# Diffusive suction and extrusion of atoms by a light field

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It is shown that during the absorption of radiation by gas atoms that then undergo transitions from the ground state the density of the absorbing atoms in the light beam changes: it increases or decreases in comparison with the equilibrium density. The effect is due to collisions with the buffer gas, and the extent of its manifestation depends on the relation between the transport cross sections for the excited and ground states. This phenomenon can form the basis of a method for measuring the transport cross sections for excited states. It can also be used in the solution of the problem of the separation and purification of isotopes.

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## 1. INTRODUCTION

In the overwhelming majority of cases the interaction of optical radiation with atoms and molecules occurs under conditions of spatial homogeneity with respect to the tranverse coordinates of the light beam. In other words, the radiative processes are brought about by the local characteristics of the radiation field. These conditions are fulfilled on account of the relatively short lifetimes of the excited states: the atom does not have enough time during the lifetime to penetrate to points where the field characteristics are appreciably different. The spatial inhomogeneity was taken into consideration only in the analysis of long-lived systems at low pressures (see, for example, the paper by Rautian and one of the present authors[1]).

Previously, when dealing with radiative processes involving the ground state and at sufficiently short lifetimes of the excited states, the analysis of the processes was performed under the assumption of spatial homogeneity. Thus, the simplest problem of monochromatic-radiation absorption was described by the following well-known equations for the density matrix:

$$\begin{gathered}(\partial/\partial t+\Gamma_m)\rho_m=-2\mathrm{Re}(iG^*\rho),\\ (\partial/\partial t+\Gamma-i\Omega)\rho=iG(\rho_n-\rho_m),\\ \Omega=\omega-\omega_{mn},\ G=Ed_{mn}/2\hbar,\ \rho=\rho_{mn},\ \rho_j=\rho_{jj};\ j=m,\ n.\end{gathered}\tag{1.1}$$

Here $E$ and $\omega$ are the amplitude of the electric field and the frequency of the radiation; $\omega_{mn}$ and $d_{mn}$ are the frequency and matrix element of the dipole moment of the $m$–$n$ transition; $\Gamma_m$ is the constant for the decay of the excited state $m$ into the ground state $n$; $\Gamma$ is the half-width of the luminescence line.

The system of equations (1.1) is not closed, and it is supplemented by the relation

$$\rho_m+\rho_n=N_0,\tag{1.2}$$

where $N_0$ is the particle density in zero field. Under steady-state conditions, from (1.1) and (1.2) we obtain, in particular, the well-known expression for the field energy,

$$P=-2\hbar\omega\,\mathrm{Re}(iG^*\rho)=2\hbar\omega|G|^2N_0\frac{\Gamma}{\Omega^2+\Gamma^2(1+4|G|^2/\Gamma\Gamma_m)},\tag{1.3}$$

which is often called the Karplus-Schwinger formula.[2]

Let us note, however, that the relation (1.2) does not follow directly from fundamental principles. Only the following relation

$$\mathrm{Sp}\,\hat{\rho}=\int d\mathbf{r}[\rho_m(\mathbf{r})+\rho_n(\mathbf{r})]=VN_0,\tag{1.4}$$

where $V$ is the total volume of the system, is rigorously valid. Since the lifetime in the ground state is infinitely long, the atom interacts with the field during the entire period it is in the light beam. However, in the course of its wanderings in space it certainly reaches the periphery of the beam, i.e., it "feels" the variation of the field in the transverse direction. Consequently, the problem of the atom-field interaction involving the ground state should, strictly speaking, be considered to be spatially inhomogeneous. Moreover, as has been pointed out earlier,[3] and rigorously validated below, the relation (1.2) may be violated, and the collisions lead to a nonequilibrium density distribution for the absorbing particles. Depending on the specific details of the collision processes, the density in the neighborhood of the light beam can turn out to be either higher, or lower than the equilibrium density. Naturally, the usual formula (1.3) will also undergo a change in this case.

## 2. DIFFUSIONAL FLUXES INSIDE THE LIGHT BEAM

In this section, we determine on the basis of the fundamental properties of the collision integrals the conditions for, and the causes of, the violation of the relation (1.2), as well as the qualitative consequences that follow from this violation.

Under steady-state conditions, the absorption of a running monochromatic wave is described by the following equations for the density matrix, which allow us to take the spatial inhomogeneity into account:

$$\begin{gathered}(\Gamma_m+\mathbf{v}\nabla)\rho_m=S_m-2\mathrm{Re}(iG^*\rho),\\ \mathbf{v}\nabla\rho_n=\Gamma_m\rho_m+S_n+2\mathrm{Re}(iG^*\rho),\\ [\Gamma-i(\Omega-\mathbf{kv})+\mathbf{v}\nabla]\rho=iG(\rho_n-\rho_m).\end{gathered}\tag{2.1}$$

Here $\mathbf{k}$ is the wave vector of the external field and $S_m$ and $S_n$ are collision integrals. The collision integral for the off-diagonal element of $\rho$ has been taken into account in the relaxation constant $\Gamma$ (collisions with phase ran-

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domization). The diagonal elements $\rho_j$ are normalized such that the integral over the velocity

$$\langle \rho_j(\mathbf{r},\mathbf{v})\rangle = \int \rho_j(\mathbf{r},\mathbf{v})\,d\mathbf{v} \tag{2.2}$$

gives the particle density in the state $j$. We assume that the collisions are elastic and occur in the general case both between the absorbing particles and with the particles of the buffer gas, whose distribution function satisfies the following equation:

$$\mathbf{v}\nabla\rho_b = S_b. \tag{2.3}$$

Thus, we have a three-component gas to deal with, and according to this

$$S_i = \sum_j S_{ij};\ i,j=\{m,n,b\}, \tag{2.4}$$

where the terms $S_{ij}$ describe the collisions of the $i$-th component with the $j$-th perturbing component.

Let us discuss Eqs. (2.1) and (2.3) on the basis of only the general properties of the collision integrals. Let us multiply each equation for $\rho_j$ in (2.1) and (2.3) by the momentum $m_j\mathbf{v}$, integrate over the velocities, and add them. As a result, we obtain

$$\sum_{\alpha j}\frac{\partial}{\partial x_\alpha}P_{\alpha\beta}{}^j = \sum_j m_j\langle v_\beta S_j\rangle;\ \alpha,\beta=1,2,3,$$
$$P_{\alpha\beta}{}^j \equiv m_j\langle v_\alpha v_\beta \rho_j\rangle. \tag{2.5}$$

The quantity $P^j_{\alpha\beta}$ has the meaning of a partial-pressure tensor for the $j$-th component of the gas.[4,5] On the other hand, in accordance with the law of conservation of momentum, the collision integrals possess the following property[4,5]:

$$m_i\langle \mathbf{v}S_{ij}\rangle + m_j\langle \mathbf{v}S_{ji}\rangle = 0. \tag{2.6}$$

On the basis of this property and the relation (2.4), the right-hand side of the equality (2.5) should vanish. This means that the total pressure of the gas does not depend on the coordinates. Such a result is entirely obvious, since the external field has no effect on the translational degrees of freedom of the atoms.[1)] If the temperature does not depend on the coordinates, then, following the pressure, the total particle density will also be a constant. However, the case is somewhat different in respect to the partial pressures and densities.

Since the lifetime of the excited state is finite, the quantity $\rho_m$ is different from zero only in the neighborhood of the light beam, i.e., the density of the excited particles and their partial pressure are essentially spatially inhomogeneous. The same thing can be said of the atoms in the ground state, whose density in the neighborhood of the light beam is reduced. Consequently, there exist the forces[2)] $\mathbf{F}^m$ and $\mathbf{F}^n$:

$$F_\alpha{}^j = -\sum_\beta \frac{\partial}{\partial x_\beta}P_{\beta\alpha}{}^j;\ j=m,n, \tag{2.7}$$

which set the excited and unexcited atoms in motion. As a result, there arise the atom fluxes

$$\mathbf{j}_m = \langle \mathbf{v}\rho_m\rangle,\ \mathbf{j}_n = \langle \mathbf{v}\rho_n\rangle.$$

The total gas pressure is constant; therefore, the motion has an especially diffusive character. Under steady-state conditions the total force,

$$\mathbf{F} = \mathbf{F}^m + \mathbf{F}^n,$$

acting on the absorbing particles as a whole is, according to (2.1) counterbalanced by the force of friction with the buffer gas:

$$-\mathbf{F} = \mathbf{F}_{fr} = \mathbf{F}_{fr}{}^m + \mathbf{F}_{fr}{}^n = m[\langle \mathbf{v}S_{mb}\rangle + \langle \mathbf{v}S_{nb}\rangle],$$
$$m = m_m = m_n. \tag{2.8}$$

Here $\mathbf{F}^j_{fr}$ is the frictional force exerted on the atoms in the state $j$ by the buffer gas and the absorbing particles in the other state.

Thus, the pressure and density of the absorbing particles become spatially inhomogeneous in the presence of a buffer gas. It is easy to verify that the buffer-gas pressure also becomes coordinate dependent, and then in such a way that the total gas pressure remains constant. It is clear that under steady-state conditions the diffusional fluxes, $j_m$ and $j_n$ cancel each other out:

$$\mathbf{j}_m + \mathbf{j}_n = 0. \tag{2.9}$$

This relation, as well as the equality

$$\mathbf{j}_b = 0 \tag{2.10}$$

follows directly from Eqs. (2.1) and (2.3) on account of the well-known property of collision integrals[4,5]: $\langle S_{ij}\rangle = 0$.

Since the frictional forces are directed counter to the fluxes, $\langle \mathbf{v}S_{io}\rangle = -\nu_i\mathbf{j}_i$, where $\nu_i$ is a coefficient of proportionality with the dimensions of the reciprocal second. The formula (2.8) then assumes the form

$$\mathbf{F}_{fr} = -m(\nu_m - \nu_n)\mathbf{j}_m. \tag{2.11}$$

In the general case the laws for the scattering of particles in the excited and ground states by the buffer-gas particles are different, so that $\nu_m \neq \nu_n$, and the extent of this difference will determine the importance of the noted spatial inhomogeneity. It is also easy to see from the formula (2.11) the cases when the density of the absorbing particles is homogeneous and the relation (1.2) is valid. To wit, the frictional force (2.11) vanishes when the scattering laws for the excited and unexcited particles are the same, in the absence of a buffer gas, as well as when $\mathbf{j}_m = -\mathbf{j}_n = 0$. Clearly the last case is realized when the radiation field is uniform over its entire active volume.

Two situations are possible when $\nu_m \neq \nu_n$, $\mathbf{j}_m \neq 0$. If $\nu_m > \nu_n$, then $\mathbf{F}_{fr}$ is directed toward the center of the light beam, and, according to (2.7), the density of the absorbing particles in the beam is higher than the density outside it. And conversely, when $\nu_m < \nu_n$ the particle density in the neighborhood of the light beam is smaller than the density beyond the limits of the beam. We can thus speak of the suction of particles into the light beam ($\nu_m > \nu_n$), or the extrusion of them from it ($\nu_m < \nu_n$).

## 3. THE SPATIAL DENSITY DISTRIBUTION OF THE ABSORBING PARTICLES

Let us solve the Eqs. (2.1), and analyze the specific manifestations of the spatial inhomogeneity of the density. Let us consider the simplest case of homogeneous broadening, i.e., the case when $\Gamma \gg k\bar{v}$, where $\bar{v}$ is the mean thermal velocity. Let us also assume that the total particle density is sufficiently high, so that the local velocity distribution differs little from the Maxwellian distribution. Moreover, let the inequality

$$\Gamma_m \gg D_m/a^2, \tag{3.1}$$

where $D_m$ is the diffusion coefficient for the excited state and $a$ is the characteristic transverse dimension of the light beam, be fulfilled. The inequality (3.1) implies that the distance over which an atom diffuses during the time $1/\Gamma_m$ is significantly smaller than the diameter of the beam.

The following equations for $\langle \rho_j \rangle$ and $\langle \rho \rangle$, which are obtained after integrating Eqs. (2.1) over the velocities and discarding the small flux terms, are valid in the indicated approximations:

$$\begin{aligned} &\Gamma_m \langle \rho_m \rangle = -2\,\mathrm{Re}(iG^* \langle \rho \rangle), \\ &(\Gamma - i\Omega)\langle \rho \rangle = iG(\langle \rho_n \rangle - \langle \rho_m \rangle). \end{aligned} \tag{3.2}$$

It is quite natural that these equations coincide with the Eqs. (1.1) taken under steady-state conditions. However, to determine the additional connection between $\langle \rho_m \rangle$, $\langle \rho_n \rangle$, and $\langle \rho \rangle$, we should proceed not from the condition (1.2), but from the rigorous relation (2.9). It is easy to construct equations for the $\mathbf{j}_m$ and $\mathbf{j}_n$ fluxes on the basis of the basic equations (2.1). Let us multiply each of the Eqs. (2.1) by, and integrate over, $\mathbf{v}$. As a result, we obtain

$$\begin{aligned} &(\Gamma_m + \nu_m)\mathbf{j}_m + {}^1/_3 \bar{v}^2 \nabla \langle \rho_m \rangle = -2\,\mathrm{Re}\,(iG^* \mathbf{j}), \\ &(\Gamma - i\Omega)\mathbf{j} + {}^1/_3 \bar{v}^2 \nabla \langle \rho \rangle = iG(\mathbf{j}_n - \mathbf{j}_m), \\ &{}^1/_3 \bar{v}^2 \nabla (\langle \rho_m \rangle + \langle \rho_n \rangle) = -\nu_m \mathbf{j}_m - \nu_n \mathbf{j}_n. \end{aligned} \tag{3.3}$$

The last equation in (3.3) is equivalent to the relation (2.11), while the quantities $\nu_m$ and $\nu_n$ in the approximation used have the meaning of collision frequencies for the transport cross section. It is not difficult to verify that the diffusion coefficients are expressible precisely in terms of them[5]:

$$D_j = \bar{v}^2/2\nu_j.$$

The Eqs. (3.3) allow us to express $\mathbf{j}_m$ and $\mathbf{j}_n$ in terms of the gradients of $\langle \rho_m \rangle$, $\langle \rho_n \rangle$, and $\langle \rho \rangle$, while the condition (2.9) establishes the necessary additional connection between these quantities. The relation has a differential character, and it is this that the spaital inhomogeneity manifests itself. The corresponding expression assumes an especially simple form when $\Gamma_m \ll \nu_m, \nu_n, \Gamma$:

$$\frac{\nabla \langle \rho_m \rangle}{\nu_m + \varkappa \Gamma_m} = -\frac{\nabla \langle \rho_n \rangle}{\nu_n + \varkappa \Gamma_m}, \quad \varkappa \equiv 4|G|^2 \frac{\Gamma}{\Gamma_m(\Gamma^2 + \Omega^2)}. \tag{3.4}$$

Let us further introduce for consideration the population-density sum and difference:

$$N = \langle \rho_m \rangle + \langle \rho_n \rangle, \quad n = \langle \rho_n \rangle - \langle \rho_m \rangle. \tag{3.5}$$

From Eqs. (3.2) and (3.4) we obtain relations between $N$ and $n$ and a differential equation for the absorbing-particle density $N$:

$$n = \frac{N}{1+\varkappa}, \tag{3.6}$$

$$\frac{\nabla N}{N} = \left(\frac{\nu_m - \nu_n}{\nu_m + \nu_n}\right) \nabla \varkappa / (\varkappa + 1)\left(\varkappa + \frac{2\nu_m}{\nu_m + \nu_n}\right). \tag{3.7}$$

Since Eq. (3.7) is homogeneous with respect to the spatail derivative, the dependence of its solution on the coordinates is a parametric one. If the buffer-gas density is significantly higher than the absorbing-gas density, then the collision frequencies virtually do not depend on the coordinates. To simplify the formulas, let us make this assumption. Then as a result of the integration of Eq. (3.7), we obtain

$$N(\varkappa) = N(0)\frac{1+\varkappa}{1+\varkappa(\nu_m+\nu_n)/2\nu_m}. \tag{3.8}$$

If the transverse dimension of the active volume significantly exceeds the diameter of the light beam, then $N(0)$ is virtually equal to the equilibrium density $N_0$. If, on the other hand, the indicated dimensions are comparable, then we should use for the determination of $N(0)$ the condition

$$\int N(\varkappa(\mathbf{r}))\,d\mathbf{r} = VN_0, \tag{3.9}$$

which is equivalent to the condition (1.4).

As can be seen from (3.8), the ratio of the density at an arbitrary point inside the light beam to the density outside it depends only on the field intensity at the given point, but not on the specific intensity distribution over the cross section. If $\nu_m \neq \nu_n$ and $\varkappa \neq 0$, then the ratio $N(\varkappa)/N(0)$ is different from unity. When $\nu_m > \nu_n$ the quantity $N(\varkappa)/N(0)$ is greater than one, i.e., the absorbing particles are sucked into the light beam. When $\nu_m < \nu_n$ the particles are expelled from the beam $[N(\varkappa) < N(0)]$. The density drop increases with increasing field intensity (i.e., with increasing $\varkappa$), and ceases to depend on the intensity when $\varkappa \gg 1$:

$$\frac{N(\varkappa)}{N(0)} = \frac{2\nu_m}{\nu_m + \nu_n}. \tag{3.10}$$

As can be seen from (3.10), almost all the absorbing particles are expelled from the beam when $\nu_n \gg \nu_m$. This situation can be interpreted as follows. Under the action of the radiation field, the particles undergo a transition into the excited state, in which they rapidly diffuse beyond the limits of the light beam. The inflow of particles, on the other hand, is greatly impeded, since outside the beam they are in the ground state and, on account of the condition that $\nu_n \gg \nu_m$, possess a relatively small diffusion coefficient. After the lapse of a sufficiently long period of time, determined by the rate of diffusion in the ground state, almost all the particles are extruded from the region of the field.

Under the opposite condition $\nu_m \gg \nu_n$ the particle density inside the beam is higher than the density outside it by a factor of two. In order to interpret this case, let us direct our attention to the expressions, obtained from (3.8) with allowance for (3.5) and (3.6), for the partial densities of the excited and unexcited particles:

$$\begin{aligned} \langle \rho_m \rangle &= N(0)\frac{\varkappa/2}{1+\varkappa(\nu_m+\nu_n)/2\nu_m}, \\ \langle \rho_n \rangle &= N(0)\frac{1+\varkappa/2}{1+\varkappa(\nu_m+\nu_n)/2\nu_m}. \end{aligned} \tag{3.11}$$

In an intense field ($\varkappa \gg 1$), the values of $\langle \rho_m \rangle$ and $\langle \rho_n \rangle$ become equal (saturation effect). On the other hand, the condition $\nu_m \gg \nu_n$ implies that the excited particles practically do not diffuse, whereas the unexcited particles manage through relatively fast diffusion to eliminate the inhomogeneity in their density. As a result, the particles that are in the ground state are uniformly distributed over the volume, while in the neighborhood of the beam, on account of the equality $\langle \rho_m \rangle = \langle \rho_n \rangle$, the total density is two times higher. The dependence of the ratio $N(\varkappa)/N(0)$ on $\varkappa$ is shown in Fig. 1 for different rela-

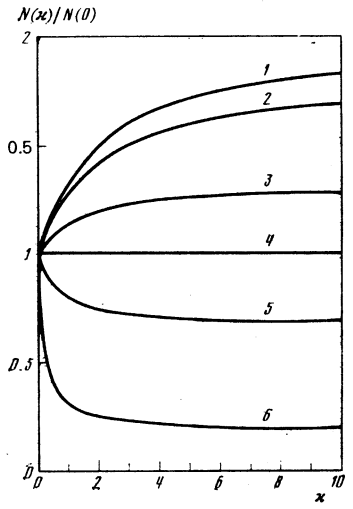


FIG. 1. Dependence of the relative absorbing-atom density $N(\mathscr{H})/N(0)$ on the saturation parameter $\mathscr{H}$ for different $\zeta = \nu_m/\nu_n$. 1) $\zeta = \infty$, 2) $\zeta = 10$, 3) $\zeta = 2$, 4) $\zeta = 1$, 5) $\zeta = 0.5$, 6) $\zeta = 0.1$.

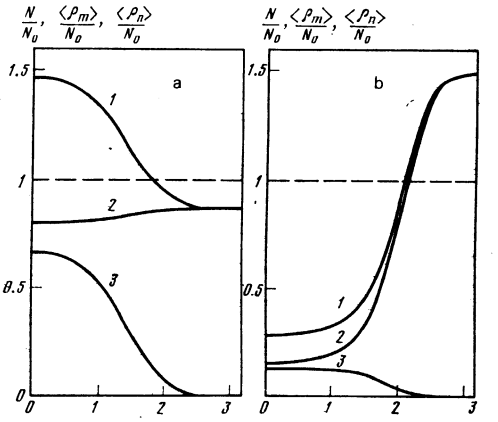


FIG. 2. Spatial distribution of the total density $N$ (1) and the partial densities $\langle \rho_n \rangle$ (2) and $\langle \rho_m \rangle$ (3) of the absorbing atoms in the case of a Gaussian beam. a) $\nu_m/\nu_n = 10$; b) $\nu_m/\nu_n = 0.1$.

tions between $\nu_m$ and $\nu_n$.

Knowledge of the spatial distribution of the intensity in the light beam is necessary for the computation of the absolute values of the density, since the quantity $N(0)$ depends on it. For a rectangular light-beam profile with a cross-sectional area $S_0$, we obtain from (3.9) the formula

$$N(0) = N_0 \left\{1 + \frac{\varkappa(\nu_m+\nu_n)}{2\nu_m}\right\} \Big/ \left\{1 + \frac{\varkappa}{2\nu_m}\left[\nu_m + \nu_n + \frac{S_0}{S}(\nu_m - \nu_n)\right]\right\}, \tag{3.12}$$

where $S$ is the cross-sectional area of the active volume. If $\varkappa$ has the following dependence on the transverse radius vector $\mathbf{r}$:

$$\varkappa(\mathbf{r}) = \varkappa_0 \exp\left[-(r/a)^2\right], \tag{3.13}$$

which characterizes a Gaussian beam, then $N(0)$ is given by the formula

$$N(0) = N_0 \Big/ \left[1 + \frac{S_0}{S}\left(\frac{\nu_m-\nu_n}{\nu_m+\nu_n}\right)\ln\left(1+\varkappa_0\left(\frac{\nu_m+\nu_n}{2\nu_m}\right)\right)\right], \tag{3.14}$$

$$S_0 = \pi a^2.$$

As it should be, at $S_0 \ll S$ the density outside the beam is equal to the euqilibrium density.

Figure 2 shows the spatial distributions of the partial densities and the total density of the absorbing particles in the case of a Gaussian beam. The plots correspond to the values $\varkappa_0 = 10$, $S/S_0 = 10$. The dashed line indicates the level of the equilibrium density.

Let us note the influence exerted by the density-redistribution effect on the field energy, which can easily be found from (3.2), using the formulas (3.6), (3.8), (3.12), and (3.14). In the simplest case, when $S_0 \ll S$, the expressions for the field energy on the axis of a Gaussian beam and in a beam of rectangular profile are identical, and have the form

$$P = 2\hbar\omega |G|^2 N_0 \Gamma \Big/ \left\{\Omega^2 + \Gamma^2\left[1 + \frac{4|G|^2}{\Gamma\Gamma_m}\left(\frac{\nu_m+\nu_n}{2\nu_m}\right)\right]\right\}. \tag{3.15}$$

The difference between this formula and the well-known formula (1.3) consists in the presence of the additional factor $(\nu_m + \nu_n)/2\nu_m$ in front of the usual saturation factor $4|G|^2/\Gamma\Gamma_m$. If $\nu_m > \nu_n$, then the saturation parameter is effectively reduced. This leads, other things being equal, to an increase in $P$ and the narrowing of the contour of the $P(\Omega)$ line. Such a result is reasonable, since a greater number of particles interact with the field when $\nu_m > \nu_n$, and this number depends on $\varkappa$ and, consequently, $\Omega$, decreasing with increasing $\Omega$. In the opposite situation, i.e., when $\nu_m < \nu_n$, the saturation parameter is effectively increased, the $P(\Omega)$ line is broadened, and the quantity $P$ is, on the whole, decreased. The explanation of this change in the field energy is similar to the explanation given for the $\nu_m > \nu_n$ case.

## 4. DISCUSSION

As follows from the above-performed analysis, the drop in the density of the absorbing particles in, and outside, the light beam, i.e., the magnitude of the effect of particle suction or extrusion by a light beam, is dependent upon the saturation parameter $\varkappa$ and the ratio, $\nu_m/\nu_n$, of the collision frequencies corresponding to the transport cross sections for collisions of the excited and unexcited particles with the particles of the buffer gas. According to the presently available data (see, for example, Refs. 6 and 7), the difference between these cross sections for the electronic states of atoms can be quite considerable ($\nu_m$ and $\nu_n$ can differ by a factor of two). Notice that, according to Ref. 7, the transport cross section for an excited state can be smaller than the cross section for the ground state. Consequently, besides the suction effect, the effect of extrusion from a light beam is, in principle, also possible.

Unfortunately, data on the transport cross sections for excited atoms and molecules are extremely scanty, and confined to metastable states. This circumstance stems from the absence of any reliable methods of measuring cross sections for short-lived states.

The extent of manifestation of the effects considered

in the present paper is to be expected to be greatest when the transitions involved are optically allowed transitions, but because of the absence of data on transport cross sections for short-lived levels, it is impossible to carry out a detailed estimate of the effect in a specific system. We can, however, hope that $\nu_m$ differs greatly from $\nu_n$ also in the case of allowed transitions. On the other hand, the effects of diffusive suction and extrusion can by themselves consititute a basis for the experimental determination of the transport cross sections for short-lived states. To wit, the relation between $\nu_m$ and $\nu_n$ can be determined from measurements of the absorbing-particle density drop, which, in its turn, can be found from the measurement of the absorption of test radiation.

Naturally, in order for the density drop in, and out of, a light beam in a specific system to attain its maximum value, it is necessary that the saturation parameter, $\varkappa$, have a large value, i.e., that $\varkappa \gg 1$. Let us estimate the radiation-power flux for which such values of $\varkappa$ are attained. At exact resonance we have from (3.4) and the definition of the flux $p$ that

$$\varkappa = \frac{6e^2}{mc^3\hbar}\frac{\lambda p}{\Gamma\Gamma_m} f = \frac{4\cdot 10^4 \bar{\lambda}\bar{p}}{\Gamma\Gamma_m} f, \tag{4.1}$$

where $e$ and $m$ are the electron charge and mass, $\bar{\lambda}$ is the wavelength in $\mu$m, $\bar{p}$ is the power flux in W/cm$^2$, $f$ is the oscillator strength, and $\Gamma$ and $\Gamma_m$ are in MHz.

For definiteness, let us consider the $D_2$ line of sodium: $\lambda = 0.589\mu$m, $\Gamma_m = 10$ MHz, and $f \sim 1$. In the case when $\Gamma \sim k\bar{v} \sim 10^3$ MHz, the quantity $\varkappa$ becomes of the order of ten when

$$\bar{p} \sim 5 \text{ W/cm}^2 . \tag{4.2}$$

Grove *et al.*[8] have investigated the resonance flourescence of sodium vapor, induced by a dye laser with radiation wavelength $\lambda = 0.589\mu$m and power in the cw regime of 40 mW (far from being a record). However, even for this power the value (4.2) for the flux is secured when the radiation is focused into a spot of $d \sim 1$ mm. Thus, the described effects can be observed in real objects with the aid of existing radiation sources. In particular, the diffusion coefficient of the $3^2P_{3/2}$ excited state of sodium can be measured.

It is of interest to compare the extent of the manifestation of the diffusion-induced suction and extrusion effects with that of the analogous electrostriction-induced effect first considered by Askar'yan.[9] The saturation parameter $\varkappa$ can be written in the form

$$\varkappa = \alpha(kT/\hbar\Gamma_m)6f, \quad \alpha = e^2E^2/m\omega\Gamma kT. \tag{4.3}$$

Here $k$ is the Boltzmann constant. The quantity $\alpha$ is a parameter ($\alpha = |U|/kT$) determining the effect of the strictional forces under resonance conditions, when the extent of their manifestation is greatest.[9] In order for the effect of the electrostriction to be appreciable, it is necessary that we have $\alpha \gtrsim 1$. It can be seen from (4.3) that the parameter $\varkappa$ exceeds the quantity $\alpha$ by a factor of $kT/\hbar\Gamma_m$. For $T = 300$ K and $\Gamma_m = 10$ MHz, we have $kT/\hbar\Gamma_m \sim 0.6\times 10^6$. Thus, the effects described in the present paper appear at considerably lower (by five-six orders of magnitude) values of the radiation intensity.

Similarly, it can be shown that the other effects connected with the direct action of a light field on the translational degrees of freedom of an atom[10] do not mask the manifestation of the diffusive suction and extrusion of atoms.

The calculations carried out in the present paper are, strictly speaking, applicable only to electron transitions in atoms. It is clear that the effect should also occur in the case of the vibrational-rotational transitions of molecules if the intensity of the field is strong enough to make a significant change in the total population of the vibrational states. Estimates of the effect in specific systems can, however, be made only after a special analysis of the corresponding problem with allowance for the fact that under normal conditions inhomogeneous broadening is characteristic of vibrational-rotational transitions and that rotational relaxation is important. The analysis of such systems falls outside the framework of the present paper.

Finally, let us note another possible practical application of the above-described phenomenon. On account of the exceptional selectivity of the effect of optical radiation on a substance, the absorbing-particle density redistribution effect can find application in the problem of isotope separation and purification. To wit, in a mixture of isotopes the influence exerted by radiation will be felt by an isotope of a definite type, and the redistribution of density over the volume will be effected only for this isotope. Here we should also note the additional positive role played by the buffer gas, which, in the present case, not only ensures the effect of spatial redistribution of the density of the absorbing isotope, but also impedes excitation transfer to other isotopes.

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[1)] Here we do not consider the fine effects connected with the light pressure.

[2)] Strictly speaking, $\mathbf{F}^j$ is a force per unit volume, but for the sake of brevity we shall simply call it force.

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# "Tangential" conical refraction in a three-dimensionally inhomogeneous weakly anisotropic medium

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The propagation of an electromagnetic wave in a three-dimensionally inhomogeneous weakly anisotropic medium with a real dielectric tensor (deformed crystal, elastically stressed isotropic material, and others) is considered. It is shown that in the vicinity of the points that would correspond in the presence of a boundary surface to internal conical reflection there should occur an intense mutual transformation of linearly polarized waves with mutually perpendicular polarization directions. For the case when the region of wave interaction is much smaller than the scale of the inhomogeneities of the medium, analytic expressions are obtained for the fields and for the transformation coefficients. It is shown that the effect takes the most complete and simplest form only in the cases when the problem has sufficient asymmetry, i.e., it is inhomogeneous, is in not fewer than two dimensions.

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## INTRODUCTION

It is known that conical (internal) reflection is produced when a plane wave is incident on a homogeneous crystal in those cases when the refractive indices for the two types of normal waves are equal to each other in the given direction of the wave vector k (Ref. 1):

$$n_1(\mathbf{k}, \hat{\varepsilon}) = n_2(\mathbf{k}, \hat{\varepsilon}); \quad (1)$$

$\varepsilon$ is the dielectric tensor of the medium. (Following Ginzburg's book,[2] we define the ordinary wave as the one corresponding to the larger refractive index.)

The purpose of the present paper is to determine the changes that occur in the classical effect if the homogeneous crystal is replaced by an anisotropic smoothly inhomogeneous medium with an inhomogeneity scale $L$ much larger than the wavelength $\lambda$. Accordingly, the condition (1) is then satisfied not in the entire volume occupied by the wave, but only at certain points, at which the wave vector $\mathbf{k}(\mathbf{r})$ and the principal axes of the tensor $\hat{\varepsilon}(\mathbf{r})$ have a suitable relative orientation (see Sec. 1). It is clear that this gives rise to a mutual linear transformation of the ordinary and extraordinary waves (see Secs. 1 and 4), and this transformation is similar in many respects to the types of transformation which occur when high-frequency electromagnetic waves pass through a magnetoactive plasma, namely, in the region of quasitransverse propagation,[3–6] as well as in a neutral current layer[7] and in the case of propagation in a direction close to that of the magnetic field.[2]

Since there are no abrupt separation boundaries in the case considered here, the ray scattering characteristic of the classical form of the effect does not occur here and its place is taken by a specific polarization picture which is connected with the wave transformation. In some implicit form, however, the effect leaves a trace also in the ray picture (see Sec. 5).

It is useful to note that effects connected with linear transformation of vibrational and wave modes are known in many divisions of physics,[8] principally in plasma physics. Optics is an exception, and the present article should fill this gap. Linear transformation effects manifest themselves most clearly in nonstationary oscillations of lumped systems[9] and accordingly in nonstationary quantum-mechanics problems connected with adiabatic perturbations.[10] The latter includes, in particular, the umklapp process in beams of polarized particles[11,12]; the optical effect considered here is similar to it in many respects.

The inhomogeneous anisotropic medium (with real $\hat{\varepsilon}$) in which the "tangential" conical refraction takes place need not necessarily be an inhomogeneously deformed crystal. It can be also inhomogeneously deformed glass, in which the optical anisotropy is due to the elasto-optical effect. Another optically inhomogeneous anisotropic material is a moving liquid with an inhomogeneous velocity field, in which the optical anisotropy is due to the Maxwell effect. By dissolving in this liquid a substance with natural optical activity (such as sugar and others) we can obtain an optically anisotropic medium with complex tensor $\hat{\varepsilon}$.

Thus, we consider primarily the case when the tensor $\hat{\varepsilon}$ is real. The presence of gyration (nonzero $\mathrm{Im}\hat{\varepsilon}$)

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