all the nF states, a ratio of 1.67 would result for the population of the $4\,^1D$ to $4\,^3D$ state due to the multiple state transfer process. The actual value of the population ratio should thus lie between 1.18 and 1.67. To compare this with our experimental data we first subtract from the measured atomic densities of $4\,^1D$ and $4\,^3D$ the amount of population produced by direct excitation and cascading from the P states. In this manner we obtain 1.4 as the ratio of the population of $4\,^1D$ vs $4\,^3D$ due to collisional excitation transfer only. The close agreement here indicates that the multiple transfer process is responsible for the major part of the pressure-dependent population of the $4\,^1D$ state.

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Complex Refractive Index of an Ideal Monatomic Gas*

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The main results of some previous work of the author on this subject are rederived in a more rigorous fashion. In particular, no appeal is made to perturbation theory or to the author's "damping operator" formalism, and closer attention is paid to the question of proper averaging over configurations of absorbers. All effects due to translational motion of absorbers are neglected, but the dipolar "resonance" interactions are included. It is shown that the formal results of the previous work, which permit a detailed calculation of the absorption line shape, are valid under the following assumptions: (a) The average optical behavior of the gas is describable in terms of a refractive index; (b) there is no correlation between the positions of different absorbers, i.e., the gas is ideal. However, it is also shown that the cutoff procedure for handling a divergent integral which appears in the theory must be modified. The modified cutoff procedure leads to a large increase in the theoretically predicted linewidth, in qualitative agreement with experiment.

I. INTRODUCTION AND SUMMARY

IN a previous pair of papers,[1] the author has presented a theory of the complex refractive index of an ideal monoatomic gas. The treatment ignored effects of translational motion of the absorbers (atoms), but included the dipolar "resonance" interactions between different absorbers. The formalism used was that of "damping operators," previously introduced by the author.[2] The calculated formula for the linewidth was in agreement with previous theories,[3] but not with recent experiments.[4]

There are two possible grounds on which the results of A and B might be questioned. First, there is the "damping operator" formalism itself. This formalism, while not particularly difficult, is nevertheless unfamiliar to most physicists, and might be regarded with suspicion by some on that account. A more solid basis for skepticism is the fact that the development of the formalism[2] depends on a number of rearrangements of the perturbation theory expansion for the true stationary state; it follows that the basic equations for the damping operators have been rigorously derived only for the case where the perturbation expansion converges absolutely, though the author feels that they are probably valid under more general conditions.

The second ground for doubt concerns the question of averaging. To make this clear, we note that in the quantum mechanical formalism the phenomenological Maxwell equations are supposed to be obeyed in some sense by those matrix elements of the various field operators which correspond to creation or destruction of a "dressed" photon. For example, the refractive index $\rho(\nu_\lambda)$ is defined by

$$4\pi\langle 0|P_i(\mathbf{r})|\bar{\lambda}\rangle=[\rho^2(\nu_\lambda)-1]\langle 0|E_i(\mathbf{r})|\bar{\lambda}\rangle, \qquad (1)$$

where $|0\rangle$ is the ground state (no absorbers excited, no photons present), and $|\bar{\lambda}\rangle$ is a dressed photon state. $P_i(\mathbf{r})$ and $E_i(\mathbf{r})$ are the operators for the i-component of dipole moment density and electric field, respectively, at the point $\mathbf{r}$. (For further details on notation, see Secs. A-II and A-IV.) The matrix elements appearing on both sides of Eq. (1) are to be interpreted as averages of some kind; and in the case of a gas with translational motion neglected, the average should be over all allowed spatial configurations of the absorbers. Once this average is taken, there is no need for the customary further averaging over a "physically small" region; the necessary "smoothing out" is already accomplished

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[1] C. A. Mead, Phys. Rev. **120**, 854, 860 (1960), hereafter denoted by A and B, respectively. Equations from these papers will be referred to by expressions such as (A-3) meaning Eq. (3) of A; sections will be similarly denoted; e.g., Sec. B-II is Sec. II of B.
[2] C. A. Mead, Phys. Rev. **112**, 1843 (1958).
[3] See, e.g., R. G. Breene, Jr., Revs. Modern Phys. **29**, 94 (1957).
[4] J. Tomiser, Acta Phys. Austriaca **8**, 198 (1953); **8**, 276 (1954); **8**, 323 (1954); **9**, 18 (1954). H. Moser and H. Schultz, Ann. Physik **7**, 243 (1959).

by averaging over configurations.[5] The procedure used in A and B, however, was rather careless about the averaging, and it is not clear under what conditions, if any, the results represent the correct average behavior of the gas.

The aim of the present paper is to rederive the key equations of A and B without appealing to the damping operator formalism or to perturbation theory; and with proper treatment of the averaging. We will make two assumptions:

(a) The average behavior of the gas can be described by a refractive index; i.e., Eq. (1) holds for all $\mathbf{r}$ in the region occupied by the gas with the correct choice of ρ and with the matrix elements interpreted as averages over configurations.

(b) There is no correlation at all between the positions of different absorbers, so that each may be averaged independently, i.e., the gas is ideal. This will lead to divergences due to the singular character of the dipole-dipole interaction as the distance between absorbers becomes small. The divergence will be ignored at first, and later a cutoff procedure will be introduced.

It will be shown that the formal equations by which the refractive index was evaluated in A and B remain unchanged, but that the cutoff procedure used in Sec. B-III for handling the divergences must be modified. The new cutoff procedure appears to greatly improve the agreement with experiment. In addition to bringing the results of A and B into closer agreement with experiment, and making them more rigorous, it is hoped that (by avoiding explicit use of the damping operator method) these results will be made intelligible to a wider audience. Moreover, the results of the present paper should strengthen our confidence in the damping operator method for other situations in which a detailed justification is more difficult.

The plan of the paper is as follows: In Sec. II some notation is introduced and some preliminary matters discussed. Section III is concerned with the formal derivation of the key equations of A and B, ignoring temporarily the fact that divergences might appear. In Sec. IV, the cutoff procedure is discussed, and the results briefly and qualitatively compared with experiment.

II. NOTATION, ETC.

The notation will be the same as that described in Sec. A-II, with the following changes: Instead of quantizing the radiation field with periodic boundary conditions in a finite volume, it will be treated as a continuum. Hence, sums over photon states will be replaced by integrals; these will be denoted by $\int d\eta$, and it will be understood that this includes a sum over polarizations as well as an integration over bare photon wave numbers. We consider a very large number N of absorbers distributed over a fixed volume, v, with the average density $N/v=\mathfrak{N}$. We are interested in the limit $v\to\infty$ with $\mathfrak{N}$ constant. The procedure, in principle, is to calculate the desired properties of the dressed photon state $|\bar{\lambda}\rangle$ for a fixed but arbitrary configuration of absorbers (arbitrary except that all absorbers, of course, must be within the volume v), and then take averages of quantities of interest to us over all configurations. In a given configuration, the position of each absorber is treated as a fixed c-number, but all other degrees of freedom are treated quantum mechanically.

Averages over configurations will be denoted by curly brackets, followed by an expression in square brackets indicating which absorbers, if any, are to be held fixed during the averaging. For example,

$$\{\langle 0|E_i(\mathbf{r})|\bar{\lambda}\rangle\}[A(\mathbf{r}')]$$

denotes the average of $\langle 0|E_i(\mathbf{r})|\bar{\lambda}\rangle$ over all configurations for which absorber A is located at the point $\mathbf{r}'$. If there are no square brackets, the average is over all configurations without restriction.

We can write the total Hamiltonian as

$$\mathfrak{IC}_{\text{tot}}=W+\mathfrak{IC},$$

where W is completely diagonal and $\mathfrak{IC}$ has no diagonal elements. After performing the Arnous-Bleuler transformation (cf. Sec. A-III), the only states that need be considered are of the form $|A_i\rangle$ (atom A excited, "polarization" in i direction, no photons present) and $|\eta\rangle$ (all atoms in ground state, photon of type η present), where the photon state $|\eta\rangle$ is on the energy shell, i.e.,

$$|\nu_\eta-\nu_0|<\epsilon\ll\nu_0,$$

(ν_0=resonance frequency). We shall always ignore terms which are $O(\epsilon/\nu_0)$. Henceforth, all integrals over photon states ($\int d\eta$) will be understood to include only those states on the energy shell, unless otherwise indicated.

Apart from trivial modifications brought about by treating the field as a continuum, the matrix elements $\langle A_i|\mathfrak{IC}|\eta\rangle$ and $\langle A_i|\mathfrak{IC}|B_j\rangle$ are given through the second order in the dipole matrix element μ by Eqs. (A-8) and (A-9) or (A-9a). Although the detailed expressions are not actually needed in the present work, we reproduce them for the convenience of the reader, in a form modified to take account of the treatment of the field as a continuum:

$$\langle A_i|\mathfrak{IC}|\lambda\rangle = -\frac{i}{\pi\sqrt{2}}\nu_0\mu\epsilon_{\lambda i}\left(\frac{\hbar}{2\nu_\lambda}\right)^{1/2}\exp(i\boldsymbol{\kappa}_\lambda\cdot\mathbf{r}_A); \tag{A-8}$$

[5] Averaging over a physically small region containing a large number of absorbers is necessary only if one wishes to make the fluctuations small. In this paper we are concerned only with the averages, and make no attempt to evaluate the fluctuations.

$$\langle A_i|\mathcal{H}|B_j\rangle$$
$$=\frac{1}{2\pi^2}\mu^2\int\frac{\kappa_i\kappa_j}{\kappa^2}\exp[i\boldsymbol{\kappa}\cdot(\mathbf{r}_A-\mathbf{r}_B)]d^3\boldsymbol{\kappa}$$
$$+\frac{1}{2\pi^2}\nu_0^2\mu^2\int''\frac{\epsilon_{\eta i}\epsilon_{\eta j}\exp[i\boldsymbol{\kappa}_\eta\cdot(\mathbf{r}_A-\mathbf{r}_B)]d\eta}{\nu_0^2-\nu_\eta^2}$$
$$-\frac{1}{4\pi^2}\nu_0^2\mu^2\int'\frac{\epsilon_{\eta i}\epsilon_{\eta j}\exp[i\boldsymbol{\kappa}_\eta\cdot(\mathbf{r}_A-\mathbf{r}_B)]d\eta}{\nu_\eta(\nu_0+\nu_\eta)};\quad\text{(A-9)}$$

$$\langle A_i|\mathcal{H}|B_j\rangle$$
$$=\frac{1}{2\pi^2}\mu^2\int\frac{\kappa_i\kappa_j}{\kappa^2}\exp[i\boldsymbol{\kappa}\cdot(\mathbf{r}_A-\mathbf{r}_B)]d^3\boldsymbol{\kappa}$$
$$+\frac{1}{2\pi^2}\nu_0^2\mu^2\int''\frac{\epsilon_{\eta i}\epsilon_{\eta j}\exp[i\boldsymbol{\kappa}_\eta\cdot(\mathbf{r}_A-\mathbf{r}_B)]d\eta}{(\nu_\lambda+i\xi)^2-\nu_\eta^2}$$
$$-\frac{1}{4\pi^2}\nu_0^2\mu^2\int'\frac{\epsilon_{\eta i}\epsilon_{\eta j}\exp[i\boldsymbol{\kappa}_\eta\cdot(\mathbf{r}_A-\mathbf{r}_B)]d\eta}{\nu_\eta(\nu_\lambda+i\xi+\nu_\eta)}.\quad\text{(A-9a)}$$

In these expressions, μ is the absolute value of the dipole moment matrix element, $\boldsymbol{\kappa}_\lambda$ and $\boldsymbol{\epsilon}_\lambda$ are, respectively, the wave-number vector and polarization unit vector associated with photons of the type λ, and the i components of these vectors are denoted by subscripts. The position vector of absorber A in a particular configuration is denoted by $\mathbf{r}_A$. A single prime on an integral means that the range of integration is restricted by the requirement $|\nu_\eta-\nu_0|<\epsilon$, while a double prime means $|\nu_\eta-\nu_0|\geq\epsilon$. The limit $\xi\to+0$ is always understood. Equation (A-9a), the expression which is actually used, differs from the exact expression (A-9) only by terms of relative order of magnitude $|\nu_\lambda-\nu_0|/\nu_0$.

The matrix elements of W are

$$\langle A_i|W|A_i\rangle=\hbar\nu_0,$$
$$\langle\lambda|W|\eta\rangle=\hbar\nu_\lambda\delta(\lambda-\eta),\quad(2)$$

where $\delta(\lambda-\eta)$ includes a Kronecker δ for the polarizations as well as a Dirac δ function for the wave numbers.

There are also matrix elements $\langle\eta|\mathcal{H}|\lambda\rangle$, originating both from the $\mathcal{A}^2$ term in the Hamiltonian ($\mathcal{A}$=vector potential) and from the second-order Arnous-Bleuler transformation. However, these are all of the form

$$\langle\eta|\mathcal{H}|\lambda\rangle=\text{const}\sum_A\exp[i(\boldsymbol{\kappa}_\lambda-\boldsymbol{\kappa}_\eta)\cdot\mathbf{r}_A].$$

The constant is zero if the polarizations associated with η and λ are mutually perpendicular. For N and v large, and for almost all configurations, moreover, the sum is very small except for a sharp peak where the two wave-number vectors are nearly equal. As v, N become infinite, this sharp peak approaches a delta function for all but a small fraction of the allowed configurations, and this fraction goes to zero. Hence, these matrix elements really only give a correction to the diagonal matrix elements of W. This correction is small, its effect on the final results being of the same order of magnitude as that of higher order corrections to the off-diagonal matrix elements. All these corrections essentially give the effect on the refractive index of virtual nonresonance processes, and may be included in a straightforward manner by perturbation theory. We will simply ignore them here, as was done in A and B. Hence, all the matrix elements we shall use are those given by (A-8), (A-9), (A-9a), and (2).

For a particular configuration of absorbers, we are interested in constructing a clothed photon state on the energy shell $|\bar{\lambda}\rangle$, defined by

$$\mathcal{H}_{\text{tot}}|\bar{\lambda}\rangle=\hbar\nu_\lambda|\bar{\lambda}\rangle\quad(3)$$

$$|\lambda\rangle=|\lambda\rangle+\int|\eta\rangle d\eta\langle\eta|\lambda\rangle+\sum_A|A_i\rangle\langle A_i|\lambda\rangle.\quad(4)$$

(We always sum over repeated indices.)

We shall have to deal with certain matrix elements of the electric field operator. Later manipulations will be facilitated by noting that, in Coulomb gauge,

$$\langle 0|E_i(\mathbf{r})|\bar{\lambda}\rangle=\langle 0|E_{ci}(\mathbf{r})|\bar{\lambda}\rangle+(i\nu_\lambda/c)\langle 0|\mathcal{A}_i(\mathbf{r})|\bar{\lambda}\rangle,$$

where $E_{ci}(\mathbf{r})$ is the i component of the electrostatic Coulomb field operator, and $\mathcal{A}_i(\mathbf{r})$ is the i component of the transverse vector potential operator. Hence we have

$$\langle 0|E_i(\mathbf{r})|\bar{\lambda}\rangle=\langle 0|\mathcal{E}_i(\mathbf{r})|\bar{\lambda}\rangle,\quad(5)$$

where

$$\mathcal{E}_i(\mathbf{r})=E_{ci}(\mathbf{r})+(i\nu_\lambda/c)\mathcal{A}_i(\mathbf{r}).\quad(6)$$

We also note that, apart from terms of relative order of magnitude ϵ/ν_0,

$$\langle A_i|\mathcal{H}|\eta\rangle=-\mu\langle 0|\mathcal{E}_i(\mathbf{r}_A)|\eta\rangle,$$
$$\langle A_i|\mathcal{H}|B_j\rangle=-\mu\langle 0|\mathcal{E}_i(\mathbf{r}_A)|B_j\rangle.\quad(7)$$

Equation (7) is obvious so far as the contribution of the electrostatic Coulomb interaction is concerned. As for the transverse contribution, we note that Eq. (A-8), for example, may be written

$$\langle A_i|\mathcal{H}|\eta\rangle=-\frac{i\mu\nu_0}{c}\langle 0|\mathcal{A}_i(\mathbf{r}_A)|\eta\rangle$$
$$=-\mu\frac{\nu_0}{\nu_\lambda}\langle 0|\mathcal{E}_i(\mathbf{r}_A)|\eta\rangle$$
$$=-\mu\langle 0|\mathcal{E}_i(\mathbf{r}_A)|\eta\rangle+O\left(\frac{\nu_\lambda-\nu_0}{\nu_0}\right).$$

The procedure is similar for the transverse contributions to (A-9) or (A-9a).

Now with the aid of (5), Eq. (1) may be written in more precise form:

$$4\pi\{\langle 0|P_i(\mathbf{r})|\bar{\lambda}\rangle\}=[\rho^2(\nu_\lambda)-1]\{\langle 0|\mathcal{E}_i(\mathbf{r})|\bar{\lambda}\rangle\}.\quad(8)$$

The dipole moment density operator $P_i(\mathbf{r})$ may be expressed in detail as

$$P_i(r)=\sum_A \mu_{Ai}\delta(\mathbf{r}-\mathbf{r}_A),$$

where μ_{Ai} is the operator for the i component of the electric dipole moment of absorber A. We now make the following definitions:

$$\begin{aligned}\mathfrak{E}_i(\mathbf{r})&=\{\langle 0|\,\mathcal{E}_i(\mathbf{r})\,|\bar{\lambda}\rangle\};\\ \mathfrak{P}_i(\mathbf{r})&=\{\langle 0|\,P_i(\mathbf{r})\,|\bar{\lambda}\rangle\};\\ F_i(\mathbf{r})&=\{\langle A_i|\bar{\lambda}\rangle\}[A(\mathbf{r})].\end{aligned}\tag{9}$$

By using (9), one can easily see that

$$\mathfrak{P}_i(\mathbf{r})=\mathfrak{N}\mu F_i(\mathbf{r}).\tag{10}$$

Equation (8) now takes the form

$$4\pi\mathfrak{P}_i(\mathbf{r})=4\pi\mathfrak{N}\mu F_i(\mathbf{r})=[\rho^2(\nu_\lambda)-1]\mathfrak{E}_i(\mathbf{r}).\tag{11}$$

We are now ready to evaluate the refractive index, and in what follows we will make frequent use of Eqs. (7) and (11). Equation (11) now expresses assumption (a) of Sec. I. That is, we assume that (11) holds for all r and v, but we make no *a priori* assumptions about the value of $\rho(\nu_\lambda)$, except that it is independent of $\mathbf{r}$.

III. EVALUATION OF REFRACTIVE INDEX

By substituting (4) into (3), taking the inner product with $\langle\eta|$, and using (2), one finds (for a particular configuration)

$$\hbar(\nu_\lambda-\nu_\eta)\langle\eta|\bar{\lambda}\rangle=\sum_B\langle\eta|\mathcal{H}|B_j\rangle\langle B_j|\bar{\lambda}\rangle.$$

This has the solution

$$\langle\eta|\bar{\lambda}\rangle=\sum_B\frac{\langle\eta|\mathcal{H}|B_j\rangle\langle B_j|\lambda\rangle}{\hbar(\nu_\lambda-\nu_\eta+i\xi)},\tag{12}$$

in which the limit $\xi\to 0+$ is understood. This guarantees that our solution will be the correct "retarded" one. Similarly, if one takes the inner product with $\langle A_i|$, one finds

$$\hbar(\nu_\lambda-\nu_0)\langle A_i|\lambda\rangle=\langle A_i|\mathcal{H}|\lambda\rangle+\sum_{B\neq A}\langle A_i|\mathcal{H}|B_j\rangle\langle B_j|\lambda\rangle+\int\langle A_i|\mathcal{H}|\eta\rangle d\eta\langle\eta|\bar{\lambda}\rangle,$$

which, when combined with (12), becomes

$$\hbar(\nu_\lambda-\nu_0)\langle A_i|\lambda\rangle=\langle A_i|\mathcal{H}|\lambda\rangle+\sum_{B\neq A}\left[\langle A_i|\mathcal{H}|B_j\rangle+\int\frac{\langle A_i|\mathcal{H}|\eta\rangle d\eta\langle\eta|\mathcal{H}|B_j\rangle}{\hbar(\nu_\lambda-\nu_\eta+i\xi)}\right]\langle B_j|\bar{\lambda}\rangle-i\hbar\sigma_{ij}\langle A_j|\lambda\rangle.\tag{13}$$

Here

$$-i\hbar\sigma_{ij}=\int\frac{\langle A_i|\mathcal{H}|\eta\rangle d\eta\langle\eta|\mathcal{H}|A_j\rangle}{\hbar(\nu_\lambda-\nu_\eta+i\xi)}\cong-i\hbar\sigma\delta_{ij},\tag{14}$$

where σ is the natural linewidth [cf. Eq. (B-19)].

By means of (7), (13) may be rearranged as follows:

$$\hbar(\nu_\lambda-\nu_0)\langle A_i|\lambda\rangle=-\mu\mathcal{E}_i^0(\mathbf{r}_A)-\mu\sum_{B\neq A}\mathcal{E}_{ij}(\mathbf{r}_A,\mathbf{r}_B)[\langle B_j|\bar{\lambda}\rangle(A)+\delta\langle B_j|\bar{\lambda}\rangle(A)]-i\hbar\sigma_{ij}\langle A_j|\bar{\lambda}\rangle,\tag{15}$$

where

$$\mathcal{E}_i^0(\mathbf{r}_A)=\langle 0|\,\mathcal{E}_i(\mathbf{r}_A)\,|\lambda\rangle;$$

$$\mathcal{E}_{ij}(\mathbf{r}_A,\mathbf{r}_B)=\langle 0|\,\mathcal{E}_i(\mathbf{r}_A)\,|B_j\rangle+\int\frac{\langle 0|\,\mathcal{E}_i(\mathbf{r}_A)\,|\eta\rangle d\eta\langle\eta|\mathcal{H}|B_j\rangle}{\hbar(\nu_\lambda-\nu_\eta+i\xi)}.\tag{16}$$

$\langle B_j|\bar{\lambda}\rangle(A)$ is the value that $\langle B_j|\bar{\lambda}\rangle$ would have if atom A were removed, and

$$\delta\langle B_j|\bar{\lambda}\rangle(A)=\langle B_j|\bar{\lambda}\rangle-\langle B_j|\bar{\lambda}\rangle(A).$$

We now take the average of (15), with $\mathbf{r}_A$ fixed at the point $\mathbf{r}$. The averaging of the terms in the summation over B can be carried out as follows: First, consider atom B also fixed at some position $\mathbf{r}'$, and average over the positions of all the absorbers except A and B. Then to average and sum over B, just multiply by $\mathfrak{N}$ and integrate the resulting expression with respect to $\mathbf{r}'$ over the volume v. Also notice that, since in the limit of large N the removal of one atom from the gas will not have any effect on averaged quantities (though it may have a large effect on some quantities referring to a particular configuration), and since according to assumption (b) the presence or absence of atom A has no effect on the allowed configurations of the other atoms,

$$\{\langle B_j|\bar{\lambda}\rangle(A)\}[A(\mathbf{r}),B(\mathbf{r}')]=F_j(\mathbf{r}').\tag{17}$$

Now, using (9), (16), and (17), we find for the above-mentioned average of (15):

$$\hbar(\nu_\lambda-\nu_0)F_i(\mathbf{r})=-\mu\mathcal{E}_i^0(\mathbf{r})-\mu\mathfrak{N}\int_v\mathcal{E}_{ij}(\mathbf{r},\mathbf{r}')F_j(\mathbf{r}')d^3r'+\hbar\Delta_{ij}(\mathbf{r})F_j(\mathbf{r}),\tag{18}$$

where

$$\hbar\Delta_{ij}(\mathbf{r})F_j(\mathbf{r})=-i\hbar\sigma_{ij}F_j(\mathbf{r})-\mu\mathfrak{N}\int_v\mathcal{E}_{ij}(\mathbf{r},\mathbf{r}')\delta F_j(\mathbf{r}',\mathbf{r})d^3r'\tag{19}$$

and

$$\delta F_j(\mathbf{r}',\mathbf{r})=\{\delta\langle B_j|\bar{\lambda}\rangle(A)\}[B(\mathbf{r}'),A(\mathbf{r})].\tag{20}$$

To find an expression for the matrix element of $\mathcal{E}_i(\mathbf{r})$, we use (4), (12), and (16), and obtain (for a

particular configuration)

$$\langle 0|\,\mathcal{E}_i(\mathbf{r})\,|\bar{\lambda}\rangle=\mathcal{E}_i^0(\mathbf{r})+\sum_B \mathcal{E}_{ij}(\mathbf{r},\mathbf{r}_B)\langle B_j|\bar{\lambda}\rangle. \quad (21)$$

Averaging (21) over all configurations, one finds with the aid of (9):

$$\mathfrak{E}_i(\mathbf{r})=\mathcal{E}_i^0(\mathbf{r})+\mathfrak{N}\int_v \mathcal{E}_{ij}(\mathbf{r},\mathbf{r}')F_j(\mathbf{r}')d^3r'. \quad (22)$$

Equation (22) substituted into (18) gives

$$\hbar(\nu_\lambda-\nu_0)F_i(\mathbf{r})=-\mu\mathfrak{E}_i(\mathbf{r})+\hbar\Delta_{ij}(\mathbf{r})F_j(\mathbf{r}), \quad (23)$$

which is compatible with (11) only if

$$\Delta_{ij}(\mathbf{r})=\Delta\delta_{ij}, \quad (24)$$

with Δ a constant independent of r, and

$$\rho^2(\nu_\lambda)-1=-\frac{4\pi\mathfrak{N}\mu^2}{\hbar(\nu_\lambda-\nu_0-\Delta)}. \quad (25)$$

Equation (25) is formally identical with Eqs. (A-17) and (A-20), although it has not yet been demonstrated that Δ has the same meaning in both cases. Equation (A-18) is equivalent to a special case of (18) with (24) substituted in. For this special case, it is shown in paper A that the integral Eq. (18) has a solution corresponding to the proper combination of incident, transmitted, and reflected waves. This solution is exhibited in Eq. (A-19).

We must now show that the procedure for evaluating Δ used in Sec. B-III is correct. First, we define a quantity $\mathfrak{D}_{jk}(\mathbf{r}',\mathbf{r})$ by

$$\delta F_j(\mathbf{r}',\mathbf{r})=\frac{\mathfrak{D}_{jk}(\mathbf{r}',\mathbf{r})F_k(\mathbf{r})}{\hbar(\nu_\lambda-\nu_0-\Delta)}. \quad (26)$$

Equations (14), (19), (24), and (26) can now be combined to give

$$\hbar\Delta\delta_{ij}=-i\hbar\sigma\delta_{ij}-\frac{\mu\mathfrak{N}}{\hbar(\nu_\lambda-\nu_0-\Delta)}\int_v \mathcal{E}_{ik}(\mathbf{r},\mathbf{r}')\times\mathfrak{D}_{kj}(\mathbf{r}',\mathbf{r})d^3r'. \quad (27)$$

Now the quantity $\delta F_j(\mathbf{r}',\mathbf{r})$ may be thought of as the change in $F_j(\mathbf{r}')$ brought about by inserting a single fixed absorber into the "medium" at the point $\mathbf{r}$. The insertion of this fixed absorber will also produce a change $\delta\mathfrak{E}_j(\mathbf{r}',\mathbf{r})$ in $\mathfrak{E}_j(\mathbf{r}')$. To calculate this, we first write down the change in (21) due to the additional absorber:

$$\delta\langle 0|\,\mathcal{E}_i(\mathbf{r}')\,|\bar{\lambda}\rangle(A)=\mathcal{E}_{ij}(\mathbf{r}',\mathbf{r}_A)\langle A_j|\bar{\lambda}\rangle+\sum_B \mathcal{E}_{ij}(\mathbf{r}',\mathbf{r}_B)\delta\langle B_j|\bar{\lambda}\rangle(\mathrm{A}).$$

Now we take the average of the above expression, holding the added absorber A fixed at the position r. This gives the result

$$\delta\mathfrak{E}_i(\mathbf{r}',\mathbf{r})=\mathcal{E}_{ij}(\mathbf{r}',\mathbf{r})F_j(\mathbf{r})+\mathfrak{N}\int_v \mathcal{E}_{ij}(\mathbf{r}',\mathbf{r}'')\delta F_j(\mathbf{r}'',\mathbf{r})d^3r''. \quad (28)$$

But, according to our assumption (a), the electric field and polarization matrix elements are related through the refractive index, and this should remain true when a fixed absorber is inserted in the medium. *Note added in proof:* Actually, (29) represents a stronger assumption than (11), since the functions appearing in (29) are rapidly varying and large in value when $\mathbf{r}'$ is close to $\mathbf{r}$. We, therefore, require, analogously to (11):

$$4\pi\mathfrak{N}\mu\delta F_i(\mathbf{r}',\mathbf{r})=[\rho^2(\nu_\lambda)-1]\delta\mathfrak{E}_i(\mathbf{r}',\mathbf{r}). \quad (29)$$

Equations (25), (26), (28), and (29) may now be combined to give

$$\mathfrak{D}_{ij}(\mathbf{r}',\mathbf{r})=-\mu\mathcal{E}_{ij}(\mathbf{r}',\mathbf{r})-\frac{\mathfrak{N}\mu}{\hbar(\nu_\lambda-\nu_0-\Delta)}\int_v \mathcal{E}_{ik}(\mathbf{r}',\mathbf{r}'')\times\mathfrak{D}_{kj}(\mathbf{r}'',\mathbf{r})d^3r''. \quad (30)$$

Equation (30) is equivalent to (B-22). In comparing the two, one should note that the matrix elements of the operator $\mathfrak{K}$ defined by Eq. (A-15) are given by

$$\langle A_i|\,\mathfrak{K}\,|B_j\rangle=-\mu\mathcal{E}_{ij}(\mathbf{r}_A,\mathbf{r}_B).$$

Also,

$$\langle A_i|\,\mathfrak{D}\,|B_j\rangle=\mathfrak{D}_{ij}(\mathbf{r}_A,\mathbf{r}_B).$$

The three equations [(25), (27), and (30)] are sufficient to determine the refractive index. Equation (25) is to be compared with (A-17) and (A-20), Eq. (27) with (B-21), and Eq. (30) with (B-22). The only difference is that (B-21) and (B-22) have sums in place of integrals, but these are replaced by integrals in the process of solving these equations. In solving (27) and (30), the integrals over v are to be extended to infinity, corresponding to the neglect of edge effects. The solution is carried out in Sec. B-III with the aid of a cutoff for the divergent integral, ending with Eq. (B-39) for the refractive index:

$$x+\Gamma/(\rho^2-1)=-i\rho+a\Gamma[(1-\rho^2)/\rho^2]. \quad (\text{B-39})$$

Here

$$x=(\nu_\lambda-\nu_0)/\sigma,$$

$$\Gamma=4\pi\mathfrak{N}\mu^2/\hbar\sigma,$$

and

$$a=1/(6\pi r_0{}^3\mathfrak{N}),$$

where r_0 is a cutoff radius which will be discussed more fully in the next section. As pointed out in Sec. B-III, Eq. (B-39) can easily be solved numerically to give the detailed dependence of refractive index on frequency.

IV. CUTOFF PROCEDURE

The divergent integral encountered in the solution of Eqs. (27) and (30) is expressed as a sum in Eq. (B-28). It is due to the singular character of the electrostatic dipole-dipole interaction at small distances. Clearly some cutoff is called for, but there seems to be no justification from our present point of view for the procedure employed in B of cutting off at the average distance between nearest neighbors. Actually, the

cutoff should be due to the fact that in a real gas, two absorbers are not permitted to approach one another more closely than a distance of the order of an atomic radius, so that interactions between absorbers at smaller distances than this should not be allowed to contribute. This can be accounted for at least approximately without introducing any explicit correlation (which would spoil our formalism) by simply doctoring the interaction in such a way that two absorbers are nor allowed to interact if they are closer together than an atomic radius. This means that r_0 should be of the ordte of an atomic radius and independent of $\mathfrak{N}$, and that a is proportional to $1/\mathfrak{N}$.

The solution of Eq. (B-39) leads to an absorption linewidth of order of magnitude

$$\delta\nu \sim a^{1/2}\Gamma\sigma.$$

If a is of the order of unity, as assumed in B, then this result is in qualitative agreement with previous theories,[3] but too small by a factor of the order of 10^3 when compared with the most recent experiments.[4] The observed linewidth is much too large to be accounted for by the Doppler effect. With our revised cutoff procedure, however, order-of-magnitude agreement can be obtained. For example, under the conditions of Tomiser's experiments with sodium vapor,[4] we have

$$\mathfrak{N} \sim 10^{15}\ \text{cm}^{-3}.$$

If we assume $r_0 \sim 5\times 10^{-8}$ cm, then we find

$$a^{1/2} \sim 500,$$

in rough agreement with the experimental results. The only difference is that the theoretical linewidth is now proportional to the square root of the density, while the observed dependence appears to be linear. Nevertheless, there is at least order-of-magnitude agreement with all the actual experimental points, and the discrepancy which does exist can apparently be largely removed if there is a slit width correction as large as 0.1 Å. This situation will be discussed more fully in a forthcoming paper, in which the effect of translational motion of the absorbers is also taken into account.

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Polarization of the $\lambda = 5876$ Å and $\lambda = 6679$ Å Lines in Helium Excited by Electrons*

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The polarization of the lines $\lambda = 5876$ Å and $\lambda = 6679$ Å resulting from the excitation of helium by electrons was measured as a function of the bombarding electron energy. The dependence of the polarization on the helium pressure is also reported for these lines. The observations reported here conflict with the theory of the polarization of collision-induced radiation at the threshold energy for excitation.

INTRODUCTION

RECENTLY, the authors of this paper reported on the polarization of light resulting from the excitation of helium by a beam of monoenergetic electrons.[1] The reader is referred to this work for a discussion of the details of the experimental arrangement, a review of the theory, and references to previous work reported on this subject by other authors. The polarization as a function of the electron energy for eight lines in helium was reported previously. It is the purpose of this present work to report on one additional line, $\lambda = 6679$ Å, as well as to present improved results on $\lambda = 5876$ Å.

The theory on the polarization of light resulting from the excitation of helium by electrons, as reviewed by Percival and Seaton,[2] predicts a unique maximum value for the polarization of light at threshold. The predicted threshold value does not depend upon a detailed calculation of the collision process, but only on some knowledge of the description of the states of helium and on the conservation of angular momentum. As noted previously,[1] experimental results indicate that, instead of approaching this unique threshold value, the polarization definitely tends toward zero. The results presented in this paper on the polarization of the $\lambda = 5876$ Å and $\lambda = 6679$ Å lines do not yield the predicted threshold value but, again, indicate a trend towards zero polarization at threshold.

EXPERIMENTAL ARRANGEMENT

Although the details of the experimental arrangement have been described in a previous paper, essentials

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[1] R. H. McFarland and E. A. Soltysik, University of California Radiation Laboratory Report UCRL-6749, 1962 (unpublished); Phys. Rev. **126**, 2090 (1962).

[2] I. C. Percival and M. J. Seaton, Phil. Trans. Roy. Soc. London **113**, 251 (1958).