

# Light-induced diffusion of gases in the field of a nonmonochromatic wave

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Recently, Ref. 1 predicted and Ref. 2 recorded an experimental observation of the appearance of a macroscopic diffusional flow of absorbing atoms in a buffer gas medium in the field of a monochromatic wave of laser radiation. A change in the direction of flow was observed when there was a change in sign of the radiation frequency detuning relative to the atomic resonance. The phenomenon has extensive applications to the physics of selective action of radiation on matter and to the study of the processes of atomic and molecular collisions.

In the present work, we studied the possibilities of achieving light-induced diffusion (LID) in a field of nonmonochromatic radiation. The study led to the important conclusion that it is advantageous to use nonmonochromatic radiation, which substantially broadens the possibilities of studying and utilizing LID.

The LID phenomenon essentially consists in the following. A traveling wave of monochromatic radiation, entering into a resonant interaction with a gas, excites only atoms with certain velocities dependent on the magnitude and sign of the frequency detuning of the radiation relative to the atomic resonance. As a result, flows of excited and unexcited atoms offsetting each other and having different directions appear in the absorbing medium. Since the transport collision cross sections are different for the excited and unexcited atoms, the compensation of flows in the buffer gas medium is disrupted, and a macroscopic flow of gas as a whole takes place.

We will study the LID phenomenon in a nonmonochromatic field. This radiation will be represented as a set of independent modes with frequency difference  $\Delta$ , spectral halfwidth  $\delta$ , and a Lorentzian distribution of amplitude squared:

$$E = \sum_j E_j \exp[-i(\omega_j t - kx)], \quad E_j^2 = E_0^2 \frac{\delta^2/\pi}{\delta^2 + (s\Delta)^2}, \quad \omega_j = \omega_0 + s\Delta. \quad (1)$$

The integrated intensity is  $I = c\delta E_0^2/8\pi\Delta$ . We assume the relation between the impact width ( $\Gamma$ ) and Doppler width ( $k\bar{v}$ ) to be arbitrary and solve the problem with the aid of the strong-collision model.

It can be shown that the steady flux  $J$  of absorbing particles as a whole is expressed in terms of the particle fluxes at the upper and lower energy levels as follows:

$$J = \frac{\nu_m - \nu_n}{\nu_m + \nu_n} (j_n - j_m) = \frac{\nu_m - \nu_n}{\nu_m + \nu_n} \int v [p_m(v) - p_n(v)] dv. \quad (2)$$

$\nu_m$  and  $\nu_n$  being the frequencies of collisions with the buffer gas at the upper and lower energy level, respectively,  $v$  being the velocity of the absorbing atoms, and  $\rho$  being the density matrix.

Under steady-state conditions, we have the following expression for flux  $J$ :

$$J = \frac{\nu_m - \nu_n}{\nu_m + \nu_n} \frac{\bar{\Gamma} x_1 \langle vY(v) \rangle}{1 + (\xi - 1)\bar{\Gamma} x_1 \langle Y(v) \rangle} N, \quad (3)$$

$$Y(v) = \frac{\bar{\Gamma}}{\bar{\Gamma}^2(1+x_1) + (\Omega - kv)^2}; \quad x_1 = \frac{1}{\xi} \frac{8\pi |d_{mn}|^2}{\hbar^2 c \bar{\Gamma} \Gamma_m} I,$$

$$\xi = \frac{2\nu_n(\Gamma_m + \nu_m)}{(\nu_n + \nu_m)\bar{\Gamma}_m}; \quad \Omega = \omega - \omega_{mn},$$

where  $\bar{\Gamma} = \delta$  in the nonmonochromatic case ( $\delta \gg \Gamma$ ) and  $\bar{\Gamma} = \Gamma$  in the monochromatic case ( $\delta \ll \Gamma$ ),  $N$  is the concentration of absorbing particles,  $d_{mn}$  is the matrix element of the dipole moment,  $\Gamma_m$  is the radiation relaxation constant of level  $m$ , and the  $\langle \dots \rangle$  angle brackets denote averaging with a Maxwellian distribution.

The quantity  $\bar{\Gamma}(1 + \kappa_1)^{1/2}$  represents the width of the Bennett dip in the velocity distribution of the population difference. For a uniform broadening  $\bar{\Gamma}(1 + \kappa_1)^{1/2} \gg k\bar{v}$ ; it then follows from Eq. (3) that

$$J = \frac{k}{k} v \frac{(\nu_m - \nu_n)}{(\nu_m + \nu_n)} \frac{1}{\xi} \frac{x_0}{1 - x_0} \frac{\Omega k v N}{\bar{\Gamma}^2(1+x_1) + \Omega^2}, \quad (4)$$

$$x_0 = \frac{8\pi |d_{mn}|^2}{\Gamma_m \hbar^2 c} I \frac{\bar{\Gamma}}{\bar{\Gamma}^2 + \Omega^2}.$$

This formula naturally extends (with the aid of the substitution  $\bar{\Gamma} \rightarrow \delta$ ) the expression for the flux for uniform broadening in a monochromatic field<sup>1</sup> to the case of nonmonochromatic radiation in the form of Eq. (1).

The flux reaches its maximum value when  $\Omega \sim \bar{\Gamma}(1 + \kappa_1)^{1/2} \sim k\bar{v}$ ,  $\kappa \geq 1$ ,  $\xi \leq 1$ . It is evident from the expression for  $\xi$  that  $\xi \leq 1$  if at least one of the collision frequencies is smaller than or of the order of  $\Gamma_m$ . In the opposite case ( $\nu_m, \nu_n \gg \Gamma_m$ ),  $\xi$  becomes greater, leading to a decrease in flux. This is natural, since at high collision frequencies, the relative fraction of the non-equilibrium part in the velocity distribution decreases.

Thus from the standpoint of the magnitude of  $J$ , it is necessary to confine oneself to buffer gas pressures at which  $\nu_j \lesssim \Gamma_m$ . It is well known, however, that the line-width  $\Gamma$  and collision frequencies  $\nu_j$  are not independent. As the impact width increases (due to pressure),  $\nu_j$  also increase. In many situations, when  $\Gamma \sim k\bar{v}$  the inequality  $\nu_m, \nu_n \gg \tau_m$  simultaneously applies. Therefore, if monochromatic radiation is used, in the case of homogeneous broadening, the parameter  $\xi$  is large, and the flux  $J$  does not reach its maximum possible value. In the nonmonochromatic case we have the opportunity of selecting the optimum  $\delta$  [so that  $\delta(1 + \kappa_1)^{1/2} \sim k\bar{v}$ ] without at the same time taking  $\xi$  out of the region of optimum values, since these parameters are independent. Hence, by using nonmonochromatic radiation, one can achieve the conditions for the maximum particle flux. Within

the framework of applicability of Eq. (4), the advantage over other cases may be by a factor of  $\xi \gg 1$ .

Even greater advantages of the use of nonmonochromatic radiation are evident under conditions of inhomogeneous broadening. In this limit [ $\Gamma(1 + \kappa_1)^{1/2} \ll k\bar{\nu}$ ], we obtain from Eqs. (3)

$$J = \frac{k}{k} \rho \frac{\nu_a - \nu_m}{\nu_a + \nu_m} \frac{\Omega}{k\bar{\nu}} \frac{\pi^{1/2} \frac{\Gamma(1 + \kappa_1)^{1/2}}{k\delta} \frac{x_1}{1 + x_1} e^{-(\Omega/k\delta)^2 N}}{1 + (\xi - 1) \pi^{1/2} \frac{\Gamma(1 + \kappa_1)^{1/2}}{k\delta} \frac{x_1}{1 + x_1} e^{-(\Omega/k\delta)^2}} \quad (5)$$

It is apparent that in this case, the optimum values of the parameters are  $\Gamma(1 + \kappa_1)^{1/2} \sim k\bar{\nu}$ ,  $\kappa_1 \geq 1$ , and  $\xi \leq 1$ . It is obvious from the definition of  $\kappa_1$  that large values of  $\xi$  lead to small values of  $\kappa_1$ . Even when one assumes that values of  $\kappa_1 \geq 1$  can be maintained by suitably increasing the intensity, the flux at  $\xi \gg 1$  nevertheless becomes small [because of the presence of the factor  $(\xi - 1)$  in the denominator of Eq. (5)].

As follows from Eq. (5) and the expression for  $\kappa_1$ , in weak fields ( $\kappa_1 \ll 1$ ) the magnitude of the flux is independent of the spectral width of the radiation and grows lin-

early with  $I$ . When  $\kappa_1 \geq 1$ , the linear growth is replaced by a slower dependence. As  $I$  grows, the flux  $J$  increases until the width of the Bennett dip  $\Gamma(1 + \kappa_1)^{1/2}$  reaches values close to  $k\bar{\nu}$ , corresponding to the optimum conditions for the magnitude of the flux  $J$ . For nonmonochromatic radiation, the linear increase is retained up to higher intensity values, and the maximum of  $J$  is reached at values of  $I$  that are  $\delta/\Gamma$  smaller than for monochromatic radiation.

Thus, the analysis of the LID phenomenon under conditions of inhomogeneous broadening opens up new advantages of nonmonochromatic radiation.

From the above study, one can conclude that nonmonochromatic radiation is useful in the realization of LID.

<sup>1</sup>F. Kh. Gel'mukhanov and A. M. Shalagin, Pis'ma Zh. Eksp. Teor. Fiz. 29, 773 (1979) [JETP Lett. 29, 711 (1979)].  
<sup>2</sup>V. D. Antsygin, S. N. Atutov, F. Kh. Gel'mukhanov, G. G. Telegin, and A. M. Shalagin, Pis'ma Zh. Eksp. Teor. Fiz. 30, 262 (1979) [JETP Lett. 30, 243 (1979)].