavelength $\lambda = 7665$ Å. For the density $n = 10^{12}$ atoms/cm$^3$ the experimental width is $\Gamma_{3/2} = 0.13$ Å while (31) gives $\Gamma_{3/2} = 0.14$ Å. As the density increases, the agreement becomes somewhat worse, and for $n = 1.45 \times 10^{14}$ atoms/cm$^3$ we have $\Delta\lambda_{3/2} = 0.6$ Å and $\Gamma_{3/2} = 1.7$ Å.

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