

Collisional orientation of atoms and molecules in a resonance light field

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It is shown that as a result of interaction with circularly polarized light the absorbing particles as a whole acquire an orientation induced by collisions with the buffer gas. At high pressures and large rotational quantum numbers the collisional orientation dominates over the known optical orientation.

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Ideas that collisions in a gas system eliminate nonequilibrium, destroy order and create disorder have a wide currency. These ideas, however, do not have the nature of a strict law and, therefore, situations can exist in which nonequilibrium may arise from collisions as a result of constant external influence. It was shown in a recent paper⁽¹⁾ that a gas, which is capable of resonance absorption of radiation and initially has a Maxwell velocity distribution, acquires a directional motion (light-induced diffusion) due to collisions with the buffer gas. This effect is based on two factors: a velocity-selective radiation due to the Doppler effect and the distinction between the velocities of translational relaxation in the ground and excited states.

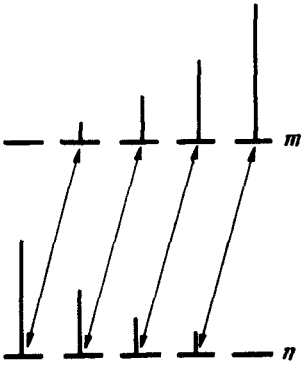


FIG. 1. Transitions caused by radiation of circular polarization and distribution of populations in the magnetic sublevels (vertical lines).

It appears that rotational motion should have a similar effect. The analog of the Doppler effect in this case is the selective interaction of the circularly polarized light with the atoms (molecules) whose angular momentum has different directions. In quantum terms, this means that the population distributions in the magnetic sublevels M in the excited (m) and ground (n) states are asymmetric (Fig. 1). In one of the states the particles are oriented principally in the direction of the wave vector \mathbf{k} and in the other state they are oriented in the opposite direction to \mathbf{k} . Let us assume that the relaxation rate of orientation of the excited state is higher than that of the ground state. Thus, in the steady state the particle as a whole is oriented in accordance with the orientation of the ground state.

Let us perform appropriate calculations. We shall use a simple model of strong disorienting collisions. In case of a uniform broadening, the interaction of circular polarization with radiation is described by the following equations for the density matrix in the M representation

$$\begin{aligned}
 (2\gamma_m + \nu_m) \rho_{mm}(M) &= \frac{\nu_m}{2J_m + 1} \sum_{M'} \rho_{mm}(M') - 2 \operatorname{Re} [iG^* a(M) \rho_{mn}(M)]; \\
 \nu_n \rho_{nn}(M) &= \frac{\nu_n}{2J_n + 1} \sum_{M'} \rho_{nn}(M') + 2 \operatorname{Re} [iG^* a(M+1) \rho_{mn}(M+1)] \\
 &\quad + 2\gamma_m \sum_{M'} \langle J_n M+1 \sigma | J_m M' \rangle \rho_{mm}(M');
 \end{aligned} \tag{1}$$

$$(\Gamma - i\Omega) \rho_{mn}(M) = iG a(M) [\rho_{nn}(M-1) - \rho_{mm}(M)];$$

$$a(M) = (-1)^{J_n - M + 1} \langle J_m M J_n - M + 1 | 11 \rangle; \quad \Omega = \omega - \omega_{mn};$$

$$G = E d_{mn} / 2\hbar.$$

Here ν_m and ν_n are the frequencies of the disorienting collisions, E is the amplitude of the electric field of the wave, d_{mn} is the matrix element of the dipole moment, $2\gamma_m$ is

the constant for radiative decay of the m state to the n state, and $\langle \dots | \dots \rangle$ is the coefficient of vector addition.

Let us confine ourselves to the solution of Eqs. (1) by the iteration method and calculate the orientation of the particle as a whole by using the following expression for the polarization moments of the levels:

$$\rho_{jj}(\kappa, 0) = \sum_M (-1)^{J_j - M} \langle J_j, M, J_j, -M | \kappa, 0 \rangle \rho_{jj}(M). \quad (2)$$

The orientation of the state is determined by the quantity $\rho_{jj}(1, 0)$ and the orientation of the particles as a whole is $N(1, 0) = \rho_{mm}(1, 0) + \rho_{nn}(1, 0)$. In a first approximation of the field intensity we have:

$$\begin{aligned} J_m = J_n = J: \quad N(1, 0) &= |G|^2 \frac{N}{2J+1} \frac{1}{2\gamma_m + \nu_m} \frac{\Gamma}{\Gamma^2 + \Omega^2} \sqrt{\frac{3}{J(J+1)}} \\ &\times \left[1 - \frac{\nu_m}{\nu_n} - \frac{2\gamma_m}{\nu_n} \frac{1}{J(J+1)} \right], \\ J_m = J-1, J_n = J: \quad N(1, 0) &= |G|^2 \frac{N}{2J+1} \frac{1}{2\gamma_m + \nu_m} \frac{\Gamma}{\Gamma^2 + \Omega^2} \\ &\times \sqrt{3} \frac{J+1}{J} \left[\sqrt{\frac{(J-1)(2J+1)}{(J+1)(2J-1)}} - \frac{\nu_m}{\nu_n} - \frac{2\gamma_m}{\nu_n} \frac{1}{J} \right]. \end{aligned} \quad (3)$$

Here N is the density of the absorbing particles. The terms in the square brackets in Eqs. (3) and (4), which contain the factor $2\gamma_m/\nu_m$, are responsible for the usual optical orientation.¹²⁾ The remaining terms describe the collisional orientation. It is easy to see that the collisional orientation becomes dominant over the optical orientation with increasing pressure and rotational quantum number. Specifically, in the classical limit ($J \rightarrow \infty$) there is a collisional orientation but no optical orientation.

There are molecules (for example, benzene), which do not have a dipole moment in the electronic ground state but acquire a dipole moment in the excited state. If they are used as absorbing particles and the gas of the dipole molecules is used as the buffer, then the orientation will relax the absorbing particles in the excited state much faster than those in the ground state (because of the dipole-dipole interaction), i.e., we get $\nu_M \gg \nu_n$ and the collisional orientation is very strong.

Finally, we note that collisions can induce not only orientation but also polarization moments of a higher rank. In particular, a collisional alignment may occur in absorbing particles as a whole as a result of interaction of the linear polarization with radiation.

¹F. Kh. Gel'mukhanov and A. M. Shalagin, *Pis'ma Zh. Eksp. Teor. Fiz.* **29**, 773 (1979) [*JETP Lett.* **29**, 711 (1979)].

²A. Kastler, *J. Phys. Radium* **11**, 255 (1950).