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DIELECTRIC CONSTANT OF A RESONANCE MEDIUM

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An expression is obtained for the dielectric constant of a medium consisting of a set of identical two-level quantum objects. The extreme cases of weak and strong fields are investigated in detail. The dependence of the dielectric constant on the amplitude of the electromagnetic field is found in the latter case. A general solution of the closed set of equations for the vector potential, polarization current and level overpopulation is obtained without taking into account pumping and relaxation losses. The frequencies of the natural oscillations in a resonant medium in weak and strong fields are determined.

FOR many physical applications it is very important to understand the kinetics of the emission and absorption of quanta by a system consisting of a very large number of identical two-level quantum objects. Such objects may be, for example, a system of spins in an external field or gas molecules with the two levels in question, or impurity atoms in the field of a crystal lattice. For the sake of brevity we shall refer to the identical two-level objects simply as molecules. Such a problem was investigated from various points of view by many workers. We have narrowed down the formulation of the problem, excluding from consideration the losses of quanta, relaxation processes, and pumping, in order to make the simplified problem amenable to a correct solution. In view of the great complexity of the calculations, certain physical simplifications were made from the very outset. For example, we have neglected the collision of the molecules in the gas of gaseous substances, the level spread broadening due to the Stark effect in the alternating field of the oscillating lattice, temperature effects, etc. We hope that allowance for these effects will not change the principal aspect of our results. We have thus arrived at a model problem involving the interaction of a radiation field with a system of two-level molecules within a bounded volume V with perfectly reflecting walls. The final results will apply equally well to an unbounded medium with a specified density of the molecules in question.

In an earlier paper [1] we investigated in part the oscillations of photon density $n(t)/V$ in similar circumstances, under the condition that at the initial instant there are no quanta and the distribution of the molecules among the levels is fixed. In the present paper we consider the general case, with an arbitrary number of quanta n_0 and an arbitrary distribution N of molecules among the levels specified at the initial instant of time. On the basis of the closed system of equations for the vector potential, the polarization current, and the change in level overpopulation, we obtained a general expression for the dielectric constant of a resonant medium and investigated in detail some limiting cases. Knowledge of the dielectric constant makes it possible to approach the solution of the main problem by a different, phenomenological method. However, the use of the dielectric constant of a resonant medium is apparently not always convenient (for example, in a weak field, if the number of molecules in the excited state is large at the instant when the field is turned on, $N^0_- \gg n_0$, $N^{02}_- \gg N^0_2$). In such cases, the electromagnetic oscillations are best described directly by the system of equations proposed below for the vector potential, the polarization current, and the level overpopulation.

From the equation for the vector potential with the determined dielectric constant we have obtained the frequencies of the natural oscillations of the field-intensity vector and the oscillations of the photon density in the resonant medium. It turned out that to each mode of photon-density oscillation there correspond oscillations of the field intensity vector with two frequencies in the case of weak fields, and with three frequencies in the case of strong fields. In the strong field one is the fundamental harmonic and the other two are satellites with small amplitude. Addition of electromagnetic oscillations at these frequencies leads to beats, which are manifest in the photon density oscillation mode. In this connection, we also investigated the beats from a different point of view, using the procedure of the earlier paper[1]. As expected, the equations obtained by the two methods for the photon-density oscillations coincide. In the case of sufficiently strong positive initial overpopulation ($N_-^0 \gg n_0$, $N_-^{02} \gg N_2^0$) the photon-density oscillation is in the form of exponentially growing (in time) bursts, which coincide with those considered earlier in[1]. In weak and strong fields ($|n - n_0| \ll |n_0 - N_-^0|$) each type of photon-density oscillation has a harmonic character. At the same time, the usual balance equation[2], written out for our model problem, leads to a different law of oscillation of $n(t)$. This means that the balance equations cannot be used here. Consequently, they are likewise not applicable in the general case when account is taken of pumping and of relaxation losses.

The investigation carried out of the system of equations of the vector potential, the polarization, current, and the level overpopulation is of interest also in principle, since it leads to a definite conclusion concerning the limits of applicability of an analogous system of equations obtained in several papers (see, for example [3-5]) by a semiclassical method. In addition, the procedure here, and also the solution for our model problem, can serve as the basis for further investigations of more complicated equations for the vector potential, with allowance for the pumping and for relaxation losses.

The Hamiltonian H of a system of N identical two-level molecules and a radiation field, contained in a volume V, will be taken in the same form as in[1]. However, unlike in[1], we retain all the terms of the Hamiltonian, writing them in the form

$$H = \frac{\hbar\omega_0}{2} N_- + \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}} n_{\mathbf{k}} - \frac{1}{c} \sum_{\mathbf{k}} (\mathbf{A}(\mathbf{k})\, \mathbf{j}(-\mathbf{k}) - \frac{\omega_L{}^2}{8\pi c} \mathbf{A}(\mathbf{k})\, \mathbf{A}(-\mathbf{k})), \tag{1}$$

$$N_- = \sum_j \sigma_z{}^j, \qquad n_{\mathbf{k}} = \sum_\lambda c_{\mathbf{k}\lambda}{}^+ c_{\mathbf{k}\lambda},$$

$$\mathbf{A}(\mathbf{k}) = (2\pi\hbar c^2/\omega_{\mathbf{k}})^{1/2} \sum_\lambda (c_{\mathbf{k}\lambda} \mathbf{l}_{\mathbf{k}}{}^\lambda + c_{-\mathbf{k}\lambda}{}^+ \mathbf{l}_{-\mathbf{k}}{}^\lambda), \tag{2}$$

$$j_\alpha(\mathbf{k}) = -V^{-1/2} \sum_{j\beta} (\delta_{\alpha\beta} - k_\alpha k_\beta/\mathbf{k}^2)\, (\sigma_+{}^j M_\beta{}^j(-\mathbf{k}) + \sigma_-{}^j M_\beta{}^{*j}(\mathbf{k}))\, e^{-i\mathbf{k}\mathbf{x}_j}, \tag{3}$$

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

where the first and second terms in (1) are the energy operators of the isolated molecules and of the free radiation field, respectively, and the last term is the operator of their interaction energy; $\hbar\omega_0$—energy difference of the upper and lower levels of the isolated molecule; $\omega_L^2 = 4\pi e^2 ZN/mV$, where e and m—charge and mass of the electron, respectively, and V—number of electrons in the molecule. Further, $c_{\mathbf{k}\lambda}$ and $c_{\mathbf{k}\lambda}^+$ are the operators of absorption and emission of a photon with momentum $\hbar\mathbf{k}$ and polarization $\mathbf{l}_{\mathbf{k}}^\lambda$, respectively, with

$$c_{\mathbf{k}\lambda} c^+{}_{\mathbf{k}'\lambda'} - c^+{}_{\mathbf{k}'\lambda'} c_{\mathbf{k}\lambda} = \delta_{\mathbf{k}\mathbf{k}'} \delta_{\lambda\lambda'},$$
$$\lambda = 1, 2, \qquad \omega_{\mathbf{k}}{}^2 = \mathbf{k}^2 c^2.$$

Finally, $\mathbf{x}_j$—coordinate of the center of gravity of the j-th molecule, and $\mathbf{M}^j(\mathbf{k})$—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 of momentum $\hbar\mathbf{k}$.

The Fourier component $\mathbf{A}(\mathbf{k})$ of the operator of vector potential $\mathbf{A}(\mathbf{x})$

$$\mathbf{A}(\mathbf{k}) = V^{-1/2} \int \mathbf{A}(\mathbf{x})\, e^{-i\mathbf{k}\mathbf{x}}\, dV,$$

and the operator $\mathbf{j}(\mathbf{k})$ are connected with the Fourier component of the polarization current $\mathbf{j}'(\mathbf{k})$ by the relation

$$\mathbf{j}'(\mathbf{k}) = \mathbf{j}(\mathbf{k}) - \omega_L{}^2 \mathbf{A}(\mathbf{k}) / 4\pi c, \tag{4}$$

which follows from the fact that in the absence of conduction or extraneous currents the operators $\mathbf{A}(\mathbf{k})$ and $\mathbf{j}'(\mathbf{k})$ should satisfy, in accordance with Maxwell's equations, the following equation:

$$\ddot{\mathbf{A}}(\mathbf{k}) + \omega_{\mathbf{k}}{}^2 \mathbf{A}(\mathbf{k}) = 4\pi c\, \mathbf{j}'(\mathbf{k}).$$

In the case of dipole radiation of the molecules, the polarization current (4) is none other than the current due to the transition of atomic electrons in the presence of an "external" electromagnetic field with potential **A**. If the radiation is of multipole type, then the intuitive meaning of (4) disappears. In addition, the polarization vector **P**, defined by the relation $\dot{\mathbf{P}} = \mathbf{j}'$, does not reduce here to a sum of the dipole moments of individual molecules.

We introduce the dielectric constant ϵ of the medium in the usual way, using the equality

$$4\pi \mathbf{j}'(t) + \frac{\partial}{\partial t} \mathbf{E}(t) = \frac{\partial}{\partial t} (\hat{\varepsilon} \mathbf{E}(t)), \tag{5}$$

where $\mathbf{E} = -\dot{\mathbf{A}}/c$ and $\hat{\epsilon}$—integral operator to be determined (we use throughout a gauge in which the scalar potential vanishes identically and $\operatorname{div} \mathbf{A}(\mathbf{x}, t) = 0$).

Thus, to determine the dielectric constant of the medium it is sufficient to find the connection (5) between the polarization current and the vector potential. In order to establish this connection, we make use of the rule for the differentiation of operators in quantum mechanics, and find the equations satisfied by the operators of the vector potential and of the current j. Simple calculations for an isotropic medium yield

$$\ddot{A}_\alpha(\mathbf{k}) + (\omega_\mathbf{k}^2 + \omega_L^2) A_\alpha(\mathbf{k}) = 4\pi c j_\alpha(\mathbf{k}), \tag{6}$$

$$\ddot{j}_\alpha(\mathbf{k}) + \omega_0^2 j_\alpha(\mathbf{k}) = -\frac{\omega_0^2}{4\pi c\tau_0^2} N_- A_\alpha(\mathbf{k}) - \sum_{\mathbf{k}'\alpha'}{}' B_{\mathbf{k}\alpha,\,\mathbf{k}'\alpha'} A_{\alpha'}(\mathbf{k}'), \tag{7}$$

$$B_{\mathbf{k}\alpha,\,\mathbf{k}'\alpha'} = \frac{\omega_0}{\hbar c V} \sum_{j\beta} \sigma_z^j \left(\delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{\mathbf{k}^2}\right) (M_\beta^{*j}(\mathbf{k}) M_{\alpha'}^j(\mathbf{k}') + M_\beta^j(-\mathbf{k}) M_{\alpha'}^{*j}(-\mathbf{k}')) e^{i(\mathbf{k}'-\mathbf{k})\mathbf{x}_j}, \tag{8}$$

$$\tau_0^{-2} = 2\pi c^3 W/\omega_0^2 V,$$

where W—probability that a single isolated molecule emits one quantum per unit time, and the prime at the summation sign denotes the condition $\mathbf{k}' \neq \mathbf{k}$.

We first determine ϵ in the presence of a weak electromagnetic field

$$n = \sum_\mathbf{k} n_\mathbf{k} \ll N,$$

assuming that prior to application of the electric magnetic field all the molecules were in the ground state. Then, when taking the matrix elements of the operators in (7), we can confine ourselves to the linear approximation in the number of photons, putting $\sigma_z^j = -1$ (see also [1]). Expanding the operator matrix elements of the vector potential $\mathbf{A}(\mathbf{k}, t)$ and current $\mathbf{j}(\mathbf{k}, t)$ in a Fourier integral with respect to the variable t, for example

$$\mathbf{A}(\mathbf{k}, t) = \frac{1}{2\pi} \int \mathbf{A}(\mathbf{k}, \omega) e^{-i\omega t} d\omega,$$

we obtain

$$(\omega_\mathbf{k}^2 + \omega_L^2 - \omega^2) A_\alpha(\mathbf{k}, \omega) = 4\pi c j_\alpha(\mathbf{k}, \omega), \tag{9}$$

$$(\omega_0^2 - \omega^2) j_\alpha(\mathbf{k}, \omega) = \frac{\omega_0^2 N}{4\pi c\tau_0^2} A_\alpha(\mathbf{k}, \omega) + \sum_{\mathbf{k}'\alpha'}{}' B'_{\mathbf{k}\alpha,\,\mathbf{k}'\alpha'} A_{\alpha'}(\mathbf{k}', \omega), \tag{10}$$

where $B'_{\mathbf{k}\alpha,\mathbf{k}'\alpha'}$ differs from (8) in that it does not contain σ_z^j.

It is easy to determine in (10) the contribution of the off-diagonal terms $B'_{\mathbf{k}\alpha,\mathbf{k}'\alpha'}$ with $\mathbf{k}' \neq \mathbf{k}$. To this end we express A_α in the primed sum of the right side of (10) in terms of j_α with the aid of (9), and replace the quantities $B'_{\mathbf{k}\alpha,\mathbf{k}'\alpha'}$ and j_α by their sums in accordance with (3) and (8). Only the principal terms with $j = j'$ need be retained in the resultant double sum over the indices j and j′. The discarded terms contain additional rapidly-oscillating factors of the type $\exp[i\mathbf{k}\cdot(\mathbf{x}_j - \mathbf{x}_{j'})]$, and therefore make a negligible contribution. In addition, we assume that the molecule radiation is of the dipole type, the wavelength is small compared with the linear dimensions of the volume V, and $\omega_L^2 \ll \omega_0^2$. Then the primed sum in (10) is equal to

$$\left(i\omega W + \frac{\omega_0}{\pi} \int_0^\infty \frac{W_{\omega'} d\omega'}{\omega' - \omega}\right) j_\alpha(\mathbf{k}, \omega), \tag{11}$$

where $W_{\omega'}$—probability of dipole emission of a photon of frequency ω' by a single isolated molecule, $W_{\omega_0} \equiv W$, and the integral is taken in the sense of the principal value. The second term in (11) is the radiation level shift of the isolated molecule, an account of which in (10) leads to an insignificant change in the value of ω_0^2. The radiative level shifts will therefore not be written in explicit form in what follows.

Thus, as was already noted in [1], the off-diagonal terms of the operator (8) make a contribution that is due to the radiative damping of the individual molecules (as if the latter were isolated).

Relations (10) and (11) enable us to express j in terms of **A**. If we now use formulas (4) and (5), we obtain the sought-for connection between the polarization current and the vector potential, which yields

$$\hat{\varepsilon}\mathbf{E}(t) = \frac{1}{2\pi} \int \varepsilon(\omega) \mathbf{E}(\omega) e^{-i\omega t} d\omega,$$

$$\varepsilon(\omega) = 1 - \frac{\omega_L^2}{\omega^2} + \frac{8\pi\omega_0 \sum_j |\mathbf{M}^j|^2}{3\hbar\omega^2 V(\omega_0^2 - \omega^2 - i\omega W)}. \tag{12}$$

At high frequencies ω, expression (12) coincides with the known result [6]. At low frequencies $\omega \to 0$, the contribution to the dielectric constant of the medium will be made not only by the virtual transitions of the molecule between the two levels in question, but also by virtual transitions to other levels. For example, in the case of dipole radiation, neglecting attenuation, allowance for the other levels can be readily made by making in (12) the substitutions $\omega_0^2 \to \omega_{m0}^2$ and $\mathbf{M}^j \to ie\omega_{m0}x_{m0}$ with subsequent summation over the indices m, which number the levels of the j-th molecule. Then as $\omega \to 0$ we obtain

$$\varepsilon = 1 - \frac{\omega_L^2}{\omega^2} + \frac{\omega_L^2}{\omega^2} \sum_m \frac{\omega_{m0} f_{m0}}{\omega_{m0}^2 - \omega^2} = 1 + \frac{4\pi e^2 N}{mV} \sum_m \frac{f_{m0}}{\omega_{m0}^2},$$

which is likewise in agreement with the universally known result (here $f_{m0} = 2m\omega_{m0} \times |x_{m0}|^2/3\hbar$ is the oscillator strength). If electromagnetic waves with frequencies ω close to ω_0 propagate through the medium, then the largest contribution to the dielectric constant of the medium will be made by the transitions between the two resonant levels of the molecule in question, and the dielectric constant (12) will then take on the form

$$\varepsilon = 1 + \frac{\pi \bar{\lambda}_0^3 WN/V}{\omega_0 - \omega - iW/2}, \tag{13}$$

where $\bar{\lambda}_0 = c/\omega_0$.

For further investigation of magnetic oscillations of arbitrary amplitude it is necessary to obtain a closed system of equations for $\mathbf{A}(\mathbf{k})$ and $\mathbf{j}(\mathbf{k})$. For simplicity we shall neglect in (6) and (7) the nondiagonal terms $B_{\mathbf{k}\alpha,\mathbf{k}'\alpha'}$, which are proportional to W, and the small quantity $\omega_L^2 \ll \omega_0^2$. To obtain a closed system of equations it is necessary to add to (6) and (7) an equation for the operator N_- of the level overpopulation

$$\dot{N}_- = \frac{2}{\hbar\omega_0 c}\sum_{\mathbf{k}} \mathbf{A}(\mathbf{k})\,\dot{\mathbf{j}}(-\mathbf{k}). \tag{14}$$

The solution of the resultant system of equations (6), (7), and (14) is made difficult by the large number of functions $\mathbf{A}(\mathbf{k})$ with different values of $\mathbf{k}$. We therefore consider a simplified problem, with only one arbitrary oscillation mode occurring in the volume V. Such a situation arises, for example, in a cylindrical cavity in which the active molecules are located near the axis. In this case the molecules interact intensely only with one mode determined by a zero-order Bessel function.

In this connection we rewrite (6), (7), and (14) in such a way that they contain only the standing waves $\mathbf{A}_1 = (\mathbf{A}(\mathbf{k}) + \mathbf{A}(-\mathbf{k}))/\sqrt{2}$ and $i\mathbf{A}_2 = (\mathbf{A}(\mathbf{k}) - \mathbf{A}(-\mathbf{k}))/\sqrt{2}$, which are characterized by the parameter ω_k^2, and the corresponding polarization current

$$\mathbf{j}_1 = (\mathbf{j}(\mathbf{k}) + \mathbf{j}(-\mathbf{k}))/\sqrt{2}, \qquad i\mathbf{j}_2 = (\mathbf{j}(\mathbf{k}) - \mathbf{j}(-\mathbf{k}))/\sqrt{2},$$

namely:

$$\ddot{\mathbf{A}}_s + \omega_k^2\mathbf{A}_s = 4\pi c\mathbf{j}_s, \tag{15}$$

$$\ddot{\mathbf{j}}_s + \omega_0^2\mathbf{j}_s = -\frac{\omega_0^2}{4\pi c\tau_0^2}N_-\mathbf{A}_s, \tag{16}$$

$$\dot{N}_- = \frac{2}{\hbar\omega_0 c}\sum_s \mathbf{A}_s\dot{\mathbf{j}}_s, \tag{17}$$

where s = 1, 2 and the quantity W, which enters in (16) via the parameter τ_0^2, denotes here the probability of not only dipole but of any arbitrary multipole radiation of the molecule. Equations (15)—(17) are valid for operators in either the Schrödinger or the Heisenberg representation.

Inasmuch as the expansion used in quantizing the free electromagnetic field was in terms of plane waves, the standing waves are in this case harmonic. However, the conclusions that follow are general in character and hold also when the free electromagnetic field is expanded in arbitrary eigenfunctions $\mathbf{A}_{\omega\mathbf{k}}(\mathbf{x})$ of the resonator, satisfying within the volume V the equation

$$c^2\nabla^2\mathbf{A}_{\omega_k}(\mathbf{x}) + \omega_k^2\mathbf{A}_{\omega_k}(\mathbf{x}) = 0 \tag{18}$$

with boundary conditions determined by the formulation of the concrete problem. This statement follows from the fact that the final results depend only on the initial energy of the radiation field, on the initial distribution of the particles over the levels, and on ω_k^2.

Assume that at the initial instant t = 0 the number of molecules in the upper level was $N_2(0) = N_2^0$, and in the lower level $N_1(0) = N_1^0$, while the number of photons $n = n_k + n_{-k}$ with given ω_k^2 was then equal to n_0. Let us determine the frequencies of the field-intensity vector and the oscillation of the photon density n/V.

According to [1], the overpopulation $N_-(t)$ and the number of photons $n(t)$ are slow functions of t. We shall therefore seek a solution of (15)—(17) in the form

$$\dot{\mathbf{A}}_s - i\omega_0\mathbf{A}_s = \mathbf{X}_s(t)e^{-i\omega_0 t}, \tag{19}$$

$$\dot{\mathbf{j}}_s - i\omega_0\mathbf{j}_s = \mathbf{Y}_s(t)e^{-i\omega_0 t}, \tag{20}$$

where $X_s(t)$ and $Y_s(t)$—slow functions of t. Then, leaving out the rapidly oscillating terms, and also terms of order $|n_0 - N_-^0 + 1/2|^{1/2}/\omega_0\tau_0 \ll 1$, we obtain

$$2i\omega_0\dot{\mathbf{X}}_s = (\omega_k^2 - \omega_0^2)\mathbf{X}_s - 4\pi c\mathbf{Y}_s, \tag{21}$$

$$2i\omega_0\dot{\mathbf{Y}}_s = \frac{\omega_0^2}{4\pi c\tau_0^2}N_-\mathbf{X}_s, \tag{22}$$

$$2i\hbar\omega_0^2 c\dot{N}_- = \sum_s(\mathbf{X}_s^+\mathbf{Y}_s - \mathbf{Y}_s^+\mathbf{X}_s). \tag{23}$$

Combining (21) with (23), and then (22) with (23), and finally (23) with (21) and (22), we obtain the respective integrals of motion

$$\sum_s \mathbf{X}_s^+\mathbf{X}_s + 4\pi\hbar\omega_0 c^2(N_- - I_1) = 0, \tag{24}$$

$$8\pi\tau_0^2\sum_s \mathbf{Y}_s^+\mathbf{Y}_s + \hbar\omega_0^3(N_-^2 - 2N_- - I_2) = 0, \tag{25}$$

$$\sum_s(\mathbf{X}_s^+\mathbf{Y}_s + \mathbf{Y}_s^+\mathbf{X}_s) + \hbar\omega_0(\omega_k^2 - \omega_0^2)c(N_- - I_3) = 0, \tag{26}$$

where the constants I_1, I_2, and I_3 are determined from the initial conditions. Expressions (24)—(26) represent essentially the adiabatic invariants of the fundamental problem (15)—(17). From (21)—(26) we have

$$4\tau_0^2\ddot{N}_- - 3N_-^2 + 2(I_1+1)N_-$$
$$+ 4\tau_0^2(\omega_{\mathbf{k}} - \omega_0)^2(N_- - I_3) + I_2 = 0.$$

An analogous equation is obtained also for the number of photons $n = n_{k} + n_{-k}$ with given ω_{k}^2, since it follows from (24) that

$$2n + N_- = I_1. \tag{27}$$

All the relations (19)—(27) are valid not only for the average quantities or matrix elements of the transition, but also for the corresponding Heisenberg operators[1]. Therefore, in interpreting the integrals of motion (24)—(26), we shall make use of formulas (2) and (3), assuming that the subsystems (the molecules and the radiation field) did not interact at the initial instant of time and were described by their own wave functions. Taking into consideration the definition of the operators $\mathbf{X}_{\mathrm{S}}$ and $\mathbf{Y}_{\mathrm{S}}$

$$\mathbf{X}_s^+\mathbf{X}_s = \dot{\mathbf{A}}_s^2 + \omega_0^2\mathbf{A}_s^2 + i\omega_0(\mathbf{A}_s\dot{\mathbf{A}}_s - \dot{\mathbf{A}}_s\mathbf{A}_s), \tag{28}$$

$$\mathbf{Y}_s^+\mathbf{Y}_s = \dot{\mathbf{j}}_s^2 + \omega_0^2\mathbf{j}_s^2 + i\omega_0(\mathbf{j}_s\dot{\mathbf{j}}_s - \dot{\mathbf{j}}_s\mathbf{j}_s) \tag{29}$$

and the initial conditions, we find

$$I_1 = 2n_0 + N_-^0,$$

$$I_2 = N_-^{02} - 2N_-^0 + 16N_2^0, \quad I_3 = N_-^0, \tag{30}$$

where $N_-^0 \equiv N_-(0)$—initial overpopulation.

Thus, the sought-for connection between the polarization current $\mathbf{j}$ and the vector potential $\mathbf{A}$ is determined in the general case by relation (16), in which the overpopulation $N_-(t)$ is a solution of the equation

$$\ddot{N}' - \frac{3}{4\tau_0^2}N'^2 + \left(\frac{n_0 - N_-^0 + 1/2}{\tau_0^2} + (\omega_{\mathbf{k}} - \omega_0)^2\right)N'$$

$$+\frac{n_0N_-^0 + 4N_2^0}{\tau_0^2} = 0, \tag{31}$$

where $N' \equiv N_-(t) - N_-^0$, and the initial condition (30) was taken into account.

Solution (31) is expressed in terms of an elliptic function. Consequently, the overpopulation $N_-(t)$ has a definite period $T \equiv 2\pi/\Omega$. It is consequently convenient to represent it in the form of a Fourier series

$$N_-(t) = \sum_{m=-\infty}^{\infty} N_m e^{im\Omega t}. \tag{32}$$

Using (32), (16), and (5) we easily obtain the dielectric constant of the resonant medium

$$\hat{\varepsilon}\mathbf{E}(t) = \frac{1}{2\pi}\int d\omega e^{-i\omega t}\sum_{m=-\infty}^{\infty}\varepsilon_m(\omega)\mathbf{E}(\omega + m\Omega),$$
$$\varepsilon_m(\omega) = \delta_{m0} + \omega_0^2N_m / \omega(\omega + m\Omega)(\omega^2 - \omega_0^2)\tau_0^2. \tag{33}$$

The integral operator $\hat{\epsilon}$ is written in explicit form as

$$\varepsilon\mathbf{E}(t - t_0) \equiv \int_{t_0}^{t}\varepsilon(t - t', t' - t_0)\mathbf{E}(t' - t_0)\,dt',$$

$$\varepsilon(t - t', t' - t_0) = \frac{1}{2\pi}\int d\omega e^{-i\omega(t-t')}\sum_{m=-\infty}^{\infty}e^{im\Omega(t'-t_0)}\varepsilon_m(\omega),$$

where t_0—instant when the magnetic field is turned on (in our problem $t_0 = 0$). Thus, the kernel $\epsilon(t_1, t_2)$ of the integral operator is expanded in a Fourier integral with respect to the variable t_1 and in a Fourier series with respect to the variable t_2.

For concreteness, we consider the region of weak and strong fields at $N_-^0 = -N$, for which the roots of the characteristic equation

$$(N' - \alpha_1)(N' - \alpha_2)(N' - \alpha_3) = 0$$

satisfy the inequality

$$\alpha_1 > \alpha_2 > \alpha_3, \qquad \alpha_2 \geqslant N'(t) \geqslant \alpha_3,$$

where $\alpha_3 = 0$, and α_1 and α_2 are respectively the following numbers:

$$n_0 + N + (\omega_{\mathbf{k}} - \omega_0)^2\tau_0^2$$
$$\pm\left[(n_0 + N + (\omega_{\mathbf{k}} - \omega_0)^2\tau_0^2)^2 - 4n_0N\right]^{1/2}.$$

Then the solution of (31) takes the form

$$N_-(t) = -N + \alpha_2\,\mathrm{sn}^2(2Kt / T, (\alpha_2 / \alpha_1)^{1/2}). \tag{34}$$

Here $\mathrm{sn}(2Kt/T, (\alpha_2/\alpha_1)^{1/2})$—elliptic sine, and T—period of the function $N_0(t)$, equal to

$$T = 4\tau_0\left(\frac{2}{\alpha_1}\right)^{1/2}K, \quad K \equiv F\left[\frac{\pi}{2}, \left(\frac{\alpha_2}{\alpha_1}\right)^{1/2}\right],$$

where K—complete elliptic integral of the first kind.

Relation (34) enables us to calculate the Fourier component N_m of the expansion (32), after which the general expression for the dielectric constant (33) assumes the concrete form

$$\varepsilon_0(\omega) = 1 + \frac{\omega_0^2[N - \alpha_1(1 - E / K)]}{\omega^2(\omega_0^2 - \omega^2)\tau_0^2},$$

$$\varepsilon_{\pm m}(\omega) = \frac{m\pi^2q^m\alpha_1\omega_0^2}{(1 - q^{2m})K^2\omega(\omega \pm m\Omega)(\omega_0^2 - \omega^2)\tau_0^2}, \tag{35}$$

where m = 1, 2, . . . , and E and q are well known

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[1] By definition, relations (19) and (20) for the operators are understood in the sense that the result is the same when the wave function of the system is acted upon from the left by both sides of the equation.

functions [7]

$$E = E\left[\frac{\pi}{2}, \left(\frac{\alpha_2}{\alpha_1}\right)^{1/2}\right], \quad q = \exp(-\pi K'/K),$$

$$K' = F\left[\frac{\pi}{2}, \left(\frac{\alpha_1 - \alpha_2}{\alpha_1}\right)^{1/2}\right].$$

If $\mathbf{E}(t)$ is a superposition of waves with different ω_k^2, then the dielectric constant (35) will, generally speaking, change.

For example, in a weak field $n_0 \ll N$, the dielectric constant (35) in the first nonzero approximation in the parameter n_0/N coincides with expression (13), provided we discard in the latter the imaginary component in the denominator. In the next approximation we have

$$\begin{aligned} \varepsilon_0(\omega) &= 1 + f_\omega[1 - n_0/(N + (\omega_\mathbf{k} - \omega_0)^2\tau_0^2)], \\ \varepsilon_{\pm 1}(\omega) &= \omega n_0 f_\omega / 2(N + (\omega_\mathbf{k} - \omega_0)^2\tau_0^2)(\omega \pm \Omega_\mathbf{k}), \\ \varepsilon_m(\omega) &= \varepsilon_{-m}(\omega) = 0, \qquad m = 2, 3, \ldots, \end{aligned} \tag{36}$$

where

$$f_\omega = 2\pi c^3 WN / \omega^2(\omega_0^2 - \omega^2)V.$$

The numerical value of $\epsilon_{\pm m}(\omega)$ with $m = 2, 3, \ldots$, in a weak field is equal, in order of magnitude, to $(\epsilon_0 - 1)(n_0/N)^m$.

In a strong field $n_0 \gg N$ far from resonance $(\omega_k - \omega_0)^2\tau_0^2 \gg N$ we obtain in the approximation that is linear in the parameter N/n_0

$$\varepsilon_0(\omega) = 1 + (\omega_\mathbf{k} - \omega_0)^2 f_\omega / [(\omega_\mathbf{k} - \omega_0)^2 + n_0\tau_0^{-2}], \tag{37}$$

and the expression for $\epsilon_\pm(\omega)$ assumes the form (36), in which N in the denominator is replaced by n_0, and $\epsilon_m(\omega) = \epsilon_{-m}(\omega) = 0$ for $m = 2, 3, \ldots$

The numerical value of $\epsilon_{\pm m}(\omega)$ for $m = 2, 3, \ldots$ in a strong field is equal in order of magnitude to $(\epsilon_0 - 1)(N/n_0)^m$. In a strong field $n_0 \gg N$ the dielectric constant changes near resonance $N \gtrsim (\omega_k - \omega_0)^2\tau_0^2 \gg W^2$. For example, in place of (37) we have

$$\varepsilon_0(\omega) = 1 + f_\omega[8(\omega_\mathbf{k} - \omega_0)^2\tau_0^2 - N] / 8n_0. \tag{38}$$

In all the presented formulas it is assumed that the strong field remains smaller than atomic, so as not to violate the applicability of the Hamiltonian (1). Under reasonable assumptions concerning the molecule density N/V, the indicated region for the strong field takes place when

$$e^2V / \hbar\omega_0 a^4 \gg n_0 \gg N,$$

where a—Bohr radius.

Knowing the dielectric constant (33) we obtain in the usual manner the natural frequencies of the oscillation and the Fourier components of the vector potential, with the aid of the equation

$$\begin{aligned} &\omega_\mathbf{k}^2\mathbf{A}_m - (\omega + m\Omega)\sum_{m'=-\infty}^{\infty}[\omega + (m + m')\Omega] \\ &\quad\times \boldsymbol{\varepsilon}_{m'}(\omega + m\Omega)\,\mathbf{A}_{m+m'} = 0, \end{aligned} \tag{39}$$

$$\mathbf{A}_m \equiv \mathbf{A}(\omega + m\Omega), \quad m = 0, \pm 1, \pm 2, \ldots.$$

If we now change over from the Fourier components to the time domain and use the initial conditions, we obtain the time dependence of the vector potential $\mathbf{A}(t)$. Its dependence on the coordinates is determined by the solution of (18).

Let us consider the case of a single unpolarized standing wave, characterized by a parameter ω_k. For a weak field $n_0 \ll N$ we have from (39), in the first nonvanishing approximation,

$$\begin{aligned} A(t) &= a_1 \sin \omega_1 t + a_2 \sin \omega_2 t, \\ \omega_{1,2} &= (\omega_\mathbf{k} + \omega_0 \pm \Omega_\mathbf{k}) / 2, \\ a_{1,2} &= (2\pi\hbar c^2 n_0 \omega_0^{-1})^{1/2}[1 \pm (\omega_\mathbf{k} - \omega_0) / \Omega_\mathbf{k}], \end{aligned} \tag{40}$$

where it becomes necessary in the expression

$$\Omega_\mathbf{k}^2 = (\omega_\mathbf{k} - \omega_0)^2 + (n_0 + N)\tau_0^{-2}$$

to discard n_0 in weak fields and N in strong fields (see below).

In a strong field $n_0 \gg N$, far from resonance $(\omega_k - \omega_0)^2\tau_0^2 \gg N$, we obtain from (37) and (39)

$$\begin{aligned} A(t) &= (a_0 - a_1 - a_2) \sin \omega t + a_1 \sin \omega_1 t + a_2 \sin \omega_2 t, \\ \omega_{1,2} &= \omega \pm \Omega, \quad a_0 = (8\pi\hbar c^2 n_0 \omega_0^{-1})^{1/2}, \\ a_{1,2} &= a_0 n_0 N / 8\tau_0^4\Omega_\mathbf{k}^2(\omega_{1,2} - \omega_\mathbf{k})(\omega_{1,2} - \omega_0), \end{aligned} \tag{41}$$

where Ω is equal in this approximation to Ω_k, and the frequency ω is expressed in terms of ω_k by a relation that can be regarded as a dispersion equation for the strong field $a_0 \sin \omega t$:

$$\omega_\mathbf{k}^2 - \omega^2 = \frac{\omega_0^2 N (\omega_\mathbf{k} - \omega_0)^2}{(\omega_0^2 - \omega^2)[(\omega_\mathbf{k} - \omega_0)^2\tau_0^2 + n_0]}. \tag{42}$$

The frequency ω of the fundamental harmonic of the strong field (41) coincides with great accuracy with the frequency ω_k of the free field. The second root of Eq. (42) is not related to the electromagnetic oscillations of the vector $\mathbf{A}$ which are considered here, for in the limit as $N \to 0$ this root tends to a value $\omega^2 = \omega_0^2$, whereas for the free electromagnetic field we should have $\omega^2 = \omega_k^2$. In the next higher approximations the value of ω^2 is made more precise by adding terms containing the small parameter N/n_0 raised to the proper power. With this there appear in (41) harmonics with frequencies $\omega \pm m\Omega$, $m = 2, 3, \ldots$, with amplitude of the order of $a_0(N/n_0)^m$.

Thus, far from resonance $(\omega_k - \omega_0)^2\tau_0^2 \gg N$ the dielectric constant of the resonant medium, for a strong field $a_0 \sin \omega t$, is equal, in the approximation considered here, to

$$\varepsilon(\omega) = 1 + \frac{\pi \lambda\!\!\!^{-}{}_0^3 (\omega_0 - \omega) WN/V}{(\omega_0 - \omega)^2 + n_0 \tau_0^{-2}},$$

which agrees with the results of Karplus and Schwinger [8], and also of Basov and Prokhorov [9], provided we set the relaxation time equal to infinity in the cited papers. However, near resonance $N \gtrsim (\omega_k - \omega_0)^2 \tau_0^2 \gg W^2$ the dielectric constant takes a different form:

$$\varepsilon(\omega) = 1 + \frac{\pi \lambda\!\!\!^{-}{}_0^3 WN}{(\omega_0 - \omega) V} \frac{8(\omega - \omega_0)^2 \tau_0^2 - N}{8 n_0}.$$

According to (40) and (41) each type of electromagnetic oscillations, characterized by a parameter ω_k^2, is a superposition of two field-intensity oscillations with nearly equal frequencies in the case of weak fields, and three oscillations in strong fields. The superposition of these oscillations leads to beats which are manifest in a periodic variation of the average photon density n/V in time, with

$$n = n_0 - \frac{n_0 N}{\tau_0^2 \Omega_k^2} \sin^2 \frac{\Omega_k t}{2}, \qquad |n - n_0| \ll |n_0 + N|. \tag{43}$$

The result (43) can also be obtained directly from (34), using (27). Formula (43) together with the particle-number conservation law (27) yields the oscillation of the average number $N_2(t)$ of the molecules at the upper level in an external field of frequency ω. In a strong field $n_0 \gg N$, the function $N_2(t)$ takes the form

$$N_2(t) = \frac{n_0 N}{n_0 + (\omega - \omega_0)^2 \tau_0^2} \sin^2 \{[n_0 \tau_0^{-2} + (\omega - \omega_0)^2]^{1/2} t/2\}, \tag{44}$$

which coincides with the well known result [10]. In a weak field $n_0 \lesssim N$ formula (44) is greatly modified.

In order to obtain also the coordinate dependence of the vector potential, it is necessary to multiply (40) and (41) by the eigenfunction of (18) corresponding to the parameter ω_k^2 and normalized to unity. The vector potential $\mathbf{A}(\mathbf{x}, t)$, obtained in this manner must be taken in the sense of classical electrodynamics, so that the quantity

$$(\dot{\mathbf{A}}^2(\mathbf{x}, t) + \omega_k^2 \mathbf{A}^2(\mathbf{x}, t)) / 8\pi c^2 \tag{45}$$

represents the distribution of the electromagnetic-field energy density in the volume V at each instant of time t. The quantity (45) divided by $\hbar\omega$ is the photon distribution density as a function of x and t, whereas formula (43), like the formulas from [1], describe the oscillation of only the total number n(t) of the photons in the volume V or the average density n(t)/V. This circumstance may be useful in the construction of a theory of quantum generators and amplifiers, where the question of the spatial distribution of the photons is vital.

If the parameter ω_k corresponds to two harmonic eigenfunctions of (18), then

$$\mathbf{A}(\mathbf{x}, t) = A(t) V^{-1/2} (\mathbf{l}_1 \cos \mathbf{kx} + \mathbf{l}_2 \sin \mathbf{kx}),$$

where A(t) is given in (40) and (41), while the unit vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ lie in a plane perpendicular to the vector **k**. Since the unit vectors are arbitrary, when $\mathbf{A}(\mathbf{x}, t)$ is substituted in (45) it is necessary to average (45) over the directions of the vectors $\mathbf{l}_1$ and $\mathbf{l}_2$.

It is easy to obtain from (39) the weak harmonics of the vector potential, with frequencies that differ from the fundamental frequency ω by integer Ω. Upon addition, these harmonics give weak oscillations of the photon density with multiple frequencies $m\Omega$, m = 2, 3, ..., the amplitude of which is smaller than the amplitude of the fundamental oscillation of the photon density with frequency $\Omega \cong \Omega_k$, by a factor $(N/n_0)^m$ in the case of a strong field and $(n_0/N)^m$ in weak fields. The solution of (15)—(17) in the next higher approximations contains also harmonics of the vector potential, with frequencies that differ from the fundamental frequency ω by an odd number of times. However, their amplitudes are quite small. For example, in a strong field the amplitude of the harmonic with frequency 3ω is of the order of $a_0 N/(\omega_0 \tau_0)^2$.

In the general case, a given type of oscillation $n(t) = n_k(t) + n_{-k}(t)$, characterized by a parameter ω_k^2, is described in accordance with (27) and (31) by the following equation ($n' \equiv n - n_0$)

$$\ddot{n}' + \frac{3}{2\tau_0^2} n'^2 + \left(\frac{n_0 - N_-^0 + 1/2}{\tau_0^2} + (\omega_k - \omega_0)^2 \right) \times n' = \frac{n_0 N_-^0 + 4 N_2^0}{2\tau_0^2}, \tag{46}$$

which coincides when $n_0 = 0$ and $N_-^0 \gg 1$ with the result of [1], if we recognize that in the derivation of (46) we took into consideration photons with **k**, λ and −**k**, λ (λ = 1, 2).

In the case of one particle in an electromagnetic field ($N = 1$, $N_2^0 = 0$), Eq. (46) coincides with the analogous equation of Jaynes and Cummings [11]. However, the deductions drawn in [11] for a system of N molecules on the basis of the single-particle problem are in error. In particular, it is proved in [11] that when $n_0 = 0$ the presence of N excited molecules changes only the pre-exponential factor in the formula for the growth of the photon cascade obtained for N = 1.

Yet it follows from Eq. (46) with $n_0 = 0$ and $N_-^0 = N$ that in the case of N excited molecules a significant change takes place also in the exponential factor itself, which determines the effective radiation time of the system of N molecules. This means that the radiation of a system of N molecules has a collective character (see also [1]). In addition, unlike in [11], we have used in the derivation of (46) a decomposition of the average quantum mechanical quantities

$$\langle nn \rangle = \langle n \rangle \langle n \rangle \equiv n^2,$$

which are quadratic combinations of the operators $c_{k\lambda}$ and $c_{k\lambda}^+$. When n is large this decomposition is valid with a high degree of accuracy. Yet the results of [11] are based on the substitution

$$\langle N_- \mathbf{A} \rangle = \langle N_- \rangle \langle \mathbf{A} \rangle, \quad \langle \mathbf{Aj} \rangle = \langle \mathbf{A} \rangle \langle \mathbf{j} \rangle, \qquad (47)$$

where all angle brackets denote quantum-mechanical averaging.

It must be noted in this connection that in accordance with (28)—(31) and (46), the substitutions (47) in (15)—(17) do not lead to an error only if $n_0 \gg 1$ and the particles at the initial instant of time are either overwhelmingly at the upper level $N_2^0 \gg N_1^0$, or mostly at the lower level $N_2^0 \ll N_1^0$. If $N_2^0 \gtrsim n_0 N_-^0$, then the initial conditions are quantum-mechanical and the problem cannot be solved on the basis of (15)—(17) with the substitution (47), since the substitution (47) essentially reduces the problem to a classical analysis. Indeed, let us take for example the case $N_2(0) = N_2^0$ and $n_0 = 0$. According to classical theory we have $j_s^2(0) \equiv |\mathbf{Y}_s(0)|^2 = 0$, since the medium is isotropic and the average dipole moment of the molecule is equal to zero. Yet a quantum-mechanical analysis yields

$$|\mathbf{Y}_s(0)|^2 = \hbar\omega_0^3 N_2^0 / \pi\tau_0^2,$$

where s = 1, 2. This remark is of fundamental importance, because the operator relations (15)—(17) lead upon making the substitution (47) to the same equations for the mean values of A and j as were obtained in many papers (see, for example [3-5]) by a semiclassical method2). Consequently, the semiclassical equations of [3-5] are applicable only in the region where (47) is valid.

Following [3-5], it is easy to introduce into Eqs. (15)—(17), with the substitution (47), terms that take into account relaxation processes, losses, and pumping. The equations obtained in this manner have a phenomenological character and their value is determined by a comparison of the results of the solution with the experiment. We emphasize in this connection that the introduction of a relaxation term in (16), to take account of spontaneous radiation of molecules in weak fields, was justified by us rigorously [see (10)].

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2) In this case it is necessary to neglect pumping and the relaxation terms in the corresponding equations of[3-5].

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