Theory of the Complex Refractive Index

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A quantum-mechanical formalism is developed for representing the complex refractive index of a gas, including both dispersive and absorptive parts. In analogy with some previous work by the author, it is shown that the phenomenological Maxwell equations are obeyed as equations between certain matrix elements of operators representing the field quantities. The method makes use of a canonical transformation due to Arnous and Bleuler, and also of the "damping operators" previously introduced by the author. It is shown that the calculation of the complex refractive index reduces to the calculation of a single diagonal matrix element of a damping operator. The actual evaluation of this matrix element is taken up in the following paper.

I. INTRODUCTION AND SUMMARY

IN a previous paper (hereafter denoted as I),[1] the author has developed a quantum-mechanical theory of the refractive index of a gas in the absence of absorption. The method was essentially that of ordinary perturbation theory, and the refractive index was represented in terms of the self-energy of the stationary states of the system consisting of radiation field plus matter. It was shown that the phenomenological Maxwell equations were obeyed as equations between matrix elements of the various field operators corresponding to creation or destruction of a "dressed" (physical) photon.

However, if absorption takes place, the theory of I is not applicable, since the zero-order system is now highly degenerate, and effects of radiative damping must be taken into account. A formalism has been developed by the author for finding the stationary states of such systems in a second paper,[2] which from now on will be referred to as II. In the present paper, we use the method of II to develop a theory of the complex refractive index of a gas analogous to that of I for the case where no absorption takes place.

Now consider the situation in which, to zero-order approximation, all the atoms of the gas are in their ground states, and a photon is present whose frequency is close to one of the resonant absorption frequencies of the gas. If a true stationary state exists to which this is a zero-order approximation, it will contain large admixtures of states in which the photon has disappeared and one of the atoms has become excited to the level with which the photon is in resonance, and also of states in which the photon has been replaced by another photon of nearly the same frequency. All these states have nearly the same energy, so it is possible for "real" transitions to take place between them. We may refer to these as "real" states; they will form the main contribution to the true stationary state wave function. However, there will also be smaller admixtures of "virtual" states, with energies appreciably different from that of the zero-order state. For instance, the photon may be absorbed with an atom becoming excited to a level which is not in resonance with the original photon. Or an atom may become excited to the resonance level, but a new photon may be emitted rather than the original one being absorbed. These states will only make a small contribution to the wave function, which may be evaluated by means of perturbation theory.

The method we shall use, therefore, is as follows: First, the "virtual" states will be eliminated, through the second order in the interaction, by perturbation theory, leaving a Hamiltonian with matrix elements corresponding to transitions only between the "real" states. Then the methods of II will be applied to complete the solution of the problem. In this paper, only the general theory will be developed, and it will be shown how to represent the complex refractive index of a gas by means of the "damping operators" of II. As in I, it is shown that the phenomenological Maxwell equations are obeyed as equations between creation and destruction matrix elements for "dressed" photons. In contrast to I, however, this theory leads to some new results of practical interest, since the refractive index may actually be calculated as a function of frequency, taking into account the dipolar interactions between the atoms. This leads to a theory of the shape of the "resonance-broadened" absorption line, which is given in the following paper.

II. NOTATION AND HAMILTONIAN

As usual, we quantize our field in a box of volume $V=L^3$. We assume that a fraction α of the volume is filled with gas atoms, uniformly distributed with density $\mathfrak{N}$. We choose the plane $z=0$ as the boundary between the empty and the filled portions. Thus, the density of atoms is $\mathfrak{N}$ for $0\leq z\leq \alpha L$, and is zero for $(\alpha-1)L\leq z<0$. For the sake of definiteness, we assume that the excited state in which we are interested is a triply degenerate p state, which may be thought of as consisting of three components with transition dipole moment matrix elements directed along the x, y, and z axes of our coordinate system.

We denote the frequency of the transition by ν_0. It

[1] C. A. Mead, Phys. Rev. **110**, 359 (1958).
[2] C. A. Mead, Phys. Rev. **112**, 1843 (1958).

is assumed that ν_0 is defined in such a way that it includes those self-energy effects which do not depend on the presence of the other atoms. The effect of other excited states will be neglected. If the frequencies of transition between these excited states and the ground state differ from ν_0 by an amount large compared with the width of the absorption line, their contribution to the refractive index will be small, and may easily be evaluated, if desired, by the methods of I. We also neglect the translational motion of the atoms, assuming them to be fixed.[3]

Capital Latin letters will be used to represent states in which an atom is excited, and small Latin subscripts will denote the direction of "polarization" of the excited state. Thus $|A_i^0\rangle$ denotes a state in which atom A is excited, with the dipole moment matrix element between the excited state and the ground state being directed along the i axis ($i=x, y, z$); all the other atoms are in the ground state. The superscript $(^0)$ indicates that the state is defined in the zero-order representation, in which all interactions between different atoms, and with the radiation field, are neglected. A state in which all the atoms are in the ground state and a photon is present is denoted by a small Greek letter, λ and η being used for this purpose. For example $|\lambda^0\rangle$ denotes a state in which all atoms are in the ground state and a bare photon of wave number vector $\boldsymbol{\kappa}_\lambda$ and polarization $\boldsymbol{\varepsilon}_\lambda$ is present. A general state will be denoted by the Greek letters α, β, γ. The state in which all atoms are in the ground state and no photons are present is denoted by $|0\rangle$.

Following the procedure of II, we write the Hamiltonian as follows[4]:

$$\begin{aligned}\mathcal{H}^{\text{tot}}&=E+\mathcal{H}^{\text{int}}=W+\mathcal{H},\\ \mathcal{H}&=\mathcal{H}^{\text{int}}-\mathcal{H}^{\text{self}},\end{aligned}\tag{II-2}$$

where E is the diagonal zero-order Hamiltonian, and W is a diagonal matrix whose elements include the self-energies. That is, if there exists a true stationary state corresponding to the zero-order state $|\alpha\rangle$, the diagonal element W_α is just the energy of this state. If no such stationary state exists, the value of W_α may be assigned arbitrarily, but of course anything added to W must also be added to $\mathcal{H}^{\text{self}}$.

[3] The neglect of translational motion is expressed mathematically by omitting from the Hamiltonian the term representing the kinetic energy of center-of-mass motion of each atom, so that the position of the center of mass becomes a constant of the motion. Thus, the derivations which follow will be valid if the atoms are sufficiently heavy so that their kinetic energy may be neglected. More precisely, it must be possible to localize the atom in a region small compared with wavelengths of interest without the resulting Doppler broadening of the absorption line arising from translational momentum uncertainty being comparable with the width of the line. This condition, which is assumed to hold in what follows, is sufficient for the validity of the results of this and the following paper, but, in the author's opinion, probably not necessary. However, the problem of whether and to what extent this condition may be relaxed will not be taken up here.

[4] (II-2) means Eq. (2) of II.

The matrix elements of $\mathcal{H}$ in which we are interested are as follows:

$$\langle A_i^0|\mathcal{H}|\lambda^0\rangle=-\frac{2i\pi^{\frac{1}{2}}}{V^{\frac{1}{2}}}\nu_0\mu\epsilon_{\lambda i}\left(\frac{\hbar}{2\nu}\right)^{\frac{1}{2}}\exp(i\boldsymbol{\kappa}_\lambda\cdot\mathbf{r}_A),\tag{1}$$

$$\langle A_i^0|\mathcal{H}|B_j^0\rangle=\frac{4\pi}{V}\mu^2\sum_\kappa\frac{\kappa_i\kappa_j}{\kappa^2}\exp[i\boldsymbol{\kappa}\cdot(\mathbf{r}_A-\mathbf{r}_B)],\tag{2}$$

in which μ is the absolute value of the transition dipole matrix element, and $\mathbf{r}_A$ is the position vector of atom A. The summation in (2) goes over all vectors $\boldsymbol{\kappa}$ satisfying the periodic boundary conditions in the box, to which the $\boldsymbol{\kappa}_\lambda$ are also subject.

It is easily seen that (1) represents the usual absorption (or emission) matrix element in dipole approximation. For details the reader is referred to I or to the standard text by Heitler.[5] Equation (2) is the dipole-dipole interaction between the atoms, represented as a Fourier series. There are also some diagonal matrix elements of $\mathcal{H}$, coming from the A^2 term in the Hamiltonian. We neglect these entirely, since their contribution to the refractive index is of the same order of magnitude as that of the other excited states, which we have already decided to neglect.

Finally, we fix our zero of energy by setting $W_0=0$. For the energies of the other states, we put

$$\begin{aligned}W_\lambda&=\hbar\nu_\lambda=\hbar c\kappa_\lambda,\\ W_{A_i}&=\hbar\nu_0.\end{aligned}\tag{3}$$

It will be seen later that these energy assignments are self-consistent, i.e., they lead to a solution in which the correct energies agree with those originally assumed.

III. THE ARNOUS-BLEULER CANONICAL TRANSFORMATION

Our first task is to eliminate the "virtual" transitions. This can be done by means of the "finite energy shell" transformation first developed by Arnous and Bleuler.[6] Here we only summarize those of their results that we will need.

Any operator O may be represented as the sum of two operators,

$$O=O'+O'',$$

by means of the relations

$$\begin{aligned}\langle\alpha^0|O'|\beta^0\rangle&=0, \quad |W_\alpha-W_\beta|\geq\hbar\epsilon,\\ \langle\alpha^0|O''|\beta^0\rangle&=0, \quad |W_\alpha-W_\beta|<\hbar\epsilon.\end{aligned}$$

We choose ϵ in such a way that

$$\nu_0\gg\epsilon\gg\Delta\nu,\tag{4}$$

where $\Delta\nu$ is the width of the absorption line. It is also assumed that (4) would continue to hold if ν_0 were replaced by $|\nu'-\nu_0|$, where ν' is the frequency of one of

[5] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1954), 3rd ed., Chaps. II and V.

[6] E. Arnous and K. Bleuler, Helv. Phys. Acta **25**, 581 (1952).

the other transitions of the atoms of the gas. We now introduce the unitary transformation

$$|\alpha\rangle = \mathcal{S}|\alpha^0\rangle,$$

where $\mathcal{S}$ is determined by the condition

$$(\mathcal{S}^\dagger \mathcal{H} \mathcal{S})'' = 0,$$

i.e., in the representation of the state vectors $|\alpha\rangle$, the Hamiltonian has matrix elements only between states on the same energy shell, or in other words, only for real transitions. $\mathcal{S}$ may be expanded in powers of μ:

$$\mathcal{S} = 1 + \mathcal{S}^{(1)} + \mathcal{S}^{(2)} + \cdots. \tag{5}$$

One obtains the following values for the matrix elements of $\mathcal{S}^{(1)}$:

$$\mathcal{S}^{(1)\prime} = 0,$$

$$\langle \alpha^0 | \mathcal{S}^{(1)\prime\prime} | \beta^0 \rangle = \frac{\langle \alpha^0 | \mathcal{H}^{(1)\prime\prime} | \beta^0 \rangle}{W_\beta - W_\alpha}. \tag{6}$$

The Hamiltonian matrix elements in the new representation are, through the second order,

$$\langle \alpha | \mathcal{H} | \beta \rangle^{(1)} = \langle \alpha^0 | \mathcal{H}^{(1)\prime} | \beta^0 \rangle, \tag{7a}$$

$$\langle \alpha | \mathcal{H} | \beta \rangle^{(2)} = \langle \alpha^0 | \mathcal{H}^{(2)\prime} | \beta^0 \rangle + \sum\nolimits_\gamma \frac{\langle \alpha^0 | \mathcal{H}^{(1)} | \gamma^0 \rangle \langle \alpha^0 | \mathcal{H}^{(1)\prime\prime} | \beta^0 \rangle}{W_\beta - W_\gamma}. \tag{7b}$$

Of course, (7a) and (7b) only hold if $|W_\alpha - W_\beta| < \hbar\epsilon$. Otherwise, the Hamiltonian matrix elements are zero. There are two terms omitted from (7b), neither of which is of importance to us. One of these is important only near the edge of the energy shell, in which we are not interested; the other is smaller than those of (7b) by a factor $\sim \epsilon/\nu_0$. For further details, the reader should consult the original work of Arnous and Bleuler.[6]

To obtain the Hamiltonian for our problem, we substitute (1) and (2) into (6) and (7), and make use of (3) for the values of W. The result (through the second order in μ) is

$$\langle A_i | \mathcal{H} | \lambda \rangle = -\frac{2i\pi^{\frac{1}{2}}}{V^{\frac{1}{2}}} \nu_0 \mu \epsilon_{\lambda i} \left(\frac{\hbar}{2\nu_\lambda} \right)^{\frac{1}{2}} \exp(i\boldsymbol{\kappa}_\lambda \cdot \mathbf{r}_A), \tag{8}$$

$$\langle A_i | \mathcal{H} | B_j \rangle = \frac{4\pi}{V} \mu^2 \sum\nolimits_\kappa \frac{\kappa_i \kappa_j}{\kappa^2} \exp[i\boldsymbol{\kappa} \cdot (\mathbf{r}_A - \mathbf{r}_B)] + \frac{4\pi}{V} \nu_0^2 \mu^2 \sum\nolimits_\eta{}'' \frac{\epsilon_{\eta i} \epsilon_{\eta j} \exp[i\boldsymbol{\kappa}_\eta \cdot (\mathbf{r}_A - \mathbf{r}_B)]}{\nu_0^2 - \nu_\eta^2} - \frac{2\pi}{V} \nu_0^2 \mu^2 \sum\nolimits_\eta{}' \frac{\epsilon_{\eta i} \epsilon_{\eta j} \exp[i\boldsymbol{\kappa}_\eta \cdot (\mathbf{r}_A - \mathbf{r}_B)]}{\nu_\eta (\nu_0 + \nu_\eta)}. \tag{9}$$

It is clear that (8) only holds if $|\nu_\lambda - \nu_0| < \epsilon$, with the matrix element being zero otherwise. The double prime on the first sum in (9) means that only terms for which $|\nu_\eta - \nu_0| \geq \epsilon$ are to be included, while the sum with the single prime is restricted to terms for which $|\nu_\eta - \nu_0| < \epsilon$. There are also some diagonal elements, which are omitted for the same reason that the diagonal matrix elements were omitted from the Hamiltonian in the zero-order representation.

Now in the next section we will be interested in finding a stationary state of the system corresponding to the zero-order state in which a photon is present with $|\nu_\lambda - \nu_0| \lesssim \Delta\nu$. In view of (4), the change in (9) will be insignificant if we replace ν_0 in the denominators by $\nu_\lambda + i\xi$, where we will understand always the limit $\xi \to +0$. With this change, (9) becomes

$$\langle A_i | \mathcal{H} | B_j \rangle = \frac{4\pi}{V} \mu^2 \sum\nolimits_\kappa \frac{\kappa_i \kappa_j}{\kappa^2} \exp[i\boldsymbol{\kappa} \cdot (\mathbf{r}_A - \mathbf{r}_B)] + \frac{4\pi}{V} \nu_0^2 \mu^2 \sum\nolimits_\eta{}'' \frac{\epsilon_{\eta i} \epsilon_{\eta j} \exp[i\boldsymbol{\kappa}_\eta \cdot (\mathbf{r}_A - \mathbf{r}_B)]}{(\nu_\lambda + i\xi)^2 - \nu_\eta^2} - \frac{2\pi}{V} \nu_0^2 \mu^2 \sum\nolimits_\eta{}' \frac{\epsilon_{\eta i} \epsilon_{\eta j} \exp[i\boldsymbol{\kappa}_\eta \cdot (\mathbf{r}_A - \mathbf{r}_B)]}{\nu_\eta (\nu_\lambda + i\xi + \nu_\eta)}. \tag{9a}$$

Equations (8), (9a) give the Hamiltonian which we shall use from now on.

IV. SOLUTION BY DAMPING OPERATORS

Consider the state $|\lambda\rangle$, in which $\boldsymbol{\kappa}_\lambda$ is directed along the positive z axis, and $|\nu_\lambda - \nu_0| \lesssim \Delta\nu$. We wish to find the true stationary state $|\bar{\lambda}\rangle$ corresponding to this, if such a state exists: $(W + \mathcal{H})|\bar{\lambda}\rangle = \hbar\nu_\lambda |\bar{\lambda}\rangle$. For this purpose, one may use the "damping operators," introduced in II. For convenience, we summarize a few of their important properties here.

The transformation matrix S is defined by

$$S|\alpha\rangle = |\bar{\alpha}\rangle \text{ if } |\bar{\alpha}\rangle \text{ exists}, \quad = 0 \text{ otherwise}. \tag{II-4}$$

S may be represented as

$$S = (1+Q)N, \tag{II-6}$$

where N has only diagonal matrix elements, which may be considered as normalizing factors, and Q has only nondiagonal elements. The damping operator $\mathfrak{D}^{(ij\cdots)}(\epsilon)$ associated with the unperturbed states $i, j, \cdots$ and the energy ϵ is defined by

$$\mathfrak{D}^{(ij\cdots)}(\epsilon) = \mathcal{H} + \mathcal{H}(1 - P^{(ij\cdots)}) \times \zeta(\epsilon - W) \mathfrak{D}^{(ij\cdots)}(\epsilon). \tag{II-13}$$

Here $P(i,j,\cdots)$ is a projection operator on the linear manifold spanned by $i, j, \cdots$, and

$$\zeta(x) = \lim_{\xi \to +0} (x + i\xi)^{-1}.$$

It can then be shown that

$$\langle\beta|Q|\alpha\rangle=\frac{\langle\beta|\mathfrak{D}^{(\alpha,\beta)}(W_\alpha)|\alpha\rangle}{W_\alpha-W_\beta-\langle\beta|\mathfrak{D}^{(\alpha\beta)}(W_\alpha)|\beta\rangle}. \quad \text{(II-15)}$$

In our problem, the superscript on the $\mathfrak{D}$ operator is unimportant. It merely requires that a finite number of states be left out of certain summations. But we have so many states with essentially identical properties that the omission of a small number of them from a summation cannot have any significant effect. We will, therefore, omit the superscript from now on. If we do this, it can be shown that

$$\langle\beta|\mathfrak{D}(\epsilon)|\alpha\rangle=\langle\beta|\mathfrak{K}|\alpha\rangle+\sum_\gamma\frac{\langle\beta|\mathfrak{K}|\gamma\rangle\langle\gamma|\mathfrak{D}(\epsilon)|\alpha\rangle}{\epsilon-W_\gamma-\langle\gamma|\mathfrak{D}(\epsilon)|\gamma\rangle}. \quad \text{(II-19a)}$$

It is thus seen that the solution of the problem requires the evaluation of certain matrix elements of the damping operator, $\mathfrak{D}(\hbar\nu_\lambda)$, which from now on we write simply as $\mathfrak{D}$. We seek a solution such that

$$\langle A_i|\mathfrak{D}|\lambda\rangle=-\frac{2i\pi^{\frac{1}{2}}}{V^{\frac{1}{2}}}\nu_0\mu\epsilon_{\lambda i}\left(\frac{\hbar}{2\nu_\lambda}\right)^{\frac{1}{2}}\varphi(z_A), \quad (10)$$

$$\langle\eta|\mathfrak{D}|\eta\rangle=-i\xi, \quad \xi\to+0, \quad (11)$$

$$\langle A_i|\mathfrak{D}|A_i\rangle=\hbar\Delta, \quad (12)$$

where $\mathrm{Im}\Delta\leq 0$, and the value of Δ is independent of A and i, but may, of course, depend on ν_λ. By using (3), (II-19a), (11), and (12), we find

$$\langle A_i|\mathfrak{D}|\lambda\rangle=\langle A_i|\mathfrak{K}|\lambda\rangle+\sum_\eta{}'\frac{\langle A_i|\mathfrak{K}|\eta\rangle\langle\eta|\mathfrak{D}|\lambda\rangle}{\hbar(\nu_\lambda+i\xi-\nu_\eta)}+\sum_{B,j}\frac{\langle A_i|\mathfrak{K}|B_j\rangle\langle B_j|\mathfrak{D}|\lambda\rangle}{\hbar(\nu_\lambda-\nu_0-\Delta)}. \quad (13)$$

The matrix element $\langle\eta|\mathfrak{D}|\lambda\rangle$ may be eliminated from (13) by means of (II-19a), giving

$$\langle A_i|\mathfrak{D}|\lambda\rangle=\langle A_i|\mathfrak{K}|\lambda\rangle+\sum_{B,j}\frac{\langle A_i|\mathfrak{K}|B_j\rangle\langle B_j|\mathfrak{D}|\lambda\rangle}{\hbar(\nu_\lambda-\nu_0-\Delta)}, \quad (14)$$

where

$$\langle A_i|\mathfrak{K}|B_j\rangle=\langle A_i|\mathfrak{K}|B_j\rangle+\sum_\eta{}'\frac{\langle A_i|\mathfrak{K}|\eta\rangle\langle\eta|\mathfrak{K}|B_j\rangle}{\hbar(\nu_\lambda+i\xi-\nu_\eta)}. \quad (15)$$

With the help of (8) and (9a), the matrix element of $\mathfrak{K}$ can be evaluated, with the result

$$\langle A_i|\mathfrak{K}|B_j\rangle=\frac{4\pi}{V}\mu^2\sum_\kappa\frac{\kappa_i\kappa_j}{\kappa^2}\exp[i\boldsymbol{\kappa}\cdot(\mathbf{r}_A-\mathbf{r}_B)]+\frac{4\pi}{V}\nu_0^2\mu^2\sum_\eta\frac{\epsilon_{\eta i}\epsilon_{\eta j}\exp[i\boldsymbol{\kappa}_\eta\cdot(\mathbf{r}_A-\mathbf{r}_B)]}{(\nu_\lambda+i\xi)^2-\nu_\eta^2}, \quad (16)$$

where the summation over η is now unrestricted. One can now substitute (8), (10), and (16) into (14), and cancel out a common factor $-(2i\pi^{\frac{1}{2}}/V^{\frac{1}{2}})\nu_0\mu(\hbar/2\nu_\lambda)^{\frac{1}{2}}$ to obtain

$$\epsilon_{\lambda i}\varphi(z_A)=\epsilon_{\lambda i}\exp(i\kappa_\lambda z_A)+\frac{4\pi}{V}\frac{\mu^2}{\hbar(\nu_\lambda-\nu_0-\Delta)}\sum_{B,j}\left\{\sum_\kappa\frac{\kappa_i\kappa_j}{\kappa^2}\exp[i\boldsymbol{\kappa}\cdot(\mathbf{r}_A-\mathbf{r}_B)]+\nu_0^2\sum_\eta\frac{\epsilon_{\eta i}\epsilon_{\eta j}\exp[i\boldsymbol{\kappa}_\eta\cdot(\mathbf{r}_A-\mathbf{r}_B)]}{(\nu_\lambda+i\xi)^2-\nu_\eta^2}\right\}\epsilon_{\lambda j}\varphi(z_B).$$

We now replace summation over B by integration over the uniform distribution of atoms: $\sum_B\to\mathfrak{N}\times\int dx_B dy_B dz_B$. The x and y integrations can be carried out immediately, giving (due to the imaginary exponential factor) zero for all terms in the summations except those for which $\boldsymbol{\kappa}$ is directed along the z axis. For these remaining terms, the first summation vanishes on summing over j, since $\epsilon_{\lambda z}=0$. As for the second summation, we are left only with terms for which $\boldsymbol{\epsilon}_\eta=\boldsymbol{\epsilon}_\lambda$. We can now cancel a common factor $\epsilon_{\lambda i}$ and obtain

$$\varphi(z_A)=\exp(i\kappa_\lambda z_A)+\frac{\Lambda\nu_0^2}{L}\int_0^{L\alpha\to\infty}\left[\sum_\eta{}^*\frac{\exp[i\kappa_\eta(z_A-z_B)]}{(\nu_\lambda+i\xi)^2-\nu_\eta^2}\right]\varphi(z_B)dz_B,$$

where

$$\Lambda=4\pi\mathfrak{N}\mu^2/\hbar(\nu_\lambda-\nu_0-\Delta). \quad (17)$$

The asterisk on the summation in the above expression indicates that the summation is restricted to those η for which $\boldsymbol{\kappa}_\eta$ is directed along the z axis, and $\boldsymbol{\epsilon}_\eta=\boldsymbol{\epsilon}_\lambda$. We can now replace summation over η by integration:

$$\sum_\eta\to\frac{L}{2\pi}\int_{-\infty}^{\infty}d\kappa_\eta.$$

The integration is elementary, and leads to the final result[7]

$$\varphi(z_A)=\exp(i\kappa_\lambda z_A)-\frac{i\kappa_\lambda\Lambda}{2}\int_0^\infty\exp[i\kappa_\lambda|z_A-z_B|]\varphi(z_B)dz_B. \quad (18)$$

[7] Strictly speaking, there is a slight difficulty in passing from summation over η to integration in the derivation of (18). If we had kept the superscripts on $\mathfrak{D}$, it would have been necessary to omit $\eta=\lambda$ from the summation; but it is just this singular point that gives the main contribution to the integral. However, in a real situation, the volume occupied by the gas will always be finite, while the radiation field is quantized in a universe of infinite volume. Hence, the integration over x_B and y_B above does not really result in restricting the sum over η to $\kappa_{\eta x}=\kappa_{\eta y}=0$, but only restricts these components to a small but finite range about zero. Therefore, the summation really includes an infinite number of η for which ν_η is arbitrarily close to ν_λ, but for which the direction of $\boldsymbol{\kappa}_\eta$ is slightly different from $\boldsymbol{\kappa}_\lambda$. In this situation, omission of one term from the sum (one point from the integration) makes no difference.

In arriving at (18), we have also made use of (4) in setting $\nu_0/\nu_\lambda \cong 1$.

Now it is easily seen that the integral equation (18) has the solution

$$\varphi(z)=\frac{2}{\rho+1}\exp(i\rho\kappa_\lambda z), \quad z>0$$
$$=\exp(i\kappa_\lambda z)-\left(\frac{\rho-1}{\rho+1}\right)\exp(-i\kappa_\lambda z), \quad z<0, \quad (19)$$

where

$$\rho^2-1=-\Lambda, \quad \mathrm{Im}\rho\geq 0. \quad (20)$$

The value of φ for $z<0$ is unimportant in the present context, but its meaning will become clear presently.

It is now possible to verify directly that (11) is satisfied. This is done in the Appendix. According to (11), in the case $\eta=\lambda$, the self-energy of the state $|\lambda\rangle$ is zero, in agreement with the first Eq. (3). In the following paper, Δ is evaluated. In general, it has a nonvanishing imaginary part, so that, in accordance with the criterion of II, no real self-energy can be defined for the state $|A_i\rangle$, and there is no true stationary state corresponding to it. Hence W_{A_i} is at our disposal, and, in particular, may be given the value of the second Eq. (3).

We are now interested in evaluating the matrix elements of the vector potential $\mathbf{A}(\mathbf{r})$ corresponding to creation or destruction of a physical photon. We have

$$\langle\bar{0}|\mathbf{A}(\mathbf{r})|\bar{\lambda}\rangle=\langle 0|\mathbf{A}(\mathbf{r})|\bar{\lambda}\rangle=\langle 0|\mathbf{A}(\mathbf{r})S|\lambda\rangle$$
$$=\langle 0|\mathbf{A}(\mathbf{r})(1+Q)|\lambda\rangle N_\lambda, \quad (21)$$

in which use is made of the fact that, since the ground state $|0\rangle$ is alone on its energy shell, $|\bar{0}\rangle=|0\rangle$. We have also used (II-4) and (II-6). Writing out the matrix elements, we find

$$\langle 0|\mathbf{A}(\mathbf{r})|\bar{\lambda}\rangle=\{\langle 0|\mathbf{A}(\mathbf{r})|\lambda\rangle+\textstyle\sum_\eta{}'\langle 0|\mathbf{A}(\mathbf{r})|\eta\rangle\langle\eta|Q|\lambda\rangle$$
$$+\textstyle\sum_{B,i}\langle 0|\mathbf{A}(\mathbf{r})|B_i\rangle\langle B_i|Q|\lambda\rangle\}N_\lambda. \quad (22)$$

To evaluate approximately the matrix elements of $\mathbf{A}$, we use (1), (3), (6) and (22):

$$\langle 0|\mathbf{A}(\mathbf{r})|\eta\rangle=\langle 0^0|\mathbf{A}(\mathbf{r})|\eta^0\rangle=\frac{2c\pi^{\frac{1}{2}}}{V^{\frac{1}{2}}}\left(\frac{\hbar}{2\nu_\eta}\right)^{\frac{1}{2}}\boldsymbol{\varepsilon}_\eta$$
$$\times\exp(i\boldsymbol{\kappa}_\eta\cdot\mathbf{r}); \quad (23)$$

$$\langle 0|\mathbf{A}(\mathbf{r})|B_i\rangle=\textstyle\sum_\eta\langle 0^0|\mathbb{S}^\dagger|(B_i,\eta)^0\rangle\langle(B_i,\eta)^0|\mathbf{A}(\mathbf{r})|B_i^0\rangle$$
$$+\textstyle\sum_\eta{}''\langle 0^0|\mathbf{A}(\mathbf{r})|\eta^0\rangle\langle\eta^0|\mathbb{S}|B_i^0\rangle$$
$$=\frac{4c\pi}{V}i\nu_0\mu\textstyle\sum_\eta{}''\dfrac{\boldsymbol{\epsilon}_{\eta i}\boldsymbol{\varepsilon}_\eta}{(\nu_\lambda+i\xi)^2-\nu_\eta^2}$$
$$\times\exp[i\boldsymbol{\kappa}_\eta\cdot(\mathbf{r}-\mathbf{r}_B)]$$
$$-\frac{2c\pi}{V}i\nu_0\mu\textstyle\sum_\eta{}'\dfrac{\boldsymbol{\epsilon}_{\eta i}\boldsymbol{\varepsilon}_\eta}{\nu_\eta(\nu_\lambda+i\xi+\nu_\eta)}$$
$$\times\exp[i\boldsymbol{\kappa}_\eta\cdot(\mathbf{r}-\mathbf{r}_B)]. \quad (24)$$

In deriving (24), we have replaced ν_0 by $\nu_\lambda+i\xi$, as before. We can now substitute (23) and (24) for the $\mathbf{A}$ matrix elements in (22), and (II-15) for the Q elements. Just as in the derivation of (18), we use (10), (11), and (12), eliminate $\langle\eta|\mathfrak{D}|\lambda\rangle$ by means of (II-19a), and combine terms. Making use also of (18), we obtain

$$\langle\bar{0}|\mathbf{A}(\mathbf{r})|\bar{\lambda}\rangle=\left\{\frac{2c\pi^{\frac{1}{2}}}{V^{\frac{1}{2}}}\left(\frac{\hbar}{2\nu_\lambda}\right)^{\frac{1}{2}}\boldsymbol{\varepsilon}_\lambda\varphi(z)\right\}N_\lambda. \quad (25)$$

According to (25), the matrix element of $\mathbf{A}$ for destruction of a physical photon has the form of an attenuated wave in the medium with complex refractive index given by ρ. Referring to (19), we see that outside the medium ($z<0$), the vector potential consists of an incident and reflected wave, and it is easily verified that the various coefficients are the same as one would obtain in the classical theory by applying the appropriate boundary conditions. Hence, analogously to the case treated in I, the phenomenological Maxwell equations are obeyed by the destruction (or creation) matrix elements of the field operators in the representation of the true stationary states.[8] If more than one photon is present in the zero-order state, the result will be the same, except insofar as the refractive index depends on the intensity of the beam.

V. DISCUSSION

Equations (17) and (20) show that the refractive index may be calculated if Δ is known. The evaluation of Δ as a function of ν_λ is taken up in the next paper. Since the absorption coefficient is essentially given by the imaginary part of ρ, this leads to a formally exact theory of the absorption line shape. Since Δ is in general complex, it follows from the results of II that no true stationary state exists corresponding to $|A_i\rangle$. The stationary state $|\bar{\lambda}\rangle$ does exist, however.

We have therefore found some of the true stationary states of the system, but apparently not all of them; the states $|\bar{\lambda}\rangle$ do not form a complete set. (For instance, the state $|A_i\rangle$ cannot be expanded in terms of them if atom A is located well inside the medium.) If the other stationary states could be found, it would lead, among other things, to a theory of Čerenkov radiation in the region of anomalous dispersion which would be analogous to that given in I for the case of no absorption.

A limitation on the method used here is the use of

[8] There is one difference between the situation here and that in I. In I, the "physical" photons had the same wave numbers as the corresponding "bare" photons, with different frequencies, while here the reverse is true: The frequencies are the same, but the wave numbers of the physical photons are different from those of the bare ones, and are in general complex. An attempt to reproduce the situation of I in the case considered here would have resulted in the frequencies being complex, in contradiction to the assumption that the physical photons represent true stationary states.

Eq. (II-19a), and the replacing of summation over atoms by integration. (II-19a) assumes that the omission of any one or two atoms from a sum makes no difference, while the use of an integration requires that the distribution is, on the average, uniform. Thus, the method would appear to be justified in the case of a random distribution (as in a gas), but not in that of a regular array, as in a crystal.

APPENDIX: VERIFICATION OF (11)

Without loss of generality, we may assume that κ_η is directed along the positive z axis. Then if we put

$$\langle A_i|\mathfrak{D}|\eta\rangle=-\frac{2i\pi^{\frac{1}{2}}}{V^{\frac{1}{2}}}\nu_0\mu\epsilon_{\eta i}\left(\frac{\hbar}{2\nu_\eta}\right)^{\frac{1}{2}}\psi(z_A), \tag{A-1}$$

we arrive at the integral equation

$$\psi(z)=\exp(i\kappa_\eta z)-\frac{ia\kappa_\eta\Lambda}{2}\int_0^\infty \exp[ia\kappa_\eta|z-z'|]\psi(z')dz', \tag{A-2}$$

where $a=\nu_\lambda/\nu_\eta$. The derivation of (A-2) is exactly like that of (18). Its solution is

$$\psi(z)=p\exp(i\kappa_\eta z)+q\exp(ia\rho\kappa_\eta z), \tag{A-3}$$

where, except for terms of relative order of magnitude $(1-a)$,

$$p=(a^2-1)/(\rho^2-1),\quad q=2/(\rho+1). \tag{A-4}$$

We can now combine (II-19a), (8), (12), (A-1), and (A-3) to obtain

$$\langle\eta|\mathfrak{D}|\eta\rangle=\frac{2\pi}{V}\frac{\nu_0^2\mu^2}{\nu_\eta(\nu_\lambda-\nu_0-\Delta)}\times\sum_A\{p+q\exp[i(a\rho-1)\kappa_\eta z_A]\}. \tag{A-5}$$

In (A-5), we can now replace summation by integration, and make use of (A-4), (17), and (20). If we again neglect terms of relative order of magnitude $(1-a)$, we find

$$\langle\eta|\mathfrak{D}|\eta\rangle\approx\tfrac{1}{2}h\nu_0\left\{\alpha(1-a^2)-\frac{2i}{L\kappa_\eta}\right\}. \tag{A-6}$$

The imaginary term in (A-6) has the desired form (11), since we are interested in the limit $L\to\infty$. The real term is proportional to α, the fraction of the volume of our normalization box which is filled with matter. Now any actual sample of matter must be of finite size, while the normalization box has significance only in the limit $L\to\infty$. Therefore we should take the limit $\alpha\to 0$, in which case the real term in (A-6) vanishes. Of course, this does not interfere with taking the limit $\alpha L\to\infty$, as in (18). What this means is that the dimensions of our sample are very large compared with all wavelengths, etc., but very small compared with the dimensions of the "universe" in which the quantization of the field is carried out. This completes the verification of (11).

It is interesting to observe that the spurious nature of the real term in (A-6) might not have been noticed if we had considered our box to be completely filled with matter. This, together with the point mentioned in footnote,[7] would seem to indicate that some care must be taken in the treatment of "infinite media" by this method, lest unphysical terms appear in the results.