Theory of optically induced diffusion of gases

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A theoretical analysis is presented of optically induced diffusion of gases. In this phenomenon the atoms, upon absorbing radiation from a traveling monochromatic wave, can be displaced by collisions with the buffer gas either along or against the wave vector, depending on certain conditions. The effective force that causes this directional motion exceeds the light-pressure force by several orders. The phenomenon can be used to separate isotopes or isomers, to investigate the transport cross sections by excited states, etc.

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1. INTRODUCTION

We have previously reported briefly[1] the optically induced diffusion (OID) of gases, a phenomenon that appears in the field of a traveling light wave whose frequency is close to the resonant frequency of the transition in the atom. Because of the Doppler effect, the excitation of the atoms by the light is selective in velocity, as a result of which opposing fluxes of excited and unexcited atoms are induced. Since the dimensions of the atom in different states are different, these fluxes encounter different resistances in the buffer gas (Fig. 1), and as a result the absorbing gas is set in motion as a whole.

A task-oriented experiment on the observation of OID was performed recently.[2] The results of the experiment, which so far are only approximate, have shown that the effect manifests itself very strongly.

In this paper we present a theoretical analysis of the OID phenomenon, determine its dependence on the characteristics of the medium and of the radiation, and present some estimates and typical applications. We consider a very simple system corresponding to the electronic transitions in the atoms.

2. ANALYSIS OF KINETIC EQUATIONS

The interaction of atoms with a traveling light wave of frequency ω close to the frequency ω_{mn} of the transition from the ground[1] (n) into an excited state is described by the following kinetic equations for the density matrix:

$$(\partial/\partial t + v\nabla + 2\gamma_m)\rho_{mm}(v) = S_m(v) - 2\mathrm{Re}\,[iG^*\rho_{mn}(v)],$$
$$(\partial/\partial t + v\nabla)\rho_{nn}(v) = S_n(v) + 2\gamma_m\rho_{mm}(v) + 2\mathrm{Re}\,[iG^*\rho_{mn}(v)], \quad (2.1)$$

$$[\partial/\partial t + v\nabla + \Gamma - i(\Omega - kv)]\rho_{mn}(v) = iG[\rho_{nn}(v) - \rho_{mm}(v)],$$
$$\Omega = \omega - \omega_{mn}, \quad G = Ed_{mn}/2\hbar.$$

Here E and $\mathbf{k}$ are respectively the amplitudes of the electric field and the wave vector of the radiation, $2\gamma_m$ is the decay constant of the excited states, S_m and S_n are the integrals of the elastic collisions, and d_{mn} is the dipole-moment matrix element. We assume that in the collisions the states m and n are quite differently excited so that the collisions lead for the off-diagonal element ρ_{mn} of the density matrix to impact broadening (Γ) and to a shift (if necessary, the latter can be regarded as included in the quantity ω_{mn}). Each of the

"diagonal" collision integrals (S_m, S_n) consists of three parts. For example,

$$S_m(v) = S_{mm}(v) + S_{mn}(v) + S_{mb}(v), \quad (2.2)$$

where S_{mm} describes the collisions of the excited particles with one another, S_{mn} corresponds to collisions with unexcited atoms, and S_{mb} corresponds to collisions with particles that do not interact with the radiation (the buffer gas), for which the following kinetic equation holds

$$(\partial/\partial t + v\nabla)\rho_{bb}(v) = S_b(v), \quad S_b = S_{bb} + S_{bm} + S_{bn}. \quad (2.3)$$

The collision integrals have the following properties, which follow from the particle-number and momentum conservation laws in elastic scattering (see. e.g., Refs. 3 and 4):

$$\int S_{il}(v)\,dv = 0, \quad \int v[M_i S_{il}(v) + M_l S_{li}(v)]\,dv = 0, \quad (2.4)$$

where the subscripts i and l take on the values m, n, and b, while M_i is the mass of particle of sort i.

When account is taken of relations (2.4), we can obtain the hydrodynamics equations from the kinetic equations (2.1) and (2.3). To this end we consider the following hydrodynamic characteristics: the particle-number densities, ρ_m, ρ_n, and ρ_b, the particle fluxes j_m, j_n, and j_b, the pressure tensors $P_m^{\alpha\beta}$, $P_n^{\alpha\beta}$, and $P_b^{\alpha\beta}$ ($\alpha, \beta = x, y, z$), and the internal friction forces F_m, F_n, and F_b. These characteristics are expressed in the following manner in terms of the distribution functions and the collision intergals[3,4]

$$\rho_i = \int \rho_{ii}(v)\,dv, \quad j_i = \int v\rho_{ii}(v)\,dv,$$

$$P_i^{\alpha\beta} = M_i \int v_\alpha v_\beta \rho_{ii}(v)\,dv, \quad F_i = M_i \int vS_i(v)\,dv. \quad (2.5)$$
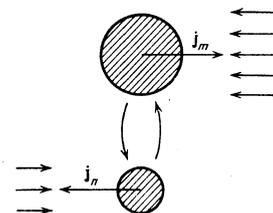
FIG. 1. Illustrating the onset of a flux of absorbing particles. The bent arrows represent the radiative transitions.

For absorbing atoms we shall consider also the total particle density $N = \rho_m + \rho_n$, the total flux $J = j_m + j_n$, the pressure tensor $P^{\alpha\beta} = P_m^{\alpha\beta} + P_n^{\alpha\beta}$, and the friction force $F = F_m + F_n$.

We add the first two equations of (2.1) and integrate with respect to the velocities. As a result, taking the first relation of (2.4) into account, we obtain the continuity equation for the absorbing particles

$$\frac{\partial}{\partial t} N + \operatorname{div} J = 0. \tag{2.6}$$

It follows similarly from (2.3) that

$$\frac{\partial}{\partial t} \rho_b + \operatorname{div} j_b = 0. \tag{2.7}$$

Multiplication of Eqs. (2.1) and (2.3) by v, followed by integration with respect to velocity with account taken of (2.4), yields an equation of the Euler type. For the absorbing particles as a whole and for the buffer-gas particles, we have

$$M \frac{\partial}{\partial t} J^\alpha + \sum_\beta \frac{\partial P^{\beta\alpha}}{\partial \beta} = F^\alpha, \quad M \equiv M_m = M_n, \tag{2.8}$$

$$M_b \frac{\partial}{\partial t} j_b^\alpha + \sum_\beta \frac{\partial}{\partial \beta} P_b^{\beta\alpha} = -F^\alpha. \tag{2.9}$$

The friction force F, whose components enter in these equations, is given by

$$F = F_m + F_n = M \int v [S_{mb}(v) + S_{nb}(v)] dv, \tag{2.10}$$

i. e., it is governed by the collisions of the excited and unexcited particles with the buffer-gas particles. Obviously, the partial friction forces F_m and F_n are directed opposite to the corresponding fluxes j_m and j_n. In particular,

$$\int v S_{mb}(v) dv = -\nu_m j_m, \quad \int v S_{nb}(v) dv = -\nu_n j_n, \tag{2.11}$$

where we have introduced the proportionality coefficients ν_m and ν_n, which have the dimensionality and meaning of collision frequencies. The expression for the force F can consequently be represented in the form

$$F = M[(\nu_n - \nu_m) j_m - \nu_n J]. \tag{2.12}$$

If there is no radiation, then $j_m = 0$, and the equations (2.8) and (2.9), together with (2.6) and (2.7), describe the ordinary diffusion processes in a two-component medium: at constant total pressure, the spatial inhomogeneity of the density of the components leads to diffuse currents which, in turn, equalize the density of each of the components. Under stationary conditions we have simultaneously $F = 0$ and $J = 0$.

On the other hand, as noted in the preceding section (see also Ref. 1), the radiation induces a flux j_m. Since the interaction of the buffer gas with the excited atoms is in general different from that with the unexcited ones, we have $\nu_m \neq \nu_n$, and according to (2.12), simultaneous vanishing of the friction force F and of the flux of the absorbing atoms J is impossible.

If the gas mixture was initially at equilibrium, then at the first instant after the field is turned on we have $J = 0$, but with increasing population of the excited state a flux j_m is produced, and with it also a force F, which

in turn produces the flux J of the absorbing atoms as a unit. The buffer gas is then acted upon by a force of the same magnitude but of opposite direction, producing an opposing flux. If the absorbing cell is open on both ends and is contained in the reservoir with the original mixture, then after the lapse of a certain time a stationary flux is established, wherein

$$F = 0, \quad J = \left(\frac{\nu_n - \nu_m}{\nu_n}\right) j_m. \tag{2.13}$$

If the ends of the absorbing cells are closed, then a stationary regime is established in the course of time, such that

$$J = 0, \quad F = M(\nu_n - \nu_m) j_m, \tag{2.14}$$

and the friction force (2.14) is offset by the produced pressure gradient in accordance with (2.8). In other words, the absorbing particles flow over to one end of the cell or to the other (depending on the direction of j_m). The buffer gas, on the other hand, flows to the opposite end in accordance with (2.9).

It follows from (2.8) and (2.9) that the established gas pressure is uniform over the volume, if the gas was at equilibrium prior to the application of the field. This is due to the fact that the processes of momentum transfer (light pressure) and energy transfer from the radiation in the medium are not reflected in Eqs. (2.1).

Since the transfer process considered here takes place at a constant total pressure (flow of the different gas components through each other), it is related to the diffusion process and we shall therefore call it self-induced diffusion.

The question of the concrete values of the flux J in (2.13) and the force F in (2.14) reduces to the calculation of the flux j_m of the excited atoms. We consider first homogeneous-broadening conditions, when the problem can be solved at an arbitrary field intensity.

3. HOMOGENEOUS BROADENING

If the luminescence-line half-width Γ exceeds the Doppler width

$$\Gamma > k\bar{v}, \tag{3.1}$$

then the interaction of the atoms with the field has a low velocity selectivity. The dependence of the density-matrix elements $\rho_{ii}(v)$ in (2.1) is then close to Maxwellian and can be represented as a sum of a Maxwellian distribution $W(v)$ and an anti-symmetrical increment:

$$\rho_{ii}(v) = \left[\rho_i + \frac{2}{\bar{v}^2} v j_i\right] W(v) \quad (i = m, n),$$

$$\rho_{mn}(v) = \left[\rho + \frac{2}{\bar{v}^2} v j\right] W(v). \tag{3.2}$$

On the basis of (2.1) we can easily derive equations for the zeroth moments (ρ_i, ρ) and the first moments (the fluxes j_i, and j) of the density matrix.

We confine ourselves henceforth to the stationary regime. Integrating (2.1) with respect to velocity, we ob-

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tain

$$2\gamma_m\rho_m+\mathrm{div}\,\mathbf{j}_m=-2\mathrm{Re}\,(iG^*\rho),$$
$$(\Gamma-i\Omega)\rho+(i\mathbf{k}+\mathrm{div})\mathbf{j}=iG(\rho_n-\rho_m). \qquad (3.3)$$

In place of the second equation (2.1), we shall use the continuity equation (2.6), which yields

$$\mathbf{J}=\mathbf{j}_m+\mathbf{j}_n=\mathrm{const}. \qquad (3.4)$$

Multiplication of (2.1) by $\mathbf{v}$ followed by integration with respect to $\mathbf{v}$ leads to the equations

$$(\nu_m+2\gamma_m)\mathbf{j}_m+{}^{1}\!/{}_{2}\bar{v}^2\nabla\rho_m=-2\mathrm{Re}\,(iG^*\mathbf{j}),$$
$$(\Gamma-i\Omega)\mathbf{j}+{}^{1}\!/{}_{2}\bar{v}^2(i\mathbf{k}+\nabla)\rho=iG(\mathbf{j}_n-\mathbf{j}_m), \qquad (3.5)$$

and in place of the equation for $\mathbf{j}_n$ it is convenient to use Eq. (2.8), which reduces in the approximation (3.2) to the form

$$\nabla N=2\bar{v}^{-2}[\,(\nu_n-\nu_m)\mathbf{j}_m-\nu_n\mathbf{J}]. \qquad (3.6)$$

The collision frequencies ν_m and ν_n are governed in the approximation (3.2) by the corresponding transport cross sections σ_m^{tr} and σ_n^{tr}:

$$\nu_i=\frac{8\pi^2}{3}\rho b\frac{\mu}{M}\left(\frac{\mu}{2\pi KT}\right)^{5/2}\int_0^\infty u^5 e^{-\mu u^2/2KT}\sigma_i^{\mathrm{tr}}(u)\,du, \qquad (3.7)$$

$$\mu=MM_b/(M+M_b),$$

where u is the relative velocity of the colliding particles.

The system (3.3)—(3.6) is closed and makes it possible to find all the quantities of interest to us. We assume here that the density of buffer gas is much higher than the density of the absorbing particles, and that the intensity of the radiation does not depend on the coordinates (an optically thin medium). In this case only the sought functions are spatially inhomogeneous, but not the coefficients of the employed equations.

The simplest solutions of the equations are obtained in the stationary-flux regime, when the terms with the spatial derivatives in (3.3)–(3.6) vanish and the equations reduce to algebraic. As a result of their solution, accurate to corrections of order $(k\bar{v}/\Gamma)^2$, we find the steady-state flux $\mathbf{J}$ of the absorbing particles as a whole:

$$\mathbf{J}=\frac{\mathbf{k}\bar{v}}{k}\frac{\nu_n-\nu_m}{\nu_n+\nu_m}\left(\frac{\Omega k\bar{v}}{\Gamma^2+\Omega^2}\right)\frac{\varkappa N}{1+\varkappa}\left[\varkappa+\frac{\nu_n}{\nu_n+\nu_m}\left(\frac{2\gamma_m+\nu_m}{\gamma_m}\right)\right]^{-1},$$
$$\varkappa=\frac{2|G|^2}{\gamma_m}\frac{\Gamma}{\Gamma^2+\Omega^2}. \qquad (3.8)$$

Here $\varkappa$ is the so called saturation parameter and characterizes the degree of equalization of the level populations: at $\varkappa\ll1$ the populations of the ground and excited states are approximately equal.

We now discuss the result. The flux of atoms can be directed either along the wave vector or in the opposite direction, depending on the sign of the difference $\nu_n-\nu_m$ of the collision frequencies and on the sign of the detuning $\Omega=\omega-\omega_{mn}$ of the radiation from resonance. For example, at $\Omega>0$ and $\nu_m>\nu_n$ the atoms move opposite to the light flux (counter to $\mathbf{k}$). With increasing $|\Omega|$, the flux increases, reaches a maximum at $|\Omega|$

$\sim\Gamma$, and then again begins to decrease (even at a fixed saturation parameter). The decrease of $\mathbf{J}$ is connected with the fact that at large $|\Omega|$ the interaction with the field has low velocity selectivity, and this leads to a small asymmetry of the distribution $\rho_{mm}(\mathbf{v})$, and consequently to a low value of the flux $\mathbf{j}$. In this respect, a large departure from resonance is equivalent to a large homogeneous broadening: the velocity selectivity decreases with increasing Γ and, as a consequence, the OID effect decreases. The dependence of $\mathbf{J}$ on the radiation intensity (on $\varkappa$) is qualitatively similar to the dependence on $|\Omega|$: at small $\varkappa$ the number of excited atoms is small, and consequently the flux $\mathbf{J}$ is also small, since it is proportional to $\varkappa$. In a very intense field ($\varkappa>(2\gamma_m+\nu_m)/2\gamma_m$) the flux again decreases, since the field broadening comes into play and leads again to a decrease of the selectivity of the interaction.

We determine now the optimal conditions for the manifestation of the OID effect within the framework of the applicability of Eq. (3.8). With respect to Ω and these conditions are obvious: $|\Omega|\sim\Gamma$, $\varkappa\sim1$. Since the effect decreases with increasing Γ, it is necessary to take the minimum possible Γ allowed by the approximation (3.1), i.e., $\Gamma\sim k\bar{v}$. It is also seen from (3.8) that to obtain the optimal $\mathbf{J}$ it is necessary to satisfy the condition $\nu_m\ll2\gamma_m$. The flux $\mathbf{J}$ then ceases to depend on γ_m (if the value of $\varkappa$ is fixed). We note finally that the transport cross sections for the ground and excited electronic states of atoms and molecules can differ substantially.[5] In the estimates we shall assume concretely that the cross sections differ by a factor of two ($\nu_m/\nu_n=2$).

For the presented values of the parameters it follows from (3.8) that the directional velocity of the absorbing atoms $\mathbf{u}=\mathbf{J}/N$ has the following value:

$$u\sim5\times10^{-2}\bar{v}, \qquad (3.9)$$

which amounts to a noticeable fraction of the mean thermal velocity.

We examine now the solution of Eqs. (3.3)–(3.6) in an absorbing cell with closed ends, when $\mathbf{J}=0$. Accurate to corrections of order $(k\bar{v}/\Gamma)^2$, we can discard the terms $\mathrm{div}\,\mathbf{j}_m$ and $(i\mathbf{k}+\mathrm{div})\mathbf{j}$ in Eqs. (3.3). We assume also the condition $\gamma_m\ll\Gamma$, which allows us to neglect the term $\nabla\rho$ in the second equation of (3.5). Thus, the spatial derivatives remain only in the first equation of (3.5) and in (3.6). From (3.3)–(3.5) we obtain $\mathbf{j}_m$ as a function of N and ∇N, after which we obtain in lieu of (3.6) the following differential equation for the density of the absorbing particles:

$$\frac{\nabla N}{N}=\frac{2k(\nu_n-\nu_m)\varkappa\gamma_m\Omega/(\Gamma^2+\Omega^2)}{\nu_m+{}^{1}\!/{}_{2}(\nu_m+\nu_n)\varkappa+2\gamma_m(1+\varkappa)^2}. \qquad (3.10)$$

Expression (3.10) describes the exponential variation of the density along the wave vector. The direction in which the density increases, just as in the case of the flux tensor (3.8), is determined by the sign of the difference $\nu_n-\nu_m$ and by the sign of Ω. The character of the dependence of $|\nabla N/N|$ on Ω, Γ, and $\varkappa$ is the same as for the stationary flux $\mathbf{J}$ in (3.8). At $\Omega\sim\Gamma\sim k\bar{v}$,

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$\varkappa\sim 1$, $\nu_m/\nu_n = 2$, $2\gamma_m\sim \nu_m$ we get from (3.10)

$$|\nabla N/N|\sim 10^{-1}\nu_m/\bar{v}. \qquad (3.11)$$

This means that the characteristic dimension l of the layer in which the absorbing atoms are gathered amounts to approximately 10 mean free paths. For $\bar{v}\sim 10^5$ cm/sec and $\nu_m\sim 10^8$ sec^{-1} we have $l\sim 10^{-2}$ cm.

We note that in both analyzed cases we have assumed the radiation field to be uniform over the cross section of the absorbing cell. If the diameter of the light beam is less than the cell diameter, then circular fluxes should arise. In addition, effects of "transverse" diffusion come into play,[6] i. e., the picture becomes complicated and will not be considered here.

4. INHOMOGENEOUS BROADENING

Under conditions of inhomogeneous broadening

$$\Gamma < k\bar{v}, \qquad (4.1)$$

the interaction of the atoms with the field is highly selective in velocity, the velocity distributions contain Bennett dips and peaks, thus complicating the solution of (2.1) at arbitrary field intensities. If, however, we assume the condition

$$\left(1+\frac{\nu_m}{\nu_n}\right)\frac{\Gamma}{\Gamma^2+\Omega^2}\frac{|G|^2}{(2\gamma_m+\nu_m)}\ll 1, \qquad (4.2)$$

then in this case it becomes quite simple to calculate the sought characteristics

In homogeneous broadening, as follows from the foregoing, the scale of the spatial inhomogeneity produced on account of the OID greatly exceeds the wavelength of the radiation. This result is valid in the general case, so that in Eq. (2.1) for $\rho_{mn}(\mathbf{v})$ we can neglect the term with the spatial derivative compared with the term containing the Doppler shift $\mathbf{k}\cdot\mathbf{v}$. Thus,

$$\rho_{mn}(\mathbf{v}) = \frac{iG}{\Gamma-i(\Omega-\mathbf{kv})}[\rho_{nn}(\mathbf{v})-\rho_{mm}(\mathbf{v})]. \qquad (4.3)$$

The solution for the diagonal elements in (2.1) will be sought in the form

$$\rho_{ii}(\mathbf{v}) = R_{ii}(\mathbf{v})+r_{ii}(\mathbf{v}),$$
$$R_{ii}(\mathbf{v}) = [\rho_i+2\bar{v}^{-2}\mathbf{v}\mathbf{j}_i]W(\mathbf{v}). \qquad (4.4)$$

The Bennett peaks and dips are contained in this case in the increments $r_{ii}(\mathbf{v})$. This subdivision is somewhat arbitrary and we eliminate it by assuming that the R_{ii} satisfy the following equations:

$$(2\gamma_m+\mathbf{v}\nabla)R_{mm}(\mathbf{v}) = S_m[R_{mm}(\mathbf{v})]$$
$$+\gamma_m(\rho_n-\rho_m)W(\mathbf{v})[\langle\varkappa'\rangle+2\bar{v}^{-2}\mathbf{v}\langle\mathbf{v}\varkappa'\rangle],$$
$$\mathbf{v}\nabla[R_{mm}(\mathbf{v})+R_{nn}(\mathbf{v})] = S_m[R_{mm}(\mathbf{v})]+S_n[R_{nn}(\mathbf{v})]. \qquad (4.5)$$

We have introduced here the symbol

$$\varkappa' = \frac{2|G|^2}{\gamma_m}\frac{\Gamma}{\Gamma^2+(\Omega-\mathbf{kv})^2}, \qquad (4.6)$$

while the angle brackets denote averaging over the velocities with Maxwellian distributions. To avoid misunderstandings, the written-out arguments of the collision integrals are the corresponding distribution functions.

From the initial Eqs. (2.1), with allowance for (4.4) and (4.5), follow the following equations for $r_{ii}(\mathbf{v})$:

$$(2\gamma_m+\mathbf{v}\nabla)r_{mm}(\mathbf{v}) = \gamma_m\varkappa'[r_{nn}-r_{mm}+2\bar{v}^{-2}\mathbf{v}(\mathbf{j}_n-\mathbf{j}_m)W(\mathbf{v})]$$
$$+\gamma_m(\rho_n-\rho_m)W(\mathbf{v})[\varkappa'-\langle\varkappa'\rangle-2\bar{v}^{-2}\mathbf{v}\langle\mathbf{v}\varkappa'\rangle]+S_m[r_{mm}(\mathbf{v})],$$
$$\mathbf{v}\nabla[r_{mm}(\mathbf{v})+r_{nn}(\mathbf{v})] = S_m[r_{mm}(\mathbf{v})]+S_n[r_{nn}(\mathbf{v})]. \qquad (4.7)$$

For $r_{ii}(\mathbf{v})$ we confine ourselves to the first term of the expansion in the field intensity (in $\varkappa'$). In the absence of a field, r_{ii} and $\mathbf{j}_i$ are equal to zero, and therefore the terms containing $r_{nn}-r_{mm}$ and $\mathbf{j}_n-\mathbf{j}_m$ in (4.7) are proportional to $\varkappa'^2$ and will be neglected. It can be shown that discarding these terms means the use of condition (4.2). Recognizing also that the scale of the spatial inhomogeneity as larger than the mean free path, we neglect the term $\mathbf{v}\nabla r_{mm}$ in (4.7). We see then that $r_{mm}(\mathbf{v})$ has the following properties:

$$\int r_{mm}(\mathbf{v})d\mathbf{v}=0, \quad \int \mathbf{v}r_{mm}(\mathbf{v})d\mathbf{v}=0. \qquad (4.8)$$

Similar relations are valid also for $r_{nn}(\mathbf{v})$ and indicate, in particular, that the quantities ρ_i and $\mathbf{j}_i$ in (4.4) have respectively the meaning of the total population of the level i and the total flux in the state i.

Thus, to find the fluxes and the spatial distribution of the absorbing particles in the approximation (4.2), it suffices to consider only Eqs. (4.5). On their basis, in analogy with the preceding section, we derive the equations for the total populations and fluxes:

$$2\gamma_m\rho_m+\text{div }\mathbf{j}_m=\gamma_m\langle\varkappa'\rangle(\rho_n-\rho_m), \quad \rho_m+\rho_n=N, \qquad (4.9)$$

$$(2\gamma_m+\nu_m)\mathbf{j}_m+\tfrac{1}{2}\bar{v}^2\nabla\rho_m=\gamma_m(\rho_n-\rho_m)\langle\mathbf{v}\varkappa'\rangle. \qquad (4.10)$$

In addition, relations (3.4) and (3.6) remain in force. In the stationary-flux regime it follows from (3.6), (4.9), and (4.10) that

$$\mathbf{J}=\langle\mathbf{v}\varkappa'\rangle\frac{N}{1+\langle\varkappa'\rangle}\left(\frac{\nu_n-\nu_m}{\nu_n}\right)\frac{\gamma_m}{2\gamma_m+\nu_m}, \quad N=\text{const.} \qquad (4.11)$$

For an absorbing cell with closed ends we obtain the following differential equation for the density of the absorbing particles (the term $\text{div }\mathbf{j}_m$ in (4.9) is neglected, since its weight is determined by the ratio of the mean free path to the scale of the spatial inhomogeneity)

$$\frac{\nabla N}{N}=2\langle\mathbf{v}\varkappa'\rangle\frac{(\nu_n-\nu_m)}{2\gamma_m+\nu_m+\tfrac{1}{2}\langle\varkappa'\rangle(\nu_m+\nu_n)}\frac{\gamma_m}{\bar{v}^2}. \qquad (4.12)$$

We note that in the derivation of (4.11) and (4.12) we did not use the relation (4.1) and confined ourselves only to the condition (4.2). Thus, the obtained formulas are valid at any ratio of Γ and $k\bar{v}$ within the framework of the condition (4.2). In particular, if $\Gamma\gg k\bar{v}$, then

$$\langle\varkappa'\rangle=\varkappa, \quad \langle\mathbf{v}\varkappa'\rangle=k\bar{v}^2\varkappa\Omega/(\Gamma^2+\Omega^2), \qquad (4.13)$$

and we obtain Eqs. (3.8) and (3.10) in the approximation of (4.2).

For inhomogeneous broadening ($\Gamma\ll k\bar{v}$) we have

$$\langle\varkappa'\rangle=\frac{2\pi^{1/2}|G|^2}{k\bar{v}\gamma_m}\exp\left\{-\left(\frac{\Omega}{k\bar{v}}\right)^2\right\}, \quad \langle\mathbf{v}\varkappa'\rangle=\frac{\Omega}{k^2}\langle\varkappa'\rangle\mathbf{k}. \qquad (4.14)$$

Despite the limitation (4.2) with respect to the field, the quantity $\langle\varkappa'\rangle$ can be quite large if subject to sat-

isfaction of the condition $\Gamma(2\gamma_m + \nu_m)/2\gamma_m k\bar{v} \gg 1$, which can be ensured by the large collision frequency ν_m.

At $\Gamma \ll k\bar{v}$ expressions (4.11) and (4.12) as functions of Ω have an extremum at $|\Omega| \sim k\bar{v}$. Practical interest attaches also to the dependence on the pressure of the buffer gas. In inhomogeneous broadening, only ν_m and ν_n in (4.11) and (4.12) depend on the pressure. As seen from (4.11), the flux increases with decreasing ν_m and assumes a constant value at $\nu_m < 2\gamma_m$. It might seem that this result contradicts the obvious premise that when the pressure tends to zero there should be no OID effect. However, expression (4.11) was obtained by implicitly assuming that the length of the absorbing cell is unlimited and that collisions must take place over its length. Actually it is sufficient to require that the length of the cell greatly exceed the mean free path. The density gradient (4.12) decreases with decreasing pressure, as it should, and at $\nu_m > 2\lambda_m$ it is practically independent of pressure until the line half-width Γ becomes comparable with $k\bar{v}$. If the pressure is such that $\Gamma > k\bar{v}$, then further growth of the pressure, as shown in the preceding section, leads again to a decrease of $|\nabla N/N|$.

5. SELF-CONSISTENT EQUATIONS FOR THE PARTICLE DENSITY AND IRRADIATION INTENSITY

We have considered above the OID phenomenon in the given-field approximation. Moreover, we have assumed that the intensity of the radiation remains practically unchanged when it passes through the absorbing cell (optically tenuous medium). If the radiation absorption is substantial (optically dense medium), then new interesting features of the OID appear, one of which consists in the following. Let the absorbing cell be closed on both ends and let the radiation be absorbed over a length l_{ph} much less than the cell length L. At the initial instant after it is turned on, the radiation is absorbed in a region adjacent to the entrance end of the cell. At $\Omega(\nu_n - \nu_m) > 0$ the absorbing particles in this region are displayed by the OID in the direction of the wave vector. The particles move away from the entry window and compress the remaining gas of absorbing particles. At the same time, the region of effective radiation absorption also moves in the k direction. The process continues until the density drop over the effective absorption length compensates for the force due to the OID. Thus, the described process is equivalent to the action of a piston (optical "piston") that compresses the absorbing gas in the wave-vector direction. In contrast to the ordinary piston, the optical "piston" has selectivity—it acts only on one component of the gas.

Obviously, in the analysis of the OID in an optically dense medium it is necessary to solve the self-consistent equations for the particle density and radiation intensity. We shall consider this problem under conditions of homogeneous broadening ($\Gamma > k\bar{v}$) for a cell closed on both ends, and assume that the intensity of the traveling monochromatic wave is uniform over the cell cross section.

From the kinetic equations for the density matrix and

for the associated gasdynamic equations, which were analyzed above, we obtained the following equation for the density N of the absorbing particles (the z axis is directed along the wave vector k):

$$\frac{dN}{dz} = \frac{\nu_n - \nu_m}{\nu_m + 2\gamma_m(1+\varkappa)}\left[\left(\frac{2\gamma_m k\Omega}{\Gamma^2 + \Omega^2}\right)\frac{\varkappa N}{1+\varkappa} - \frac{1}{2}\frac{d}{dz}\left(\frac{\varkappa N}{1+\varkappa}\right)\right]. \quad (5.1)$$

We assume next that the collision frequencies ν_m and ν_n and the quantity Γ do not depend on the coordinates. This corresponds to the assumption that the buffer-gas density is much higher than the absorbing-gas density. In this case the coordinate dependence is concentrated in the quantities N and $\varkappa$.

In the medium is optically tenuous, then the change of the saturation parameter in (5.1) can be neglected, and we return to Eq. (3.10): the particle density has an exponential dependence on z, and its maximum is either at the entrance or at the exit window of the cell, depending on the sign of the combination $\Omega(\nu_n - \nu_m)$.

The radiation intensity is represented in (5.1) by the quantity $\varkappa$, for which we can deduce, on the basis of the abbreviated Maxwell's equations and Eqs. (3.3) (the terms which spatial derivatives and the term $i k \cdot j$ are discarded in the latter because they are small) the following differential equation:

$$\frac{d\varkappa}{dz} = -\sigma\frac{\varkappa N}{1+\varkappa}, \qquad \sigma = \frac{6\pi}{k^2}\frac{\gamma_m \Gamma}{\Gamma^2 + \Omega^2}, \quad (5.2)$$

where σ has the meaning of the photon-absorption cross section.

We obtain finally the spatial distribution of the particle density and of the radiation intensity it is necessary to add to (5.1) and (5.2) the relation (the condition for the conservation of the total number of particles)

$$\int_0^L N(z)\,dz = N_0 L, \quad (5.3)$$

where N_0 is the density of the absorbing particles in the absence of radiation.

To simplify the analysis we confine ourselves to fields of moderate intensity in accord with the condition (4.2). Within the framework of this condition, the saturation parameter may be not small if the buffer-gas pressure is large enough ($\nu_m > 2\gamma_m$).

When the condition (4.2) is satisfied, Eq. (5.1), with allowance for (5.2), has a rather simple integral of motion, which connects algebraically the particle density with the saturation parameter:

$$N\left(\frac{1+B\varkappa}{1+\varkappa}\right) + A\varkappa \equiv I = \text{const},$$
$$A = \left(\frac{\nu_n - \nu_m}{2\gamma_m + \nu_m}\right)\frac{\Omega}{\Gamma}\frac{k^3}{3\pi}, \qquad B = \frac{2\gamma_m + (\nu_m + \nu_n)/2}{2\gamma_m + \nu_m}. \quad (5.4)$$

The sign of the coefficient A is determined by the sign of the combination $\Omega(\nu_n - \nu_m)$. The parameter B can take on values in the range from one half (at $\nu_m \gg \nu_n$, $2\gamma_m$) to (at $\nu_n \gg \nu_m$, $2\gamma_m$). It is actually bounded from above by values of the order of unity. If $\nu_m = \nu_n$ then $B = 1$ and in this case $A = 0$.

If the radiation is absorbed over a distance much shorter than the cell length L, then the integral of motion (5.4) has the rather simple meaning of the asymptotic value of the particle density N_∞ beyond the absorption region ($\varkappa \to 0$). It follows then from (5.4) that

$$N = \frac{1+\varkappa}{1+B\varkappa}(N_\infty - A\varkappa). \tag{5.5}$$

Since N is positive, at $A > 0$ we should have $N_\infty \geqslant A\varkappa_0$, where $\varkappa_0$ is the value of the saturation parameter at the entry to the cell ($z = 0$). This already leads to the possibility of the optical "piston" effect. In fact, assume that in the absence of the field the particle density N_0 was less than the value $N_{cr} \equiv A\varkappa_0$. Then, to ensure the condition $N_\infty \geqslant A\varkappa_0$, the absorbing-particle gas must be compressed in the k direction, and this means a piston effect, which thus takes place when the initial particle density is lower than a certain critical value.

In the case of an optically tenuous medium at exact resonance ($\Omega = 0$), the equilibrium value of the density N is not disturbed. On the other hand if we put $\Omega = 0$ in (5.5), which means the vanishing of the coefficient A, then N still can depend on the coordinates to the extent that $\varkappa$ is spatially homogeneous and to the extent that the coefficient B differs from unity (the latter is connected with the relation between ν_m and ν_n). The noted spatial inhomogeneity of the density has a direct counterpart—the effect of drawing particles in and out of a light beam, dealt with in Ref. 6. The effect considered in the present paper can be explained as follows. The concentration of the excited particles is larger in that region where the field intensity is higher, so that there exists a diffusion flux of excited particles from the region of increased intensity of the field. The unexcited particles diffuse in the opposite direction, since the field makes their deficit larger the higher its intensity. Thus, in this case, too, we have opposite fluxes of excited and unexcited particles. Consequently, owing to the difference between the transport cross sections in the states m and n, the collisions with the buffer gas produce an absorbing-particle flux as a whole. If $\nu_m > \nu_n$, then the particles are drawn into the region of increased field intensity, and at $\nu_m < \nu_n$ they are pushed out of it. The maximum density drop occurs at $\varkappa_0 \gg 1$ and is determined by the ratio.

6. OPTICAL "PISTON" AND OPTICAL "PLUG"

We examine now in greater detail the solutions of Eqs. (5.1) and (5.2) at $A > 0$, when the OID effect causes a displacement of the absorbing particle in the direction of the wave vector. In addition, we assume that $N_0 < N_{cr} = A\varkappa_0$.

The solution of (5.2), with (5.4) taken into account, is of the form

$$\ln\frac{\varkappa_0}{\varkappa} + \left[1 + \frac{B}{A}N_\infty\right]\ln\frac{N_\infty - A\varkappa}{N_\infty - A\varkappa_0} = \sigma N_\infty z. \tag{6.1}$$

Under the assumed conditions, the absorbing particles near the input window ($z = 0$) are practically nonexistent, and consequently $N_\infty \to A\varkappa_0$, in accord with Eq. (5.4),

which can thus be rewritten in the form

$$N = \frac{1+\varkappa}{1+B\varkappa}(\varkappa_0 - \varkappa)A. \tag{6.2}$$

Some of the plots of N against $\varkappa$, describing this relation, are shown in Fig. 2.

As $N_\infty \to A\varkappa_0$, a logarithmic divergence appears in (6.1), which we shall reduce, to simplify the analysis, to a different form. We separate on the z axis the point z' at which the radiation intensity decreases by a factor of 2. By identity transformations we can obtain from (6.1), taking (6.2) into account,

$$\ln\frac{\varkappa_0}{2\varkappa} + (1+B\varkappa_0)\ln 2\left(1 - \frac{\varkappa}{\varkappa_0}\right) = \sigma A\varkappa_0(z - z'). \tag{6.3}$$

We determine now the characteristic regions of the variation of $\varkappa$ on the right and on the left of the point z'. We denote by l_+ the value of the difference $z - z'$ at which $\varkappa = \varkappa_0/4$, and by l_- the difference $z' - z$, where z is the point at which $\varkappa = 3\varkappa_0/4$. Obviously, l_+ and l_- determine the dimensions of the corresponding regions, and it follows for them from (6.3) that

$$l_+ = \frac{1}{\sigma A\varkappa_0}\left[\ln 2 + (1+B\varkappa_0)\ln\frac{3}{2}\right],$$
$$l_- = \frac{1}{\sigma A\varkappa_0}\left[\ln\frac{3}{2} + (1+B\varkappa_0)\ln 2\right]. \tag{6.4}$$

The values of l_+ and l_- are practically the same, and in the case of practical importance when $B \sim 1$ and $\varkappa_0 \gtrsim 1$ they are determined by the quantity $1/\sigma A$. The asymptotic behavior of $\varkappa$ (at $|z - z'| \gg l_+, l_-$) is described by an exponential dependence on the coordinates:

$$\varkappa = \varkappa_0 \varkappa 2^{B\varkappa_0} \exp\{-\sigma A\varkappa_0(z - z')\} \quad (z - z' \gg l_+); \tag{6.5}$$

$$\varkappa_0 - \varkappa = \varkappa_0 \varkappa 2^{-B\varkappa_0/(1+B\varkappa_0)} \exp\left\{-\frac{\sigma A\varkappa_0}{1+B\varkappa_0}(z' - z)\right\} \quad (z' - z \gg l_-). \tag{6.6}$$

From (6.2) and from the plots of Fig. 2 it follows that the particle density, just as the radiation intensity, changes substantially only in the region adjacent to the point $z = z'$. The characteristic dimension of the region of variation of N is described completely by expressions (6.4), and the asymptotic behavior is given by the formulas

$$A\varkappa_0 - N = A\varkappa_0\{1 + \varkappa_0(B-1)\}\varkappa 2^{B\varkappa_0} \exp\{-\sigma A\varkappa_0(z - z')\} \quad (z - z' \gg l_+);$$

$$N = A\varkappa_0\frac{1+\varkappa_0}{1+B\varkappa_0}\varkappa 2^{-B\varkappa_0/(1+B\varkappa_0)} \exp\left\{-\frac{\sigma A\varkappa_0}{1+B\varkappa_0}(z' - z)\right\} \quad (z' - z \gg l_-). \tag{6.7}$$
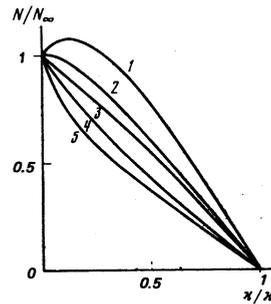
FIG. 2. Examples of the dependence of N on $\varkappa$ at $N_\infty = A\varkappa_0$: curve 1—$\varkappa_0 = 5$, $B = 1/2$; 2—$\varkappa_0 = 5$, $B = 3/4$; 3—$\varkappa_0 = 1/2$, $B = 1/2$; 4—$\varkappa_0 = 1$, $B = 3/2$; 5—$\varkappa_0 = 5$, $B = 3/2$.
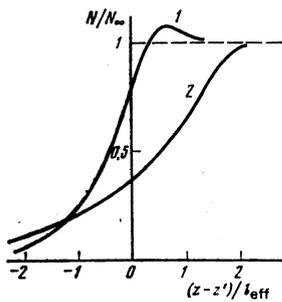
FIG. 3. Plot of the density against the coordinates in the transition region: curve $1 - \varkappa_0 = 5$, $B = 1/2$; $2 - \varkappa_0 = 5$, $B = 3/2$; l_{eff} $1/\alpha A$.

On both sides of z', the function $N(z)$ has an exponential asymptotic behavior: the density tends exponentially to zero on the left of z' and to $A\varkappa_0$ on the right.

Figure 3 shows two characteristic $N(z)$ curves calculated from formulas (6.2) and (6.3). The curves differ in shape and this difference is due to the influence of the spatial inhomogeneity of the field on the particle density. At $B < 1$ ($\nu_m > \nu_n$) the particles are drawn into the region of increased intensity (the curve with the extremum), while at $B > 1$ ($\nu_m < \nu_n$) they are pushed out. This is precisely why the curve is less steep in the second case.

Thus, both from qualitative considerations and from a formal analysis of the equations and their solutions it follows that there exists a situation in which the region of substantial absorption of the radiation is not adjacent to the entrance end of the absorbing cell, but is situated near a point z' located at a certain distance from the entrance. Simultaneously, in the vicinity of z' the density of the absorbing particle changes from zero to the asymptotic value $N_\infty = A\varkappa_0$.

The characteristic dimension of this (transition) region is determined according to (6.5) by the quantity $(1 + B\varkappa_0)/\sigma A\varkappa_0$, which sets the scale of the spatial inhomogeneity of the density also for optically tenuous media (see Sec. 3). Consequently, the thickness of the optical piston can reach 0.1 mm, which is less by a large factor than the length of the customarily employed absorbing cells.

The coordinate z' can be easily obtained with the aid of relation (5.3). Neglecting the dimension of the transition region, we obtain

$$z' = L(1 - N_0/A\varkappa_0). \qquad (6.8)$$

Since the quantity $A\varkappa_0$ depends on the frequency and on the intensity of the radiation, we can shift the position of the optical piston along the z axis by varying these parameters. This obviously changes also the value of the density on the right of z', equal to $N_\infty = A\varkappa_0$.

Let us estimate the density N_∞ to which the optical piston can compress the gas of absorbing particles. At $\nu_m = 2\nu_n$, $\Omega = \Gamma$, $2\gamma_m \sim \nu_m$, $\varkappa_0 \sim 1$ we have $N_\infty \sim 10/\lambda^3$, which amounts to 10^{13}–10^{14} cm^{-3} for the optical region of the spectrum. We note that at this density the radiation is absorbed within a very short distance ($\sim 10^{-2}$ cm).
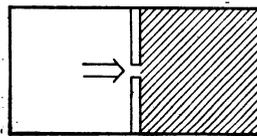
FIG. 4. Optical plunger.

We can consider now the following situation. Let the vessel containing the mixture of absorbing and buffer gases be separated by a partition with an opening (Fig. 4) through which the radiation (shown by an arrow in the figure) passes. If the thickness of the partition is chosen to be larger than or of the order of the thickness of the optical piston, then the absorbing gas can under stationary conditions be "locked" in the right-hand (shaded) part of the vessel. Thus, on account of the OID, the radiation can assume the role of a plug. The density N_∞ of the absorbing particles retained by the optical plug was estimated above. The radiation is then absorbed within the limits of the partition thickness, which can be decreased to $\sim 10^{-2}$ cm.

We recall that both the optical piston and the optical plug are impermeable only to the absorbing component of the mixture. The remaining components can diffuse freely through them.

7. CONCLUSION

The OID effect is characterized as one of the effects wherein the radiation influences the translational motion of particles. We note, however, that previously known examples of such an influence (the usual, presently called spontaneous light pressure,[8, 7] stimulated light pressure,[7] striction forces[7, 9]) are connected with the direct force action exerted by the radiation on the individual particles. In this respect OID occupies a special position. The onset of OID does not call at all for dissipation of the radiation energy, it suffices to assume that the absorption of the radiation is accompanied by its isotropic scattering without change of the frequency. The role of the radiation consists of sorting the particles by velocities (selective excitation), and the remainder is done by the collisions. Thus, the energy of the direction of motion of the particles, produced in the case of OID, is drawn from the thermal energy of the gas. This decreases, of course, the entropy of the gas mixture, but this decrease is offset of the entropy of the radiation produced when it is scattered in the gas.

The same effect as the OID is produced qualitatively also by spontaneous light pressure, i.e., it produces fluxes of absorbing particles in the gas phase, and leads to a spatial redistribution of the density. We compare now the degrees of manifestation of the OID and of spontaneous optical pressure. In the case of homogeneous broadening, the force due to the spontaneous optical pressure is given by the expression (see, e.g., Ref. 7).

$$F_L = \hbar k \gamma_m N \varkappa/(1 + \varkappa). \qquad (7.1)$$

We compare this force[2] with the force (2.14), using the result (3.10). For each of the effects we choose the conditions that are optimal for its manifestation and as a result we get

$$\left|\frac{F}{F_L}\right| \sim \frac{M\bar{v}}{\hbar k}\left|\frac{v_n - v_m}{2\gamma_m + v_m}\right|. \tag{7.2}$$

The principal factor that determines this relation is $M\bar{v}/\hbar k$ —the ratio of the thermal momentum of the particle to the photon momentum. For the optical region of the spectrum and at room temperatures $M\bar{v}/\hbar k \sim 10^4$. Thus, the forces causing the OID can exceed by three or four orders of magnitude the force of the spontaneous light pressure. This is precisely why we have neglected in the initial equations the recoil effect in absorption and emission of photons.

The OID phenomenon can find extensive applications in scientific research. We must emphasize particularly in this connection the possibility of investigating the transport cross sections of short-lived states of atoms and molecules, for which there are no reliable methods at present. Since the OID effect is directly sensitive to the difference between the transport cross sections, it becomes possible to register in experiment even small differences of these cross sections, which are characteristic, for example, for vibrational–rotational transitions of molecules.

Owing to the selectivity of the resonant action of the radiation on matter, the OID phenomenon can be used to separate isotopes and isomers, to obtain ultrapure chemical substances, etc., i.e., in problems calling for separation of one of the components of a gas mixture.

Let us assess the possibility of using OID for isotope separation. In a number of cases a suitable scheme for this purpose is based on the optical-piston effect (Fig. 4). We assume that a mixture of two isotopes enters in the right-hand side of the vessel. One of them is locked in this part by the optical plug, and the other diffuses freely through the opening. We estimate now the flow $Q = DS\nabla N_2 \sim DSN_2/l$ of the second isotope ($D = \bar{v}^2/2v_n$ is the diffusion coefficient, S is the area of the opening cross section, l is the partition thickness, and N_2 is the concentration of the second isotope in the right-hand part of the vessel). Substituting in this relation the value $l \sim 10^{-2}$ cm and using the estimate for the maximum density of the first isotope contained by the optical plug, we obtain for $S = 1$ cm^2

$$Q \sim \xi \times 10^{18} \text{ sec}^{-1}, \quad \xi = N_2/N_1, \quad \bar{v} \sim 10^5 \text{ cm/sec},$$
$$\lambda = 5 \times 10^{-5} \text{ cm}, \quad 2v_n \sim 10^8 \text{ sec}^{-1}, \tag{7.3}$$

where ξ is the ratio of the isotope concentrations in the initial mixture. Estimates for l and for the contained density of the first isotope were made for a saturation parameter $\varkappa \sim 1$. On the other hand, this value of $\varkappa$ is ensured by a power flux ~ 1 W/cm^2 (see, e.g., Ref. 6). Thus, the isotope flux (7.3) is made possible by a radiation power ~ 1 W. For the optical region of the spectrum, a power of 1 W corresponds to a photon flux $\sim 10^{19}$ sec^{-1}. Let, for example, $\xi = 1$. Then the estimate (7.3) means that ~ 10 photons are needed to separate one atom of the isotope. This estimate inspires optimism with respect to the ability of the isotope-separation method, based on the OID, with other methods, including laser methods.

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[1] The transition from the ground state is considered because in this case the OID effect is maximal.
[2] The forces of stimulated optical pressure can be higher by several orders of magnitude,[7,10] but they do not manifest themselves under the conditions of our problem.

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