Light-induced diffusion of gases

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A new effect of light-induced diffusion of gases is reported. This effect basically can be described as follows: a monochromatic traveling wave, upon absorbing atoms in the transition from the ground state, induces a macroscopic diffusion flux of absorbing atoms moving in the direction of or opposite to the light flux. As a result, the atoms collect at the end of the absorbing container in a $\leqslant$1-mm-thick layer.

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The resonance absorption of light by atoms in the transition from the ground state has a unique characteristic: the time the atom interacts with the field is rather long and is limited only by the time the atom stays in the light beam. This produces a number of interesting effects, one of the most striking of which— the light-induced diffusion of gases— is reported in this paper. This effect basically can be described as follows. A traveling light wave produces a macroscopic flux of absorbing particles, if the latter are mixed with a buffer gas. The flux, depending on specific circumstances, is directed either toward the propagating wave or away from it. The estimates show that the forces producing this flux, which can exceed the force of light pressure by several orders of magnitude, make it possible to collect the absorbing particles from the entire cell of arbitrary length and concentrate them in a very thin layer ($\sim$1 mm or less) at its end.

The effect can be described as follows. If the radiation frequency ω differs slightly from the frequency ω_{mn} of the m-n atomic transition, then, because of the Doppler effect, the velocity distributions $\rho_{mn}(\mathbf{V})$ and $\rho_{mn}(\mathbf{V})$ of the populated excited (m) and ground (n) states become asymmetric. Therefore, there exist population fluxes $\mathbf{j}_{ll} = \langle \mathbf{v}_{\rho ll}(v) \rangle$ ($l = m, n$; the angle brackets denote integration over the velocities). This produces internal frictional forces[1,2] directed against the flux:

$$\mathbf{F}_l \equiv M \langle \mathbf{v} S_l(\mathbf{v}) \rangle = - M \nu_l \mathbf{j}_{ll}; \qquad (l = m, n) \tag{1}$$

where M is the mass of the absorbing atom, $S_l(\mathbf{v})$ is the collision integral, and ν_l is the coefficient of the collision frequency, which is governed by the interaction character of the atom in the state l as a result of collisions with the surrounding particles. If the buffer gas is missing, then $\langle \mathbf{v} S_m(\mathbf{v}) \rangle + \langle \mathbf{v} S_n(\mathbf{v}) \rangle = 0$ according to the momentum conservation law (see Refs. 1 and 2). Therefore, the force $\mathbf{F} = \mathbf{F}_m + \mathbf{F}_n$ acting from the direction of the field on the absorbing atoms as a whole, as expected, is equal to zero.[1)] The situation is different if there is a buffer gas in addition to the absorbing gas in the volume. Of course, the force acting on the gas as a whole in this case in also equal to zero. However, the force $\mathbf{F} = \mathbf{F}_m + \mathbf{F}_n$ acting only on the absorbing atoms, which sets them in motion, generally is nonvanishing. This is clearly evident when the density of the buffer gas is much higher than that of the absorbing gas. Thus, ν_m and ν_n depend upon the interaction of the excited and nonexcited atoms, respectively, with the buffer

gas. If the velocity distributions $\rho_{ll}(\mathbf{v})$ differ little from the Maxwellian distribution $w(\mathbf{v})$, then the collision frequencies ν_m and ν_n depend on the relevant transport cross sections. It is clear that since generally the transport cross sections are different,[2)] it follows from Eq. (1) that

$$\mathbf{F} = M[(\nu_n - \nu_m)\mathbf{j}_m - \nu_n \mathbf{J}]; \qquad \mathbf{J} \equiv \mathbf{j}_m + \mathbf{j}_n, \tag{2}$$

where $\mathbf{J}$ is the flux of the absorbing particles. It is clear from Eq. (2) that a simultaneous vanishing of the force $\mathbf{F}$ and the flux $\mathbf{j}$ is impossible. If the field intensity is homogeneous along the transverse cross section of the cell,[3)] then two steady-state modes can exist: steady-state flux mode when the force $\mathbf{F}$ is equal to zero and steady-state mode for a closed volume with the absence of particle flux but when the force $\mathbf{F}$, which is compensated for by the density gradient, is nonvanishing.

In the specific calculation we used the electronic transitions of atoms under conditions of homogeneous broadening when the velocity dependence of the elements of the atomic density matrix is close to the Maxwellian:

$$\rho_{lk}(\mathbf{v}) = [\rho_{lk} + \frac{2}{v^2}\mathbf{v}\mathbf{j}_{lk}]w(\mathbf{v}). \tag{3}$$

We can easily formulate equations for ρ_{lk} and $\mathbf{j}_{lk}$ from the ordinary equations for the density matrix, which describe the interaction with the monochromatic traveling wave. Their solution in the steady-state flux mode leads to the following results:

$$\mathbf{J} = \mathbf{k}\bar{v}^2 \frac{2\gamma_m}{\nu_n}\left(\frac{\Omega}{\Gamma^2 + \Omega^2}\right)\frac{a}{1 - 2\gamma_m a(1+\kappa)/\nu_n} N, \tag{4}$$

where the following notation are introduced:

$$\Omega = \omega - \omega_{mn}; \qquad a = \frac{1}{2}\frac{\nu_n - \nu_m}{\nu_m + 2\gamma_m(1+\kappa)}\left(\frac{\kappa}{1+\kappa}\right);$$
$$\kappa = \frac{|E|^2 d_{mn}^2}{2\hbar^2\gamma_m}\left(\frac{\Gamma}{\Gamma^2 + \Omega^2}\right); \qquad N = \rho_{mm} + \rho_{nn}. \tag{5}$$

Here $\mathbf{k}$ is the wave vector, $\bar{v}$ is the average thermal velocity, γ_m is the decay constant of the excited level, Γ is the half width of the luminescence line, E is the amplitude of the electric radiation field, d_{mn} is the matrix element of the dipole moment, and N is the density of the absorbing particles.

As seen in Fig. 4, the direction of the flux depends on the sign of Ω— the difference between the radiation frequency and the transition frequency— and on the sign of the difference between ν_m and ν_n. If $\nu_m > \nu_n$, then for a positive Ω the flux of the absorbing atoms is in the opposite direction to that of the light flux. Let us estimate the velocity of the flux $u = J/N$ for typical atomic characteristics under typical experimental conditions:

$$2\gamma_m \sim 10^7 \text{ Hz}, \quad \nu_n \sim \nu_m \sim 10^8 \text{ Hz}, \quad \lambda = 2\pi/k \sim 10^{-4} \text{ cm}, \quad \Gamma \sim 10^9 \text{ Hz},$$
$$\bar{v} \sim 5\times 10^4 \text{ cm/sec}. \tag{6}$$

If $(\nu_m - \nu_n)/\nu_m \sim 1$, then at $\Omega = \Gamma$ and $\kappa \sim 1$ (such values of the saturation parameter κ are attainable) it follows from Eq. (4) that $u \sim 10^3$ cm/sec, which is a rather large value.

For a closed volume in the steady-state mode ($\mathbf{J} = 0$), we obtain from the equations for ρ_{lk} and $\mathbf{j}_{lk}$ the following equation for the density of the absorbing particles:

$$\frac{\nabla N}{N} = k \frac{a}{1+a} \frac{4\Omega\gamma_m}{\Gamma^2 + \Omega^2} . \tag{7}$$

For the same characteristic parameters we have:

$$\left| \frac{\nabla N}{N} \right| \sim 10^2 \text{ cm}^{-1}. \tag{8}$$

This means that the absorbing atoms are accumulated almost entirely in a $\sim 10^{-2}$-cm-thick layer at one or the other end of the absorbing cell (depending on the sign of Ω and on the difference between ν_n and ν_m).

The experiments on absorption of light in the transition from the ground state of the atoms and molecules have been conducted for a long time and the impressive magnitude of the effect described above leads us to suspect that it already manifested itself in some of the experiments. In fact, a literature search turned up[4)] the work of Ashkin *et al.*[4] conducted in 1975, in which the study of light pressure in sodium vapor showed that at $\Omega > 0$ the vapor density increases in the opposite direction to that of the light flux. This effect could not be explained by Ashkin *et al.*[4] We can assume with great confidence, however, that Ashkin *et al.*[4] were dealing with light-induced diffusion of gases. Of course, since the conditions for observation of the indicated effect[4] were far from optimum, the hope of getting a much more pronounced effect is fully justifiable.

In conclusion, we note that the effect of light-induced diffusion can be effectively used for separation of isotopes. If, for example, two isotopes must be separated, then by proper selection of laser frequency we can have one of them flow to one end of the container and the other to the opposite end.

[1)] We omitted here the effects of light pressure and dissipation of the radiation energy, which is justifiable for optical transitions in atoms.
[2)] The difference between them can be rather large.[3]
[3)] Otherwise convection currents will occur.
[4)] We thank G.I. Surdutovich for pointing out this reference.

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